

Interactive comment on “Uncertainty in aerosol hygroscopicity resulting from semi-volatile organic compounds” by Olivia Goulden et al.

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Q: One pressing issue is that results shown in the paper seems inconsistent. The paper first showed that the inclusion of SVOCs leads to a lower hygroscopicity (κ), which implies that neglecting SVOCs would overestimate κ , and thus CCN number as well. However, the paper later showed the effective κ is higher when including SVOCs. This means that neglecting SVOCs would underestimate κ , and also CCN number. These two conclusions seems contrast each other. Can the authors clarify this? A: The inclusion of SVOCs increases the size of particles as well as changing the chemical composition and so the hygroscopicity has a more complicated relationship to CCN concentration as would be the case for involatile particles. Ignoring composition, larger particles activate at lower supersaturations and one may expect more CCN as a

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result of the increased size of particles with the SVOCs. The hygroscopicity, however, is also inversely proportional to the critical diameter cubed (equation 1). Hence, if the particles that activate are larger, the resulting hygroscopicity is smaller.

Q: Page 3, last paragraph. The introduction of the three single-parameter measures of the hygroscopicity seems abrupt, causing confusion without further explanations. I suggest placing this introduction in the beginning of section 4: methodology including the effects of SVOCs A: This paragraph has been moved to the methodology section, as suggested.

Q: Before moving to the main body of paper, please briefly lay out the structure of the following content, telling readers what they would expect in the coming paper. A: This has been added at the start of the final paragraph of the introduction.

Q: Reorganize the main body of the paper. The current section form starts from methodology, then to results, and jump back to methodology and results, which I think is not fluent. Two ways to fix it. 1) put all methodology parts into one section, followed by the results section; 2) Combine the section 2 and section 3 into one part as for involatile aerosol with section 2 and 3 as sub-sections, and combine the section 4 and 5 into the other part as for including the effects of SVOCs. A: The sections on the involatile aerosol (method and results) was presented first to allow the reader to understand the Monte-Carlo methodology when applied to the simpler and familiar definition of hygroscopicity. In addition, experimental results are available for comparison in this case. We the go on to describe the addition of the SVOCs and discuss that this more complicated due to the variation in size and chemical composition with RH. There are no experimental results for comparison and so this builds on the current theory and understanding.

Q: Page 2, line 28. How about the recent IPCC results (AR5)? A: Citation has been updated to Myhre et al. (2013)

Q: Page 3, line 69-72. What did the author mean by 'dynamic condensation'? That

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is inconsistent with ‘equilibrium absorptive partitioning’ stated in the beginning of this sentence. A: In dynamic parcel model simulations by Connolly et al. (2014), it was found that the SVOCs approximately reached equilibrium between the vapour and total condensed phase by cloud base. The condensed mass on each individual particle within the polydisperse aerosol, however, was not in equilibrium. Consequently, the total condensed mass was calculated using equilibrium absorptive partitioning theory but this mass is distributed between the different particle sizes within a lognormal mode by changing the median diameter and geometric standard deviation. This change in lognormal parameters is used to “simulate the condensed phase of SVOCs after undergoing dynamic condensation”.

Q: Page 3, line 91. According to the equilibrium absorptive partitioning theory, the primary factors controlling the gas/particle partitioning are the vapor pressure of SVOCs, atmospheric temperature, and the total mass of existing particles, without RH, although RH is relative to the temperature. Can the authors explain more why they particularly chose RH? A: We have used the equilibrium absorptive partitioning model of Barley et al. (2006) that includes condensed water in the mass of the particle phase. The mass of water is controlled by the RH. Near 100% RH the mass of water in the particle phase increases dramatically and this induces significant condensation of SVOCs.

Q: Page 4, line 109-111. This sentence seems odd to me. “Many source of uncertainties” in the first part is logically disconnected to the second part of this sentence. A: The sentence is trying to say that we’re encapsulating the many sources of uncertainty into a single parameter with associated uncertainty. This has been reworded to make it clearer.

Q: Page 5, line 166. A little confusion here. How did the authors obtain the “12%” value? A: The 12% resulted from the method of randomising the size distribution described in the Appendix A (now in the supplement). In the interest of reducing the length of the paper, as requested by both reviewers, this paragraph has been removed

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Q: Page 8, Line 210. "Table 1". Did the authors mean Table 3? A: Yes, this has been corrected in the text.

Q: Page 11, Line 268. Any specific reason that 50% of RH was chosen for the integration of aerosol size distribution? Not 60%? Any effect on the derived hygroscopicity if using different RH? A: Air is often dried to 50% RH before measuring the aerosol size distribution. Figure 7 (Now Figure S7 in the Supplement) shows the effects of using higher RH values with little difference in the hygroscopicity below 95% RH.

Q: Page 11, line 287. Why does it have to be between 0.1 and 0.5? A: These are typical ranges measured for organic fraction of aerosol.

Q: Page 12, line 291. Can the authors remind the readers what the parameterization of Connolly et al. (2014) is? Since it is first introduced in the Introduction Section, which is quite far away from here. A: It is not that easy to briefly describe the parameterisation and a major criticism of the paper was its length. Adequate references to the original parameterisation are included.

Q: Page 13, line 313-315. The smaller uncertainty for k_{SVOC} than k_{nv} is quite surprising, because the uncertainty for k_{SVOC} includes the uncertainty associated with not only the non-volatile particles but also SVOCs volatilities and masteries, while the uncertainty for k_{nv} reflects only the non-volatile particles related uncertainty. Do the authors know why? A: The 10% is for levoglucosan while the other 3 compounds the difference is more like 5%, the text has been changed to reflect this. For ammonium sulphate, sodium chloride and sulphuric acid the hygroscopicity is large and small changes in the size of the particles can make a big difference to the number of CCN. The organics, however, have a hygroscopicity of between 0.18 and 0.27 (based on Table 5), which is "low" regardless of what value it actually takes in this range. Overall, the combination of the involatile constituents and the organic compounds will result in relatively low hygroscopic particles regardless of the particular properties of the organics. Furthermore, the uncertainty in organic mass fraction will have a large effect on

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the chemical composition of the particles, however, this is mitigated by the changes in sizes of the particles due to the SVOCs.

Q: Page 14, line 325. "50% RH" is different from "0%RH" stated on line 323. Which one is right? A: "0%" is a typo and has been corrected to 50%

Q: Page 16, line 368. Shouldn't it be wet aerosol size distribution, because at 70% RH, for example, the aerosol can absorb water? A: The equation for the hygroscopicity, (1), depends on the dry particle size, as defined in Petters and Kreidenweis (2007). In the case with SVOCs, the dry size neglects the condensed water but includes the associated condensed SVOCs at the particular RH.

Q: Figure 7. What are the red + in the top of figure? A: These are the data point that MATLAB deems outliers and are the points that are more than 1.5 times the interquartile range below and above the 25th and 75th percentiles, respectively.

Q: Figure 8. Why the line of 100% shows a different trend than other lines at high logC bins? A: The partitioning of the SVOCs into the condensed phase depends on the condensed water. At higher RH there is more condensed water and therefore more SVOCs in the condensed phase. There is significantly more water condensed at 100% RH than below 95% RH and so the total condensed mass of SVOCs will be significantly higher. By 95% humidity, the organics in the lower 8 volatility bins have all partitioned into the particle phase and so the additional condensed SVOC mass at 100% RH must come from the higher 2 volatility bins.

Q: Section 6. As a large portion of the paper concentrates on the uncertainty of hygroscopicity associated with involatile aerosol size distribution and SVOCs mass and chemical compositions, I think the authors should add the findings about this uncertainty part, which can also echo the title of the paper. A: The focus of this paper on the effect of SVOCs. The involatile section (Section 2) is included to allow some comparison with existing experimental data that, in comparison to when SVOCs are included, is more straight forward to measure. We are therefore presenting a method of including

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SVOCs that does not have the pit-falls associated with measuring SVOCs. Additionally, reviewer 2 felt there was already too much focus on the involatile case. (Point 1)

Q: Section 6. The results are derived from the assumption that SOA is the result of the equilibrium absorptive partitioning of SVOCs, but some experimental results indicate that aerosol particles containing SOA can exist in highly viscous states (e.g., Vaden et al., 2011 PNAS), breaking the equilibrium partitioning. Would the viscous states of particles change the results of this paper? A: This is true but is an active area of research. Some numerical models investigating the effect of diffusion within aerosol particles (Zobrist et al., 2011; Smith et al., 2003) indicate that glassy particles can transition into a liquid phase above about 50% relative humidity. As this paper focuses on relative humidities above 50% the effect of viscosity may not be important.

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