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 that 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 hydrophopic 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. It's 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 indented, it is much more hygroscopic in supersaturated conditions than other organic matter in the model (κ=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 approximative formula to be a good compromise between these two extremes. We have added "approximative" 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 **approximative** 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)."

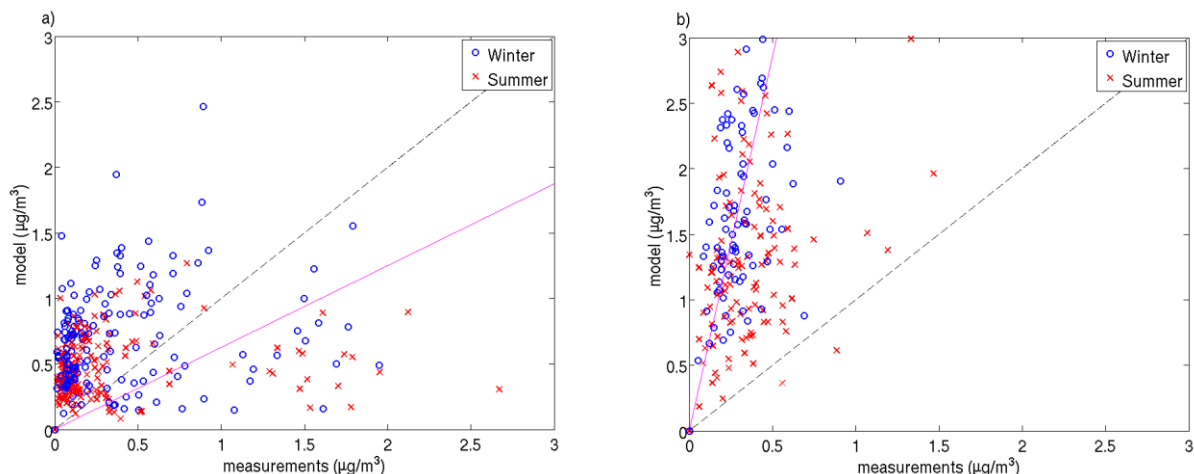We have also added the following figure to the supplementary material:

Fig. S1. Comparison of measured (EMEP stations) and modelled (simulation *default-salt*) monthly mean sodium ion concentration in **(a)** PM2.5 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 in their model against AOD obs where the model had too much AOD in midlatitudes by making a temperature dependence of sea-spay 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 trough 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:
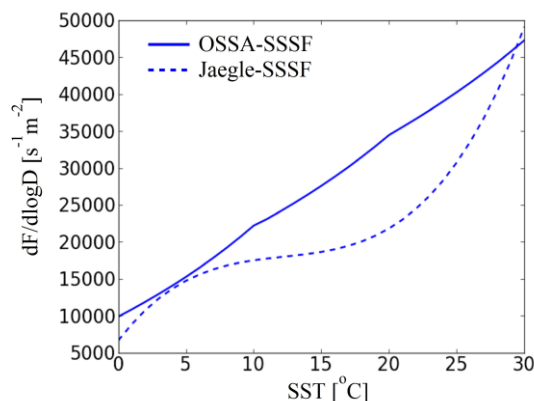

Fig. 12 from Ovadnevaite et al., 2014. Comparison of the effect of sea surface temperature (SST) on the particle production for using the OSSA-SSSF and the formulation by Jaeglé et al. (2011) for Dp=1 μm particles at 9 m s^{-1} wind speed. To eliminate the effect of the wave state, which was incorporated into the OSSA-SSSF, the constant values of C_d=1.3 $*10^{-3}$ 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."
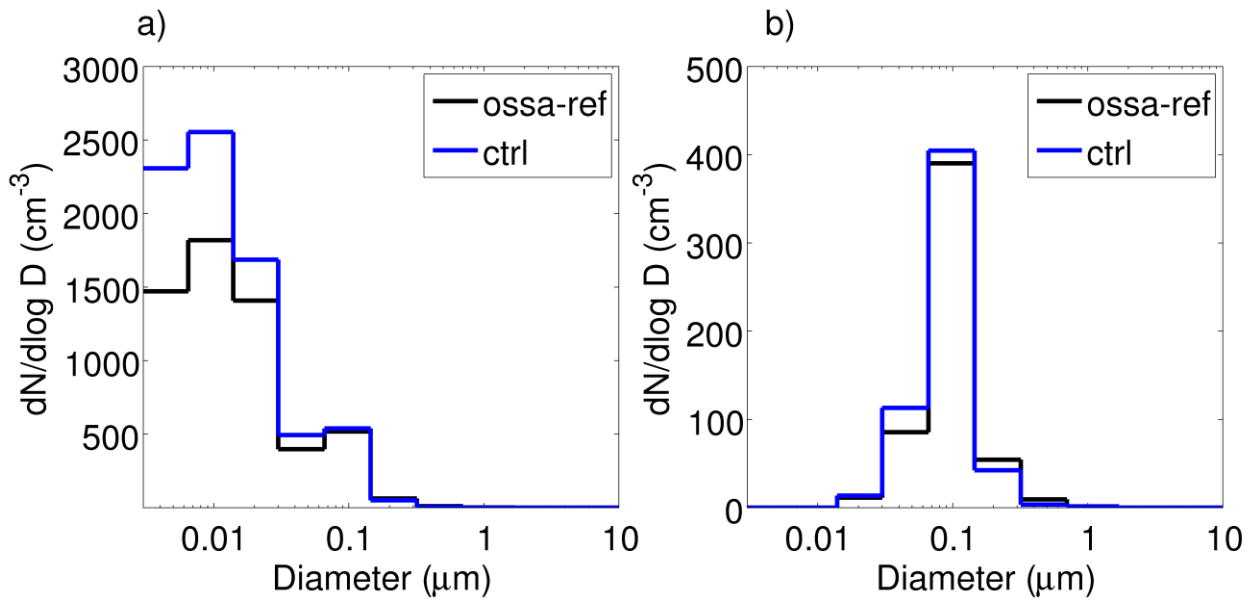
Figure 15. Five-year mean size distribution over the Southern Ocean in the five lowest model levels (about up to 1 km) a) of all aerosol particles, and b) of particles activated to cloud droplets. The diameter refers to dry size in both panels.