Review of the manuscript #acp-2013-543

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"A sea spray aerosol flux parameterization encapsulating wave state"

Atmospheric Chemistry and Physics (ACP)

Summary:

The paper describes the use of two sets of in situ data for sea spray aerosol concentrations to obtain a sea spray source function (SSSF) in terms of Reynolds number R_H which involves friction velocity, significant wave height, and kinematic viscosity of seawater. Formulation of SSSF as a function of R_H is expected to account for more forcing factors than just the wind speed; through R_H, effects of wave field history, seawater temperature, and salinity (both used to obtain the seawater viscosity) could be included implicitly. The data and the approach used to derive the new parameterization are described. The performance of the new SSSF is evaluated with an independent in situ data set. Results obtained with the proposed SSSF are compared to existing SSSFs and used to estimate global SSA flux. How the use of R_H helps to account for sea state and environmental variables is discussed.

Significance:

In the last decade or so, the air-sea interaction (ASI) community has been on a quest to find forcing parameter(s) that is (are) more suitable than wind speed (e.g., U_{10}) and friction velocity to parameterize ASI processes. Recognizing that wind speed alone cannot explain, and thus parameterize, the flux to the atmosphere of sea spray aerosols formed by breaking waves, we all try to account for one or more additional variables, either explicitly or implicitly (with nondimensional parameters such R_H). Proposing a SSSF that incorporates the effect of wind, wave field, and the environment is necessary and useful not only for the ASI community but also for atmospheric chemistry, climate, and air quality studies.

Evaluation:

The work described in the manuscript is undoubtedly significant. The authors have the expertise, data, and motivation to derive and propose a new SSSF. The manuscript is generally well written. There are, however, aspects/points of the manuscript that need improvement; also, numerous inconsistencies in notations and terminology should be sorted out. Therefore, I recommend that this manuscript be accepted for publishing in ACP after major revision.

Major comments:

1. According to the title of the manuscript, "encapsulating wave sate" seems to be the main improvement in the new SSSF. However, there is more discussion on the seawater temperature effect than on the wave field throughout the manuscript. The only figure that gives some idea how the use of Reynolds number improves the new SSSF is in the supplement, not in the main text. This needs to be rectified. In my opinion, all figures in the supplement are good to go in the main text.

2. The discussion on the wave state seems to rely mostly on the increase/decrease of the wind speed as a proxy for undeveloped and developed wave state. Then an extended presentation or discussion of the connection between increased/decreased wind and the sea state is needed.

3. The stated goal of the study presented in this manuscript is to "derive a new sea spray source function" (p. 4, lines 1-5). However, the current organization of the manuscript places the description of the approach in section 3 and extremely brief description of the derivation in the preamble of section 4; the description of the data (section 2) takes precedence. In my opinion, this organization is appropriate to present new data and evaluate their use for estimating sea spray flux. However, if one is to follow the stated goal, the organization has to emphasize the derivation of the new SSSF parameterization. The flow of the text then should be something in the line: section 2 Approach; section 3 Data; section 4 Derivation; section 5 Results. Section "Approach" should start with general expressions needed for the new SSSF and establish what kind of data you need to derive the terms in these expressions. Then section "Data" comes naturally to show what data you have used, in situ and model. Section "Derivation" is important because you have to convince the reader that your derivation is sound before offering estimates of the global production flux.

4. Possible flow of section "Approach" is as follows:
   a. Give general form of the SSSF dF/dlogD as a product of shape (size distribution) function and scaling factor that depends on forcing factors; e.g., equation (3) in de Leeuw et al. (2011) can be used.

   b. Justify the use of lognormal distribution (as opposed to other functional forms) for the size distribution, and inform that several modes can be present (references needed). Then give the specific form of dF/dlogD as in equation (4) in the manuscript.

   c. Introduce the possibility to control the contribution of each lognormal mode to the overall shape of the size-resolved flux with its own scaling factor that can be parameterized in terms of R_H. This will establish that you need to derive $F_i(R_H)$.

   d. Show how flux F for each mode can be derived from equation like equation (3) in the manuscript (lines 1-7 in your section 3.2)

   e. Show how R_H can introduce several forcing factors (your section 3.1 up to line 11 on p. 8.

5. The use of Reynolds number R_H rather than the breaking-wave parameter R_B (which uses the peak angular frequency of **wind waves** ω_p) is justified with the availability of data for significant wave height H_s from the ECMWF wave model. The distinction is important because to obtain ω_p for R_B, one should determine the wind sea part of the wave spectrum excluding swell components (Zhao et al., 2003, p. 481). Likewise, to control the possible contribution of swell components to R_H, one needs to have wave spectra so that the wind sea part is separated and the H_s corresponding to this wind sea part is determined. However, often H_s values, especially those from altimeter or wave model, are determined for the entire wave spectrum, and these H_s values most likely contain some swell components. This point should be clarified and its influence on the performance of the new SSSF discussed. Reference: Zhao et al., 2003, Tellus, 55B, 478–487.

Specific comments

p. 2, line 16: "inherently includes a sea surface temperature dependence"—what about salinity?

p.4, line 5: "sea surface water temperature"—it should be either "sea surface temperature" or "seawater temperature" (i.e., bulk seawater, below the cool skin) and use it consistently; introduce

the acronym SST on first encounter. Figure S2 shows that you use SST for Figure 7. What temperature do you use for the viscosity for the figures with in situ data? Bulk (measured during the experiments) or SST (from ECMWF)? If both bulk seawater temperature and SST are used, you should mention how much difference between them you expect and how this could affect the uncertainty of the new SSSF.

p. 4, Section 2: As noted in the major comments, a section describing the data comes more logically after you have described your approach and established what kind of data this approach would need to derive terms used in the expressions. Consider exchanging the places of sections 2 and 3.

p. 4, lines 9-10: two in situ data (Mace Head and SEASAW)—what are the seawater temperatures and salinity during the measurements? Salinity at Mace Head and in open ocean could be different because of the aging of the particle arriving at Mace Head, while SEASAW measures at the ambient salinity. What salinity then should be used to determining the seawater viscosity for each of these two data sets? How does the salinity for the in situ data compare to the assumed salinity of 35 psu for the global calculations (p. 14, lines 4-5)?

p. 4, title of section 2.1 and p. 6 title of section 2.2: Because the data you use aim to combine sea spray aerosol in two size regions (small and large or submicron and supermicron), it is more important to convey with the title the sizes of the particles that each dataset provides, not the place of observation. Thus, these two titles could be changes to "Small sea spray particles" and "Large sea spray particles" or the likes. This will also avoid using SEASAW acronym in a title.

p.6, title of section 2.3: Consider changing it to "Wave field" or something in this sense. The type of data that you need for you approach is important. Where these data comes from (i.e., ECMWF) can be stated in the text. In this way you also avoid using undefined acronym in a title.

p. 7, section 3: As noted in the major comments, the description of your approach should come before the description of the data.

p. 8, lines 12-20 belong to a section that describes the specifics of your derivation (see major comments). Once you have established in your "Approach" section that R_H is to be used, and identified in your "Data" section what data is available for this purpose, in a "Derivation" section you show how exactly you computed your R_H.

p. 9, line 8 onward to p.11, line 22: This text is more appropriate in "Derivation" section (see major comments). Your section "Results" should start with line 23 on p. 11.

p. 9, line 27: "contributes only 2-4% to the total flux." How was this determined? From your data or from literature review?

p. 10, section 3.3—Again, this subsection is more appropriate in a section describing your derivation.

p. 10, line 14: How ΔH_s and ΔU_{10} were determined? You did it from the data you used or ECMWF gives them?

p. 10, line 17: Here for the first time the symbol dF/dlogD appears for the SSSF which is the main subject of your study. You should start with it in a section "Approach" right after the Introduction (see major comments).

p. 10, lines 22-24: How did you connect the two size ranges when they differed? Averaging? Interpolating?

p. 10, line 15: "five lognormal size distributions" which you suggest are associated with different physical mechanisms (p. 11, lines 12-13). The reference to Monahan et al. (1986) here is not enough to support your suggestion as Monahan et al. recognize two processes, bubble bursting and spume droplets torn from the wave crests. You do not consider spume production here. So what are the other four processes of production? Perhaps it is not different processes, but different contributions of the same process to different sizes?

p. 12, lines 6-7: "the most common one in the real ambient environment"—needs a reference that 8 m/s is the most common one.

p. 13, Figure 6: Suggest exchanging the places of figures 5 and 6. Currently, your figures 4 and 6 compare the newly-derived SSSF with your own data. It is thus more logical to show them as figures 4 and 5. Then you expand the evaluation of the new SSSF by comparing it to external SSSFs in Figure 6.

p. 13, section 4.2: Because your new SSSF is based on a limited data set and needs validation before it can be used with confidence on a global scale, you need to start this section with a "disclaimer" in this sense.

p. 13, lines 26-27: "simple modeling tool"—is this tool developed by the authors? Or is it from ECMWF?

p. 14, line 1: "Supplement (Fig. S2)"—again, I suggest all supplement figures to come in the main text (see major comment 1).

p. 14, lines 9-10: "constant viscosity (not show)"—pity. A difference map showing reduced and enhanced production with say blue and red colors, respectively, would have been a good case for showing the seawater temperature effect and thus boost the potential this parameterization would have when well validated with more data.

p. 15, lines 5-10: All these are speculations, a possible interpretation. The data do not show unambiguously that it is the wave state that changes the spread of the data in the panels on the right and left. So you have to state this.

p. 15, line 8: "Aitken"—this is the first time that Aitken mode appears as a term. Would be helpful to mention Aitken mode when the corresponding particle sizes are introduced for the first time.

p. 16, line 21: change to "kinematic viscosity of seawater"

p. 16, line 23-24: needs a reference for the statement here.

p. 17, lines 8-18: Some advantages, yes, but still need to mention that the data set used is limited and more data for validation of OSSA-SSSF are necessary.

p. 17: Should include a paragraph discussing similarities and contrasts with the only other SSSF in terms of R_H, that of Norris et al. (2013, GRL). I understand that this paper appeared after you submitted the manuscript; still the comparison would be useful.

p. 26, Figure 1: The title on the abscissa in panel (d) should be "Wind speed, U_{10}" not "ECMWF." That U_{10} is from ECMWF should be said in the captions.

p. 30, Figure 5: Why the SSSF of Norris et al. (2012) is higher than OSSA-SSSF? You use the same SEASAW data for the large sea spray aerosol.

Editorial

p.2, line 21: Here "sea spray aerosol," on p. 3, line 21 you use sea-spray aerosol." Suggest using "sea spray aerosol" (no hyphen) consistently throughout the text.

p. 3, line 2: Monahan et al., 1986 is not in the reference list

p. 3, line 11—you use "whitecap fraction;" in line 24, you use "whitecap coverage." Suggest using "whitecap fraction" consistently throughout the text.

p. 3, line 14: Here "white cap," also "white-capping" (p.11, line 20). Suggest using "whitecap" and "whitecapping" consistently throughout the text.

p. 4, line 14: change to "in November"

p. 5, lines 5-6: You use acronyms "HR-ToF-AMS" and "AMS" for the High Resolution Time of Flight Aerosol Mass Spectrometer. Suggest introduce here only "AMS" and use one acronym throughout the text.

p. 5, line 14: remove dash after "elevated"

p. 6, line 13: New paragraph for "Flux estimates…"

p. 6, line 15: Here and figures 3a and 4, you used "dry particle diameter (D_{dry})." You also use D (p. 9, line 7 and figure 2), and D_p (p. 13, line 1 and figure 5). Make a choice and use the chosen symbol consistently in the text and figures.

p. 6, line 15: Here you introduce a symbol in parentheses "(D_{dry})". On p. 7, line 23, you introduce a symbol with commas ",Re_H,". Choose one way of introducing new symbols and use it consistently throughout the text.

p. 6, lines 17-18: Remove sentence "More details…(2012)." You said this already in lines 3-4 above.

p. 6, line 18: Comma after "SSSF"—sentence beginning with an introductory clause.

p. 6, line 27: Here and on p. 14 (lines 2 and 3) and in the caption of figure 1—revise to avoid "hly"; should be unit for hour "hr"

p. 7, line 1: "should be "basis at a 0.5x0.5"

p. 7, lines 4-5: Do you need to mention JASON-1, JASONN-2, and ENVISAT? Knowing the names of the missions whose data WAM assimilates is not adding anything to clarify your presentation. You can write just "…from satellite altimetry data (Abdalla et al., 2010)" and rely on the reference to provide details for the interested readers.

p. 7, lines 8-10: You use here "break" for everything--wave breaking, air breaking, and bubble breaking. Better use "waves break," "bubbles burst," and "air ruptures"

p. 7, line 12: Suggest changing "the fraction of whitecaps covering the ocean surface" to "the fraction of the ocean surface covered by whitecaps"

p. 9, line 13: typo "mass flux"

p. 9, line 14: Suggest change to "was representative of open ocean conditions"

p. 10, lines 6-7: Here "number size distribution (N)"—is this not the same as number concentration $N(D)$ on p. 9, line 4? If different, make the difference clear; if not, better choose one term and symbol and use them consistently. Same for H (p. 10, line 7) and H_{MBL} (p. 9, line 5).

p. 10, line 17: Here and many other places in the text and in Table 1 and in the captions of the figures you give order of magnitude in the form $1e^5$. All figures give order of magnitude with 10^5. Use the 10-base form consistently in the text, table, and figures.

p. 11, line 17: "were not unexpected"–why don't you say "were expected"?

p. 11, line 21: Here acronym SST appears. See specific comment on the surface versus bulk temperatures.

p. 11, line 24: Here you say "d$F(D)$/dlog(D) vs D" but Figure 4 shows dF/dlog$D$ vs Ddry. See previous comments on using symbols consistently. Also, "d$F(D)$/dlog(D) " should be "dF/dlog$D$" for consistency with previous use and symbols in the figures.

p. 12, line 4: Introduce acronym OSSA-SSSF here, on first encounter. Remove the sentence "Below..." on p. 11, lines 7-8.

p. 12, lines 23-24: Suggest change "periods which are more manifested in the winter season" to read "periods usually occulting in winter."

p. 13, line 20: Remove "the particle vacuum aerodynamic diameter," you already defined D_{va} (p.13, line 13). On the same line, suggested removing D_m; yet another diameter symbol, which is never used later.

p. 14, line 4: Change "sea surface temperature (SST)" to "SST." This acronym is already introduced by this point.

p. 14, line 10: For consistency, use "OSSA-SSSF" instead of "OSSA source function"

p. 14, line 28: This sentence should start a new paragraph.

p. 15, lines 5 and 10: Use either "OSSA-SSSF" or "OSSA SSSF." Consistency!

Acronyms: All these must be spelled in full and introduced as acronyms on first encounter: SEASAW (p. 2 and p. 4; acronyms must be spelled and introduced in both the abstract and the main text), ECMWF, CLASP, ERA, WAM, NOAA, EMEP, IFS, PM.