Interactive comment on "A sea spray aerosol flux parameterization encapsulating wave state" *by* J. Ovadnevaite et al.

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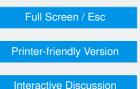
At the section for Atmospheric Science, ITM, Stockholm University we hold bi-weekly paper seminars of interest to the group. The following comments represent an overview of the comments made by the group.

In what is a generally well written and well presented manuscript the authors present a new sea spray flux parameterisation as a function of the wave Reynolds number rather than the often used wind speed measured at 10m height. The wave Reynolds number as proposed by Zhao and Toba (2001) incorporates the friction velocity, the significant wave height, and a viscosity term which can be either that of air or seawater. To date the kinematic viscosity of seawater has been deemed more appropriate given that it is

conceptually more robust (e.g. Woolf, 2005; Goddijn-Murphy, 2011) and the authors of this manuscript opt to use this version.

It appears likely that use of the wave Reynolds number may account for more variability in primary sea spray fluxes through its inclusion of both wind and wave state (e.g. Norris et al., 2013). However, statements contained within the manuscript that the parameterisation presented here includes the effects of seawater temperature and salinity appear, at the very least, to be premature given the limited data used to generate the parameterisation. In other words, we welcome the formulation because it includes wave height but tone down the discussion on how well it represents temperature effects.

The authors combine data from two discreet observational sets to obtain their fluxes. In the larger size range a previously published, eddy covariance flux dataset derived from the open ocean of the North Atlantic is used (Norris et al., 2012) whilst at smaller sizes fluxes are inferred indirectly using SMPS concentration data collected from one day at the Mace Head station on the west coast of Ireland. The method by which the authors convert this concentration data to flux data is known as the statistical wet deposition method and inherent to this technique are a number of assumptions which deserve greater attention in the manuscript. For instance implicit in the method are assumptions that i) no aerosol remains following the precipitation event which proceeds the measurements; ii) following this first rain event there is no further rain/drizzle; iii) that the depth of the MBL is constant across the footprint of the flux; iv) that there is no transformation by cloud processing or particle-gas interactions and v) that dry deposition is negligible.

Considering the dominance of a mode in their derived flux parameterisation at r80~20nm, the assumption that the SMPS concentration measurements are solely due to primary SSA production comes into question. Considering recent reviews (e.g de Leeuw et al., 2011) there appears to be no appreciable physical mechanism for the generation of so many small particles relative to the number of particles at

r80∼100nm. Indeed such a 20nm sea spray mode has not been observed in any previous in situ or laboratory measurements. Could this mode not simply derive from late nucleation/young Aitken mode following post-rain event gas to particle oxidation and nucleation? Since a consequence of the method used is that it simply transfers any features from the observed aerosol number concentration size distribution to the derived size dependent "emissions" we feel the authors should at least discuss this aspect in the manuscript. It should also be borne in mind that such a small mode would contain very little mass and its origin would not have been easily attributed by the mass-spectrometer.

The method also assumes that dry deposition is negligible which may well be the case. However the authors imply that the process is constant across the size distribution which it is likely not. Thus we suggest the application of a size dependent dry deposition model (e.g. Zhang et al., 2001; Nho-Kim et al., 2004). The use of such a model would take into account the fact that the smallest and largest particles would have a higher dry deposition due to Brownian diffusion and sedimentation respectively, and may be important for the shape of the resulting source function.

In terms of error propagation it appears that no sizing errors for either of the methods (SMPS derived or the flux data from Norris et al., 2012) are included and this should be considered by the authors. Furthermore, the MBL height as measured by ground-based LIDAR (presumably at Mace Head) is assumed to be constant over the entire trajectory. The validity of this needs some discussion as well as whether any associated error in this measurement was propagated.

Assuming that the SMPS data presented extends to sizes larger than those currently presented in the manuscript (the EBAS database shows that SMPS measurements are made up to 500nm at Mace Head), it would be useful to see a comparison of the overlap between the direct eddy covariance derived flux data at larger sizes (those from Norris et al., 2012) and the indirect SMPS derived fluxes as a function of wave Reynolds number. Looking at the data as it is currently presented, it appears that the

best agreement is derived from wave Reynolds number at moderate wind speeds but that there is a factor of 2 difference at higher and lower wind speeds.

Throughout the manuscript, the authors state that the use of the wave Reynolds number means that effects of seawater salinity and temperature are included in their parameterisation. Whilst it is true that this parameter does include kinematic viscosity, the fact that these fluxes were derived from a single event means that variation in temperature and salinity will have been very small. Thus statements that this parameterisation includes the effects of temperature and salinity need to be moderated or the parameterisation needs to be validated over a range of temperature and salinity. As it stands the trend of increasing fluxes with increasing seawater temperatures that this new parameterisation infers flies in the face of the majority of the laboratory studies on the subject. The authors do make vague reference to this in the first paragraph of section 5, however we feel this discussion needs improving. It is true that there are inconsistencies in the laboratory effects of seawater temperature on sea spray production but all the studies the authors cite (Mårtensson et al., 2003; Sellegri et al., 2006; Zábori et al., 2012) show increasing production of small particles with decreasing temperature despite the use of seawater of diverse provenance, manifold artificial seawater recipes, several methods of air entrainment and diverse methods of aerosol enumeration. While we do not suggest that the laboratory studies are correct over the field data, the very different trends are worthy of further discussion in the manuscript. The temperature dependence that the authors include in their parameterisation is only one of many possible processes through which the sea spray production may depend on temperature. This must be considered when comparing the model results with observations of actual production over a wide range of temperatures, where it is likely that several different processes contribute to the temperature dependency.

Regarding the fitting of the observed flux with five lognormal modes. This is introduced in a somewhat arbitrary way with little discussion of the physical processes underlying the various modes. A statistical model is strongest when it has some bases in physical

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processes. The F5 mode of the parameterisation appears to be strongly leveraged by only a single data point. The F2 mode seems to be of little use. We wonder if the authors could comment on this since this is the size region where the optical properties of the flux will be strongest (in the case of the F5 mode specifically).

The errorbars on the direct eddy covariance data (from Norris et al., 2012) appear to be constant in magnitude across all sizes whilst we would expect the counting errors to increase as particle sizes increase due their absolute number decreasing. We appreciate that this data has been published previously but some explanation of this would be useful (or reference to the relevant section of the cited paper).

References

Nho-Kim et al. 2004 Parameterization of size-dependent particle dry deposition velocities for global modeling. Atmospheric environment 38 1933-1942

Zhang et al. 2001 A size-segregated particle dry deposition scheme for an atmospheric aerosol module. Atmospheric environment 35 549-560

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