

Final response on the referee comments

A.-I. Partanen et al.

Our replies to the three referees are given below. During the revision process, we discovered a coding error in the box model that was used to tune the cloud activation efficiency of primary marine organic matter. This issue is resolved and discussed in the reply to Referee #2.

We have also made some editorial changes in the revised manuscript, and corrected Figure 5 with respect to organic matter concentration in the run with default sea salt source function.

Reply to Prof. J. R. Pierce

A.-I. Partanen et al.

We thank Prof. Pierce for his evaluation of our manuscript. Our point-by-point responses to his comments are given below.

P4544 L9: Please explain what “significant height” is.

We have added the following explanation in parenthesis after the word significant wave height: “four times the standard deviation of sea surface elevation or roughly the average height of the highest one third of the waves”

P4545 L32: What wavelengths (or what wavelength range) do you use? This is important for knowing if using a single ref index is reasonable.

Model calculates radiation between wavelengths of 0.28 μm and 4 μm . We added this information to the manuscript.

P4546 L15-22: So the PMOM is more hygroscopic than sea salt? (at least this is what it seems like). This is weird enough that you probably should discuss this a bit more (I realize you do say that it is to match observations), but it’s probably worth pointing out just how hygroscopic the PMOM is.

In subsaturated conditions, PMOM is hydrophobic as described by Vaishya et al. (2013). For example a particle consisting of only PMOM would have a Growth Factor (GF) of 1.27 at a relative humidity of 90% (GF=2.3 for NaCl particles at the same RH with this parameterization). We have added the following sentence to discuss the hygroscopicity in subsaturated conditions:

“For example, growth factors at a relative humidity of 90% for pure PMOM and pure sea salt particles were 1.3 and 2.3, respectively.”

In supersaturated conditions, the hygroscopicity of PMOM is actually slightly lower than that of sea salt in the model in contrast what was stated in the manuscript previously. This misconception rose

from a coding error in the box version of the activation parameterization, which was used to tune the dissociation constant of PMOM. Its CCN-activity-derived κ (Eq. 10 by Petters and Kreidenweis, 2007) is 0.78 and lower than that of NaCl (1.33 from the same equation). Although CCN activity of PMOM was set to a lower value in the model simulations than originally intended, it is much more hygroscopic in supersaturated conditions than other organic matter in the model ($\kappa=0.24$). Some of the results needed to, however, be reinterpreted and analyzed in more detail as replacing some of the salt with PMOM actually lowered the CCN activity of sea spray (comparison of ossa-salt and ossa-ref simulations). See reply to Referee #2 for more details.

Equation 5: Is there a physical or observational basis for this dependence of the chlorophyll dropping off with latitude with this dependence? There is no citation given.

Missing observations outside the satellite range presented a challenge, and we did not find a observationally based solution to fill the gaps. Filling missing values in the winter hemisphere with summer-time values resulted in very high chlorophyll-a concentrations that looked totally unrealistic. The other trivial option of setting missing values to zero would have lead to an underestimation of the chlorophyll-a concentrations. Therefore, we consider our approximate formula to be a good compromise between these two extremes. We have added “approximate” to the sentence:

“Outside of this latitude range, the chlorophyll concentration in a given grid cell (C_i) was then set according to the following **approximate** formula.”

Section 2.4.2: Is the model AOD taken as an average over clear-sky conditions only? Since POLDER and AEROSOL will only retrieve AOD values under these conditions, it is important to also sample the model this way (because aerosols may be different between clear sky and cloudy conditions).

This is an important point and was missing from the original manuscript. Model AOD is calculated for each model time-step independent of the cloud conditions. This creates some additional uncertainty as there is certainly some correlation with cloud cover and aerosol concentrations. However, the model does not provide AOD diagnostics for cloud-free conditions (i.e. mapping only time-steps with no cloud cover) nor mapping only when PARASOL or AERONET stations have observations. Improving the AOD diagnostics of the model to calculate AOD only when PARASOL or some AERONET station has observations would be fairly complicated and time-consuming, and therefore outside the scope of this paper. We have added the following paragraph to Section 2.4.2 to discuss this issue:

“AOD observations from both AERONET and PARASOL are retrieved under clear-sky conditions, whereas the modelled AOD is calculated over all time-steps. This difference may cause additional uncertainty in the model-measurements comparison as aerosol concentrations and cloud fields depend partly on each other for example through precipitation and wet deposition. However, large-scale patterns and long-term averages are affected considerably less by this uncertainty than local transient values.”

Section 2.4.2: Why are you correcting the AOD of the measurements for the wavelength that you use in the model (550 nm)? Since in the model you have the aerosol size distribution, you should be able to calculate the AOD at any wavelength you want. Probably no need to change

at this point, but you might want to think of this in the future so that you don't need to use monthly mean angstrom exponents to fudge the observations.

It is true that we could have set the model diagnostics to calculate AOD at the wavelengths used by PARASOL and AERONET, and thus avoided using Ångström exponent. However, we had set up and initiated the model runs before we made the final decisions about which satellite product to use and started collecting the satellite and sun photometer data. Therefore, it was too late to change the model diagnostics at that point, and calculating AOD from the mean size distributions off-line would have caused some error too. We'll keep this issue in mind for our future studies.

P4552 L17-22: Are you multiplying just the submicron part of the emissions by 0.4 and 1.6 or the entire distribution (the first sentence of the paragraph talks about uncertainties in the submicron part).

We assumed the same uncertainty in the whole size range, and used the same multipliers also for the supermicron range. This multiplication was implicitly extended also over the six micrometer size as the source function by Monahan (1986) was matched with the OSSA source function at this size. We have clarified this issue in the manuscript by rewriting the last sentence of that paragraph to:

“Therefore, to test the sensitivity of our results to these uncertainties, we **assumed the same uncertainty in the whole size range** and set up two sensitivity runs (*ossa-lowflux* and *ossa-highflux*) in which the sea spray aerosol flux from the extended OSSA source function was multiplied by 0.4 and 1.6, respectively.”

Figure 9: How different did these comparisons look when using the default param? Can you compare some metrics (e.g. bias and correlation)?

All-year mean normalized mean biases in the default-salt run were 37.5% and 83.8% for PM2.5 and PM10, respectively (-65.6% and 31.9% *ossa-ref*). Respective correlations were 0.14 and 0.60 for default-salt and 0.03 and 0.55 for *ossa-ref*. We have included these numbers in the manuscript. They are also listed in a table (see response to Referee #2) in the supplementary material along with Figure 1. We added the following text to the manuscript:

“All-year normalized mean bias for PM10 in *ossa-ref* was 32%. The correlation of PM2.5 was weak (0.03) but better (0.55) for PM10 in *ossa-ref*.”

Both PM2.5 and PM10 concentrations were overestimated using the model default sea spray source function (Fig S1; normalized mean biases of 38% and 84%, respectively, in default-salt). The correlations of PM2.5 and PM10 between the model and the measurements were slightly better in default-salt than in *ossa-ref* (0.14 and 0.60).”

We have also added the following figure to the supplementary material:

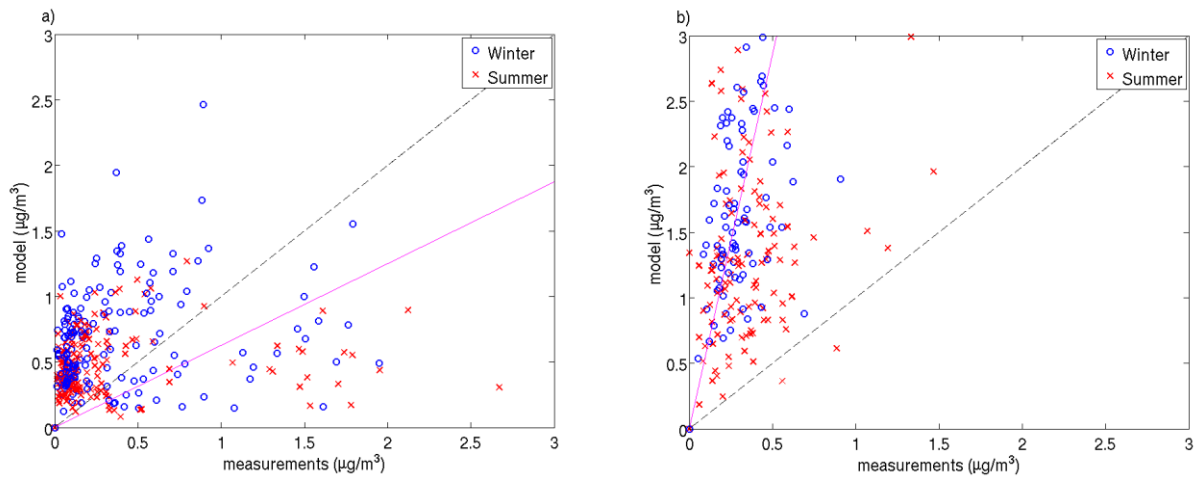


Fig. S1. Comparison of measured (EMEP stations) and modelled (simulation *default-salt*) monthly mean sodium ion concentration in (a) PM_{2.5} and (b) PM₁₀ at various sites for the years 2006–2011. Blue circles indicate boreal winter months (October–March) and red crosses indicate boreal summer months (April–September).

The discussion of Figure 10: In “Jaeglé, L., P.K. Quinn, T. Bates, B. Alexander, and J.-T. Lin (2011), Global distribution of sea salt aerosols: New constraints from in situ and remote sensing observations, *Atmos. Chem. Phys.*, 11, 3137-3157, doi:10.5194/acp-11-3137-2011.” the authors corrected a similar bias in marine AOD in their model against AOD obs where the model had too much AOD in midlatitudes by making a temperature dependence of sea-spray emissions. In your manuscript you do this by switching to a new source parameterization that includes wave height. It might be worth some discussion of how these two techniques might be related. E.g. is there a physical linking between temperature and wave height (at a fixed wind speed)?

The OSSA source function includes an implicit temperature dependence through water viscosity, what results in a quite similar OSSA-SSSF Flux-temperature dependency to Jaeglé et al. (2011), see Fig. 12 in Ovadnevaite et al. (2014) or replicated figure below:

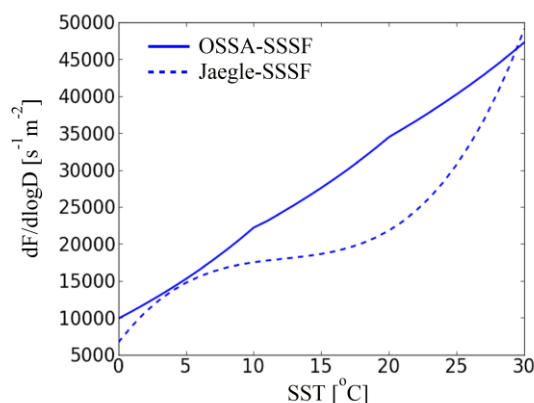


Fig. 12 from Ovadnevaite et al., 2014. Comparison of the effect of sea surface temperature (SST) on the particle production for using the OSSA-SSSF and the formulation by Jaeglé et al. (2011) for $D_p=1 \mu\text{m}$ particles at 9 m s^{-1} wind speed. To eliminate the effect of the wave state, which was incorporated into the OSSA-SSSF, the constant values of $C_d=1.3 \cdot 10^{-3}$ and $H_s=1.5$ were used in the calculation of the production fluxes.

We added the following sentence to the model description to discuss this issue:

“This implicit temperature dependence of the OSSA source function is similar to the results of Jaeglé et al. (2011) (Ovadnevaite et al., 2014).”

Figure 11 and P4562 L17: Can you quantify the correlation coefficient so that you have a metric of how well the model is capturing the variability?

The correlation coefficients for Mace Head and Point Reyes were 0.32 and 0.13, respectively for ossa-ref, and 0.15 and 0.16 for default-salt. We have added the correlation coefficients of ossa-ref in the manuscript and listed them for both runs in Table S1 (see response to Referee #2).

Figure 12: How did the default model do? Can you compare some metrics (e.g. bias and correlation)?

We have added the following paragraph to the manuscript:

“The simulation default-salt had slightly larger normalized mean biases than ossa-ref (-16% vs. 8% for coastal stations and 19% vs. -16% for island stations. All-year correlation was slightly higher in default-salt than in ossa-ref for coastal stations (0.91 vs. 0.83) and slightly lower for island stations (0.67 vs. 0.74). See Table S1 for more details.”

P4564-4565, the discussion of reduced CDNC due to sea salt. Can you add a plot showing (1) the mean aerosol number size distributions over the southern ocean with and without sea-spray and (2) the mean minimum activating diameters over the same region w/ and w/o sea spray. This would allow us to see how the distribution changed due to sea spray and nucleation/growth feedbacks as well as see how much the sea spray suppressed activation of smaller particles.

See the left panel of the figure below for the mean aerosol size distribution over the Southern Ocean with and without sea spray aerosol. The model diagnostics do unfortunately not include minimum activating diameter so we cannot plot that without adding new diagnostics and rerunning the experiments. However, the activated aerosol distribution is shown in the right panel of the figure below. The figure shows that the number of activated particles is lower in the small sizes (below 145 nm in dry diameter) in the simulation ossa-ref than in the control run. The revised manuscript includes the figure and the following text:

“The effect of these two mechanisms on the positive indirect effect is supported by the fact that there were clearly less particles in the model size bins below 145 nm in ossa-ref than in ctrl (Fig 15a). While slightly more large sea spray particles (dry diameter above 145 nm) activated in ossa-ref compared to ctrl, the lower number of activated small particles (below 145 nm) meant that the total CDNC was lower in ossa-ref (Fig. 15b). The total number of soluble particles with dry diameter larger than 30 nm was 7% lower in ossa-ref than in ctrl in the lowest five model levels (up to about 1 km) above the ocean.”

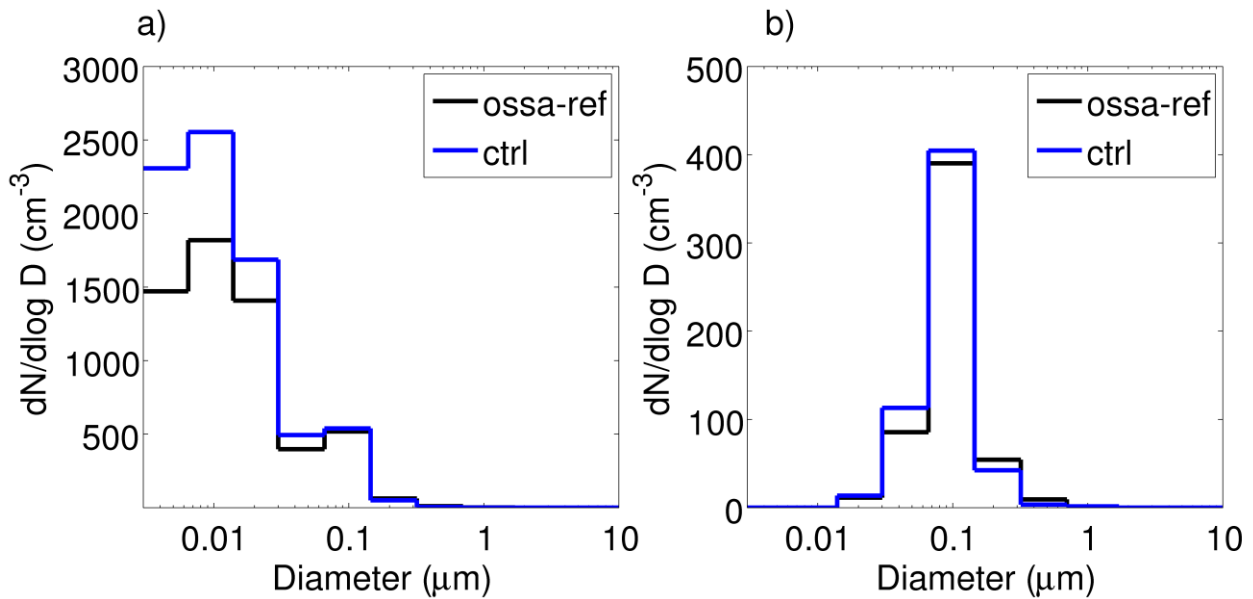


Figure 15. Five-year mean size distribution over the Southern Ocean in the five lowest model levels (about up to 1 km) a) of all aerosol particles, and b) of particles activated to cloud droplets. The diameter refers to dry size in both panels.

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 ($\kappa=0.24$), 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 ($\kappa=0.24$), 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 Tg yr^{-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 10.1029/2011jd0173

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.

			ossa-ref				default salt				
	Meas.			NMB	NMB				NMB	NMB	
	mean	Mean	NMB	O-M	A-S	r	Mean	NMB	O-M	A-S	r
In-situ measurements											
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 (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 (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

Reply to Referee #1

A.-I. Partanen et al.

We thank the referee for the critical and valuable comments on our manuscript. Our point-by-point responses to the referee's comments are given below.

Partanen et al. (2014) evaluate global-scale influences of marine aerosols on optical properties and radiative forcing based on a source function for marine aerosol published recently by Ovadnevaite et al. (2014). This function incorporates a parameterization for internal mixtures of marine-derived primary organic material (annotated "PMOM") based on Rinaldi et al. (2013). Production fluxes of PMOM are forced with global observations of surface ocean Chlorophyll-a. The system was tested within the ECHAM-HAMMOZ aerosol-climate model system. The development and testing of a marine aerosol flux function based on wave parameters represents a step forwards and I commend the authors for their effort. However, the parameterization of PMOM suffers from fundamental deficiencies that render conclusions suspect. First, recent work by Quinn et al. (2014; *Nature Geosci.*) clearly demonstrates that PMOM emissions do not vary as a linear function of Chlorophyll a. Marine aerosol produced artificially from productive and oligotrophic waters (mean chlorophyll a concentrations of 7.1 and 0.03 $\mu\text{g L}^{-1}$, respectively) in the western North Atlantic Ocean exhibited similar size-resolved enrichment factors for organic matter. Bates et al. (2012; *JGR*) also observe no systematic variability in organic abundance as a function of chlorophyll a concentrations in the north western Pacific Ocean. In addition, as reported by Long et al. (2011, *ACP*), organic enrichments in fresh marine aerosol produced from oligotrophic waters in the western North Atlantic at Bermuda (mean chlorophyll a $\sim 0.2 \mu\text{g L}^{-1}$; Keene et al., 2007, *JGR*) were only slightly less to those for marine aerosol produced from productive waters in the eastern North Atlantic (mean chlorophyll a of $\sim 1.4 \mu\text{g L}^{-1}$; Facchini et al., 2008, *GRL*). These results suggest that marine aerosols are enriched in PMOM by roughly similar amounts everywhere, which is inconsistent with parameterizations that scale linearly with chlorophyll a as employed by Partanen et al. It is evident that extrapolation of weak correlations between PMOM production and corresponding chlorophyll a concentrations in productive waters of the northeastern Atlantic underestimate PMOM fluxes under more oligotrophic conditions that are typical of most of the surface ocean globally. Second, in another recent paper that was not available at the time of this manuscript's discussion publication, Long et al. (2014; *GRL*) show evidence that bubble plumes in both productive and oligotrophic seawater are coated with organic matter. Wurl et al. (2009 *J. Mar. Chem.*; 2011 *Biogeochem.*) also report that organic microlayers form in both productive and oligotrophic waters, which is consistent with the presence of surfactant coatings on bubble surfaces in both productive and oligotrophic water as observed by Long et al. (2014) and similar organic enrichments of particles produced when those bubbles burst as measured in parallel by Quinn et al. (2014). Were the authors to consider the above studies, they would likely come to different conclusions with respect to simulated PMOM emissions, concentrations, and impacts. The assumed linear regression between PMOM production and chlorophyll a concentration results in low PMOM emission fluxes from oligotrophic waters, which are clearly inconsistent with a growing body of measurements by other groups. For example, such under-predictions may explain the low simulated PMOM concentrations relative to observations in the low chlorophyll a region surrounding Amsterdam Island. Lastly, Quinn et al. (2014; *Nat. Geosci.*) also report that freshly produced and ambient marine aerosol in

regions of high and low chlorophyll a exhibit no systematic differences in CCN activity, which again is inconsistent with results reported by Partanen et al. Because the fundamental inconsistencies noted above have important implications for the major conclusions of this modeling study, it is essential that they be addressed explicitly before publication.

We agree that chl-a is not an ideal proxy for PMOM emissions. However, this is the best parameterization currently available as Quinn et al. (2014) did not provide any parameterization. Moreover, the lack of correlation or linear relationship between the in-situ chl-a and PMOM may not be contradictory as Rinaldi et al. (2013) was based on the weekly averages as well as long term measurements (several years) and introduced the lag between chl-a concentration and resulting OM enrichment. Meanwhile studies of Quinn et al. (2014) and Long et al. (2014) were based on short term field campaigns and attempted to correlate the instantaneous local chl-a measurements with PMOM emissions, which may well be unrelated. However, to address this important issue, we have added the following paragraph to model description to discuss the uncertainties involved in predicting the organic fraction of sea spray:

“The parameterization for the organic fraction of sea spray is derived from long-term data in the North Atlantic, which show that chlorophyll-a concentration with an 8-day time lag are a useful proxy of organic enrichment in this region. The usefulness of chlorophyll-a proxy for medium time-scales on larger areas was also shown by Gantt et al. (2012). However, the parameterization has not yet been evaluated in other regions and may not capture localized events, such as typically observed during short-term campaigns that do not fulfill the eight-day time lag criterion. Measuring such localized or short-term events, Gantt et al. (2012) as well as Quinn et al. (2014) have recently shown that the correlation between the chlorophyll-a concentration and organic enrichment is not necessarily present. This complex relationship between oceanic biological activity and organic enrichment calls for more long-term data sets from different regions of the world’s oceans to improve the parameterizations.”

Line 26 through 28 on Page 4556: This sentence implies that the measurements may be unreliable because they diverge from simulated values. This sentence should either be removed or revised to eliminate this unsubstantiated suggestion.

We find it hard to see that this sentence could be misinterpreted to imply that measurements may be unreliable simply based on divergence from simulated values. However, to avoid confusion, we have reformulated the sentence to:

“The difference in the spatial patterns of sea spray burdens and emissions imply that it is very difficult to use in situ measurements of sea spray aerosol mass or remote sensing measurements of AOD to directly deduce information about the magnitude of local sea spray emissions.”

Line 21 through 25 on Page 4566: The parameterization of Rinaldi et al. (2013) does not account for mixing of organic material between different layers of the ocean. To my knowledge, there are no regions in the ocean that can be considered organic-poor (e.g., see Hansell, 2013, Ann. Rev. Mar. Sci.), nor would a simple linear parameterization be able to account for the highly non-linear and poorly understood dynamics of marine organic cycling in the surface ocean

We agree that dynamics of marine organic cycling are yet poorly understood and that linear parameterization does not describe the many non-linear processes perfectly. On larger spatial and temporal scales, the Rinaldi et al. (2013) parameterization matches with observations in the North Atlantic and one purpose of this study is to evaluate its performance in other ocean regions.

We have added the following sentence to Section 3.1:

“Note that the formation of PMOM is based on the Rinaldi et al. (2013) parameterization which was derived using data from the North Atlantic but has not been evaluated for other regions by lack of representative experimental data.”