Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-19-RC3, 2019

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Liquid-liquid phase separation in secondary organic aerosol particles produced from α-pinene ozonolysis and α-pinene photo-oxidation with/without ammonia" *by* Suhan Ham et al.

Anonymous Referee #3

Received and published: 15 March 2019

This work investigate the liquid liquid phase separation of alpha-pinene derived SOA from both ozonolysis and photo-oxidation in the presence or absence of ammonia gas. This work shows that only reaction products originated from ozonolysis of alpha-pinene undergo phase separation at a very high humidity. The authors suggest that bulk elemental composition (e.g. O/C ratio) could be a good proxy for determining the phase separation of alpha-pinene derived SOA formed from ozonolysis and photo-oxidation. This work provide valuable data which allow us to better understand the phase state and morphology of ambient SOA. I have a few comments for the authors' consideration.

Comments:

Abstract, "LLPS occurred when the O:C ratio was less than ∼0.44 and LLPS did not occur when the O:C ratio was greater than ∼0.40. When LLPS was observed". Since the elemental ratios of the SOAs have not been measured in this work, it may not be appropriate to put down the O/C ratios in the abstract.

Experiment section, my major comments are how the high gas-phase concentration of alpha-pinene, ozone, OH, ammonia and aerosol mass loading used in this study affect the molecular composition of the SOAs, which would ultimately govern the phase separation of the aerosols. The authors should give more discussion on these aspects.

Page 5, line 13, "During a humidity cycle, RH was reduced from ∼100% to 0%, and then, it was increased to ∼100% at a rate of 0.5–1.0% RH·min-1 if LLPS was not observed. If LLPS was observed, the RH was reduced from ∼100% to ∼5–10% lower than the RH at which the two liquid phases merged into one phase, followed by an increase to ∼100% RH at a rate of 0.1–0.5% RH·min-1." Could the authors elaborate how they could confirm an equilibrium state could be achieved for all systems under these RH increasing or decreasing rate?

Page 6, line 17, As shown Table 1, the phase separation was observed at very high RH. Could the authors explain why this happens for the investigated systems?

Also, in addition to the comments on the O/C ratios, could the authors explain why the phase separation does not occur for the SOAs generated from photooxidation of alpha-pinene (e.g. from a molecular insight or perspective)?

Page 9, line 16, "The range of O:C ratio corresponding to the absence of LLPS is wider 17 than that reported by a previous work (Song et al., 2017). The difference could be attributed to the fact that the SOA particles were generated from different types of VOCs." Could the authors elaborate this point a more?

Page 10, line 19, "Moreover, LLPS occurred in the SOA particles at high RH (as high as

$\sim$100%), implying that these results can provide additional information toward the CCN properties of organic particles." Could the authors elaborate what addition information could be gained from the results of this work? How this information would help us to better understand the CCN properties of the particles?

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-19, 2019.