General comment:

The manuscript by Antossian et al. uses trans-aconitic acid (AA) as a model system for atmospheric organic aerosol and investigates how different types of aging affect key particle properties, namely dynamic viscosity and hygroscopicity, using an electrodynamic balance (EDB) setup. As aging types, UV-aging is considered and compared to simultaneous aging of AA aerosols to UV light and ozone (O3). Such experiments considering the combined and possible synergistic effects of different aging agents provides a key advancement compared to previous studies that have often aged aerosols individually by either exposure to UV or O3 and investigated effects on particle properties. The authors demonstrate that aging led to significant enhancement of particle viscosity, while hygroscopicity decreased slightly. The results are interesting and the topic fits well within the scope of the journal. The manuscript is well written, and the arguments can be clearly followed. Below I list some specific comments that I encourage the authors to address in a revised version of their manuscript, to further clarify their observations.

Specific comments:

L50: Change to “O’Brien…”. Also on e.g., L289, L302

L52: Formatting. Change to: “e.g. Kalberer et al (2004).”

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

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

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

L79: Add: “low” relative humidities

L81: Consider adding something like: “… UV light and ozone, as simultaneously encountered in Earth’s atmosphere during daytime.”

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?

L99: Add “possible” synergistic effects …

L110: Change to “Milli-Q”

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

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?

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

L183: “yields”

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

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

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?

L209: “O’Meara”

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

L315: Can you quantify the “slightly different particle sizes”?

L342: Please clarify what you mean with “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. 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?

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.

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

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

L387: Consider citing (Petters and Kreidenweis, 2007)

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.

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)?

L471: “presence of water typically slows down oligomerization”. Please add appropriate reference.

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

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?
- I would like to encourage the authors to comment on possible "limitations" of their experiments.
  - E.g., UV and or UV + O3 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 O3 aging become kinetically limited at lower RH?
- To what extent does the relatively large particle size of several micrometer used in these experiments affect the results? I would expect O3 aging to be sensitive to particle surface-to-volume ratio, but UV-aging not.
- 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.

Chenyakin, Y., Ullmann, D. A., Evoy, E., Renbaum-Wolff, L., Kamal, S., and Bertram, A. K.: Diffusion coefficients of organic molecules in sucrose–water solutions and comparison with Stokes–Einstein predictions, Atmos. Chem. Phys., 17, 2423–2435, https://doi.org/10.5194/acp-17-2423-2017, 2017.

Kuwata, M. and Martin, S. T.: Phase of atmospheric secondary organic material affects its reactivity, Proceedings of the National Academy of Sciences of the United States of America, 109, 17354–17359, https://doi.org/10.1073/pnas.1209071109, 2012.

Marcolli, C., Luo, B. P., and Peter, T.: Mixing of the organic aerosol fractions: Liquids as the thermodynamically stable phases, Journal of Physical Chemistry A, 108, 2216–2224, https://doi.org/10.1021/jp036080l, 2004.

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Zaveri, R. A., Easter, R. C., Shilling, J. E., and Seinfeld, J. H.: Modeling kinetic partitioning of secondary organic aerosol and size distribution dynamics: representing effects of volatility, phase state, and particle-phase reaction, Atmos. Chem. Phys., 14, 5153–5181, https://doi.org/10.5194/acp-14-5153-2014, 2014.

Zelenyuk, A., Imre, D., Beranek, J., Abramson, E., Wilson, J., and Shrivastava, M.: Synergy between Secondary Organic Aerosols and Long-Range Transport of Polycyclic Aromatic Hydrocarbons, Environ. Sci. Technol., 46, 12459–12466, https://doi.org/10.1021/es302743z, 2012.