

We have revised our manuscript according to the suggestions of the Referee's comments and the responses to the comments are as following. For clarity, the Referee's comments are reproduced in blue, authors' responses are in black and changes in the manuscript are in red color text.

Anonymous Referee #1

The work of Li et al. presents a detailed investigation of photochemical aging of atmospheric relevant organic films at the air-aqueous interface. The stability and oxidized products of organic film under UV irradiation were studied with the aid of Langmuir trough and Infrared reflection absorption spectroscopy (IRRAS). This work represents a significant advance, as the water soluble organic compounds extracted from atmospheric and chamber samples were used in the Langmuir experiments for the first time. These methods play an actual role in researching the photochemical aging of organic-coated aqueous aerosols, and the further atmospheric implications. The area expansion of DOPC film was revealed by relaxation curves and was further confirmed by IRRAS spectra. The authors illustrated the mechanisms for the photosensitizing reaction of organic film consisted by unsaturated lipid and brown carbon. The authors have established a representative model for organic aerosol coating with advanced instruments. The methods are appropriate and properly conducted, and the experimental data presented in this work is of high quality. The manuscript is well written, although the authors need to make some small modifications (see details below). This manuscript can be published in Atmospheric Chemistry and Physics after revisions in consideration of the following comments:

Page 4: Why did the authors mix photosensitizers with the artificial seawater? The effect of artificial seawater in the experiment should be described.

Response:

The components of artificial seawater are approximately equal to fresh sea spray. According to our previous studies, artificial sea salts also play a role in stabilizing the organic films (*Atmos. Environ.*, **2019**, 200, 15–23; *Environ. Pollut.*, **2018**, 242, 626–633). We have added some description about the effect of artificial seawater in the experimental section on Page 4:

“The effect of artificial seawater on stabilizing organic monolayer has been confirmed in previous studies (Li et al., 2018; Li et al., 2019).”

Page 5, line 1: The chamber experiment needs to be more specific on the drying conditions.

Response:

The chamber experiment was performed at RH 20%, which is lower than the crystallization RH (35%) of ammonium sulfate (*Atmos. Chem. Phys.*, **2007**, 7, 3909–3922). The seed particle was kept at solid phase.

We have added some discussion on Page 5:

“Relative humidity was about 20% throughout the experiments, which is lower than the crystallization RH (35%) of (NH₄)₂SO₄ (Ng et al., 2007). The seed particle was kept at solid phase.”

Page 7, line 14: The surface state of DOPC monolayers underwent a transition from the gas phase to the liquid phase. The authors should describe gas and liquid phase of monolayer.

Response:

Surface pressure–area isotherms show phase transition-like behaviour of the Langmuir films. In the gas phase, there is minimal pressure increase for a decrease in area. The first transition from gas phase to liquid phase occurs at the lift-off area. There is a proportional increase in surface pressure with decreasing area. Moving into the solid region of monolayer is accompanied by another sharp transition to a more severe area dependent pressure. This trend continues up to a point where the molecules are packed closely and have very little space to move. Applying an increasing pressure at this point causes the monolayer to become unstable and destroy the monolayer. We have added some description of phase transitions and the insert of monolayer behaviours at the various stages in the surface pressure-area isotherm on Page 8:

“When the surface area of the DOPC monolayer was larger than $125 \text{ \AA}^2/\text{molecule}$, the distance between the DOPC molecules was quite large and the intermolecular force was quite weak. The surface pressure of a DOPC monolayer on artificial seawater started to increase from $125 \text{ \AA}^2/\text{molecule}$, where the DOPC monolayer surface state underwent a transition from the gas to the liquid-condensed phase. After the first phase transition, there is a proportional increase in surface pressure with decreasing area. This caused condensation and ordering at the interface, increasing the surface pressure of the organic monolayer. This trend continued up to a point where the DOPC molecules were packed closely and have very little space to move. Finally, the DOPC monolayer collapsed at 46 mN/m (Pereira et al., 2018). Applying an increasing pressure at the collapse pressure caused the monolayer to become unstable and destroy the monolayer. The packing and phase behaviours of DOPC monolayer at different phases were also shown in the inserts in **Fig. 2(a)**.”

Page 12: The y-axes in Figure 5 that are being directly compared had different ranges. It might make more sense to compare A and C in the same ranges (so, switch the y-axes for subplots C and D).

Response:

We have switched the y-axes for A and B in Figure 5.

Page 16: Organic coatings on the aerosols are also important for multiphase chemistry in the atmosphere (i.e. N_2O_5 , HNO_3 uptake). A more expansive discussion of the effect of organic film on the multiphase aerosol chemistry in the atmospheric implication section is encouraged.

Response:

We have added some discussion about the effect of organic film on the multiphase aerosol chemistry on Page 3:

“The packing order and stability of films on the surface also facilitate further discussion of the uptake efficiency of marine aerosols toward atmospheric trace gases such as N_2O_5

and HNO₃ (Bertram et al., 2018).”