

## ***Interactive comment on “Do organic surface films on sea salt aerosols influence atmospheric chemistry? – A model study” by L. Smoydzin and R. von Glasow***

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I received these comments from a fourth reviewer and I would like to post them as they enrich the discussion:

This paper clearly presents a very interesting contribution, attempting to describe the effects of organic film-forming compounds. Such organic films may indeed change the chemistry of aerosols especially in the marine boundary layer as the oceans are largely covered by organic films.

This study tries to take advantage of the currently growing literature about the chemistry of oleic acid as being a proxy of unsaturated fatty acid in aerosol.

To my knowledge this is one of the first attempts trying to model the coupled effects of hindered mass transfer to the aerosols and condensed phase chemistry.

From their simulations, the authors draw a few conclusions:

1. Oleic acid is not a representative substance for atmospheric long chain fatty acids.
2. The real composition of the surfactant coating which contains a lot of different organic compounds alters the uptake behaviour in such a way that the reactive uptake coefficients from laboratory studies cannot be applied under atmospheric conditions.
3. Only a very small fraction of the organic surfactants react with O<sub>3</sub> and the largest fraction reacts with OH or with other oxidants like NO<sub>3</sub>.

However, my main concern about this paper is linked to the strength of these conclusions. The latter are written in a very general way but are the underlying data strong enough to allow such general conclusions to be made? Shouldn't the conclusion and some part of the manuscript be rewritten more specifically in order to list the modellers' need in terms of input data? Basically, firmly established data are missing and therefore the conclusions may appear as weak. Adding specific request in terms of data need would be of major importance.

First, it is absolutely obvious that the importance of oleic acid as an atmospheric proxy needs to be questioned, especially for those being produced in the marine boundary layer! Indeed, strong evidences that oleic acid is important are finally sparse.

In this context, the first conclusion of this manuscript is relatively important as it opens a debate on the necessity to investigate the chemistry of other possible proxies. This is in line with the second conclusion, which requires that more complex systems are studied. Especially what about surface humic acids being transported into the marine aerosols? Recently, Stemmler et al showed that the chemistry of humic acids is important. What about translating such processes in the marine boundary layer? Such humic compounds are certainly much more relevant to the marine environment than

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oleic acid.

In addition most experiments involving oleic acid have been done on solid substrates whereas most the marine boundary layer aerosols are liquids! Can a underlying liquid phase change the chemistry of surfactants? Certainly as it could change the mobility of molecules and therefore its trapping efficiency (in the case of oleic acid, an in-coming oxidant could be trapped in the long carbon chain before reacting) but also its chemistry.

This brings to my second point.

I did not really understand how the condensed phase chemistry is treated during these simulations. Basically, a liquid droplet is a highly reactive medium where the surfactant can be oxidised from both phases (gas and liquid). But what is finally the most efficient process? I do believe that the model used could also simulate condensed phase chemistry. In this context what is the level of radicals (OH or NO<sub>3</sub>) at the interface due to in coming gases and in situ produced radicals? Indeed, a particle with high nitrate content could produce non negligible levels of aqueous nitrate radicals... Can this be of any importance? These radicals would interact with surfactant differently (possibly efficiently) in the aqueous phase compared to the gas phase.

Basically I would request that the manuscript is slightly revisited as no firm conclusion can be drawn for this study, but more interestingly insisting on new data needs.

Stemmler, K., Amman, M., Donders, C., Kleffmann, J., and George, C.: Photosensitized reduction of nitrogen dioxide on

humic acid as source of nitrous acid, *Nature*, 440, 195-198, doi:10.1038/nature04603, 2006.

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