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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling marine emissions and atmospheric distributions of halocarbons and DMS: the influence of prescribed water concentration vs. prescribed emissions" *by* S. T. Lennartz et al.

Anonymous Referee #4

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Overall comments:

This manuscript describes numerical experiments assessing the uncertainty in emissions of DMS and halocarbons, conducted using recent climatologies of ocean water DMS concentrations (Lana et al., 2011) and halocarbons (Ziska et al., 2013), and online calculation of emissions using a parameterization of the air-sea transfer velocity. Simulations with online emissions are compared that use eight different parameterizations of the transfer velocity (2-year simulations + 1 year spin-up); prescribed emissions

are also compared with default online emissions (23-year simulations). Careful intercomparisons of model parameterizations within the same model system are highly valuable and important in improving understanding of differences in performance of different parameterizations.

The study is well-designed and has been carefully and thoughfully carried out. The results are mostly well-presented and discussed, but some additional information is needed to clearly show the results of the model-observation comparison. In particular, the presentation of error metrics should be improved, and alternative error metrics for characterizing the model performance should be considered. Also, a direct comparison of the observations and subsampled model output should be provided in a figure.

After these and the remaining detailed comments below have been addressed, I would recommend this paper for publication.

Major comments:

1. p. 17564, l. 11-12: The simulations comparing the effects of different transfer velocity parameterizations are each two-year simulations, with one year of spin-up. Two years may not be long enough to obtain a good statistics. Please provide results from simulations lasting at least five years (plus spin-up time), or justify why two years is sufficient for this study.

2. p. 17564, l. 19 - p. 17565, l. 2: Please provide some brief information for the reader about how these different transfer velocity parameterizations were developed, e.g., are they based on laboratory or field observations?

3. When model resolution is increased, a greater amount of wind gustiness can be represented. For emission parameterizations with a non-linear dependence on wind speed, this leads to resolution dependencies in the emissions. How much do the online-calculated transfer velocities in this study depend on model resolution, and were these parameterizations previously developed / tuned for use at a particular spatial res-

olution?

4. Table 5: I am finding the row labels in this table confusing – it needs to be explained better what the numbers represent. For instance, I expected the row "Total ship" to equal the sum of the four rows above it, but it doesn't. It is unclear which rows are absolute differences expressed as ppt, and which are relative differences expressed as percentages. This may seem comparatively minor, but I am listing it as a "major issue" here because it makes it difficult to understand what results were obtained. The caption seems to indicate that some of the statistics presented here are "normalized mean bias" (i.e., sum (model – obs) / sum (obs) x 100%). If that's correct, please use this standard terminology for clarity. The normalized mean bias suffers from the difficulty that it is asymmetric with regards to overestimation (which is unbounded) and underestimation (which is bounded by 100%). It would be valuable (and should require minimal additional effort) to also provide additional a performance statistic such as the mean normalized fractional bias (Yu et al., 2006), which is a statistic of relative bias that is symmetric to relative values of overestimation and underestimation.

Yu, S., Eder, B., Dennis, R., Chu, S.-H. and Schwartz, S. E. (2006), New unbiased symmetric metrics for evaluation of air quality models. Atmosph. Sci. Lett., 7: 26–34. doi: 10.1002/asl.125

5. Please add one or more scatterplots showing subsampled model output versus observations. This is especially important for the ship and aircraft observations, since the paper currently doesn't include any figure showing the values of these observations, or how they compare with the model. However, it would be useful for the ground-based observations as well.

6. Comparison of various transfer velocity parameterizations: how well does each parameterization compare to observations? Can any conclusions be drawn about which parameterization is most realistic and which should be used?

7. Model setup: What feedbacks processes of the VSLS compounds onto climate

(via radiation, clouds) are represented in the model configuration used? Do the atmospheric chemistry reaction mechnisms used here interact with aerosols, and which aerosol model / microphysical and chemical representations were used?

Minor and technical comments:

p. 17557, l. 18: "Compared to. . ." -> "In contrast to"

p. 17559, l.4: "EMAC/MESSy" -> "ECHAM/MESSy"

p. 17559, l. 11: The model resolution used was T42L39, which is reasonable but at the lower end of the resolutions typically used for global modelling – how sensitive are the processes modelled here (air-sea gas exchange, atmospheric transport and chemistry) anticipated to be to increases in model resolution?

p. 17559, l. 24-26: "Photolysis rates for VSLS were calculated by the TOMCAT CTM. . ." – Since the results will depend strongly on these rates, please describe this in a little bit more detail here, so that readers can get a quick idea of what was this photolysis rate product represents. In particular, what is the time/spatial resolution (e.g., are the rates that are used monthly means?), and are photolysis rates entirely prescribed, or is there some ability for them to respond to online, prognostically calculated variables (particularly radiative transfer)? How is the use of prescribed (rather than online-calculated) photolysis rates expected to affect the results?

p. 17563, l. 8: please mention whether the regridding performed using the "extensive" regridding algorithm (conserving global mass). Please also mention the spatial resolution of the original datasets, and their time resolution. When applying the datasets in the online calculation of fluxes, are the prescribed ocean concentrations fields interpolated in time to the model time step?

p. 17563, l. 17-18: "opposite to"-> "as opposed to" or "in contrast to"

p. 17564, l. 19: "overview on" -> "overview of"

p. 17564, l. 10: “sensitivity towards”

p. 17565, l. 9-10: “These two parameterization for kw were added to the submodule code of AIRSEA.” Will the implementations of the parameterizations be made available to the public by contributing them back to EMAC for future released versions? (also please note the typo in this sentence).

p. 17565, l. 13: “until the wind speed of . . .” could perhaps be changed to “at wind speeds below. . .”

p. 17566, l. 6: “same location of” -> “same location as”

p. 17568, l. 19-20: “respond stronger” -> “respond more strongly”

p. 17570, l. 24: “both . . . and” -> “either . . . or”

p. 17572, l. 8: “eight 2 year” -> “eight 2-year”

p. 17572, l. 9: “Largest uncertainty” -> “The largest uncertainty”

p. 17573, l. 4: The sentence beginning with “White cap coverage. . .” needs revision.

p. 17574, l. 22: instead of “uncertainties”, the term “relative differences” (or similar) should be used for clarity. These are not really uncertainties so much as differences between the results of different parameterizations.

Table 4: why not convert the parameterization for simulation 10 into cm/h for better comparability?

Figure 6: Are these zonal means? Please clarify.

Figure 7: Are the standard deviations here the standard deviations of monthly mean values? Please clarify. Please also remind the reader here (i.e. in the caption) how many / which years of observations were used.

Figure 10: Taylor diagrams are calculated from centered statistics and can obscure information about the mean bias. Please also print the mean bias and uncentered

RMSE on the plot for the reader’s information / reference.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 17553, 2015.