The authors thank the Anonymous Referee #3 for the comments, which will help us to improve the manuscript.

**Specific comments:**

*L50: Change to "O'Brien…". Also on e.g., L289, L302*

This error will be corrected in a revised manuscript.

*L52: Formatting. Change to: "e.g. Kalberer et al (2004)."*

This error will be corrected in a revised manuscript.

*L63: For condensed phase reactivity, consider adding (Kuwata and Martin, 2012)*

This reference will be added in a revised manuscript.

*L63: For effects on gas-particle partitioning, consider adding (Zaveri et al., 2014)*

This reference will be added in a revised manuscript.

*L70-72: Consider adding (Zelenyuk et al., 2012)*

This reference will be added in a revised manuscript.

*L79: Add: "low" relative humidities*

We thank the reviewer for pointing this out. "low" relative humidities will be added in a revised manuscript.

*L81: Consider adding something like: "… UV light and ozone, as simultaneously encountered in Earth's atmosphere during daytime."*

This phrase will be added to the sentence in line 81 in a revised manuscript.

*L88-91 & L99: Further information should be provided how trans-aconitic acid was chosen as a model system for atmospheric SOA? Has this been identified in organic aerosol in the atmosphere? How does its light absorption compare to other atmospheric organics?*

*Trans*-aconitic acid was chosen because of its favorable physicochemical properties and not because of its presence in the atmosphere. Due to the constraints of our experimental set-up, only limited number of organic compounds can be studied. First, the organic compound must have a carbon-carbon double bond to react with $O_3$. Second, it must be water soluble and must not crystallize easily. It is important to have a liquid droplet to enable the measurement of its size by analyzing its TAOS pattern. Third, it must have low vapor pressure so it can stay levitated in the EDB for a long time without significant evaporation. Non-volatile compounds allow a more precise measurement of size because the change in size of the particle would be attributed solely to aging and not to loss of the pure compound by evaporation. Fourth, it must have low viscosity to prevent mass transfer limitations before aging occurs. Fifth, it must have a high reaction rate coefficient with $O_3$ to allow significant aging in reasonable experimental time spans. Finally, it must photolyze either directly at wavelengths longer than 300 nm or indirectly by using a photosensitizer.

Several acids were tested including shikimic acid, maleic acid, citraconic acid, and *trans*-aconitic acid. The reaction of shikimic acid with $O_3$ and UV was very slow, maleic acid crystallized very quickly, while citraconic acid was volatile. Since *trans*-aconitic acid fulfilled most of the mentioned criteria, it was chosen as a proxy system.

For the question concerning the absorption of AA, we refer to our answer to a related question by reviewer #1:

“We calculated the absorption cross section of 4-nitrocatechol based on the absorption spectrum given by (Cornard et al., 2005). The cross section of 4-nitrocatechol is about 3 orders of magnitude higher compared to trans-aconitic acid between 300 and 600 nm.

We will add a sentence comparing the two cross sections in a revised version of the manuscript to put the absorbance of AA into perspective.”

*L99: Add “possible” synergistic effects …*

“possible” will be added in a revised manuscript.

*L110: Change to “Milli-Q”*

“Milli-Q” will be corrected in a revised manuscript.

*L111: Please indicate an approximate droplet size range used in your experiments in the text.*

According to the technical specification of the inkjet cartridge, the size of the ejected droplets is ~37 µm. We start with a 4 wt-% AA solution with water content equilibrating quickly to RH in the environmental cell. Depending on RH, the size range in the experiments is from ~6 µm under dry conditions after mass loss due to reaction to ~20 µm at high RH before reaction.

We will add size range in a revised manuscript.

*L116-118: Please elaborate how “the hygroscopic response of NaCl particles” were used to check on RH. Was the 1.5% uncertainty in RH determined from the NaCl experiments?*

The humidity sensor used (SHT85) is factory calibrated to ± 1.5% accuracy at the temperatures and relative humidities of our experiments. We checked this by measuring the deliquescence RH (DRH) of NaCl, which is well established in the literature. We found agreement with the calibration of the manufacturer.

We will modify the sentence in line 116 to make clear that these were deliquescence measurements.

L141: Formatting of reference: “as introduced by Braun and Krieger (2001)”. Also on e.g., L257, L302, L346, L380

This reference will be formatted in a revised manuscript.

*L183: “yields”*

This grammatical mistake will be corrected in a revised manuscript.

*L187: It is unclear to me what the “conventional additivity rule” is. Please clarify.*

The additivity rule used is mass fraction based and can be found in Appendix B:

$$\rho(w_s) = \left(\frac{1-w_s}{\rho_w} + \frac{w_s}{\rho_0}\right)$$

where $\rho(w_s)$ is the density of solute at mass fraction $w_s$, $\rho_w$ is the density of water, we retrieve $\rho_0$, the density of aqueous AA solute at dry conditions, i.e. the subcooled melt density.

More details on the choice of this equation can found in (Lienhard et al., 2012).

*L189: Please use the same symbol for “kappa” throughout. Compare e.g., L189 and L190; L385, …*

Same symbol for “kappa” will be used in a revised manuscript.

*Fig. 5: If the efflorescence of AA is around 59.9% (L189), why do you still observe such as strong decrease in particle mass at RH below this ERH?*

The efflorescence of uncreated AA particle is typically around 59.9%. However, this figure corresponds to AA particle, which has been reacted with UV and $O_3$. AA particles after UV and $O_3$ exposure did not exhibit any efflorescence upon drying, as mentioned in section 3.4.

*L209: “O’Meara”*

This error will be corrected in a revised manuscript.

*L229: Consider adding (Chenyakin et al., 2017) as an example for where Rdiff > Rmatrix.*

This reference will be added in a revised manuscript.

*L315: Can you quantify the "slightly different particle sizes"?*

The size of particle 1 at the beginning of the reaction is 14.6 µm, while that of particle 2 is 10.4 µm.

L342: Please clarify what you mean with "partial wetting morphology"

LLPS can exhibit different morphologies, such as core-shell and partial wetting. Core-shell occurs when one phase of the aerosol particle is totally wetted by the other, while partial wetting occurs when one phase is partially engulfed by the other, thus creating asymmetric structures (Qiu and Molinero, 2015). Schematics of different morphologies are illustrated in (Qiu and Molinero, 2015).

If LLSP occurs with core-shell morphology, the TAOS pattern remains that of spherical symmetric particle, while with partial wetting, it becomes irregular. For this reason, we suggest that if LLPS occurred, it would have partial wetting morphology.

*L340-341: Can the authors provide further reasoning why the UV-illumination of AA did not result in changes in the TAOS pattern as compared to the ozonolysis experiments? I would expect UV aging to also cause formation of small, oxidized molecules that could undergo liquid-liquid phase separation from larger oligomers.*

It is still possible that liquid-liquid phase separation occurred upon UV reaction, but if it happened with core-shell morphology, then the TAOS pattern will remain that of a spherical symmetric particle, thus we cannot observe the phase separation.

*On the other hand, you suggest that the O:C ratio before vs. after aging is similar (L435-451). Would this not go against your hypothesis of particles undergoing LLPS or am I not understanding your argument correctly?*

This section refers to particles exposed to UV irradiation or UV and $O_3$ simultaneously and not to particles exposed to $O_3$ alone. Because of the irregularity in the TAOS pattern of the particles exposed to $O_3$ alone, hygroscopicity measurements were not carried out on the particle aged with $O_3$ alone, thus we cannot deduce whether O:C ratio changed after ozonolysis.

*L350: "Precipitation of trans-aconitic acid". Is this feasible? What is the deliquescence RH of (unaged/pure) AA? I.e., do you expect solid AA at the RH of your experiments (~78%)? Please provide further details. This would also help to understand your argumentation on L371.*

The deliquescence RH of pure AA based on our experiments is about 96%. All AA particles at the beginning of the experiments are liquid, since the reactions are carried out at RH above 78%, above the highest efflorescence RH observed in our experiments (~59.9%). This can be seen clearly from the TAOS pattern, which is spherical, unlike solid particles, which exhibit irregular TAOS pattern.

If precipitation of AA occurs after an hour of ozone exposure, part of the particle could be crystallized AA within a multicomponent liquid mixture.

*Fig. 10: It could be nice to add curves for one of your UV experiments to illustrate the difference between both aging types.*

We thank the reviewer for the suggestion. Figure 10 will be modified to include an example of UV aging.

*L363: Consider citing (Marcolli et al., 2004)*

This reference will be added in a revised manuscript.

*L387: Consider citing (Petters and Kreidenweis, 2007)*

This reference will be added in a revised manuscript.

*Fig. 11: It could be helpful to cut off the y-axis at ~0.3 to improve readability of your experimental data. In addition, the comparison to ammonium sulfate is not discussed in the text.*

Putting ammonium sulfate as a reference illustrates for the reader that organics have a significantly lower κ. We will change the first two sentences starting in line 385 to:

*"Figure 11 shows the hygroscopicity results in terms of derived κ values. For reference, a selection of κ values of other organic acids and ammonium sulfate are taken from the literature. As shown in this figure, organic acids can have quite different κ values, but are significantly lower than inorganic salts."*

*L450: "Note that if..." This is an interesting thought. Could it be that the increase in O:C and increase in average molecular weight upon aging "balance" each other so that the hygroscopicity is largely unaffected (as suggested by your Fig. 11), whereas this would both promote an increase in viscosity (as suggested by your Fig. 12)?*

Yes, exactly, this is what we meant by this sentence.

*L471: "presence of water typically slows down oligomerization". Please add appropriate reference.*

(Kalberer et al., 2004)

*L498: Please specify "dry"? Do you mean 17.7%? Also, please specify the temperature for which your mixing time calculations were performed.*

Our calculations were done for the viscosity at the RH of our experiments (~17%) and at room temperature (~298 K). We know from our experiments that at RH 38%, the viscosity is already significantly lower. This would imply that at 298 K, the boundary layer RH needs to be below 20% to yield these long mixing times. However, since lower temperature increases the viscosity, in a colder boundary layer, higher RH may lead to similar mixing times.

We will add the RH and temperature to line 497.

**Section 4:**

*- I would like to see some further discussion on the atmospheric implications and relevance of your results. E.e., can you provide some thoughts on how your results affect the atmospheric processes that you motivate on L63-72? Can the characteristic mixing times (L494-498) be used to draw conclusions about e.g., PAH oxidation rates in the atmosphere?*

Since the characteristic mixing time increases after aging, we expect longer-range transport of the remaining particle. However, this is not directly related to the oxidation rate of PAHs in the atmosphere.

In addition, the lifetime of the particle in the atmosphere increases because it becomes less susceptible to washout and further chemical processing as the viscosity increases.

We will add one sentence about the potential implications of the characteristic mixing times on aerosol lifetime and long-range transport to line 498.

*- I would like to encourage the authors to comment on possible "limitations" of their experiments.*

- *E.g., UV and or UV + $O_3$ aging was done at high RH in this study. Can you provide some*

*thoughts on how performing similar experiments at lower RH would affect your conclusion?*

*Could $O_3$ aging become kinetically limited at lower RH?*

Since most unreacted AA particles effloresced at ~59.9% RH, it is not possible to perform the aging experiments at lower RH. This is because we need the TAOS pattern to measure the change in size and then deduce the viscosity. Since solid AA particles exhibit irregular TAOS pattern, quantitative analysis would be difficult.

$O_3$ aging could become kinetically limited at lower RH, based on previous studies (Berkemeier et al., 2016; Steimer et al., 2014; Kohli et al., 2024). For instance, in the case of ozonolysis of shikimic acid particles, the bulk diffusion coefficients of both ozone and shikimic acid increased by several orders of magnitude as RH increased, since the particle phase changed from amorphous solid to semisolid and liquid, suggesting that the reaction occurs at the near particle surface at low RH and is kinetically limited by slow diffusion (Berkemeier et al., 2016).

From our point of view, the biggest limitation of our study is the missing chemical information during aging, which makes it difficult to understand the differences that we observe. This limitation is already mentioned throughout the text.

*- To what extent does the relatively large particle size of several micrometer used in these experiments affect the results? I would expect $O_3$ aging to be sensitive to particle surface-to-volume ratio, but UV-aging not.*

We agree with the reviewer that ozone aging is sensitive to particle surface-to-volume ratio, while UV aging is not. We expect the rate of ozonolysis to increase with smaller particles, while the rate of UV reaction is less sensitive. However, here we are not providing any rate dependence on ozone concentration or UV intensity. We are only interested in the different properties after aging, which are independent of size because the mechanism should be similar.

*- The brief outlook could be strengthened.*

- *E.g., you would really benefit from measuring both the gas and particle phase composition during aging in future studies.*
- *Lastly, the importance of future aging studies, similar to yours, that simultaneously consider different aging types could be further clarified.*

Concentrations of products in gas phase emitted in single particle experiments are so low that we cannot measure them with current technology. However, flow tube or chamber experiments could be used.

We will elaborate the outlook according to these suggestions in a revised manuscript.

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