

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

Anonymous Referee #1

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Aerosol hygroscopicity plays a major role in determining the ability of aerosol acting as cloud condensation nuclei (CCN), which has an important effect on cloud and thus climate. Most of current existing cloud droplet activation parameterizations neglect the effect of organic compounds on hygroscopicity, despite of the large amount of organic aerosols in the atmosphere.

Goulden et al. (2017) paper examined the effect of semi-volatile organic compounds (SVOCs) on aerosol hygroscopicity and quantified its uncertainties. The paper concluded that the including SVOCs tend to decrease the aerosol hygroscopicity primarily because of the lower hygroscopicity of SVOCs than those of non-volatile aerosols. The paper also proposed a parameter, called effective hygroscopicity, to account for the effect of SVOCs on cloud droplet number concentrations. The effective hygroscopicity was shown to be higher than the original hygroscopicity of non-volatile aerosols.

C1

For sure, this paper is of great interest to the aerosol/climate community and will help us understand the effect of organic aerosol on cloud formation. Yet, it needs improvement to better present its results and to clarify its ambiguous results.

Major comments

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?

The paper was not well structured, with many sudden jumps between paragraphs and sections, causing me a lot of trouble to follow. Here are some of my suggestions to improve that.

- 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.
- 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.
- 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.

C2

Minor comments

Page 2, line 28. How about the recent IPCC results (AR5)?

Page 3, line 69-72. What did the author mean by 'dynamic condensation'? That is inconsistent with 'equilibrium absorptive partitioning' stated in the beginning of this sentence.

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?

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.

Page 5, line 166. A little confusion here. How did the authors obtain the "12%" value?

Page 8, Line 210. "Table 1". Did the authors mean Table 3?

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?

Page 11, line 287. Why does it have to be between 0.1 and 0.5?

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.

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

C3

authors know why?

Page 14, line 325. "50% RH" is different from "0%RH" stated on line 323. Which one is right?

Page 16, line 368. Shouldn't it be wet aerosol size distribution, because at 70% RH, for example, the aerosol can absorb water?

Figure 7. What are the red + in the top of figure?

Figure 8. Why the line of 100% shows a different trend than other lines at high logC bins?

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.

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?

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C4