

***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.***

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Editors of ACP:

Thank you for asking me to review the manuscript “Modeling Marine Emissions and atmospheric distributions of VSLS”, by Lennartz et al. The analysis in the paper is strong, and I personally found no technical errors. The work represents an important

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step towards fully coupled marine-atmospheric biogeochemistry modeling, which is one of the keys to building next generation Earth System Models (ESM). The authors have adopted a careful, incremental and defensible approach to the problem of computing sea-air trace gas fluxes in a more consistent manner. The paper is simple and effective. I wish I had personally conceived of such an elegant study.

By extension the research raises interesting science and modeling questions bearing on my own work, which lies in the area of biogeochemical ESM development. Let me touch upon these issues here in the sense of an on-line discussion, while noting that it is not at all critical for the authors to address them directly before publication.

General –The overall theme here is that gas fluxes computed from surface ocean concentration distributions will improve simulation consistency, relative to the usual and standard emission data sets. But dissolved concentrations need not necessarily be climatologies. The argument can be pushed a step further –if surface water distributions are computed from dynamic on-line biogeochemistry then the entire marine system becomes unified. This is in fact the major driving force for our own model development in the U.S. Department of Energy climate system code. The possibility is opened for full CLAW-like feedback studies.

Line 70 –The importance of iodine to stratospheric ozone chemistry is mentioned as a motivation. My understanding, however, is that this particular heavy element may in fact be of even greater importance in the troposphere. Its lower atmospheric relevance occurs in the context of nucleation and coastal aerosol composition. My favorite references on the subject come from the O’Dowd and Saiz-Lopez groups. Organo-iodine compounds are apparently produced with particular intensity by the ice algae, so that there may be links through polar aerosol and cloud chemistry to albedo amplification.

Lines 161 and 243 –I am gratified to see that the authors have the courage to cite classic references like Liss and Slater 1974, or even Wilke and Change 1955. I grew up with these papers and agree that the pioneers should be continually recognized.

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Line 182 and elsewhere –Beginning with the treatment of rain effects, I was reminded of a traditional obstacle to effective sea-air gas transfer modeling. A lingering question is, what processes contribute the largest uncertainties to dynamic flux estimation? I believe the answer has always been and remains the same. The effect of organics and surfactants on physical properties of the ocean interface drives error bars of order a factor of three in either direction. Hence the total uncertainty can approach an order of magnitude at some wind speeds. This difficulty is implicit in almost any transfer study and sometimes it is even stated directly. One often finds the information buried deep in a discussion section, since it is viewed partly as an intractable embarrassment. But Nelson Frew of WHOI began to unravel the real physical chemical issues involved beginning in the middle 1990s. In our DOE-ESM effort, we are now simulating global distributions of chemically resolved marine surfactants, initially for purposes of computing primary organic aerosol sources from bubble breaking. This involves the detailed simulation of generalized biomacromolecules and polymers from within the familiar DOC. We are hoping to make the Frew connections to laminar layer barrier and viscosity effects in the very near future. I would be interested in interacting with Lennartz and company on this topic.

Line 342 –the effect of including real atmospheric DMS concentrations is surprisingly and disturbingly large. But this of course is the point of the entire exercise.

To conclude, let me summarize as follows: The paper Lennartz et al. is scientifically important, complete and understandable. It aligns in several interesting ways with my own work on unified biogeochemical systems modeling, and so I have been a very receptive audience. Any criticisms or suggestions that I can offer are quite minor. As I moved through the text I found certain phrases for which the English might be improved or made more standard. These number perhaps a few per page. But in fact while I was finishing up my reading, I reflected on the potential edits and decided that they are unimportant. The work is timely to the ESM community and so it should not be delayed. I will send my long list of small recommendations only if specifically requested to do so.

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Please get this one in the literature as soon as possible, and encourage Lennartz plus coworkers to be in touch with me.

Thanks again for inclusion in the process.

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