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Interactive comment on "Cloud droplet activation of secondary organic aerosol is mainly controlled by molecular weight, not water solubility" *by* Jian Wang et al.

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Cloud droplet activation of organic compounds depends on various parameters such as solubility, molecular weight, density, and surface activity. A large number of studies have investigated the role of different parameters in cloud droplet activation using pure compounds or simple mixtures. However, since atmospheric organic aerosols are comprised of numerous unknown compounds, it remains to be challenging to understand controlling parameters in cloud droplet activation of organic aerosol. Empirical relationships between the hygroscopicity parameter kappa and O/C have been widely used to infer the effects of aging of organic aerosol on CCN activity. However, underlying mechanisms of the relationship remain uncertain. In this paper, they performed smog chamber experiments to study the role of solubility in cloud droplet activation.

In their experiments, size-selected ammonium sulfate particles were introduced into a smog chamber and varying amount of SOA was condensed according to different residence time distribution in the steady-state chamber. By analyzing the hygroscopicity parameter kappa at different diameter (and supersaturation), they evaluated the dependence of apparent organic kappa as a function of ammonium sulfate (AS) fraction that affects the amount of liquid water at the point of activation. They found that for the majority of chamber-generated SOA (except for beta-caryophyllene SOA), there was no significant dependence of apparent kappa of organics on AS fraction, suggesting that solubility-limitation is largely absent in those SOA investigated in this study. They further performed model calculations to show that apparent linear dependence of kappa of organics and O/C is primarily due to molecular weight. The experiments were carefully performed and the paper is well written. However, I would like to see more careful citations of literature to put this study into a better context. The following are specific comments.

1) The introduction gives an impression that previous study assumed solubility of SOA compounds is the only controlling parameter (although it may not be the authors' intent). Jimenez et al. (2009), Riipinen et al. (2015), and Nakao (2017) are cited as examples of studies that hypothesized that increase in solubility leads to increase in kappa (p.4 L9-19).

a. I believe Jimenez et al. only showed a correlation between kappa and O/C without proposing a mechanism.

b. Riipinen et al. showed that distribution of solubility in the range of 0.1 – 100 g L-1 (or 1e-4 to 1e-1 in volume scale if a unit density is assumed) is critical for capturing the CCN activation. Riipinen et al. recognized that some SOA are sufficiently soluble: "…the observations of the close-to-complete dissolution of SOA at activation (Huff Hartz et al., 2005; Engelhart et al., 2008) indicate that the majority of the material in the studied SOA mixtures had solubilities larger than 10 g L-1."

c. Nakao investigated the combined role of solubility and molar volume through the 2D-VBS framework (C* vs O/C): “The model calculation provides a plausible explanation on why O/C can have an impact on k. The mechanism is likely to be not just the solubility (Kuwata et al. 2013; Riipinen et al. 2015), but a combination of the solubility and volatility that determines the molecular size of dissolved solutes.”

Therefore, both Riipinen et al. and Nakao recognized that solubility plays a role only up to a certain threshold that depends on solubility distributions. I believe the value of this study is that it experimentally showed many of SOA constituents are past that threshold.

2) Kuwata et al. (2013) is cited as an example of studies on pure species, but they did also investigate alpha-pinene SOA and isoprene SOA. Isoprene SOA was found to be highly soluble. Alpha-pinene SOA was predicted to be slightly soluble based on O/C alone but observed to be highly soluble. They hypothesized that the amorphous state of organics led to continuous water uptake without deliquescence. More discussion is needed in assessing the consistency with Kuwata et al. (2013).

3) Consistency with the solubility parameterization developed by Kuwata et al. (2013), lnC = 20*[(O/C)ˆ0.402 – 1], needs to be evaluated. Can the observation be reproduced using measured O/C values in this study? For instance, using beta-caryophyllene’s measured O/C = 0.33, C is calculated to be 7.5e-4. Based on Figure S7, it seems the wider distribution case is consistent with the experiment. A discussion along this line would be helpful.

4) There can be a number of exceptions to the title. More careful clarification of potential exceptions should be discussed, or the title needs to be reconsidered. Within their experiments, beta-caryophyllene SOA is controlled by solubility up to AS 15%. They use toluene as an example of the major precursors of anthropogenic SOA. I would expect SOA produced from large alkanes (linear, branched, cyclic) potentially important in the urban atmosphere (Gentner et al. 2017) would be solubility limited. It remains

unknown whether SOA formed from larger aromatic hydrocarbons such as trimethylbenzene, naphthalene, and methylnaphthalenes are solubility-limited.

5) What are the potential impacts of nitrogen and sulfur-containing organics? Suda et al. showed that nitrates are less CCN active (Suda et al. 2014).

References

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