

Comment on acp-2021-270

Anonymous Referee #2

Referee comment on "An organic crystalline state in ageing atmospheric aerosol proxies: spatially resolved structural changes in levitated fatty acid particles" by Adam Milsom et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-270-RC2>, 2021

Review of "An organic crystalline state in ageing atmospheric aerosol proxies: spatially resolved structural changes in levitated fatty acid particles" by Milsom et al.

General comments: The manuscript presents results on the analysis of levitated droplets containing oleic acid and its sodium salt using Small-Angle and Wide-Angle X-ray scattering in addition to Raman microscopy and FTIR. The authors report that the particles show a phase gradient during humidification/dehumidification and attribute this to an inverse micellar phase on the outside with a crystalline phase in the interior. The other aging experiment that was carried out used ozone and they propose a core-shell structure where the shell protects the inner oleic acid from ozonolysis even at longer times. Overall, this is an interesting paper and presents results that will be valuable to the community of scientists focusing on aerosol phase state. However, I recommend major revisions and a re-review because there is no supplemental information provided (only raw data, no figures or discussion). A large fraction of the data is reported in these figures and I do not think we can properly evaluate the work without this included. Below I list some additional comments that I would recommend also be addressed based on the information provided in the main manuscript.

Specific comments:

- In the Methodology, please provide a little more information on the experimental set up. Where was the ozone concentration measured and using what instrument/ports? For the POM analysis, how long were the samples allowed to equilibrate?
- For Figure 1, what was the RH value for each of the time points (g-l)?
- Page 13, Figure 7 is mentioned but there is no Figure 7 in the manuscript.

- In Section 3.3 a discussion is made about the ozonolysis experiments. However, no discussion of the role of ozonolysis products is included. Is it known if the particles loss mass? Is the levitator a closed system, or could semi-volatile products evaporate out of the particle? What effect might this have on the observed rates? The raw data for the C=C peak are not provided, please include these as a supplemental figure.
- In section 3.3.3 it is stated that the weak shoulder at \sim 2854 cm⁻¹ becomes more defined during oxidation and that this indicates the oleic acid left is not involved in the acid-soap structure. Please include what this shoulder corresponds to, how does the increase in definition show this?
- Page 17 discusses inverse micellar vs. micellar. Please provide a little more information on why inverse micelles are expected for this system, even at the highest RH values.
- The data provided in the figures in the manuscript look like data for a single experiment. Were replicate experiments run for these samples? If so, which ones and how many replicates?