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.

Minor revisions:

Regarding the small spectral shifts presented as evidence for changes in DOPC conformation (e.g. P10L33, P11L7), the authors state in their response that "Spectra were averaged over 2000 scans, and IRRAS measurements were repeated at least three times to ensure reproducibility." What was the variability in the reported IRRAS peaks over the three trials? Was it smaller than the reported spectral shifts (i.e. < 2 cm-1)? I think that it would be very useful to clarify this in the text—I know that dark controls were performed, but the variability of the IRRAS bands for both the dark controls and the illuminated samples would be helpful to report.

Response:

We have taken irradiated and non-irradiated DOPC monolayer on the artificial seawater containing IC as examples and clarified the variability of the IRRAS bands for both the dark controls and the illuminated samples on Page 7. The figure of IRRAS variability have been added in the supplement:

"The variability in the IRRAS peaks over the three trials was smaller than 2 cm^{-1}, as shown in **Fig. S3**."

P1L1 "Photosensitizing compounds like brown carbon can absorb UV light"

BrC isn't a "compound"-this makes it sound like it is an individual chemical species, whereas it is actually a complex mixture of light-absorbing species.

Response:

We have modified the sentence on Page 1:

"Water-soluble brown carbon in the aqueous core of aerosol may play a role in the photochemical aging of organic film on the aerosol surface."

P1L1 "can absorb UV light and produce low volatile organic compounds (O:C ratio of 0.25 to 1)"

Where is this O:C range coming from? I wonder if a more general/"big picture" opening statement would be helpful here? Where are the low-volatility species coming from?

Response:

We have modified the sentence on Page 1:

"Water-soluble brown carbon in the aqueous core of aerosol may play a role in the photochemical aging of organic film on the aerosol surface."

P2L4

Literature citations for the fates of phospholipids would be helpful.

Response:

We have cited several references on Page 2 as suggested.

P3L5-20 "These water-soluble organic materials like IC are termed humic-like substances (HULIS) due to their similar properties to macromolecular humic substances"
I wouldn't classify IC itself as HULIS, since IC is a small-molecular-weight species, whereas HULIS are much higher in molecular weight.
Response:
We have modified the statement as suggested on Page 3.
"In addition to small-molecular-weight species like IC, water-soluble organic materials with higher molecular weight like humic-like substances (HULIS) can also absorb light. HULIS have the similar properties to macromolecular humic substances, such as their amphiphilic and polyacidic nature, aromaticity, surface active properties and light absorption ability (Gelencser et al., 2002; Graber and Rudich, 2006; Sannigrahi et al., 2006; Krivacsy et al., 2008)."

P5L7 "All size fractions of SOA we collected were involved in photosensitizing reaction."
Does this mean that all of the aluminum foil substrates were combined for extraction? This should be clarified in the text.
Response:
We have modified the sentence on Page 5-6:
"Then, all the aluminum foil substrates were combined for extraction."

P6L12-P7L6"The collapse pressure of DOPC monolayer on pure artificial seawater decreased from 46 to 28 mN/m with the addition of IC. The collapse pressure for the DOPC monolayer on the artificial seawater containing HA was even lower."
It would be helpful here to explicitly state the concentrations, or at least mention that the collapse pressure would vary with concentration of the two additives—otherwise, the inference for the reader (I think) is that HA reduces the collapse pressure for DOPC more than IC.
Response:
We have modified the sentence on Page 9 as suggested.
"The collapse pressure of DOPC monolayer on pure artificial seawater decreased from 46 to 28 mN/m with the addition of 2.5 mM IC. The collapse pressure for the DOPC monolayer on the artificial seawater containing 30 mg/L HA was even lower."

"In contrast to IC and HA, the collapse pressures of DOPC monolayer were higher in the PM2.5 and SOA samples. The inorganic ions from the PM2.5 and SOA samples may contribute to the assembly of organic monolayer."
A reference for this statement would be helpful—how would the inorganic ions affect the assembly of the monolayer (what does assembly mean in this context)?
Response:
Inorganic ions can interact with lipids and produce ion-lipid complex through chelation (*Phys. Chem. Chem. Phys.*, **2016**, 18, 32345-32357; *Sci. Total Environ.*,

2017, 580, 1155-1161; *Phys. Chem. Chem. Phys.*, **2017**, 19, 10481-10490). The assembly of organic film here means the presence of inorganic ions in the $PM_{2.5}$ and SOA sample may induce the self-organization of lipid molecules and the stability of organic monolayer.

We have added some discussion and references on Page 9:

"In contrast to IC and HA, the collapse pressures of DOPC monolayer were higher in the $PM_{2.5}$ and SOA samples. The inorganic ions from the $PM_{2.5}$ and SOA samples like metal ions may contribute to the organization of organic monolayer (Adams et al., 2016; Adams et al., 2017; Denton et al., 2019)."