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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effect of primary organic sea spray emissions on cloud condensation nuclei concentrations" *by* D. M. Westervelt et al.

D. M. Westervelt et al.

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Reviewer comment:

The work presented is of potential great significance and the reader does expect a lot while reading the manuscript. Yet, the few results presented are very briefly analyzed and even less validated. The approach in calculating the oceanic enrichment of seaspray is not the most current one, since Vignati et al. (2009), already cited in the paper, updated the O'Dowd et al. (2008) parameterization. That should not affect the conclusions in any significant way, but the most resent parameterization must be used. Also, the validation of the model is not properly done and some generalizations are unjustified. I do not suggest publication to ACP in the present form, since the results presented are neither convincing nor properly analyzed.

Response:

Vignati et al. (2009) is indeed more updated, but was not available at the time the work began. Repeating the simulations with the updated parameterization would take considerable amounts of time, with no significant changes to the conclusions (as suggested by the reviewer and the results of Meskhidze et al., 2011). We therefore would like to keep the current emissions scheme, but mention this issue in the text.

Reviewer comment:

The authors presented results in a couple of places selecting arbitrary examples. Although the cases selected are justified, they should present additional information for more cases and/or wider regions, for the reader to be able to understand how the conclusions drawn change, primarily in space. The results presented at the end of paragraph 2.3.1 are for 15% enrichment, while the parameterization used allows (and this is expected to happen as well) for enrichments higher than 50%. In these cases, the effects on CCN and kappa of the mixed particle are expected to be augmented. In addition, the authors arbitrarily selected a single grid point (45S, 0E) for the discussion in paragraph 3.4. I would strongly suggest extending this discussion in larger areas, or with additional representative grid points.

Response:

We appreciate the comments on the choice of location and we are sorry for any confusion. The choice of 45 degrees south latitude is not completely arbitraryâĂŤit was chosen because that particular point lies within the largest model predicted marine OC concentrations, a justification the reviewer acknowledges. This can be seen clearly in Figs 1c and 1d of the original manuscript. We expect CCN effects to be greatest at that location and have chosen it as a model test site.

The model does not show larger than 15% enrichment in monthly average concentrations at any of the explored locations. This is due to transport and mixing from less

biologically active areas with those with high levels of marine organics.

Reviewer comment:

The aging was found to be insignificant, at least for the CCN activation. Still section 3.1 and table 3 should have some numbers from these simulations, at least the aging budget (middle of table 3). Further, having internally mixed aerosols should affect the solubility of the mixed sea-spray particles considerably. This is not evident in table 3, where the lifetimes are listed: sea salt lifetime hardly changes, while the lifetime of hydrophobic OC increases by about 15%. Is solubility of the mixed particle being affected in the same way as kappa?

Response:

We appreciate the reviewer's interest in the aging budget information. We have not edited Table 3, but the important figures are now quoted in Sect. 3. Section 3 now contains a sentence that reads:

"The main difference between the slow aging case presented here (S-ORG/BASE) and the other two cases (F-ORG/BASE and N-ORG/BASE) is the magnitude of the aging term. The aging budget information for the N-ORG/BASE case is 0 Tg yr-1 for both simulations since those simulations have no aging occurring. For the F-ORG/BASE simulations, aging is happening quite a bit faster than in the S-ORG/BASE scenarios. In F-ORG and F-BASE cases, aging is responsible for 28 Tg yr-1 and 20 Tg yr-1, respectively. As shown in Table 3, S-ORG and S-BASE having aging budget values of 18 and 13 Tg yr-1, respectively."

Sea salt lifetime hardly changes because the relative changes in the sources and the sinks of sea salt are largely not affected by the replacement with organic carbon aerosol. On the other hand, the marine organic aerosol is a significant addition to the organic aerosol budget in the model, thus the lifetime is affected more significantly. The following has been added to the manuscript in Sect. 3.1:

"The sea salt budget is dominated by the coarse mode and therefore is largely unchanged since marine organic OC emissions were limited to submicron sizes."

Reviewer comment:

One of the weakest parts of the manuscript is the comparison with measurements. Acknowledging that remote oceanic organic aerosol measurements are extremely sparse, one should note that they do exist. Out of the three stations mentioned in the paper one of them (Azores) was not studied without any given reason. The second, Mace Head, is very difficult to compare with, since (to my understanding) the measurements were performed only when clean air masses were influencing the area. If the authors compared the model's monthly mean concentration with Mace Head's data, the comparison is not valid. For Amsterdam Island, they had a bad correlation without primary marine organics (underestimation), and they ended up with a bad correlation (overestimation). There is no improvement here, and no discussion why the model has improved, even with this overestimation. It was mentioned that the present study has a factor of 2 higher fluxes than previous studies. If one assumes for a minute that this factor of 2 is a real overestimation, the factor of 5 overestimation of Amsterdam Island will become something like factor of 2.5, which is still large. It was not mentioned though that Roelofs (2008) had almost a factor of 5 higher marine organic fluxes compared to this study.

Response:

We appreciate the reviewer noticing the mention of Azores. This location should have been removed from the manuscript. The Azores location was not compared and any mention of it is now removed from the manuscript.

When we compare to Mace Head observations (Figure 2), the Mace Head observations are not filtered for clean air masses. The observational data are unfiltered monthly mean values .Specifically, according to Yoon et al. (2007), 23 samples were taken per week for a two year period from 2002-2004. Model monthly means, while not

necessarily ideal, should be an appropriate point of comparison.

Regarding the model evaluation, we acknowledge that it is not great and appreciate the criticism. We have softened the language somewhat such that any mention in the manuscript that we have vastly improved marine OC predictions is removed. We still feel that our results show some improvement and have quantified these improvements. Particularly, for January, February, December, March, and April, the model over predicts by a factor of 2 or less. Compared to a factor of 10 underprediction in the original model, this is an improvement. See for details the response to the previous reviewer, where we add quantification of the improvements in terms of log mean normalized bias (LMNB).

The reviewer incorrectly states that we did not cite the Roelofs (2008) study. We apologize for the confusion as the study was mentioned and cited but perhaps not in the location the reviewer was expecting. When we claimed we were a factor of two larger in the emissions, we did not include that to mean the Roelofs paper as indicated by the citation list in the results section. In the final paragraph of the conclusion, we add this clarifying sentence:

"Only the Roelofs (2008) study had a higher emission source than the work presented here."

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 5757, 2011.