We thank both of the reviewers for their comments and valuable time reviewing this manuscript. There seems to have been an oversight regarding the supporting information. We believed that a PDF with figures and descriptions was uploaded at submission with the .zip file containing the raw data (please note all of the references to the Supplement *e.g.* "Fig. S6" in the main text). This was accessible when the authors originally submitted the paper (and apparently available to reviewer 1). However, this PDF file seems not to be present in the link to the Supplement. Reviewer 2 therefore has raised justifiable concerns over the apparent lack of a supporting information document. We have included this PDF document with the revised manuscript (and made it available immediately via an author comment when reviewer 2 raised this) and ask that the editor check both the PDF and .zip file are included before any final decision is made. Most of the points raised by reviewer 2 are answered, at least in part, by the Supporting Information document as outlined below.

Reviewer 1

1) As a proxy for cooking aerosol and sea spray aerosol the authors used a 1:1 wt ratio of oleic acid and sodium oleate. I am wondering how well this proxy represents cooking aerosol, or sea spray aerosol, or mixtures thereof? I assume that particles containing a 1:1 ratio of oleic acid and sodium oleate (without other organics or salts) are not present in the atmosphere. Hence, I am wondering how to extrapolate the results in the current study to the atmosphere.

We thank the reviewer for their comment regarding the relevance of this proxy to the atmosphere. We note that the mixture is a molar excess of oleic acid (see beginning of sect. 2 of the main text). The fact that this acid-soap complex still forms at a molar excess of oleic acid shows that the sample does not need to be at an exact 1:1 composition, and that there is a composition window where it can form, where the 1:1 molar complex coexists with the excess component.

Atmospheric particulate matter is not always well mixed and its composition can be rather heterogeneous (Laskin et al., 2019). Fatty acids, including oleic acid, have been characterised on the surface of marine aerosols (Kirpes et al., 2019; Tervahattu, 2002; Tervahattu et al., 2005). This therefore means that they can concentrate in a specific region of a particle, and we suggest that 1:1 acid soap complexes can exist in some regions, in co-existence with other sections of different compositions. However, these results could contribute to an explanation for the extended lifetime of oleic acid in the atmosphere compared with laboratory investigations (Robinson et al., 2006; Rudich et al., 2007).

Our results suggest that it is possible to form such a phase at room humidity (~ 50 % RH).

The addition of other components to the mixture is the subject of upcoming publications which answer the reviewer's point that there are other molecules present in aerosol particles. The addition of other molecules does indeed change the phase observed, something explored qualitatively in previous work (Pfrang et al., 2017).

We have added a paragraph to the end of the atmospheric implications section (4), addressing this point and point **2** together (see next point).

2) How sensitive are the result to the 1:1 ratio of oleic acid and sodium oleate? If different ratios are used or if other organic species or salts are added, do the authors expect the results to be completely different?

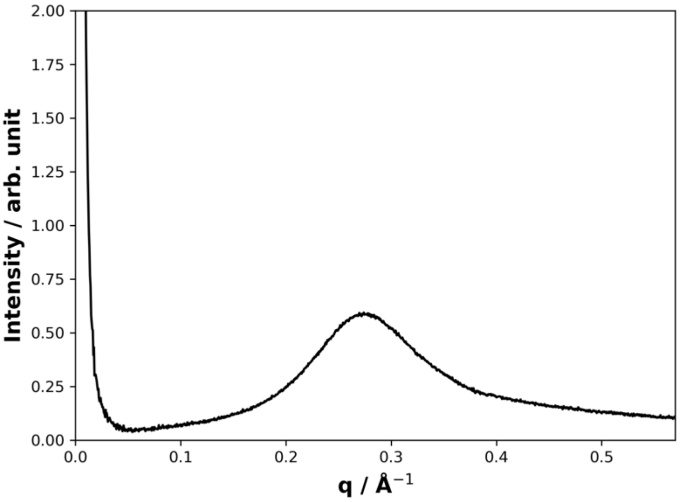
As mentioned above, the composition does not need to be exactly at a 1:1 ratio, and there is a composition window overall where the 1:1 complex co-exists with the excess component. As pH is variable in the atmosphere (Paglione et al., 2021), so too would the ratio between oleic acid and sodium oleate. Beyond the composition window, we have data from an experiment on a 2:1 (oleic acid:sodium oleate) ratio particle, which we present below and will be added to the Supplement as Fig. S14. There is now a much larger excess of oleic acid in the particle and this resulted in an inverse micellar phase. Where exactly in the phase diagram this transition would occur is difficult to determine (small changes in composition are required) and are beyond the scope of this study. However, we can say that more oleic acid (more acidic conditions) induces the inverse micellar phase to form, consistent with the literature (Seddon et al., 1990). Again, follow-on work addresses the valid question of a more complex aerosol composition.

Regarding the addition of other molecules, although we have shown previously that they alter the lyotropic phases formed, they do not prevent lyotropic phase formation (Pfrang et al., 2017); we therefore suggest that the complex in the current work could still exist in the presence of other atmospheric components.

Two paragraphs in response to point **1** and **2** of reviewer 1 added to the *Atmospheric implications* section (4) are as follows:

*[**Atmospheric particulate matter is not always well mixed and its composition can be rather heterogeneous (Laskin et al., 2019). Fatty acids, including oleic acid, have been characterised on the surface of marine aerosols*** (Kirpes et al., 2019; Tervahattu, 2002; Tervahattu et al., 2005)***. This therefore means that they can concentrate in a specific region of a particle,*** **and we suggest that 1:1 acid-soap complexes can exist in some regions, in coexistence with other sections of different compositions*****. The fact that this acid-soap complex still forms at a molar excess of oleic acid shows that the sample does not need to be at an exact 1:1 composition, and that there is a composition window where it can form,*** **where the 1:1 molar complex coexists with the excess component.** ***Our results suggest that it is possible to form such a phase at room humidity (~ 50 % RH). Formation of this phase could contribute to an explanation for the extended lifetime of oleic acid in the atmosphere compared with laboratory investigations (Robinson et al., 2006; Rudich et al., 2007).***

As pH is variable in atmospheric aerosols (Paglione et al., 2021), so too would the ratio of oleic acid and sodium oleate. SAXS data from a 2:1 wt (oleic acid : sodium oleate) levitated particle, representing more acidic conditions, demonstrate that an inverse micellar phase forms at ~ 50 % RH (Fig. S14). Where exactly in the phase diagram this transition would occur is difficult to determine, requiring small changes in composition not practicable for a beamline experiment. This observation shows that a change in aerosol pH could affect particle viscosity via a change in nanostructure. The addition of other molecules does not prevent self-assembled phase formation and composition-dependent phase changes have been qualitatively observed (Pfrang et al., 2017) and are explored further in follow-on work.]



[***Figure S14. A levitated particle of 2:1 wt (oleic acid : sodium oleate) composition demonstrating a broad peak characteristic of the inverse micellar phase with a d-spacing of 36 Å. RH ~ 50 %.***]

Minor comment: The authors include changes due to relative humidity changes in the category of "aging". I would not refer to this as an aging process.

We agree with the reviewer that this is not technically a direct ageing process. We have re-labelled section 3.2 as "**Atmospheric processing: (i) exposure to humidity changes**" and 3.3 as "**Atmospheric processing (ii): chemical ageing with ozone**". We have also amended references to "atmospheric ageing" to "atmospheric processing" when discussing humidification in section 4 (Atmospheric implications).

Reviewer 2

1) 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?

The ozoniser was calibrated offline by UV-Vis spectroscopy as described in the manuscript. We did not make this as clear as we should have done and have now addressed this point directly by adding to section 2.3:

[*The ozone concentration was kept constant at 51.9 ± 0.5 ppm and was calibrated* ***offline*** *by UV/Vis spectroscopy* ***at the outlet of the ozoniser*** *using a* ***PerkinElmer Lambda 465 Spectrophotometer*** *and the ozone absorption band at 254 nm and the absorption cross-section for ozone at this wavelength (1.137 ± 0.070 x*10^{-17} *cm*^{2}*)*]

The POM experiments are described in more detail in the Supporting Information (not originally accessible to reviewer 2). We have now included an additional description of the equilibration times in the main text in any case:

[*Samples* ***for humidity experiments were*** *deposited on microscope slides* ***and allowed to equilibrate at ~ 50 % RH over 6 days. Samples were then*** *humidified by suspending the slides above distilled water inside a small, sealed container. This provided a saturated environment for the samples to equilibrate with* ***for 7 days***. ***Samples for temperature experiments were prepared in the same way but without any humidification.***]

2) *For Figure 1, what was the RH value for each of the time points (g-l)?*

The experiment involved a step increase from ~38 to 90 % RH, with the target humidity being reached within 3 – 4 min. For the humidification plots (Fig. 1 (g) - (i)) only the first plot (g) was at 38 % RH. The rest were at 90 % RH. For the dehumidification plots (Fig. 1 (j) – (l)) again, the first plot was at 90 % RH and the rest at ~38 % RH (room RH). We apologise that this was not clear in the caption and this has now been amended accordingly:

[*Figure 1. ((a) and (b)) 1-D SAXS and WAXS patterns obtained from a dry levitated particle of the acid–soap complex – 1st, 2nd and 3rd lamellar peaks are labelled and a cartoon of the lamellar phase is presented (a). ((c) and (d)) experimental fraction of maximum water content as a function of distance from particle centre and time humidifying/dehumidifying. ((e) and (f)) modelled fraction of maximum water content – best fits to experimental data for humidification and dehumidification. 3–D surface plots of 1–D SAXS patterns plotted against distance from the particle centre for the same particle humidifying ((g) – (i)) and dehumidifying ((j) – (l)) 180 with time humidifying/dehumidifying presented at the top right of each plot (particle size: ~ 150 µm (vertical radius) x 500 µm (horizontal radius); humidification experiment: ~ 38 % (room RH)* ***((g))*** *to 90 %* ***((h) and (i))*** *RH, dehumidification experiment: 90 %* ***(j)*** *to ~ 38 % (room RH)* ***((k) and (l))*** *RH).*]

3) Page 13, Figure 7 is mentioned but there is no Figure 7 in the manuscript.

We thank the reviewer for spotting this typo carried over from an earlier figure arrangement. The figure referred to was Fig. 2(i). This has been corrected.

4) 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.

We thank the reviewer for this comment. A brief description of the ozonolysis was provided at the beginning of section 3.3, however we understand the reviewer's point about not discussing the role of reaction products specifically. Our response is summarised in an additional paragraph at the beginning of section 3.3:

[***Of the reaction products, only nonanal is known to be volatile enough to evaporate appreciably (Vesna et al., 2009). The rest of the ozonolysis products are assumed to remain in the particle phase. There is evidence that particles of oleic acid lose a small proportion of mass during ozonolysis (~ 6 % mass loss after 20 h at 2 ppm), probably due to nonanal loss (Lee et al., 2012). If this is the case, a size change in these particles is likely to be smaller than can be resolved by the X-ray beam in these experiments (~ 15 µm in diameter). Being in an open system with a constant flow of oxygen and ozone, we cannot rule out any mass loss occurring during these experiments. Compared with the effect of particle phase state, we do not expect nonanal loss to impact significantly on the reaction rate.***]

We assume that the reviewer refers to the Raman spectra for the levitated particles. The Raman spectra before and after ozonolysis are included in the Supporting Information file.

5) In section 3.3.3 it is stated that the weak shoulder at ~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?

The shoulder at ~2854 cm^{-1} corresponds to the $–CH_2$ symmetric stretching band in the free oleic acid, as opposed to the asymmetric stretching band of the ordered alkyl chains (~2887 cm^{-1}) (Tandon et al., 2001). There is a molar excess of oleic acid in the mixture, so this is to be expected. As ozonolysis proceeds this shoulder becomes a more defined peak. This area of the Raman spectrum now resembles that of liquid oleic acid. This spectrum is presented in the Supporting Information file along with a comparison with the other components of the mixture. We have amended the text accordingly to define these two bands more clearly.

[*Three key changes in the Raman spectrum are observed during ozonolysis. First, there is a clear shift of the strong acid–soap peak from ~ 2887 cm^{-1}* ***($-CH_2$ asymmetric stretching band)*** *to ~ 2897 cm^{-1} accompanied by some broadening.*]

And:

[*Secondly, the weak shoulder at ~ 2854 cm^{-1}* ***($-CH_2$ symmetric stretch)*** *becomes a more defined* ***peak*** *during oxidation.* ***This region of the Raman spectrum resembles that of oleic acid (Fig. S1(c)).*** *This is further evidence, in combination with SAXS observations (Fig. 2(j)), that the oleic acid left in the system is not involved in an acid–soap structure after ozonolysis.*]

6) 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.

We thank the reviewer for this comment and understand the need for extra clarity on this point. We have added the following to the paragraph discussing this aspect in section 4:

[…~~It is therefore likely that the micellar phase observed in this system has an inverse rather than the normal topology suggested in the preceding atmospheric literature (see Fig. 3 for a cartoon representation).~~ ***The larger hydrophobic tail region of the oleic acid molecule compared with its hydrophilic head drives interface curvature towards water, and therefore the formation of inverse phases, even in excess water. Inverted micellar and other inverted topology phases have been observed for oleic acid – sodium oleate mixtures in excess water (Seddon et al., 1990). Normal topology micelles (polar head groups at the micelle surface) form in systems with larger / charged headgroups, and are only observed within the sodium oleate – oleic acid system at high sodium oleate content (> 80 wt %) (Seddon et al., 1990). It is therefore likely that the micellar phase observed in this system has an inverse rather than normal topology suggested in the preceding atmospheric literature (see Fig. 3 for a cartoon representation).***]

7) 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?

The ozonolysis and humidity experiments were carried out in different particles. The vertical and horizontal radii are quoted in the figure captions for Fig. 1 and 2. We recognise that in the methods section 2.3 the final paragraph makes reference to the size of the particles but does not state the number studied. One particle was studied for the humidity experiment and three for the ozonolysis experiments (including the oleic acid particle) – see Fig. 2(i) in the main text.

The methods section 2.3 has been updated:

[*The levitated particles were analysed on the I22 beamline at the Diamond Light Source (UK). Solid samples, crystallised from ethanolic solutions, were placed into a node of the acoustic levitator. The particles had vertical radii of ~ 90–150 µm and horizontal radii of ~ 500 µm (determined using the attenuation of the X-ray beam).* ***One particle of proxy mixture was levitated subjected to the humidity change experiment. Two particles of the proxy mixture and one pure oleic acid particle were levitated and subjected to ozonolysis experiments.]***

The figure caption for Fig. 2 has also been updated for extra clarity:

[*Figure 2. Vertical scans through the particle showing the effect of ozonolysis on self-assembly. Each row of plots ((a) and (b), (c) and (d), (e) and (f), (g) and (h)) shows simultaneous 1–D SAXS and WAXS scattering patterns vs. distance from the particle centre (measured in µm from what was deemed the particle centre from attenuation data) at increasing time exposed to ozone (labelled at the top-right of every WAXS plot). The particle moved and possibly changed shape during the experiment, vertical movement is apparent from the SAXS and WAXS patterns. (i) Comparison of a levitated pure oleic acid droplet vs. a levitated acid–soap complex particle undergoing ozonolysis, measured by Raman microscopy - A longer ozonolysis experiment on a different levitated acid–soap complex particle is also presented,* ***totalling two ozonolysis experiments on this proxy****. (j) Evolution of the Raman* *spectra between 2750 and 3050 cm^{-1} of a levitated acid–soap complex during ozonolysis. (Particle size: ~ 85 µm (vertical radius) x ~ 500 µm (horizontal radius); [O3] = 51.9 ± 0.5 ppm).*]

An extra sentence has been added to the conclusion which clarifies that the acid-soap complex was also observed on microscope slide deposits:

[*The oleic acid/sodium oleate acid–soap complex has been identified in an unsaturated fatty acid aerosol proxy. Raman and IR spectroscopy, along with SAXS/WAXS, were used to confirm the formation of the acid–soap complex in acoustically levitated particles.* ***The acid–soap complex was also identified by Raman microscopy on microscope slide deposits.*****]**

Additional change:

We noticed a typo in line 300 referring to a "Fig. 6". We have amended this to read "**Fig. 1(d) and (f)**".

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