RESPONSE TO ANONYMOUS REVIEWER #2

We would like to thank the reviewer for his/her comments. We have done our best to address each of the points as detailed below.

Note: All reviewer comments in *italics;* all responses by the authors in normal font.

#1. *The description of how the marine POA emission mechanisms affect the emitted aerosol size distribution should be made clearer (Sec 2.2.1). If I understand correctly, OM_ss provides a size-dependent organic mass fraction of sea spray. Do the sea spray parameterisations implemented in the model (Martensson 03, Monahan 86) account for sea salt only, or for total sea-spray? If the latter, then a fraction of mass emitted should be partitioned as POA according to OM_ss, thus reducing the mass of sea salt emitted (if this was previously the only assumed component of 'sea spray'). If the former, then POA should be added as extra mass, according to OM_ss, as a fraction of the total sea salt + POA mass (as has presumably been done here). The authors should clarify that the approach used is consistent with the sea-spray parameterization used. For the internally mixed case where POA mass is added to the distribution, it is stated that addition of mass leads to 10% increase in mean modal diameter at OM_ss=0.5. It would be useful to know more details of the change across the 4 model size modes resulting from this increase in mass. Since changes in CCN are the focus, these details are crucial to the conclusions.*

The manuscript includes the following discussion: "As the Mårtensson et al. (2003) parameterization was developed for a synthetic sea-water (without organics) and in light of observational and laboratory evidence for the external mixtures of sea-salt and marine POA (Leck and Bigg, 2007; Hawkins et al., 2010; Hultin et al., 2010) and changes in number and size distribution of marine aerosol during high biological productivity (Andreae et al., 2007; Yoon et al., 2007; Fuentes et al., 2010), two different approaches are used for modeling marine POA: externally-mixed and internally-mixed (with sea-salt) emissions." In addition as a Table 5 we have also included an average change in CCN number (not mass) in each of the modes due to addition of marine organic aerosol.

#2. *In the comparison of CAM5 with observations, it is stated that "Global climate models like CAM5 give an average realization of the atmospheric state..." (Page 18870). This is only the case if they are run for several years - model internal variability would be expected to lead to differences in a simulation from year to year. It is stated that the model has been run for 5 years in each scenario, but it is not explicitly stated whether 5-year average fields are shown and used to compare with observations. In addition, it is important to show the variability over the 5-year simulation, and how this compares with the magnitude of differences shown between the model scenarios (this should be shown on Figure 2c). The statement that "Long-term observations of marine organic aerosols available from these two sites smooth out day-to-day variations and therefore are suitable for judging the accuracy of CAM5..." is somewhat misleading in the context of these comparisons. The difficulty in comparing with CAM is not necessarily day-to-day variability, but more interannual variability. Presumably the observations are still for specific years (although details are not provided), and therefore caution should be taken when*

comparing them with a free-running climate model. According to Figure 2C, the model appears to have difficulties in reproducing the shape and magnitude of both POA and SOA observations at the two sites. It is important to clarify whether this is a result of model internal variability (i.e. has only one year been shown?), or if this is the average of 5 year simulation, the variability around the mean should be shown. i.e. is this representative of the model skill on average, or does the model do much better for some years (due to e.g. variability in windspeed)? It would be useful to quantify the agreement between model and observations from the different simulations (e.g. model mean bias - see also specific comment below).

The updated manuscript explicitly states that fields shown on Figs. 2-7 are 5-year averages. Fig. 2C has been modified to show the variability over the 5-year simulation (as error bars). The updated manuscript also includes the results of the paired t-test ($P < 0.05$) between marine-source submicron OM concentration and CCN number changes that shows that the regional increases in CCN in the vicinity of biologically active regions are statistically significant (see Supplemental Fig. S1). Additional discussion is also included regarding the model difficulty reproducing the shape and magnitude of POA and SOA and that more detailed studies using CAM5 with the reanalyzed winds (for comparison with daily observations) may be needed for improved evaluation of different emission mechanisms against larger suite of observational data.

#3. - *Page 18861, line 23: Remote-sensed chlorophyll data - please provide more information on the source of this (SeaWiFS?, MODIS?, years used).*

The source of Chlorophyll-*a* is included in the updated manuscript. "In the Vignati et al. (2010) scheme which updates O'Dowd et al. (2008), the organic fraction of sea spray is determined using a positive linear relationship with ocean surface chlorophyll-*a* concentration ([Chl-*a*]) obtained from the Sea Wide Field-of-view Sensor (SeaWiFS) using the OC4v4 algorithm (O'Reilly et al., 1998) for the years 2000 – 2007."

#4. - *Page 18870, line 27/28: Underestimation of oceanic POA. It is not clear what potential problem is being highlighted by "averaging over a coarse model grid". Does this refer to problems in averaging fine-scale atmospheric features in the model, or does it refer to errors in emissions resulting from use of coarse grid-scale wind speed?*

This has been clarified in the updated manuscript. The text now reads: "Possible reasons for the model underestimation include the climatological monthly mean surface ocean [Chl-*a*], coarse model grid averaging of wind speed and [Chl-a] over highly productive surface regions off the coast of Mace Head, small but non-negligible (~20%) contribution of fossil fuel to OM classified as "marine" at Mace Head (Ceburnis et al., 2011), and uncertainties related to the surf zone impact on aerosol emissions (de Leeuw et al., 2000; Vignati et al., 2001)."

#5. - *Page 18871, lines 11/12. Please provide some reference/evidence to support the statement that "Amsterdam island can be considered to be representative for marine background".*

The reference (Sciare et al., 2009) is included in the updated manuscript.

#6. - *Page 18871, line 16: "significant improvement" of model OA. Please provide a more quantitative measure of the improvement (e.g. model mean bias compared with observations).*

A quantitative measure of the improvement is included in the updated manuscript. The text now reads: "…POA in CAM5 (either through V10 or G11 schemes) leads to an improvement (Default mean bias of -106 ng m^{-3} vs. G11 mean bias of 69 ng m^{-3}) in the predicted organic aerosol concentrations at Amsterdam Island."

#7. - *Page 18871, lines 27/28: Possible underestimation of phytoplankton emissions of monoterpenes and isoprene. The authors have simply cited Luo and Yu, (2010), but a little more detail on what the evidence is to support this possible underestimation would be beneficial.*

We have added the following detail: "The underestimation of WSOM in CAM5 may be caused by several factors including an underestimation of phytoplankton emissions of isoprene and monoterpenes with the ratios of "top-down" to "bottom-up" estimates on the order of 30 and 2000, respectively (Luo and Yu, 2010)…"

#8. *Page 18856, line 12: "important for *the* global CCN budget".*

This has been corrected in the updated manuscript.

#9. *Page 18871, line 11: "over Amsterdam Island" not "over the Amsterdam island".*

This has been corrected in the updated manuscript.