

*We thank the reviewer for thorough consideration of the MS and the constructive comments, responses are presented below.*

## **Referee 2**

### **Review of A sea spray aerosol flux parameterization encapsulating wave state by Ovadnevaite et al.**

This paper describes a new sea spray source function (SSSF) derived using two in-situ sea spray measurements: at the Mace Head coastal station and an open-ocean eddy correlation flux measurements from the Eastern Atlantic (SEASAW cruise). The new SSSF function is spanning the dry diameter range from 15nm to 6  $\mu\text{m}$  and is expressed in terms of the Reynolds number which involves significant wave height, drag coefficient, and kinematic viscosity of seawater. The use of a Reynolds number, in theory, allows a self-consistent treatment of the different environmental processes controlling sea-spray emissions. The sea spray production is parameterized in terms of 5 log-normal modes. The new SSSF is then used for an estimate of the global production flux. The effect of sea surface temperature (SST) on sea-spray particle flux is discussed. The topic is suitable for ACP, and of interest to the atmospheric chemistry/physics and climate communities.

The paper is well written with clear conclusions. However, due to some concerns with regards to the methodology and lack of details on the evaluation, I recommend major revisions before the manuscript is accepted for publication. The discussion should also be more balanced and critical in terms of the reliability and significance of the parameterization.

### **Major comments**

1) It is stated that  $u^* = C_d^{1/2} U_{10}$ . Please see Foreman and Emeis (2010) for definition of the drag coefficient in the marine atmospheric boundary layer. The definition given in text is strictly valid for neutral drag coefficient. Moreover, due to strong nonlinearity for higher velocities, a Charnock constant needs to be applied for friction velocities greater than 0.27 m s<sup>-1</sup> and wind speeds greater than 8 m s<sup>-1</sup> (Foreman and Emeis, 2010).

*Response: We thank the reviewer for pointing this reference out. It is indeed an approximation, but  $C_d$  and  $U_{10}$  are provided by ECMWF and are used by WAM.  $C_d$  is calculated based on the actual wave field. [Foreman and Emeis, 2010] provide an alternative formulation of  $u^*$ , which could have been used as an alternative to eq. 2, but it is a fit to data, which was latter updated by Andreas et al [2012]. The drawback of this*

*method is that the fit cannot account for the development of the wave field. However, it is interesting to elaborate further on the different wind-dependency for values above and below 9 m/s (e.g., [Callaghan et al., 2008] and also [Goddijn-Murphy et al., 2011]), which corresponds to the transition to rough flow. This makes the direct use of  $u^*$ , regardless of its exact parameterization, even more valuable, since  $u^*$  implicitly includes such considerations, and doesn't require different parameterization for the sea spray source function.*

*The manuscript has been updated and the arguments on the use of eq. 2 have been included along with the discussion on the parameterizations of  $u^*$  and references to the work of [Andreas et al., 2012] and [Foreman and Emeis, 2010].*

2) It is true, that the advantages of using Reynolds number (in addition to wind speed and wave energy) is that it includes temperature and salinity effects through the water viscosity. However, since constant salinity of 35 psu is used in this work, please clearly state that no salinity effects are included in the current approach.

*Response: We did not attempt deriving salinity dependency from field data as it is implicit in the Reynolds number. The effect has not been validated, though, and the statement on validation has been included into revised text:*

*“Secondly, the Reynolds number encapsulated the SST effects through the viscosity of sea water which depends on its temperature and salinity. We have not validated the dependence of the SSSF on SST or salinity.” (p16, l6-7).*

3) Please improve Fig.1a (which is already included in Ovadnevaite et al. (2012)) by overlaying 3-hr back trajectories. This will also strengthen the application of the “filling time” concept under variable wind speed conditions.

*Response: done*

4) The OSSA-SSSF evaluation by comparing the resulting sea spray mass with independent AMS measurements shows excellent agreement for wind speeds  $<15 \text{ ms}^{-1}$  (that cover the majority of the conditions over the ocean). However, I am not sure how even small (say 1-2 ng) difference in mass can be translated into a number. Since particle number distribution within a size range  $D_p = 0.03\text{--}0.58 \text{ }\mu\text{m}$  is known, I think it would be more helpful for the comparison if AMS derived mass is converted to number and plotted on the y-axis on Fig. 6.

*Response: This is a good point, but unfortunately, AMS cannot provide the sea salt mass distribution due to lower evaporation efficiency of the sea salt as compared to the typical nonrefractory material, which affects the particle time of flight and thus particles sizing. Moreover, the SMPS size distribution cannot be used in this case as this would mean validating the measurement with itself.*

5) Looking at Fig. 7 and the supplementary Fig. S2 it is not obvious to me that there is such a strong temperature gradient, “warm waters on both sides of the equator,” to explain the enhanced particle production. Although particle number flux changes more than a factor of 4, variability in temperature can only account for less than 10% changes in seawater kinematic viscosity. It looks to me that the wind speed is also higher along the equator (particularly over the Pacific and Indian Oceans), yet particle number flux is very low. I believe Fig. 7 warrants more detailed discussion.

*Response: We only claim that temperature modulation comes on top of the flux- wind speed dependency, e.g. an increase in the flux due to higher temperatures at the equator would reduce the difference between the fluxes at higher latitudes (stronger winds) and equator (weaker winds). We also argue that this modulation is considerable as the temperature change of  $\sim 20$  °C (reasonable between the equator and higher latitudes) would result in changes in the viscosity of  $\sim 80\%$  [Sharqawy et al., 2010].*

*It seems that the reviewer misinterpreted the colour scale of Fig.7 and the ones in the former supplementary, as dark blue or green areas correspond to the larger fluxes, which are in line with the higher wind areas and, in fact, wind speed is not higher along the equator.*

6) The discussion regarding the effect of temperature on sea-spray number flux needs to be revised. Comparison of sea-spray fluxes of instantaneous global data with Jaeglé et al. (2011) is somewhat misleading. Jaegle et al. derived their Eq (4) using AOD that is a poor proxy for the sea spray source function. 1) Atmospheric turbidity over the remote ocean is controlled by sea-spray particle sources as well as sinks, different meteorological parameters (e.g., relative humidity), etc., and 2) According to the size distribution given in the paper mass scattering efficiencies will be controlled by a small number of particles with  $D_{p,dry} > 0.3 \mu\text{m}$ . Comparison of sea-spray number particle fluxes with AOD may not lead to a straightforward conclusions.

*Response: We agree that the comparison with Jaeglé is not ideal; however, there is no other study, which shows temperature dependencies across the globe. Moreover, we present it as an intercomparison and not as a validation. Nevertheless, the following text has been included to highlight the drawbacks raised by reviewer:*

*‘‘ At this stage, it is difficult to say which dependency is more appropriate as the OSSA-SST dependency derives from first principles but is not (yet) compared to observational data representing different SST, while Jaeglé’s comes from the adjustment to AOD measurements; however, AOD measurements are not a good proxy for the direct particle flux since AOD is determined by both production and subsequent processes in the atmosphere, i.e., transport, removal and transformation due to chemical and physical processes which affect particle concentrations, size distributions and optical properties.’’*

*P15, l23*

7) Three lab measurements referenced in the paper (Mårtensson et al., 2003; Sellegri et al., 2006; and Zábori et al., 2012) do not show “contradictory” results. With increasing temperature all lab measurements show decrease in small particle number concentration and an increase for larger particles (with different thresholds). Therefore, lab measurements show decrease in total number of particles with increasing temperature, the result that seems to contradict the outcome of the current study. Please explain.

*Response: We stand by the point that these studies contradict each other, especially in terms of the climate effects. It is not as simple as the threshold issue: different threshold means that number concentration of 100 nm particles would increase or decrease with the temperature depending on the selection of the parameterization scheme. However, in our study the temperature dependency comes from the first principles of fluid mechanics and agrees with the current ambient observations, although, we admit that we have not validated our temperature dependency yet. Since laboratory experiments rely on different approaches with a bubble generation mechanism that may not represent real ocean conditions, this may cause the discrepancies between our function and the relationships found in laboratory studies. Therefore, we prefer to use the temperature dependency coming from the viscosity until a more robust temperature dependency based on (or consistent with) ambient observations becomes available.*

8) Explanation for the temperature dependency for SSSF as “bubbles in warmer waters will rise more quickly to the surface than in colder waters which would increase the number of smaller bubbles reaching the surface, and thus increase the production of SSA particles” is oversimplification of large number of complex processes involved in the sea-spray production. For completeness, please include some back-of-the-envelope calculation that shows how bubble spectra change in the ocean with changes in temperature and how such a change will influence sea-spray production flux.

*Response: These calculations fall out of the scope of this paper, studies providing such interpretations are cited in the manuscript. We can only provide simple calculations based on the study by [Lewis and Schwartz, 2004], which shows that bubbles of 0.04 mm reach terminal rise velocities of 0.28 cm/s and 0.53 cm/s at 0 and 20 degrees of SST, respectively. This example has been included in the revised manuscript.*

#### **Specific comments**

Pg. 23144 Ln 9. Please define CLASP.

*Response: done*

Pg. 23148, Ln. 8. “SMSP” should be “SMPS”

*Response: changed*

Pg. 23149, Ln 21. Please specify what is meant for “given wave height and sea surface temperature (SST) conditions.”

*Response: specified*

Pg. 23151. Please explain how can the “largest discrepancy between the SSA mass fluxes obtained with the OSSA-SSSF and measured with the AMS” be caused by supermicron aerosol distributions. It is stated in the manuscript that AMS measurements ranged  $D_p = 0.03\text{--}0.58 \mu\text{m}$ .

*Response: It was caused by the uncertainty in the derivation of the 5<sup>th</sup> mode, as we call it super-micron; however, part of this mode extends down to 150 nm. Any error in derivation of that mode would result in uncertainties of submicron mass.*

Pg. 23152, Ln. 11. Please explain what is meant "In addition, the wave state contributes to  $R_{eHW}$  modulating the impact of wind speed alone and thus bringing an extra dimension to the interpretation."

*Response: removed*

Pg. 23154, Ln. 2. "as regards" should be "in regards".

*Response: changed*

### **References:**

Foreman, R., and Emeis, S.: Revisiting the Definition of the Drag Coefficient in the Marine Atmospheric Boundary Layer, *J. Physical Oceanography*, 40, DOI: 10.1175/2010JPO4420.1, 2010.

### *References:*

*Andreas, E. L., L. Mahrt, and D. Vickers (2012), A New Drag Relation for Aerodynamically Rough Flow over the Ocean, J Atmos Sci, 69(8), 2520-2537, doi:10.1175/JAS-D-11-0312.1.*

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*Lewis, E. R., and S. E. Schwartz (2004), Sea salt aerosol production: mechanisms, methods, measurements and models, 413 pp., AGU, Washington, D. C.*

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