

Atmos. Chem. Phys. Discuss., referee comment RC1  
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## Comment on acp-2022-331

Anonymous Referee #1

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Referee comment on "Not all types of secondary organic aerosol mix: two phases observed when mixing different secondary organic aerosol types" by Fabian Mahrt et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-331-RC1>, 2022

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General:

The authors show that phase separation can occur when SOAs from different VOC precursors are mixed. The manuscript describes new and original data and set it well into context of previous work.

The manuscript left over some questions, however for answering that the data set is too small and too selected. Nevertheless, it a very well written and inspiring manuscript, which could induce more research in the direction of phase separation in atmospheric OA. The manuscript is suited to be published as ACP letter after the authors considered some comments.

I would like the authors to respond to the following considerations.

Hypothesis: the difference in average O/C ratio is a necessary prerequisite but not sufficient for phase separation. The width of the O/C distribution of the condensed products, thus the (non-) overlap in O/C must play a role, too. They authors addressed that a bit in the section where they discuss partial miscibility. It could be extended there. (For the moment we can neglect that O/C is not the real driver -only the indicator- for specific intermolecular interactions between differently functionalized molecules, which determines the phase behavior besides entropic aspects.)

I guess the data set is a bit lucky in this way. The aromatics are shorter in C-chain and lower in molecular weight (MW) thus need to reach a relative high O/C before condensing. In contrast SQT will condense already at low O/C, because they have already high molecular masses *per se*. So *a priori* SQT SOAs are in tendency less polar than aromatic SOAs. These are exactly the two classes that do not mix, while MT with intermediate MW and oxidation degrees will mix with both other classes.

Since all pure systems show no phase separation, different O/C bins in these cases are obviously not populated enough. As consequence O/C variation of groups of molecules within the product spectrum is too narrow. A Delta O/C = 0.47 and cannot be realized in order to undergo phase separation.

So, it is probably not only the Delta O/C but also the spread of the O/C around the average which is eventually important. From this point of view, it would be interesting to see if mixing SQT, MT and Aromatic SOA would lead to phase separation or if the MT products would "mediate" sufficient miscibility.

These considerations do not affect the results as such, but possibly a simple transfer to atmospheric mixtures and particles.

Minor comments:

Looking at all the images I don't recognize the growth with increasing RH (compare suppl. Line 265-270). Can it be that transport barriers arising by the fact that the particles are in glassy or high viscous states state help to establish phase separations?

Line 65: Anttila et al, J. Atm. Chem., 2007 show direct experimental evidence of phase separation and shell core morphology.

Technical corrections:

Line 106/Figure S11: The images of the pine SOA look different in shape compared to the

binary mixtures. It seems to wet the carrier surface? What is the reason?

Line 117/eq 1: Why "1-2" instead of "-1" ?

Line 118: m denotes the slope in which sense? That it must become  $\ll 1$  to get a step like function? Please, move that info from the caption Figure 4 (also) to here.

Figure 3/line 576: The labels of the isolines are too small and too weak. They are hard to see.

Line 241: "The RH within the particles..."? I guess, you meant "the RH the particles were exposed to"?

In the Supplement

Line 71: I guess UV radiation at 254 nm is not energetic enough to photolyse O<sub>2</sub>. Could it be that the pen-ray lamp had also a 185 nm contribution?

Line 324: Please, give a reference for the "TROPOS atomizer".