

RESPONSE TO ANONYMOUS REVIEWER #1

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. My major critique of the work is that the authors fail to address whether any of their results are statistically significant. It is common practice to run climate simulations for at least 10 years (rather than the 5 shown here) to estimate the impact of climate variability on any results, particularly any estimated differences between simulations. Thus, the results relevant to Figures 5, 6, and 7 need to be discussed in light of what differences can simply be expected from climate variability and whether any of the regional differences apparent in these figures are in fact “noise”. Five years of simulation is insufficient to judge this, thus the authors need to extend their simulations.

We thank the reviewer for bringing up this point. In the updated manuscript we include the discussion for the statistical significance (at 95th percentile) of the observed regional changes. The paired t-test shows that the regional increases in CCN (at 0.2% supersaturation) number concentration in the vicinity of biologically active oceanic regions are statistically significant, while CCN changes over the land are likely to be a noise. We agree with the reviewer that in order to capture climate variability at quasi-biennial and/or decadal time scales simulations need to be run over the time scales considerably longer than 5 years. However, in this study we hold sea surface temperature (SST) constant; therefore it is expected to see only intra-annual (seasonal) changes and the variability associated with weather-related events. Our statistical test shows that the 5-year averages used in this study are enough to capture variability caused by marine sub-micron organic aerosol emissions. Similar studies in which the aerosol effects on clouds were explored (e.g., Storelvmo et al., 2006, 2008; Liu et al., 2007; Seland et al., 2008; Hoose et al., 2009; Gettelman et al., 2010; Liu and Wang, 2010) also conducted sensitivity tests for five years (or less) with a 3-month (or one month) spin-up period.

#2. Abstract, lines 17-19, and page 18876, lines 15-20: This rosy interpretation of the model-measurement comparison is not supported by the analysis shown here (Figure 2), where WIOM is at least a factor of 2 off at both sites. The manuscript should clearly indicate that more work is required to improve the model simulation and/or compare it more extensively to observations.

The following text has been added to the modified manuscript: “However, remaining differences (often more than a factor of 2) between measured and model-predicted submicron marine organic aerosol concentrations suggest that comprehensive analysis of different marine POA emission schemes, combined with long-term measurements of size-and composition-dependent production flux of sea spray aerosol, and process-based laboratory studies are needed to improve model simulations of marine organic aerosol.”

#3. Abstract, line 25: grammar “increases and decreases”

This has been corrected in the updated manuscript.

#4. Page 18856, line 12: grammar, “important for the global”

This has been corrected in the updated manuscript.

#5. Page 18856, line 15: grammar “and the lifetime”

This has been corrected in the updated manuscript.

#6. Page 18856, line 23: grammar, “the resulting reflectivity”

This has been corrected in the updated manuscript.

#7. Page 18857, lines 20-21: Do Ma et al., estimate a radiative forcing or a radiative effect of sea salt?

Although the title of the paper by Ma et al. (2008) is “Modelling sea salt aerosol and its direct and indirect effects on climate,” authors calculate direct and indirect radiative *forcing* of sea salt.

#8. Section 2.1: The model description fails to describe how particle deposition is treated in the model.

The following discussion regarding particle wet and dry deposition are now included in the modified manuscript: “Aerosol wet removal is calculated using the wet removal routine of Rasch et al. (2000) and Barth et al. (2000) with modifications for the consistency with cloud macro- and microphysics. The routine treats in-cloud scavenging (the removal of cloud-borne aerosol particles) and below cloud scavenging (the removal of interstitial aerosol particles by precipitation particles through impaction and Brownian diffusion). Aerosol dry deposition velocities are calculated using the (Zhang et al., 2001) parameterization with the CAM5 land-use and surface layer information, while particle gravitational settling velocities are calculated at layers above the surface (Seinfeld and Pandis, 2006). Both velocities depend on particle wet size (Neale et al., 2010).”

#9. Section 2.2.1/2.2.2: The source of the chlorophyll-*a* data or the “phytoplankton abundance” used is never described. Satellite products?

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.”

#10. Page 18862, line 8: grammar “to be consistent”

This has been corrected in the updated manuscript.

#11. Page 18863, lines 4-9: Could you give some numbers for the mean aerosol concentration differences over the ocean?

Aerosol number concentration over the ocean is now given as Table 5 in the updated manuscript.

#12. *Page 18864, lines 20-25: I assume the SOA is formed irreversibly? Please clarify.*

This has been clarified in the updated manuscript. Now the text reads: “For the irreversible conversion of marine BVOC to SOA...”

#13. *Page 18870, lines 15-17: Perhaps it would be useful to justify here why simulated POA is compared to WIOM and SOA to WSOM and what uncertainties are associated with this assumption?*

The updated manuscript includes the following discussion: “It is commonly assumed that WIOM over the marine environment is predominantly associated with primary emissions from the sea surface, while WSOM is of secondary origin (Ceburnis et al., 2008; Facchini et al. 2008b). However, recent studies revealed that oxidation of marine primary OM could also lead to the formation of WSOM (Rinaldi et al., 2010; Ovadnevaite et al., 2011). This process is not included in our model simulations and could lead to underestimation of ocean derived WSOM.”

#14. *Page 18871: The discussion here of the challenges of comparing observations with a climate model, suggests that simulations with assimilated meteorology (which is possible with CAM) should have first been performed to validate simulated aerosol concentrations against a larger suite of observations before the climate model simulations described here. Assuming that the authors do not want to undertake this effort at this time, they should clearly indicate that comparisons in a CTM are required to evaluate the proposed aerosol schemes.*

The following text has been included in the updated manuscript: “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.”

#15. *Page 18874, line 20: grammar, “such” out of place?*

This has been corrected in the updated manuscript.

#16. *Page 18874, line 21: “enhance the CCN budget”*

This has been corrected in the updated manuscript.

#17. *Page 18875, lines 4-5: presumably the relative differences in the polar regions are due to very low CCN concentrations?*

It is primarily due to elevated emissions of DMS in polar regions. This point has been clarified in the updated manuscript.

#18. *Page 18877, lines 2-3: Could the authors elaborate on “the experimental evidence”? Are they primarily referring to their own work (Meskhidze and Nenes) or others?*

Few references (e.g., O’Dowd et al., 2004; Yoon et al., 2007; Fuentes et al., 2010) are included in the updated manuscript.

References:

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