Reply to referee #2

We greatly appreciate the time and energy spent by the reviewer. The comments have helped improve the paper.

1. None of the atmospheric aerosol components exist in total isolation, and thus it is very challenging to draw comprehensive conclusions based on a model simulating only sea spray. Both primary organic material co-emitted with sea spray and sulphate from condensation and aqueous phase reactions can affect the climate properties and removal processes of sea spray. The implications of this on the conclusions of this study should be discussed.

We added a paragraph to the conclusion section to discuss the additional aspects of the marine aerosol that could be simulated and possible work to be done in future.

2. For many aerosol properties, the authors compare the model only against a single data set. I understand that more suitable data may not be available; however, the uncertainties arising from this should be discussed.

We added discussions on the limitation of the SP datasets in the first paragraph of section 3.1. Comparisons of the Mulcahy et al. (2008) dataset against other dataset are available in their paper as well as Madry et al. (2011). We do not add further comment on the uncertainty of using the dataset alone.

3. Mårtensson et al. (2003) showed that the shape of the source function depends on the sea water temperature. Since the Clarke source function does not include this dependence (measured at 25 °C) and since Mårtensson and Clarke have previously been shown to agree at 25 °C, why not use the Mårtensson flux as a base of the new integrated flux? Since many of the observations used for comparison were not measured in tropical conditions, the effect of the temperature dependence on the model-measurement comparison should be discussed. Mårtensson et al. (2003) is also an option to represent the small particles instead of Clarke et al. (2006). We did not adopt Mårtensson function because 1) we would like to make a point that using a source function that includes ultra-fine sea salt improves number prediction in climate models and Clarke et al. (2006) is the simplest model that fits the demand. 2) Using the Mårtensson function will introduce another uncertainty through the source function's dependence on SST in addition to wind speed and sampling bias. Note that for particles in the 0.1 to 1 μ m size range the Mårtensson function would greatly lower the number of particles at high latitudes relative to all other functions (Fig. 1). However, looking at data such as mass does not indicate a seasonal error, which one might expect if such a strong temperature dependence occurred.

We accept the reviewer's suggestion to add more discussion on the SST effect on the model-measurement comparison in section 2.1. There is research suggesting that the SST effect could be important in high latitudes and the tropics (Jaeglé et al., 2010). However, we do not find that the observational data base is good enough to tell us if there are temperature driven differences in the number of particles in the atmosphere. We will leave such a comparison as a potentially important future study.

4. There are some technical details that I'm not totally convinced about: a) The Caffrey and CMS source functions have been multiplied with equation 1, but the Gong hasn't. Apparently this is because the Gong function is already low in the larger size range. However, now the Gong function is not anymore comparable to the other two. In my opinion, the Gong simulations should be rerun. At the very least the effect of treating the source functions differently should be thoroughly discussed.

We reran the model after multiplying the Gong function with equation 1 and corrected the numbers related to the Gong function in Table 3 and some figures. As we expected the model predicts lower mass concentration if the Hoppel

correction is applied. The results do not change our conclusions that the CMS source function does a better job than the Gong source function when comparing to the datasets we choose in this investigation.

b) How representative of the oceans are the shape and scale parameters for dust storms (page 24510)? How will the uncertainties in them affect the sea spray fluxes?

Grini and Zender's work (2004) is not applicable only for dust storms. Actually the original reference for this method is Justus et al. (1978). We correct the reference to avoid confusion. However, one can also evaluate the parameters used following the work of (method 3-mean wind speed and standard deviation, Justus et al., 1978). We independently evaluated the values of the parameters by finding the wind variance in the NCEP data we used (not discussed in the paper). This produces spatially dependent values for the parameters. However, when we compare to the SP data we find at some location the constant parameter performs better than the spatially dependent scheme, while at other locations it performs worse. This method also required that we know the wind speed for a period long enough to get the mean and standard deviation, which is not possible for the prediction of future climate. For these reasons we choose to stay with a simple constant parameter scheme.

c) Instead of normalizing the measured and modeled values in many of the plots (e.g. 14), the authors should simply show two scales on the y-axis. This way it would be possible to compare also the differences in the magnitude, not only in the shapes of the curves. It is also totally unacceptable to normalize only some part of the curve as was done in Figure 14 (or does p 24523, line 8-9 mean that all particle sizes where normalized but actual value used for normalization did not take into account the smallest particles?).

We re-plotted figure 14, showing two scales on the y-axis. For figures with more than two curves (Fig. 15, 17, where two scale on y-axis are not enough), we put down the normalizing factor for each curve in the caption. We actually

normalize the whole size range in Figure 14, 15, and 17. The description in the text is rephrased to avoid misunderstanding.

Specific comments:

5. SSA is a commonly used abbreviation for 'single scattering albedo', and thus using it for 'sea spray aerosol' is confusing especially when the optical proper ties are discussed. I recommend changing to SS.

The literatures uses SSA to represent sea salt aerosol so we have continued with this convention. However, we agree it can be confusing with the optical convention.

6. It would make it easier for other groups to adopt CMS if the source function equations were presented explicitly in this manuscript.

We now explicitly present the CMS source function in the text.

7. In the introduction, make sure to indicate which of the previous findings are purely from models and not confirmed by observations.

We separate findings from the models and observations with more caution.

8. page 24505, lines 2-4: -> differ *by up to* a factor of 2. Also, there are similar magnitudes of difference also around 200 and 500-600 nm.

We changed "The number fluxes for various source functions between 0.1 and 10 μ m are similar, except near1 μ m where they differ by a factor of about 2." To "The number fluxes for various source functions between 0.1 and 10 μ m are similar within a factor of about 2."

9. Page 24506, lines 6-10: Contradicting information about the upper limit of Clarke applied (0.6 or 1 um?). Last paragraph: For some flux functions the largest particles dominate the areas also at low wind speed. Last line: "SSA area is usually related..." is vague. Clarify in which cases the relation holds.

We considerably changed the wording on lines 6-10. We change "SSA area is

usually related to the optical properties, such as the optical depth" to "SSA area is a critical input to the optical depth calculation."

10. page 24508, lines: 9-11: is the model run truely offline or in a nudged mode? line 18: observation -> observations; lines 21-22: I don't follow what 'as they are -' refers to

The model is run in nudged mode. CAM is run every time step as is CARMA. For example, the wet deposition has to be related to the cloud scheme simulated by the model, which is not from the reanalysis data.

11. Section 2.3: The dry deposition scheme is fairly standard and thus this section can be significantly shortened.

We shortened the section by getting rid of the description of the standard scheme and highlighting the specific setting for this investigation.

12. Section 2.4: The particle wet size is highly dependent on the RH in the range 98%-99.9%. Thus setting an upper RH limit of 98% is not a good assumption. I also can't follow the 'theoretical base' part of why this is done. If Gerber's formula cannot be used at high RHs, then they should not be shown in Figure 4.

The theoretical basis follows the argument in §2.5.3, Lewis and Schwartz, (2004) and the references thereinafter: "The equilibrium vapor pressure of water above a solution drop is less than that of bulk water at the same temperature by an amount that depends on the concentration of the solute and which for small concentrations is nearly proportional to this concentration [Raoult, 1887]. As the mole fraction of water in seawater of salinity 35 is very nearly 0.98, the vapor pressure of water in equilibrium with seawater of salinity 35 is therefore expected to be 98% of the vapor pressure of water at the same temperature; experimentally this has been found to hold to good accuracy [Robinson, 1954]. Thus, at formation, a drop of seawater of salinity 35 ejected in to the atmosphere has a water vapor pressure that corresponds to 98% RH in air at the temperature of the drop." Of course clouds form in our model so portions of grid boxes have

higher humidities than 98%. However, it would be unusual for an entire GCM gridbox to be supersaturated.

The 98%-99.9% curves in the RH plot are eliminated. We also add an argument about the unrealistic hygroscopic growth from the Gerber formula for high RHs.

13. p. 24514, lines 8-13: Do I understand correctly that the user can prescribe 0-100% of in-cloud scavenging or what does the solubility factor refer to? If the former, then the first sentence about default value is slightly confusing. Please reformulate.

We have changed the statement in the text as "The CAM in-cloud scavenging scheme assumes that a soluble fraction of aerosol particles resides in the cloud water and is later removed with the fraction of cloud water that is converted to rain. This fraction is called the solubility factor, ranging from 0 to 1,"

14. Section 3: State the time period of the model simulations as well as the measurements. It is mentioned in several places that not exactly the same years are compared (which is understandable in the case of global models) but more detail is needed. When same years are not compared, do the observations represent multiannual monthly means, etc.?

We added more details to describe the model and observation time frame.

15. Section 3.1., end of first paragraph: For consistency, it would be better to treat the data sets in the same way (i.e. do the elimination based on weekly data, if this is the coarsest time resolution). Is similar elimination of data done for model results? If not, why? Given the different local wind speeds and removal in the model and in the real atmosphere (due to poor spatial resolution, different years, difficulty to model wet removal in the first place) and the relatively small differences between the source functions, is it really possible to make conclusions about the superiority of one function over the other? We now treat the data set in the same way. We average the dataset that are in daily average to weekly average. We show results where we eliminate and do not eliminate points out of one standard deviation. As we can seen by comparing the old and new fig. 6, there are times when extreme events occur at several sites (i.e. Midway and Norfolk) that dominate the mean mass for the month. The model should have picked up the extreme events unless they were local at the measuring site as opposed to grid-wide events. We then filter extreme events in the SP data by eliminating data points outside one standard deviation of the weekly data at Midway Island and Norfolk. We do not filter the model results outside one standard deviation as the model does not pick up the extreme events. The correlation coefficient and slope are actually improved by treating the dataset as weekly averages. Both CMS and Gong function did a good job in predicting the mass. However, the CMS includes the spume droplet mechanism. Gong function doesn't. This is the basis for our conclusion that CMS is more realistic than Gong.

16. p24515, lines 21: do equally -> do almost equally; lines 22-23: this is true for CMS parameterization slope but not correlation (best correlation with solubility factor 0.3). Not at all true for Gong parameterization; Line 25: very well -> reasonably well.

We correct the words we use in this section.

17. p24516, lines 3-4: this is somewhat contradictory to p 24507, lines 4-5 where it is said that it is not know if spume is crudely presented.

If the Gong source function is totally based on Manahan without adding any new observational dataset, then the spume droplets are not represented by Gong source function. As in Gong 2003, no dataset is mentioned for spume droplets.

18. p24518: What size range does 'coarse mode optical depth' correspond to? If typical definition of coarse mode (i.e. diameter larger than 1 um or 2.5 um), then only a very small fraction of SS in terms of total optical depth or number is

investigated. I'm not convinced the presented optical depth comparison is then very useful for model validation. Last sentence of section 3.2.1: is the whole range (0.1-1um) included in the optical depth calculations presented.

The AERONET coarse mode optical depths are defined optically, rather than in terms of a microphysical cutoff of the associated particle size distribution at some specific radius (O'Neill et al., 2003). The meaning of coarse mode optical depth is that it is the part of the optical depth that is more or less wavelength independent. The whole size range (0.01~15 μ m) is used to compute the modeled optical depth for comparison with AERONET coarse-mode optical depth. We compared to the AERONET coarse-mode to eliminate as much as possible the influence of fine-mode particles, such as sulfate, on the observations.

19. Reformulate Table 3: Since two totally different data sets for different locations are used, it might be clearer to present mass and optical depth in two different tables. At the very least, the table caption and heading must be clarified to highlight this fact. The second line of heading ('Model=S_M SP') is very confusing and uninformative. Give the sites/regions/networks compared to in the table caption. Is the model optical depth from only one model grid cell (Mace Head)? This is something that should be clarified also concerning comparison to other data sets.

We reformulate Table 3 according these suggestions.

20. The Mulcahy data set is one whose representativeness is not certain (one site, very limited data set due to strict requirements). Therefore I'm not sure it makes sense to compare to model 'North Atlantic' and 'Southern Ocean'. Also refer to table 4 for definition of these areas.

Comparison is available between Mulcahy, Smirnov, and Satheesh data in Madry et al. (2011) (in press). Mulcahy matches Smirnov for low wind speeds. Satheesh data is confined for the India ocean but with similar wind speed dependence. So the wind speed dependence of Mulcahy is not limited to one location at Mace Head at least at low wind speeds. While observations need to be greatly expanded, both we and Madry et al. (2011) show that models tend to produce a similar behavior over much of the world ocean.

We added "While Mulcahy's data come from a very restricted part of the oceans, it is interesting that both our model and Madry et al. (2011)'s model suggest similar behavour may occur over much of the world's ocean."

Table 4 reference added.

21. Table 2 shows 7 (not 5 ocean regions)

We correct the corresponding part in the text.

22. Any scaling or normalization done for the figures must be explicitly stated in the figure captions, as it is unlikely that all readers go through the text in detail. We stated the normalizing that we did for Figure 14 and 15.

23. How do the values in Figure 12 compare if the scaling is not done? In my opinion it would be more honest to show the unscaled values, the similar wind speed dependence should be evident also in this case.

We replot Figure 12 with two scales, one for the measurement one for the model so that the results are more honest to the reader. We choose a scale so that the curves overlap with each other. Our point here is that the wind dependence of the number concentration is well modeled. The offset is likely to represent the difference in the rainfall between the modeled and observed years.

24. page 24522: How representative is the Norris measurement?

There are uncertainties for the Norris measurement. Firstly the emission rate of Norris is not corrected for the dry deposition velocity. Secondly the measurement is taken in coastal area. This is now discussed in the text.

25. Figure 14: If the discrepancy in the volumetric size distribution is likely due to inversion of measurements, it might be better to omit 14 a) and just discuss it in the text. Many readers only scan through the abstract and figures, and as is

Figure 14 a) gives the impression that the model doesn't perform very well. Again: normalization must be mentioned in figure caption.

We think it is necessary to show the volumetric size distribution. AERONET reports the volume distributions, and we think others should be warned to be wary of them for sea salt.

26. Figure 15: In all fairness it should be stated in the text that overall the Gong function performs the best against the available data set. p. 24525, lines: 6-7: given that there are about an order of magnitude difference at some sizes, it cannot be said that they match 'very well'. I agree that the absolute concentration is (always) very small in these size ranges, but that doesn't make the match very good.

We change "very well" to be "reasonably well". This one order of magnitude is after normalizing. Lewis and Schwartz is multiplied by 2.241. So the difference is not that bad. We would like to compare the shape of the size distribution, instead of the absolute magnitude, since as we stated in the text that there are all kinds of uncertainties influencing the absolute value in the comparison.

27. p. 24525, last paragraph: Since the majority of aerosol number from CMS comes from the part described with the Clarke parametrisation, it is a little pointless to compare to the measurements behind the Clarke parameterization. At least state this explicitly. Clarke diameter space does not match the indicated radius space (0.01-0.8 vs. 0.01- 0.4); overall, this sentence is not necessary.

We think it is necessary to compare three functions with the Clarke et al. (2006) measurement, since 1) Gong is far lower than the Clarke et al. measurement 2) Although Caffrey is also base on Clarke there seems to be an overestimation on the small particles. Caffrey's function could be based more on Mårtenson function at lower temperature. We reconstructed Clarke's measurement in radius space from 0.005 ~ 4 μ m (diameter = 0.01~8 μ m) to 0.01~4 μ m since our model extended as only small as 0.01 μ m. This is not going

to change anything in Clarke's measurement since we shows a cumulative distribution. The fraction that is smaller than 0.005 μ m is going to be merged with the range that is smaller than 0.01 μ m.

28. Figure 18 and last paragraph of 3.4.2 are common knowledge and can be removed.

Although most people know that number and mass are not controlled by the same particles, we do not agree that most people realize this about area. We have been asked by others to show they are different. Also many models interested in optical depth use mass data to check their models, which is not appropriate. Hence we keep Fig. 18, and the discussion at the end of 3.4.2

29. How do the calculated CCN concentrations compare with previously published model values (e.g. Korhonen et al. (2008, JGR) present simulations without DMS for the Southern hemisphere remote oceans)? It should be straightforward to recalculated CCN at corresponding supersaturations.

We add comparison to the results from Korhonen et al. (2008) as follows. (The CCN at 0.1% supersaturation in our paper corresponds to particles with dry radius larger than 0.07 μ m. The CCN at 0.23% supersaturation in Korhonen et al. (2008) corresponds to particle with dry radius of 0.066 μ m. So they are comparable.)

"Korhonen et al. (2008) simulates the CCN (radius > 0.066 μ m) concentrations in the range from 100 to 300 cm⁻³ in January and less than 100 cm⁻² in July in the "roaring forties" with Dimethylsulfide (DMS) emission turned off. Our prediction is lower with 20~100 cm⁻³ in December, January and February and 10~100 cm-3 in June, July, and August. This difference between the models could be partially due to the Mårtensson source function used in Korhonen et al. (2008) producing more ultrafine particles in the cold high latitudes. In addition, SO₂, which is included in the Korhonen model, could contribute to the formation of CCN in the pristine Southern Ocean even without DMS. Both models shows a similar spatial pattern in the Southern Ocean with the maximum concentration in the region near 90 °E in January and between 45-90 °E in July with minimum in the south Pacific. The two models also have consistent seasonal variation in which the CCN number peaks in the summer when the precipitation is weaker. "

Technical comments:

30. Throughout the manuscript, replace Marttenson or Martenson with Mårtenson (i.e. one t, the second letter is an å not an a) Corrected.

31. page 24504, line 13: they -> there Corrected

32. In figures 2, 14, 15 and 17, use simple lines without the dots/triangles etc. which just make the plot busier and more difficult to read. Use a wider range of line colors in plots 8, 9, 11, 13, 14, 15, 16, 17 and 18 to make them easier to read. Corrected.

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