Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-318-RC1, 2020

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Liquid–liquid phase separation in organic particles consisting of α-pinene and β-caryophyllene ozonolysis products and mixtures with commercially-available organic compounds" *by* Young-Chul Song et al.

Nancy Lei (Referee)

lziying@umich.edu

Received and published: 16 April 2020

The authors investigate liquid-liquid phase separation (LLPS) of α-pinene and β-caryophyllene ozonolysis particles, as well as other atmospherically relevant organic particles. The different types of particles with different O:C ratios were studied under different relative humidity conditions. Results show that LLPS occurred to the single component organic particles with O:C smaller than 0.44, and for two-component organic particles with O:C smaller than 0.67. Overall, the results from this study present potential to improve current understanding of atmospherically relevant aerosol parti-

cles and with some revisions as noted below this should be publishable in Atmospheric Chemistry and Physics.

General Comments

The different types of particles with varying O:C show LLPS, the morphology (e.g. core-shell, engulf-coated, inclusions) of particles associated with different O:C ratios should be discussed more in details.

The figures show large size particles ~80 to 100 μm, do smaller size particles (30 μm) present same result in this study?

Author expected the inner phase of the particle mainly consist of water and outer phase consisted mainly of organic. Would it be possible to use spectroscopy to confirm the chemical composition of the particle in different phases (e.g. core and shell)?

The inclusions present in different types of particles, which is very interesting and this occurred frequently in one-component particles. What are these inclusions and how does these inclusions form? Is this something correlated to O:C ratio?

Specific comments

Introduction

Line 57: How relevant the RH ranges (95%-100%) is compare to the atmospheric condition? Do you expect O:C ratio to be lower or higher to allow the LLPS occur at RH (50%-70%)?

Experimental

Line117: Does RH continuous decrease/increase? If so, how do you know if the particles reach to the equilibrium and the optical images is representative for that specific RH?

Line 120: Particle diameter of 30–100 μm seems like a big range, is this aerodynamic diameter or the diameter after impaction? Why does author choose those sizes to study and how relevant comparing the particle sizes in the atmosphere?

Results and Discussion

Line 125: What caused other different types of particles not presenting LLPS?

Line 131: β-caryophyllinic acid and β-nocaryophyllonic acid has same O:C ratio but have very different behavior on LLPS, can author explain why?

Line 133: From Figure1g, the pinic acid at 95.6% RH also present engulf coated morphology, which is not included in discussion.

In Figure1, b, d, and e all exhibit inclusions inside the particle, what are these inclusions and how do they form?

Line 136: if the inner phase is considered as water and the outer phase are organic, do authors have data to support this statement? Most of the organic compounds are hydrophobic and wouldn't the outside organic layer prevent the water evaporation at lower RH?

Figure1f, the pinonaldehyde particles at 94.1% RH show multiple phase, looks like three layers, is that an artifact from microcopy image or that is real?

Figure 3, how many particles have been examined for each point? Do these particles has similar size? Previous study shows the size-dependent LLPS in atmospheric systems, which suggest smaller particles are likely present homogenous and large particles are likely to present LLPS. Could different sizes of particles in this study be a factor affect the results.

Line 185: It is very interesting to see the different mixture particles present LLPS at different RH, especially these two-component particles show LLPS at much lower RH. Can author explain what cause this? Is this can be triggered by high O:C ratio or large molecular weight of mixture particles?

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-318, 2020.