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Atmospheric Chemistry and Physics Discussions

Interactive comment on "OCEANFILMS sea-spray organic aerosol emissions – Part 1: implementation and impacts on clouds" *by* Susannah M. Burrows et al.

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General Comments:

The manuscript presents changes in simulated organic aerosol, CCN, and cloud microphysical/radiative properties from the OCEANFILMS emissions parameterization incorporated into the E3SM atmosphere model. Ocean properties input to OCEANFILMS are monthly mean simulated values from an unknown biogeochemistry model. A control model run with no marine organics was carried out, and the results from this run are compared to those from 4 different perturbation runs, which vary with regard to how the organic aerosol mass emissions are incorporated into one of four aerosol size

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modes. Simulation types include 1) internal vs. external mixing of the organic aerosol and 2) whether the organics add to vs. partially replace sea salt emissions (presumably on a mass basis). A major conclusion of the paper is that the ADD cases are more physically realistic than the REPLACE cases because the former seem to exhibit more pronounced seasonal variability than the latter; such seasonal variation might be expected in some regions from past observational studies (e.g., the Southern Ocean east of Cape Horn, Mace Head). The internal mixing ADD simulation is concluded to be the most realistic and is used for further analysis of simulated perturbations of cloud and radiative forcing properties (relative to the control). The heavy reliance on annual means (despite strong seasonality in modeled organic contributions for the ADD simulations) and on zonal means (despite hemispheric differences in ocean vs. land contributions) may obscure more significant differences between the OCEANFILMS simulation and the control simulation.

The work is interesting and relevant to Atmospheric Chemistry and Physics. The manuscript is well written with a nice introductory review of the literature that contextualizes the present work. However, the current manuscript also 1) lacks important details regarding the simulation experiment setup, 2) fails to adequately support some of its conclusions, and 3) references a companion paper whose relationship to the present work is unclear. I think that this is nice work, and the manuscript should be suitable for publication after the following major/minor comments are adequately addressed:

Major comments:

1) Much of the results of this study are presented in terms a percent change in aerosol, CCN, and cloud microphysical/radiative properties from the OCEANFILMS simulations relative to the control simulation (with no marine organic aerosol emissions). Because

only changes are presented, this makes it difficult to assess whether or not the model simulations are producing realistic results and how important a change of, e.g., the 30 CCN cm^{-3} mentioned in the abstract is relative to the background CCN concentration. An in preparation companion paper with some of these results is referenced on Pg. 3, Lines 25-27, but I think it's important to include them here. Please add a column to Figures 5, 6, and 7 showing the INT-ADD simulation result variables to complement the absolute and percent change of these variables. How reasonable are the simulated aerosol number, CCN number, cloud droplet number, and LWP spatial distributions as compared to other model studies or satellite observations? This is important, because ensuring that the model output is realistic is a prerequisite to quantifying the significance of changes from incorporating the new parameterization.

2) The companion paper of Burrows et al., 2017 (referenced on Pg. 3, Lines 25-27 and Pg. 14, Lines 30-32) appears to have a very similar title to the present manuscript and is also a "Part 1". I don't understand the need for a two-part manuscript, and suggest that the authors consider removing these references, removing the Part 1 from the title of the current paper, and incorporating the additional simulation results requested in 1 above into the current paper. If there's a good reason for a two-part paper, then it would be helpful for the authors to provide that rationale in their response and a copy of the other paper so that they can be reviewed together. Often, a stand alone paper followed by a later, independent paper is a better path than a two-part set of manuscripts.

3) The color scales of the figures are very large and make it difficult to discern small changes in the statistically-significant ocean regions, while emphasizing large changes over the continents. Please mask the continental portions of the maps and change the color scale extents so that the regions of interest are more discernable. Figure 6-top-right is a good example of this, where it is hard to tell if LWP varies in the one meaningful shaded region east of Cape Horn by closer to 0% or closer to 60%. Similarly, the accumulation mode number hotspot in east Africa in Fig. 5 dominates that scale at the expense of the North Atlantic and Southern Oceans (where aerosol changes appear

to be statistically-significant but close to 0 kg^{-1}).

4) Does the model provide CCN information at higher supersaturations than 0.1%, and how do the organic emissions from OCEANFILMS change the shape of the CCN spectrum (i.e., CCN concentration or activated fraction as a function of supersaturation)? How much variability in modeled supersaturation is observed in the model results? Is 0.1% representative of remote regions, or are larger supersaturations found where there are low aerosol concentrations?

5) In Section 2.3, it is stated that the sea spray emissions parameterizations of Martensson et al. (2003) and Monahan (1986) were based on lab experiments that included only inorganic species and no organic species, so it is assumed that these parameterizations do not include any contributions from organic matter. This line of reasoning doesn't make sense to me, as presumably the presence of dissolved organic matter in the seawater would contribute sea spray aerosol emissions from the same bubble bursting processes that produce the inorganic aerosol, but not necessarily in the same amount or size. Monahan's parameterization is limited to particles above 2.5 μm, where perhaps neglecting an organic mass contribution is reasonable, but Martensson et al.'s parameterization extends down to 20 nm, where the likelihood of this is assumption being true appears less convincing. On what basis can it be postulated that the emission of organic and inorganic species from the same mechanistic process (i.e., bubble bursting) can be treated independently of each other?

6) In Section 2.4, some of the key inputs from the biogeochemical model to the OCEANFILMS parameterization are mentioned briefly. While the detailed model discussion is given by Burrows et al. (2014), it would also be helpful to have a few more details given here. For example, which biogeochemistry model was used to compute the monthly mean input fields? What parameters from the E3SM atmosphere model, if any, serve as inputs to the combined OCEANFILMS and sea spray emissions parameterizations (e.g., wind speed)? If OCEANFILMS determines the OMF of the emitted sea spray aerosol, but the sea spray aerosol emissions rate is governed by parame-

terizations based on no organic emissions, how are these parameterizations blended together in the ADD vs. REMOVE simulations? Is this blending done on a mass or number basis (or even at all - did I misunderstand something here)?

7) Section 3.1 currently lacks details on how the ADD versus REMOVE simulations are set up and the meaning of what these simulations represent. For example, am I to understand that if 1 μg of MOA is added to the accumulation mode then 1 μg of NCL is subtracted from the accumulation mode in the REMOVE simulations? Under what circumstances might that make physical sense?

8) There appear to be large differences in the organic aerosol emissions associated with the ADD vs. the REMOVE simulations (Table 4). Shouldn't at least the sum of the MOA mass emissions rates across all size modes (in Gg/yr) be the same in all 4 simulations? I could see how differences in how these mass emissions are partitioned across mixing state, size, number might drive differences in mean atmospheric burdens (Figure 2 and Table 4), but I don't understand what would change the mass emissions rates themselves. Put another way - if the ocean biogeochemistry model monthly means drive OCEANFILMS, and OCEANFILMS drives the emissions rates, why would these emissions rates vary across the four simulations that incorporate the OCEANFILMS parameterizations?

9) Sections 3.1.1 and 3.1.2 seem out of place here as they provide background from previous observational studies. I think that this discussion would be better suited in the "Introduction" section rather than the "Model simulations and analysis methods" section.

10) The conclusion drawn on Pg. 15, Lines 4-5 seems very speculative based on the results presented in this manuscript. Despite the good agreement at Mace Head, the comparisons with observational data from Amsterdam Island and Point Reyes don't appear very conclusive. Given the speculativeness of the writing, I suggest that the authors strike this paragraph, and proceed with Sections 4.2 and 4.3 as an example

case for closer examination. It might also be worth noting that while the ADD-INT case is presented, the ADD-EXT case would likely yield similar results.

11) I think that the conclusions drawn on Pg. 1, Line 7 and Pg. 20, Line 3 about "best agreement" or "most realistic" are too strong and not supported by the findings of this study. Instead, I suggest that these statements be reworded to note that: the ADD organics simulations show seasonal variation in organic aerosol that is consistent with some past observational studies, while the REMOVE organics simulations lack this pronounced seasonality. The assumption of an internally or externally mixed organic aerosol appears to have a minor, secondary effect on the simulation results (Figure 2, Table 4).

12) Since the continents have a greater contribution to the zonal mean in the northern hemisphere than in the southern hemisphere, would recomputing Figure 8 with the continental grid boxes excluded reveal a stronger (larger) period of statistical significance in the Southern Ocean and some period of significance in the North Atlantic and Pacific Oceans?

Minor comments:

Pg. 2, Line 11: "of and" missing words

Pg. 2, Line 27: "ina" typo

Pg. 3, Line 26: If this paper is still in preparation, please cite it as such rather than with a publication year.

Pg. 5, Line 29: While "renaming" is the model process of making the jump from one size mode to another, it isn't really a physical process. I suggest removing it from this parenthetical list, but retaining the good, later discussion on Pg. 6, Line 13-14.

Pg. 14, Line 9: Please add a citation for the assumption that OM:OC $\sim$ 1.9.

Pg. 19, Line 28: "sea-spary" typo

Pg. 20, Line 29: May wish to update code availability statement if source code was released earlier this year.

Consider reversing Tables 1 and 2, as the abbreviations that are first defined in Table 2 are used in Table 1.

Table 1: Why does the density of MOA have four significant figures, while the other species have at most 2?

Figure 8: The blue (or black) trace in the top row of panels is very difficult to discern from the green trace. Suggest making it dashed and in front?

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