We thank the referees for their constructive comments. Please find below detailed responses to each comment or question, including notations of improvements to the manuscript. Referee comments are in blue fonts. Changes to the text are highlighted in yellow.

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.

We thank the reviewer for the comment. We have revised the introduction accordingly:

For single component aerosols and simple mixtures, the effect of increasing oxidation level on water solubility and in turn hygroscopicity is well established (Petters et al., 2009;Petters et al., 2016;Rickards et al., 2013;Kuwata et al., 2013). Water solubility is defined as the amount solute that can dissolve in water at saturation, corresponding to solid/aqueous solution equilibrium. Miscibility is related to solubility and refers to the ability of two liquid phases to mix. Miscibility limitation leads to liquid-liquid phase separation (LLPS). If LLPS occurs under supersaturated conditions, miscibility can exert similar control on droplet activation as water solubility (Petters et al., 2006;Petters et al., 2016), and is further discussed in a later section. Kuwata et al. (2013) shows that for single organic compounds, water solubility generally increases with O:C. One plausible explanation is that as organic molecules become increasingly oxidized (i.e., increased O:C ratio), they are more polar, leading to increased water solubility. As O:C increases from near zero, the regime for the activation of single organic compound transitions from insoluble, to slightly soluble, then to highly soluble, leading to a general increasing trend of hygroscopicity parameter (Kuwata et al., 2013). In the highly soluble regime, the hygroscopicity parameter is no longer limited by water solubility, but controlled mainly by molecular weight instead (e.g., Kuwata et al., 2013). Nakao (2017) modeled the hygroscopicity of secondary organic species, and the results suggest the increase of organic hygroscopicity with O:C is likely due to a combination of both increased water solubility and variation in volatility that determines the molecular size of dissolved solute. Riipinen et al. (2015) examined cloud activation of multicomponent organic particles theoretically and suggested that the organic hygroscopicity

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).

We have added the comparison with κ_{org} of isoprene and α-pinene SOA reported by Kuwata et al. (2013). In addition, we now included the reference of Kuwata et al. (2013) when discussing the amorphous state of organics. The texts are changed to:

3)   Consistency with the solubility parameterization developed by Kuwata et al. (2013), lnC = 20*[(O/C)^0.402 – 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.

We thank the reviewer for this suggestion. Based on the O:C value of 0.33, the solubility of β-caryophyllene SOA is estimated as 7.5e-4 (Kuwata et al. 2013). The measured variation of κ_{org}

with x_{AS} β-caryophyllene SOA is broadly consistent with the simulated trend for mixtures with a distribution of water solubilities shown in Fig. S7. This discussion is included in the revision.

"It is also worth noting that based on the O:C value of 0.33, the solubility of β-caryophyllene SOA is estimated as 7.5×10^{-4} (Kuwata et al. 2013). The measured variation of κ_{org} with x_{AS} for β-caryophyllene SOA is broadly consistent with the simulated trend for mixtures with a distribution of water solubilities shown in Fig. S7."

4)   There can be a number of exceptions to the title. More careful clarification of poten- tial 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 ex- pect 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 trimethyl- benzene, naphthalene, and methylnaphthalenes are solubility-limited.

We agree that there can be exceptions, and we have included the following discussion in the revision.

We note there can be exceptions to this finding. Whereas on the global scale, SOA is dominated by those formed from isoprene, monoterpene, and small aromatic hydrocarbons such as toluene, SOA produced from large alkanes and aromatics are potentially important in the urban atmosphere (Gentner et al. 2017), and their droplet activation may be limited by organic water solubility, especially when the volume fraction of inorganics in the particles is low.

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).

The experiments were carried out in a range of NO concentrations. We expect the organic nitrates formed in these experiments are broadly representative of those in the ambient. Whereas organic nitrates are generally less CCN active, results from our experiments indicate that nearly all organics are dissolved at the point of activation. This may be because (1) other functional groups present in the same organic nitrate molecules overcame the solubility/miscibility limitations and/or (2) the presence of water-soluble organics facilitates the solvation of the less water-soluble species such as organic nitrates. We expect that production of sulfur-containing organics was suppressed in our experiments due to the chamber conditions (i.e., dry ammonium sulfate seeds were used, and the seeds were fully neutralized). We are not aware of any work on

the hygroscopicity of typical sulfur-containing secondary organic species. We speculate that organosulfates are more polar compounds and their droplet activation is probably not limited by water solubility.

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