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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Ozonolysis of fatty acid monolayers at the air–water interface: organic films may persist at the surface of atmospheric aerosols" *by* Ben Woden et al.

Anonymous Referee #3

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Review of "Ozonolysis of fatty acid monolayers at the air–water interface: organic films may persist at the surface of atmospheric aerosols" by Woden et al.

In this manuscript the authors investigated the ozonolysis of monolayers of oleic acid on aqueous surfaces. The authors showed that a residual surface film forms at near-freezing temperatures, but not at room temperature. Using a surface analysis technique, they also showed that the results are consistent with the residual film being nonanoic acid and a mixture of azelaic and 9 oxononanoic acid. Although this reaction has been studied many times, these results provides additional insight. Nevertheless, the writing could be more succinct. Similar to the other referees, I suggest reducing

the length significantly. In addition, more discussion on why the films persist in some cases and not others is needed. This fundamental insight is needed to extrapolate the results to the atmosphere. Once, the authors address these results adequately, I would support publishing in Atmospheric Chemistry and Physics.

Major comments:

More discussion on why a surface film was not observed at room temperature but was observed at near-freezing temperatures in the current study is needed. Is the difference due to a difference in vapor pressures or a difference in solubility.

More discussion is needed to understand and rationalize the difference between the results for water at 3.1 degrees C, and NaCl solutions at 3.1 degrees C. A residual film was not reliably measured for NaCl solutions, but it was observed for water at 3.1 degrees C. Can this be explained by differences in solubility? How can the authors justify these results? This insight is needed to extrapolate the results to the atmosphere.

Page 5: lines 152-155. Some discussion is needed on why a residual film was not observed at room temperature in the current study but was observed in the study King et al. 2009. Did King et al. use different water volumes, different packing densities, different observation times? Can any of these variables explain the difference? Was King et al. more sensitive to material at the surface than in the current study?

Page 12, lines 350-353: The authors state that they have demonstrated that a residual film remains at the interface after ozonolysis at -2 degrees C for a NaCl aqueous solution. However, from Fig. 6, this doesn't look like the case. Also, considering the noise in Fig. 2, I am not completely convinced that the residual film reached a stable value at 4*10ˆ3 seconds for a NaCl aqueous solution. If the authors waited until 6*10ˆ3 seconds, would a film still remain on the NaCl subphase at -2 degrees C?

The experiments in the current study occurred on the timescale of 1 hour. Atmospheric time scales can be much longer. What would happen if you waited longer in your experiments? Also, the presence of a residual film may depend on the surface to volume ratio of a droplet. How does the surface to volume ratio in your experiments compare to the atmosphere? Perhaps the presence of a residual film may depend on the vapor pressure of the reaction products and the amount of material in the gas phase. How do your experimental conditions compare to the atmosphere, in this respect? A fundamental understanding is needed to extrapolate the laboratory results to the atmosphere.

Minor comments:

Page 3, lines 80-86. The secondary analysis technique, IRRAS, is mentioned, but the primary analysis technique in the study is not mentioned. If the authors mention the secondary analysis technique here, they should also mention the primary analysis technique. Section 2.1. Is the technique sensitive to the tilt angle of the surfactant at the interface, with respect to the surface normal?

Page 6: line 176, and elsewhere in the manuscript. The authors imply that a temperature of 3 degrees C is more atmospherically relevant than 21 degrees C. They are both atmospherically relevant temperatures. At the surface and over the tropics, 21 degrees C is more common. I would remove "more atmospherically relevant" from the discussion when comparing the two temperatures. If the authors want to focus on the free troposphere, then maybe "more atmospherically relevant" is appropriate.

Page 7, line 216. Here and elsewhere, the authors refer to a film impervious to further ozone. This implies that ozone cannot pass through the film, which was not shown in the current study. Please change "impervious" to "unreactive" or something similar.

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