Comments by referees are in blue.

Our replies are in black.

Changes to the manuscript are highlighted in red both here and in the revised manuscript.

Reply to referee #2

The paper by Peng et al. addresses gap knowledge of organosulphates hygroscopic properties and CCN activity. Organosulphate hygroscopic properties have not been systematically studied to date and as such paper is a significant contribution to atmospheric science. The paper is generally well written although a moderate revision is needed to meet the publication standard of ACP.

Reply: We would like to thank referee #2 for reviewing our manuscript and recommending it for publication after revision. His/her comments, which helped us largely improve our manuscript, have been carefully addressed in our revision, as detailed below.

1. Line 36. Maintain three significant digits for consistency. Even the third digit of the value is imprecise, because the standard error is changing the second digit.

Reply: Thanks for the suggestion, we have made them consistent in our revised manuscript.

2. Line 42. Quantitative numbers needed to illustrate qualitative terms like "reasonably well", especially in the Abstract.

Reply: In response to this comment, we have added quantitative numbers in the revised manuscript (Line 42-43): "For methyl- and ethyl-OS aerosols, κ_{ccn} values agree reasonably well with those derived from H-TDMA measurements (κ_{gf}) with relative differences being smaller than 25%, whereas κ_{ccn} was found to be ~2.4 times larger than κ_{gf} for octyl-OS, likely due to both solubility limit and surface tension reduction."

3. Line 157. delta GF should have reflected Kelvin effect which was not negligible for 100nm particles. The authors could use kappa method in estimating Kelvin effect against e.g., 300nm particle were Kelvin effect would be immeasurable.

Reply: We did calculations in our previous work, and the Kelvin effect is negligible for 100 nm particles. In the revised manuscript (line 402-403) we have added one sentence for further clarification: "Eq. (6) does not take into account the Kelvin effect as the effect is small for 100 nm particles (Tang et al., 2016)."

4. Figure 3. Is Figure 3 meant for illustration purposes or is the result? It is unnecessary for the former and if for the latter it should be presented in terms of SS_{crit} as a function of D_{crit} along with ammonium sulphate.

Reply: Figure 3 is used for illustration. We fully understand the referee's concern, but we feel that it is necessary for colleagues who are not very familiar with CCN measurements. Therefore, we would like to keep it in the manuscript.

5. Line 195. It is difficult to judge the significance of 11% without uncertainty error bars. Could it be due to physical spatial arrangement of 1mg mass lump?

Reply: In fact, errors bars are included in Figure 4, but they are too small to be visible. Our VSA can easily detect a relative mass change of <1%, and a change of 11% is significant. In addition, physical spatial arrangement would lead to change in morphology but not mass.

6. Line 212. Measurable, not obvious. Nothing is obvious in scientific experiment.

Reply: That is right, and we have corrected it in the revised manuscript (Line 216).

7. Line 214. suggesting the occurrence of ...

Reply: That is right, and we have corrected it in the revised manuscript (Line 218).

8. Line 218. Interestingly, that in this case the authors discount 10-20% increase, contrary to ethyl-OS increase of 11%, mentioned earlier.

Reply: The 11% increase for ethyl-OS is reproducible and reliable. The 10-20% increase for the other six potassium organosulfates was only occasionally observed (only for a few experiments) and not reproducible, probably because these chemicals we synthesized contained significant amounts of impurities and were not homogeneous.

9. Table 1. Maintain three significant digits throughout.

Reply: Thanks for the suggestion, we have made them consistent in our revised manuscript.

10. Figure 5. I believe that a) and b) were split due to methyl and ethyl OS being similar and partly overlapping, but it is exactly for the same reason they should be on the same graph and if a single graph was bigger it would exhibit those differences clearly.

Reply: As suggested by referee #2, the two panels have been merged into one panel in the revised manuscript.

11. Line 254. DMA sizing precision is at best 5% (Wiedensohler et al. 2012, AMT) and, consequently, 7% of the two DMAs. Clearly 8-9% difference can be attributed to sizing uncertainty of different HTDMA systems.

Reply: Thanks for the suggestion, we have modified this sentence in the revised manuscript (Line 257-258): "As DMA sizing typically has a relative uncertainty of 5-7% (Wiedensohler et al., 2012), our measured GFs..."

12. Line 297. That is understandable as the bulk material is present in large lump of mass (1mg is huge when compared to single particle). In order for VSA to represent microscopic particles, one should use tiny amount of substance spread as e.g., 100nm film, which is challenging and impractical, thereby limiting the usefulness of VSA for atmospherically relevant studies.

Reply: We respect but do not agree with the referee. In fact, many techniques which examine bulk materials provide important data to understand hygroscopicity of aerosol particles, as discussed in a recent review on aerosol hygroscopicity measurement techniques (Tang et al., 2019). Furthermore, in the last few years we have used our VSA instrument to investigate hygroscopic properties of a number of materials with atmospheric relevance, and published several peer-reviewed papers.

13. Line 379. The authors should emphasize that reduced hygroscopicity was measured in supersaturated conditions while in subsaturated conditions hygroscopicity was higher as revealed by HTDMA (e.g. 60-70%).

Reply: In fact, a similar trend was also observed for hygroscopicity measured under subsaturated conditions. In the revised manuscript (line 389-390) we have added one sentence to underscore it: "…and this suggests that the addition of hydrophobic hydrocarbon functional groups to OS reduced their hygroscopicity. Decrease in hygroscopicity of OS compounds with the increase in the number of carbon atoms was also observed under subsaturated conditions (Section 3.2)."

14. Line 416. ...but much less pronounced for a mixture octyl-OS/AS. ("mixture" should be emphasized)

Reply: That is right, and we have corrected it in the revised manuscript (Line 432): "...but much less pronounced for octyl-OS/AS mixed aerosol".

15. Line 417. What about the discrepancy of methyl and ethyl-OS despite both being very soluble? That should be noted and discussed, especially that their kappa GF are higher than kappa CCN.

Reply: We agree with the referee that some discrepancies between κ_{gf} and κ_{ccn} were observed for methyl- and ethyl-OS. On the other hand, the difference between κ_{gf} and κ_{ccn} was <25% for these two compounds. As pointed out by Petters and Kreidenweis (2007), if κ values vary by <30%,

the difference may not be significant when taking into account the uncertainties associated with deriving κ from measured grow factors and CCN activities.

To response to this comment, in the revised manuscript (Line 404-411) we have compared κ_{gf} and κ_{ccn} values for each compound: "Figure 8 compares κ_{ccn} and κ_{gf} values for the six types of aerosol particles examined. For pure OS, κ_{ccn} of methyl-OS (0.459 ±0.021) and ethyl-OS (0.397 ±0.010) were smaller than their κ_{gf} values (0.537-0.604 and 0.505-0.548), but the relative differences do not exceed 25%. Such a difference (<25%) may not be significant if all the uncertainties associated with deriving κ from measured hygroscopic growth and CCN activities (Petters and Kreidenweis, 2007). Octyl-OS appears to be an exception, and the average κ_{ccn} (0.206) was ~2.4 times larger than the average κ_{gf} (0.086). In addition, no significant difference was observed between κ_{ccn} and κ_{gf} for all the alkyl-OS/AS mixed aerosols."