Response to Referee #2

A.-I. Partanen et al.

We thank the referee for the comments and evaluation of our manuscript. Our point-by-point response is below.

1) (p. 4546): Please note whether any aerosol size dependence of the organic mass fraction of PMOM was included, or if all four size sections had the same fraction.
We have added the following sentence to the manuscript to clarify this:

"The same organic mass fraction was used for emissions in all four size sections (i.e., no size dependency was assumed)."

2) PMOM CCN activity (p. 4546): I have some concerns about this treatment because the high CCN activity during periods of high organic fraction coincided with high average aerosol diameter in the Ovadnevaite et al. (2011) study. Due to a lack of understanding of this topic, I don't recommend changing this formulation but to give more information about the hygroscopicity of PMOM and total sea spray aerosol with organics in the form of kappa values (Petters and Kreidenweis, 2007, ACP) or another metric more familiar to other modelers.

While addressing this referee comment, we discovered a coding error (dissociation constant of sea salt was erroneously set to one instead of two) in the box model activation parameterization that was used to tune the dissociation constant of PMOM. The error was not present in the global simulations. We calculated the kappa values for sea salt (NaCl in the model) and PMOM using the Equation 10 by Petters and Kreidenweis (2007) to be 0.78 and 1.33, respectively. This implies that although PMOM is very CCN active compared to model default organic matter (κ=0.24), its CCN activity is lower than that of sea salt. All soluble particles with D > 30 nm activated at supersaturation of 0.7% when the fraction of PMOM of sea spray was 50%. This result did not change significantly after correcting the bug, but increase of the PMOM fraction increased the critical supersaturation. The parts of the manuscript that were either ambiguous or claiming that PMOM had higher CCN activity than sea salt have now been rewritten.

First, the model description section was extended to include the following:

"With the given densities, molar masses and dissociation constants, the CCN-derived κ values (Petters and Kreidenweis, 2007) for PMOM and sea salt (assumed to be sodium chloride in the model) were 0.78 and 1.33, respectively. The difference means that although PMOM has a very high activation efficiency compared to e.g. other organic matter in the model (κ=0.24), its activation efficiency is lower than that of sea salt in the model. The activation efficiency of sea salt in the model is, on the other hand, higher than in ambient measurements due to the assumption that sea salt consists of pure sodium chloride. In supersaturated conditions, κ of PMOM is also greater than the average marine κ of 0.63 at the boundary layer height modelled by Pringle et al. (2010). This means that the presence of PMOM, on average, probably increases the activation efficiency of marine aerosol when all other components are held constant. However, as PMOM in our model setup replaces part of the sea salt in the source function, it decreases the cloud activation efficiency of sea spray. Using the CCN-derived κ of PMOM gives a growth factor of 2 at a relative humidity

of 90%. This value is larger than any growth factor measured by Ovadnevaite et al. (2011), which means that our model setup is consistent with their measurements, although the cloud activation efficiency of PMOM is lower than that of pure sea salt in the model."

We also revised a paragraph that was discussing the radiative effects of PMOM to the following:

"According to our simulations, PMOM decreased the magnitude of both the direct and indirect radiative effects of sea spray aerosol (Table 6, runs ossa-ref and ossa-salt). As described in Sect. 2.2, PMOM had a very low hygroscopicity in subsaturated conditions, and its cloud activation efficiency was high, but lower than that of sea salt. Although the hygroscopicity of PMOM was lower than that of sea salt, CDNC burden was higher in ossa-ref than in ossa-salt, which had no organic enrichment of sea spray aerosol emissions (Fig. 14b), and the positive indirect effect was 0.07 W m^{-2} lower (Table 6). The most likely reason for this result is that the low hygroscopicity of PMOM decreases the water uptake and thus size of sea spray particles, which in turn decreases the condensation sink of sulphate and enhances nucleation and condensation of sulphate on smaller particles. This was reflected in slightly higher (1–3% on average over Southern Ocean) concentrations of aerosol particles, sulphate, and activated particles with dry diameter smaller than 145 nm. As also the liquid water path over the oceans was slightly higher in ossa-ref than in ossa-salt, other processes in addition to the difference in the sulphate condensation sink probably affect the negative indirect effect of PMOM."

We also revised the part of PMOM's radiative effects on the conclusions section to:

"Averaged over the year, the PMOM had a small cooling indirect effect (global mean of -0.07 W m^{-2}) most likely due to the low water uptake of PMOM in subsaturated conditions that decreased the condensation sink of sulphate and thus increased the number of small particles. On the other hand, the low water uptake of PMOM led to a positive direct effect of PMOM (global mean of 0.03 W m^{-2})."

3) SSA emissions (p. 4554): Please note that the Gantt et al. (2012) global sea salt emissions of 73.6 Tg/yr represented the submicron emissions and that the total sea salt emissions were not listed but were probably consistent with the 4200 Tg/yr reported in Jaegle et al. (2011).

Thanks for pointing this out. We have removed the following sentence from the manuscript:

"On the other hand, the simulations by Gantt et al. (2012) provided a global sea salt emission of 73.6 Tgyr−1, which is clearly lower than values obtained in this study."

4) Comparison to concentration observations (p. 4557): While this section focuses on biases of the simulations, correlations and seasonal comparisons should also be reported for both sea-salt and organic matter. Also, please discuss how the temperature dependence and different wind speed-sea spray relationship in the new sea spray source function affect the predicted correlations and seasonal cycles.

We have added Table S1 in this reply to the supplementary material. It includes normalized mean biases for April-September and October-March seasons, and correlations between the model results and measurements. The correlation coefficients in sea salt mass concentrations are slightly lower in

ossa-ref than in default-salt, although the normalized mean biases are of lower magnitude in ossa-ref compared to default-salt. We have added a part of the correlation coefficients in the manuscript and the following paragraph in the manuscript to discuss the differences compared to the default sea spray source function:

"Overall, the extended OSSA source function decreased the magnitude of normalized mean bias in sea spray aerosol concentrations. There was however a small decrease in the correlation coefficients. See Table S1 for more details on the comparison of results between ossa-ref and default-salt."

5) (p. 4558): Please note that the organic aerosol underprediction at Mace Head may have also been affected by the selection of an adjacent "sea" grid cell.

The modelled concentration of organic matter was lower than the measurements in all grid-cells surrounding the grid-cell where Mace Head is located. It was only slightly higher in the grid-cell where Mace Head is than the grid-cell what was chosen in the manuscript. The normalized mean bias of organic matter concentration in the grid-cell where Mace Head is located is -65% (-82% in the grid-cell chosen in the manuscript.)

6) Conclusions (p. 4566). Please include additional discussion about the low values of global sea spray emissions from wave state-based parameterization relative to other parameterizations. Also, please explain how the optimized emissions in this study can be an order of magnitude lower than optimized emissions from a recently published sea spray comparison study (Grythe et al., 2014 ACP

We already state in the conclusions that our estimates of both sea salt and PMOM are considerably lower than the typical range in previous studies. Since we are uncertain what the reviewer means by additional discussion, we have left this part of the conclusions section as it was.

The difference between the extended OSSA source function and the optimised function in Grythe et al. (2014) can be explained by the data and fitting approach used in the two studies. Firstly, our parameterisation is based on measurements at Mace Head and Eastern North Atlantic Ocean, relating local emissions to local physical conditions, whereas Grythe et al. (2014) related observed concentrations with emissions from a full footprint area. In their approach, a good representation of the deposition velocity is crucial, still the deposition velocity is not so accurately known and could lead to differences in estimated emission strength between our and their approach. Secondly, our data only cover data that are representative of water temperatures around 12-15 °C and low organic matter concentrations. Thirdly, data used in Grythe et al. (2014) have a less clear cut-off, the as data from PM1, PM10, and TSM (total suspended mass) were used from various data providers. OSSA source function is, on the other hand, based on high quality measurements of two plumes with elevated sea salt concentration.

7) Tables: Please add a table summarizing the statistical evaluation of the model compared to in-situ/satellite-derived observations.
We have now included a table (Table S1 in this reply) to the supplementary material to provide this additional information.

8) Figures: Please include a trendline with the correlation and equation to Figures 9 and 12.

We have added the trendlines to the figures, and the correlations (for the whole year) are now listed in Table S1. The equations were omitted because of fairly low correlation values.

9) Figures: Please add a figure (possibly in the supplemental information) giving the size-resolved number flux at a given wind speed and the mass flux as a function of wind speed for the new and existing source functions
This information is available in Ovadnevaite et al. (2014), and we do not consider it worth replicating the figures in this publication. For size resolved number flux in the OSSA source function and other source functions, see Fig. 7 in Ovadnevaite et al. (2014). For number and mass flux with wind speed, see Fig. 6 and Fig. 11 in Ovadnevaite et al. (2014). Mass flux for other source functions is presented in Ovadnevaite et al. (2012).

References

Ovadnevaite, J., D. Ceburnis, M. Canagaratna, H. Berresheim, J. Bialek, G. Martucci, D. R. Worsnop, and C. O'Dowd (2012), On the effect of wind speed on submicron sea salt mass concentrations and source fluxes, *J Geophys Res-Atmos*, *117*(D16201), 11, :doi 10.1029/2011jd0

Table S1. Comparison of model results from the runs *ossa-ref* and *default-salt* with measurements. NMB stands for normalized mean bias (%), O-M denotes October-March, A-S denotes April-September, and r is the correlation coefficient.

	Meas. mean	*ossa-ref*					*default salt*				
		Mean	NMB	NMB O-M	NMB A-S	r	Mean	NMB	NMB O-M	NMB A-S	r
In-situ measurements (μg m^{-3})											
PM$_1$ sea salt concentration in Mace Head	0.19	0.17	-13	-22	7	0.83	0.13	-30	-40	-8	0.86
PM$_1$ organic matter concentration in Mace Head	1.00	0.18	-82	-90	-75	0.17	0.15	-85	-91	-80	0.23
PM$_1$ sea salt concentration in Amsterdam Island	0.28	0.14	-52	-39	-60	0.75	0.11	-60	-44	-69	0.70
PM$_{1-2.5}$ sea salt concentration in Amsterdam Island	0.26	1.01	283	227	351	0.34	2.46	845	791	912	0.36
PM$_{2.5-10}$ sea salt concentration in Amsterdam Island	0.26	2.62	899	698	1172	0.30	12.06	4519	3764	5540	0.35
Total carbonaceous aerosol concentration in Amsterdam Island	0.16	0.06	-68	-77	-58	-0.45	0.04	-76	-86	-66	-0.53
PM$_{2.5}$ sea salt concentration at Point Reyes	2.41	1.69	-29	-26	-31	0.55	3.61	50	74	35	0.55
PM$_{2.5}$ organic matter concentration at Point Reyes	0.80	2.62	212	108	418	0.13	2.53	201	103	395	0.14
Sodium ion concentration in PM$_{2.5}$ in EMEP data	0.36	0.22	-66	-41	-107	0.03	0.58	38	42	32	0.14
Sodium ion concentration in PM$_{10}$ in EMEP data	0.40	0.58	32	46	5	0.55	2.40	84	87	78	0.60
Satellite measurements (AOD at 550 nm from PARASOL)											
AOD over all oceans	0.16	0.10	-31	-25	-32	-0.17	0.16	13	23	11	-0.21
AOD over Southern Ocean	0.11	0.07	-31	-28	-33	0.18	0.17	55	57	57	0.19
AOD around Amsterdam Island	0.10	0.09	-14	-15	-14	0.29	0.19	89	82	95	0.26
AOD west of Mace Head	0.14	0.08	-39	-36	-40	0.32	0.16	15	42	-2	0.15
AOD west of Point Reyes	0.13	0.07	-41	-33	-48	0.13	0.14	8	36	-17	0.16
Sun photometer measurements (AOD at 550 nm from AERONET)											
AOD from coastal stations	0.14	0.12	-17	-4	-26	0.77	0.15	8	28	-8	0.73
AOD from island stations	0.14	0.12	-15	-1	-24	0.65	0.16	22	48	1	0.55