We thank the referee for the thorough review and 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.

Referees' comments:

Referee #1:

Review of

Cloud droplet activation of secondary organic aerosol is mainly controlled by molecular weight, not water solubility

Wang, Shilling, Liu et al. showcase a series of experiments measuring organic aerosol hygroscopicity expressed as κCCN, org using a condensation particle counter and a CCN counter. Their chamber experiments span common SOA precursors with varying inorganic (ammonium sulphate) to organic volume ratios. To explain the experimental results, the authors use previously published relationships for volatility to molecular weight and κCCN, org to molecular weight. They presented an analysis of the major factors affecting the hygroscopicity parameter of the organic aerosol formed, assuming complete dissolution at high relative humidity (RH). The molecular weight of the organic compounds is highlighted as a major factor in determining the apparent hygroscopicity parameter at high RH.

This work is well within the scope of Atmospheric Chemistry and Physics, and is of general interest to the community and discusses new findings. The paper is based on solid experimental work which is largely described in the Supplementary Information, some of which is suggested to be moved into the main manuscript. However, in this referee's opinion, the presentation of the findings and the logic of the explanations reveal a lack of clarity and certain flaws in the argumentation that should (and can) be addressed. Therefore, the manuscript requires major revisions and additional supporting evidence for the assumptions made. More clarity is especially needed for the definition and use of the terms 'water solubility' and 'hygroscopicity' as pointed out below. The organic hygroscopicity addressed in this paper specifically targets the high relative humidity or supersaturated regime. There needs to be an increased specificity on where, at approximately infinite dilution, the presented modeling results and the provided interpretation apply. The paper also needs more specificity in the use and validity of Eq. (2). General and specific comments are detailed in the following.

General comments

1. Clarity on Hygroscopicity Needed:

Hygroscopicity is a nebulous term which needs to be defined. Hygroscopicity can be defined as the mass of water per mass of solute at a given RH or water activity or the equivalent ratios using moles or volumes – or in a different way. Hygroscopicity is not equal to κCCN, org, but the authors implicitly assume just that (page 3 line 19) and elsewhere in the manuscript. Introducing and defining κ as a hygroscopicity parameter should be added to the introduction. Moreover, κ is not a material constant, since in reality, it may depend on the relative humidity range. For example, changes in κ as a function of RH for different SOA have been shown, among others, by Rastak et al. (2017).

The authors need to specify that their measurements are κ at CCN activation (i.e., κCCN, org) and do not directly translate to subsaturated hygroscopic growth factors (κHGF, org). Therefore, the papers title and main conclusion do not apply to subsaturated RH conditions. From the context of the measurements, I can infer that the discussion is on κCCN, org, but the reader should not have to do that.

The hygroscopicity parameter κ is now introduced in the introduction section. We also clarified that κ derived from the measurements represent values at droplet activation and may be different from those associated with hygroscopic growth under sub-saturated conditions.

2. Clarity on Solubility Needed:

Water-solubility is not currently defined in the introduction. It can either be the solute saturation point (limit of solubility) of a solid-liquid equilibrium or it could refer to the transition from liquid-liquid equilibrium (miscibility limitation as mentioned in the supplementary information) to a single aqueous phase. The solubility values of those variants likely differ and, according to equilibrium thermodynamics, depend on the other species in an aqueous solution. Providing a quantitative definition earlier in the paper (currently one needs to read until page 9 to find one), would also help distinguishing the differences between water-solubility (C), and apparent κCCN, org. The introduction would benefit from having the general definition of hygroscopicity early on. This change will make it clear to the reader that organic hygroscopicity may change over the full water activity range, complete dissolution occurs at some RH value, depending on the other mixture species, and κCCN, org is at water supersaturation RH.

Water-solubility is now defined in the introduction.

"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…"

We also clarified that hygroscopicity parameter κ may vary with relative humidity. The κ values examined in this study are at droplet activation under supersaturated conditions, and they may be different from those under sub-saturated conditions associated with particle hygroscopic growth.

On a related point, in the SI (section 6, page 12, line 20) the authors assumed miscibility transition of 0.5 O:C, does this include a salting-out effect or does it assume only water-organics phase separation? In this discussion on miscibility, the observational work performed by Renbaum-Wolff et al. (2016) on SOA liquid–liquid phase separation at high relative humidities should be included.

We referenced Renbaum-Wolff et al. (2016) in the discussion of miscibility and LLPS. The assumed transition of 0.5 O:C does not include the salting out effect. Since functional group composition is a more relevant measure than O:C, we also rephrased the sentence as follows:

Model calculations and experimental data suggest that adding a few hydroxyl and/or carboxylic acid groups is enough to remove miscibility limitations (Bertram et al., 2011; Petters et al., 2016) for pure organic compounds at water activity corresponding to supersaturated conditions, consistent with the results from our experiments.

Could the authors add some discussion on how the interpretation would change (or not) if the particles were phase-separated (water and organic-rich phases) during the CCN measurement? Especially, given the context of recently published work by Ovadnevaite et al. (2017) on phase-separated CCN activation.

This relevant discussion of the LLPS is already included in the supplementary information:

Miscibility is related to solubility and refers to the ability of two liquid phases to mix. Substances that form two liquid phases are immiscible. If a miscibility transition occurs in the supersaturated regime, miscibility limitations exert a similar control over critical supersaturation as solubility (Petters et al., 2006; Petters et al., 2016), i.e., the effective hygroscopicity κ_{org} observed from

CCN activation is reduced at lower net water contents (smaller size or less hygroscopic material present). Distinguishing between miscibility and solubility in CCN experiments is not possible because the manifestation of the effect is identical.

The above discussion is now moved to main text following the suggestion of referee.

…

Ovadnevaite et al. (2017) suggested that LLPS leads to complete or partial engulfing of a hygroscopic particle core by a hydrophobic organic-rich phase. This results in substantial decrease of surface tension without a reduction of the Raoult's effect, leading to higher CCN activity. Simulations based on this LLPS model suggest the effect of reduced surface tension on CCN activity is more pronounced for ultrafine particles, and the effect is much weaker for particles with larger diameters studied here. One prediction of the model is that particles with different organics to sulfate mass ratios (i.e. sulfate volume fraction) exhibit similar effective hygroscopicity parameter. Such behavior is explained by that a decrease in sulfate content, and therefore the Raoult's effect, is compensated for by a prolonged surface tension lowering (to larger diameters during droplet activation) due to a larger relative amount of organics (Ovadnevaite et al., 2017). This trend is very different from the results of SOA examined in this study, which show a nearly constant κ_{org} therefore lower effective hygroscopicity parameter for particles with high organics to sulfate mass ratio.

3. Community claims need support and clarification:

The statement on page 4 line 9-12 needs a justification, as well as a similar statement in the abstract (page 2 line 13-15): “Earlier studies showed organic hygroscopicity increases nearly linearly with oxidation level. Such increase of hygroscopicity is conventionally attributed to higher water solubility for more oxidized organics.”. Claiming that an increase in polarity leads to an increased solubility and then higher κCCN, org is a “long held hypothesis” needs to be corroborated by citation of relevant references. At least this referee does not share the opinion that the higher water-solubility was the main hypothesis in “the community”. Jimenez et al. (2015) showed a relation between O:C and κCCN, org , whereas Riipinen et al. (2015) showed a relation between solubility and κCCN, org. There is no citation provided linking O:C and solubility. On page 4 line 14-16, the citation Kuwata et al. (2013) does show a loose trend of O:C with κCCN, org, but the author's statement neglects the fact that molar mass as well as density are changing with O:C. In the same sentence, the Petters et al. (2009) citation is confusing; as

Petters et al. (2009) does not discuss the oxidation level, but discusses molar volume. If the authors are reinterpreting the results of Petters et al., (2009) and Petters et al., (2016) through an O:C lens and neglecting the covarying molar mass that should be stated.

An increase in organic polarity could increase water affinity, but to change solubility, the molecular size and, for solid-liquid solubility, the crystal structure, is also playing an important role (e.g. consider the well-known odd-even alteration with carbon atom number in the water-solubilities of dicarboxylic acids; e.g. Bilde et al., 2003). Changes to solubility would depend on the combination of molecular size, molecular interactions in solution, which would include polarity, and crystal structure (unless liquid-liquid miscibility is meant).

We thank the referee for this comment. The text is revised to:

Several studies show that for ambient organic aerosols and laboratory generated secondary organic aerosols (SOA), organic hygroscopicity parameter increases nearly linearly with the atomic O:C ratio (e.g., Jimenez et al., 2009;Lambe et al., 2011), a parameter commonly used to quantify oxidation level of organics. 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 parameter is strongly influenced by the distributions of species with water solubility between 0.1 and 100 g L^{-1}, i.e., the range for slightly soluble species. Whereas all the above studies suggest that the hygroscopicity parameter is strongly influenced by water solubility of slightly soluble organics, for ambient organic aerosols and laboratory generated secondary organic aerosols, the influence of water solubility on organic hygroscopicity has not been systematically examined experimentally, and the underlying mechanism for the increasing hygroscopicity with increasing O:C remains unclear.

4. More Support for Eq. 2:

The applicability and usability range of Eq. 2 needs to be discussed. This discussion is even more warranted since Sullivan et al. (2009) used Eq. 2 for mineral dust, not organic molecules.

Starting with the definition of κ from Petters and Kreidenweis (2007) (their Eq. 2), one can derive Eq. 2 shown in this manuscript as a limiting case. Note, one must substitute water activity for the water mole fraction and activity coefficient. In doing this, one can show the assumption of i = 1 is valid only under conditions of high dilution in water, when the activity coefficient and the mole fraction of water each approach one. Due to the activity coefficient being depended on the organic's O:C and molecular structure, it is likewise true that i is also dependent on those same properties in non-dilute cases. Such a derivation and pertinent discussion would make the origin and limited range of application of Eq. (2) clear. This is important since much of the argumentation about the role of molecular weight (i.e. molar mass) is built based on assuming Eq. (2) to be valid or at least to be a valid approximation.

We agree and clarified that Eq. 2 describes intrinsic hygroscopicity parameter at droplet activation. We also included the following discussion in the text:

The assumption of *i*=1 is valid only under conditions of high dilution in water, such as at droplet activation under supersaturated conditions.

The authors should add more to the discussion of the previous uses of Eq. 2. In the paper's current state, the reader could have the impression that the control of κCCN, org by Morg is a brand new discovery. That impression would be incorrect when we consider the following quotation:

"For highly soluble particles, CCN activity is dominantly controlled by molar volume, which is determined by both ρorg and 1/Morg (Eq. 5) (Petters et al., 2009a). Kuwata et al. (2012) developed a method to predict ρorg using O : C and H : C ratios. According to the method, ρorg is higher for highly oxygenated compounds. Even so, the range in ρorg values for O : C>0.6 from 1100 to 1650 kgm−3 is not sufficient to explain the variability observed for κ for highly soluble materials. Furthermore, the ρorg values of carbohydrates and synthetic polymers do not vary significantly yet the variations in κ is significant (cf. Supplement). The implication is that the dominant regulator of CCN activity is Morg for highly soluble particles."

-Kuwata et al., 2013

The authors' experimental work does extend the above result by Kuwata et al. (2013) to SOA systems. But, the authors did not place their results within the context of the previously published work, which similarly uses Eq. 2 with organic compounds.

The following text is in the original text:

In the absence of solubility limitation, κ_{org} is essentially the same as $\kappa_{org,intr,}$ and is controlled mainly by molar volume (Eq. 2). This has been shown by earlier studies for pure organic compounds with sufficient solubility (Kuwata et al., 2013; Petters et al., 2009; Petters et al., 2016).

We also added the following sentences in the introduction:

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 (Kuwata et al., 2013).

Also, it is clear that assuming density of organics to remain constant upon oxidation is only an approximation, since with increases in O:C and molar mass, density may also vary by up to ~ 40 %.

Following discussion is now included:

As shown by Kuwata et al. (2013), the variation of organic density with O:C is relatively weak and can't explain the variation in κ_{org}. For example, the density of the SOA examined in this study varied by approximately 30% from 1.12 to 1.49 g cm^{-3}, whereas the variation of κ_{org} is more than a factor of 2, from 0.07 to 0.16. Given the relatively narrow range of SOA density, the increasing κ_{org} with O:C suggests a negative correlation between the molecular weight and O:C for secondary organic species.

5. Discussion in the Supplemental Information:

There are a couple of topics that were fully discussed in the Supplemental Information that would better serve the reader in the main text. I suggest moving some of SI part 6 on liquid-liquid phase separation into the main text either in the introduction or section 3.3 – 3.4. In addition, the authors could add some of the Monte Carlo error assessment to the main text as well.

The simulations of the solubility distributions in the SI are quite informative. I suggest bringing some of that information into the main text or into Figure 2.

We thank the reviewer for this suggestion. We moved the discussion in SI part 6 on LLPS into section 3.3 of the main text. The section on uncertainty assessment is also moved to the main text. We left the simulations of the solubility distributions in the SI because first adding more information to Figure 2 likely will make it too busy. Second, the simulations based on a distribution of solubilities show similar trend of κ_{org} with increasing x_{AS} exhibited in the Figure 2 of the main text.

Specific comments

1) Page 2 Lines 7, 12, 18, 19: Is it hygroscopicity or is it hygroscopicity parameter?

We think it is appropriate to use “hygroscopicity” here.

2) Page 2 Line 12: Add the following…not limited by solubility at CCN activation RH, but …

We have clarified this as following:

“Therefore, for droplet activation, the organic hygroscopicity is not limited by solubility, but…”

3) Page 3 Line 19: Change hygroscopicities to hygroscopicity parameters.

Done.

4) Page 4 Line 7: Change hygroscopicity to hygroscopicity parameter.

Done.

5) Page 5 Line 1: Change atmospheric to atmospherically.

Thank you. Done.

6) Page 5 Line 3: Add the following…organic hygroscopicity at CCN activation RH is not limited.

We have clarified this as following:

“Therefore, for droplet activation, the organic hygroscopicity is not limited by solubility, but is…”

7) Page 5 Line 5: This statement “hypothesized earlier” needs a citation.

Please see response to general comment #3 above. We removed the “hypothesize earlier”. The text is changed to:

“Instead of increased solubility, the increase of the organic hygroscopicity with O:C is largely because…”

8) Page 5 line 4-6 as well as Page 21 line 16-20: These statements led to some confusion as to whether the authors are referring to the size of the precursor molecule or the oxidation products. In the same vein does the title refer to the precursor or the oxidation products?

It is the size of oxidation products (i.e., secondary organic aerosol species). On the other hand, the size of precursor and the oxidation products are related. As described in the text:

“SOA formed from smaller volatile organic compound (VOC) molecules tend to have lower average molecular weight…”

9) Page 6 Line 16: Add specificity…are the dry or water free volume fractions…

Done.

10) Page 6 Line 17: Add the following…hygroscopicityies parameters of the organics mixtures and…

Done

11) Page 7 Line 1: Add specificity to xas, is this dry or wet mixture and under what RH.

Please see response to comment # 9. It is dry volume fraction of ammonium sulfate.

12) Page 7 Line 2: What is the chamber RH?

The chamber RH is detailed in Table S1. For most of the experiments, the chamber RH was ~ 50%. However, the initial ammonium sulfate seeds and particles exiting the chamber passed through diffusion dryers and were classified under dry conditions. We have modified the sentence to:

"…where D_{p0} and D_p are the dry diameters of initial ammonium sulfate seeds (50 nm) and size-selected particles exiting the chamber, respectively."

13) Page 7 Line 3: Add the surface tension that was assumed when deriving κCCN, org.

It is stated in the text:

"the fraction of particles serving as CCN as a function of supersaturation, which was then used to derive particle hygroscopicity κ based on κ-Köhler theory…"

"The above analysis is based on κ-Köhler theory, in which the hygroscopic parameter κ is derived assuming the surface tension of water for activating droplets."

14) Page 11 Line 19: Add…miscibility limitation at supersaturated RH.

Done.

15) Page 13 Line 20: "same behavior as pure components" That statement is incorrect due to mutual solubility, see Marcolli et al., (2004). The same applies to the statement on line 21 – 22; it is not the mass that matters, but the mixing.

This refers to the 2nd case described on page 6309 in Riipinen et al., (2015), i.e., $\gamma_i Y_{i,wet}=1$. As water condenses and particles grow, before species *i* is completely dissolved, the chemical potential of species *i* in the undissolved phase (e.g., organic phase) is same as that in the aqueous phase. If all species are non-interacting solids, then the chemical potential of species *i* in the undissolved phase is the same as pure compound, and the dissolution behavior is the same as if no other species are present (i.e., non-interacting). If the species form a solution in the undissolved phase, the mixing reduces the chemical potential of species in the undissolved phase, which translates into lower chemical potential of species in the aqueous phase (i.e., dissolved phase) and therefore reduced effective water solubility.

We clarified this in the text as follows:

For example, in a mixture of non-interacting solids, because the chemical potential of undissolved components is the same as pure compounds, the components behave independently of one another (i.e., solubility not affected by the presence of other solids, and the dissolution behavior of each species is the same as their behavior as pure components.). This is in essence the 2nd case described in Riipinen et al. (2015) (i.e., $\gamma_i Y_{i,wet}=1$).

16) Page 14 line 2: The solubility values of the pure components may not apply, but potential miscibility limitations due to liquid-liquid phase separation still applies to amorphous physical states. Please discuss. Also, by “amorphous organic state” likely “amorphous physical state” or phase state is meant.

We have clarified this in the text and changed “amorphous organic state” to “amorphous phase state”.

“We note that potential miscibility limitations due to LLPS could still apply to an amorphous phase state.”

17) Page 14 line 6 and line 8: Add comma after …particle, and after Fig. 2,

Done.

18) Page 14, line 22: A narrow density is true, but molar volume (Morg/ρorg) is used in Eq. 2, and molar volume does vary. A statement on the relative impact that Morg and ρorg have on molar volume would provide a stronger justification here.

A following statement is included:

As shown by Kuwata et al. (2013), the variation of organic density with O:C is relatively weak and can't explain the variation in κ_{org}. For example, the density of the SOA examined in this study varied by approximately 30% from 1.12 to 1.49 g cm^{-3}, whereas the variation of κ_{org} is more than a factor of 2, from 0.07 to 0.16.

19) Page 17 line 8: "non-ideal solution effects are negligible" Are they negligible? A justification is needed.

The assumption is used to estimate the molecular weight, which is around 130-160 g mol^{-1}. Given the estimated range of molecular weight and that we are discussing droplet activation (i.e., water dilution is high), this assumption is appropriate. We have rephrased the sentence to:

The above estimate assumes organics are completely dissolved (i.e., $\kappa_{org}=\kappa_{org,\ intr}$) and that non-ideal solution effects are negligible at the point of activation.

20) Page 18 line 3: Add…due to Eq. 2, their lower…

We rephrased the sentence to:

In essence, to have sufficiently low volatility and contribute substantially to SOA, smaller organic molecules, which have higher κ_{org} values due to their lower molecular weight based on Eq. 2, need to be more oxidized (i.e., higher O:C).

21) Page 18 line 6: Change hygroscopicity to hygroscopicity parameter.

Done.

22) Page 18 Eq 3: Use log10

Done.

23) Page 18 line 12: Jargon used; specify that this is the number of atoms.

Done

24) Page 19 Line 11: Add…leading to a lower hygroscopicity parameter via Eq. 2.

Done.

25) Page 19 line 13: Change volatility to volatilities.

Done.

26) Page 19 line 14: Use log10

Done.

27) Page 19 line 21 & 20: Change hygroscopicity to hygroscopicity parameter.

Done.

28) Page 19 line 21: Add…during the recent…

Done.

29) Page 20 line 5: Add “at CCN activation, i.e. supersaturated conditions.”

Done.

30) Page 21 line 5: Change oxidize to oxidized

Done.

31) Page 21 line 11: Change hygroscopicity to hygroscopicity parameter

Done.

32) Page 21 line 13: Add… at CCN activation is controlled by…

Done.

33) Page 21 line 14: “mainly by the molecular weight of the organic species” This is the case for SOA due to the gas–particle partitioning affecting O:C and molar mass simultaneously. That concept should be added.

The concept is described in the manuscript as following:

“…due to gas-particle partitioning, SOA formed from smaller parent VOC molecules tend to have lower molecular weight and are more oxidized…”

34) Page 25 caption: …50 nm dry diameter ammonium…

…hygroscopicity parameter at high RH? is shown as a function of dry particle sulfate…

Done.

35) Figure 4 needs clarification in the x-axis label or caption that it is the carbon number of the SOA products.

Clarified.

36) The paper’s title alongside Figure 5 is confusing and needs clarification. Given the title of the paper, why is the exact opposite shown in Figure 5, i.e., increasing O:C with increasing κCCN, org. Why not plot molecular weight vs. κCCN, org? Then the spread in the data would be the O:C dependence, which would reflect the main point of the paper.

We thank the referee for this suggestion. However, molecular weight of organic species was not directly measured in these experiments, and Lambe et al (2011) did not report the relationship between κ_{org} and molecular weight (for the same reason). The main purpose of the figure is to demonstrate that the model, which is based on the absence of solubility limitation of κ_{org}, can successfully produce the observed relationship between κ_{org} and O:C.

References

Bilde, M., Svenningsson, B., Mønster, J. and Rosenørn, T.: Even-odd alternation of evaporation rates and vapor pressures of C3-C9 dicarboxylic acid aerosols, Environ. Sci. Technol., doi:10.1021/es0201810, 2003.

Kuwata, M., Shao, W., Lebouteiller, R. and Martin, S. T.: Classifying organic materials by oxygen-to-carbon elemental ratio to predict the activation regime of Cloud Condensation Nuclei (CCN), Atmos. Chem. Phys., 13(10), 5309–5324, doi:10.5194/acp- 13-5309-2013, 2013.

Marcolli, C., Luo, B. and Peter, T.: Mixing of the Organic Aerosol Fractions: Liquids as the Thermodynamically Stable Phases, J. Phys. Chem. A, doi:10.1021/jp036080l, 2004.

Ovadnevaite, J., Zuend, A., Laaksonen, A., Sanchez, K. J., Roberts, G., Ceburnis, D., Decesari, S., Rinaldi, M., Hodas, N., Facchini, M. C., Seinfeld, J. H. and O' Dowd, C.: Surface tension prevails over solute effect in organic-influenced cloud droplet activation, Nature, 546(7660), 637–641, doi:10.1038/nature22806, 2017.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity – Part 2: Including solubility, Atmos.

Chem. Phys., 7(8), 1961–1971, doi:10.5194/acp-7-1961-2007, 2007.

Petters, M. D., Kreidenweis, S. M., Prenni, A. J., Sullivan, R. C., Carrico, C. M., Koehler,

K. A. and Ziemann, P. J.: Role of molecular size in cloud droplet activation, Geophys. Res. Lett., 36(22), 1–5, doi:10.1029/2009GL040131, 2009.

Petters, M. D., Kreidenweis, S. M. and Ziemann, P. J.: Prediction of cloud condensation nuclei activity for organic compounds using functional group contribution methods, Geosci. Model Dev., 9(1), 111–124, doi:10.5194/gmd-9-111-2016, 2016.

Rastak, N., Pajunoja, A., Acosta Navarro, J. C., Ma, J., Song, M., Partridge, D. G., Kirkevåg, A., Leong, Y., Hu, W. W., Taylor, N. F., Lambe, A., Cerully, K., Bougiatioti, A., Liu, P., Krejci, R., Petäjä, T., Percival, C., Davidovits, P., Worsnop, D. R., Ekman, A.

M. L., Nenes, A., Martin, S., Jimenez, J. L., Collins, D. R., Topping, D. O., Bertram, A. K., Zuend, A., Virtanen, A. and Riipinen, I.: Microphysical explanation of the RH- dependent water affinity of biogenic organic aerosol and its importance for climate, Geophys. Res. Lett., 44(10), 5167–5177, doi:10.1002/2017GL073056, 2017.

Renbaum-Wolff, L., Song, M., Marcolli, C., Zhang, Y., Liu, P. F., Grayson, J. W., Geiger,

F. M., Martin, S. T. and Bertram, A. K.: Observations and implications of liquid–liquid phase separation at high relative humidities in secondary organic material produced by α-pinene ozonolysis without inorganic salts, Atmos. Chem. Phys., 16(12), 7969–7979, doi:10.5194/acp-16-7969-2016, 2016.

Riipinen, I., Rastak, N. and Pandis, S. N.: Connecting the solubility and CCN activation of complex organic aerosols: A theoretical study using solubility distributions, Atmos. Chem. Phys., 15(11), 6305–6322, doi:10.5194/acp-15-6305-2015, 2015.

Sullivan, R. C., Moore, M. J. K., Petters, M. D., Kreidenweis, S. M., Roberts, G. C. and Prather, K. A.: Effect of chemical mixing state on the hygroscopicity and cloud nucleation properties of calcium mineral dust particles, Atmos. Chem. Phys., 9(10), 3303–3316, doi:10.5194/acp-9-3303-2009, 2009.