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 #1

This work measures the hygroscopic properties and cloud condensation nucleation (CCN) activities of model organosulfate aerosols and allow us to better understand the environmental fates and impacts of these compounds. This work fits the scope of Atmospheric Chemistry and Physics. I have the following comments and suggestions for the authors' consideration.

Reply: We would like to thank referee #1 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.

Comments:

1. Line 72, "As OS are ubiquitous and abundant in the troposphere, it is important to understand their hygroscopic properties and CCN activities in order to assess their environmental and climatic effects." As mentioned by the authors, the atmospheric abundance of organosulfates investigated in this work is in order of ng/m^3. Can the authors further elaborate or comment the atmospheric significances of these investigated organosulfates? For instance, how the presence of these organosulfates would affect the physiochemical properties of atmospheric aerosols such as surface tension, hygroscopicity and CCN studied in this work?

Reply: We understand the referee's concern. In fact, for each single organic compound, its contribution to aerosol mass concentrations is small; however, it is still valuable to investigate physicochemical properties of these compounds. Furthermore, the mass fraction of OS in organic aerosols can reach up to 30%, as we pointed out in our original manuscript (line 57-58). To address this comments, in the revised manuscript (line 75-77) we have expanded the sentence to underscore the importance of OS: "As OS are ubiquitous in the troposphere, it is important to understand their hygroscopic properties and CCN activities in order to assess their environmental and climatic effects (Kanakidou et al., 2005; Moise et al., 2015; Tang et al., 2016; Tang et al., 2019a), especially considering that OS could contribute up to 30% of total mass of organic aerosols in the troposphere (Surratt et al., 2008; Tolocka and Turpin, 2012; Liao et al., 2015)."

2. Line 127, “As shown in Figure 2, the mass of OS samples at different RH was determined by the VSA using the following method.” In figure 2, only the results of sodium methyl sulfate were shown. What would be the VSA results for other species?

Reply: Figure 2 was used as an example (methyl-OS) to show the raw data of our VSA measurements, and thus it is unnecessary and difficult to show raw data for other species. In addition, the VSA results for all species investigated in this work have been summarized in Section 3.1 and Table 1.

3. Line 197, “the increase in sample mass at 50% and 60% RH may be because ethyl-OS were partially deliquesced at this stage.” What causes the ethyl-OS partially deliquesce? Why other OSs do not deliquesce partially?

Reply: To be honest, we do not have a definite answer to this question. Therefore, similar to many previous studies, we attributed this to partial deliquescence.

4. Line 276, “Figure 4 shows that obvious deliquescence transitions were observed for methyl-, ethyl-, and octyl-OS in the VSA experiments; in contrast, as revealed by Figure 5, continuous hygroscopic growth without obvious phase transitions was observed for methyl-, ethyl- and octyl-OS aerosol particles in H-TDMA measurements, suggesting that these aerosol particles may exist in amorphous state.” Can the authors discuss why methyl OS, ethyl-OS and octyl OS exhibited different deliquescence behaviours in H-TDMA and VSA measurements?

Reply: The different deliquescence behaviors in H-TDMA and VSA measurements were due to different states of samples used in the two experiments. In the revised manuscript (Line 281-285) we have expanded the two sentences to provide further explanation: “Figure 4 shows that obvious deliquescence transitions were observed for methyl-, ethyl-, and octyl-OS in the VSA experiments, because samples used in VSA experiments may be crystalline salts; in contrast, as revealed by Figure 5, continuous hygroscopic growth without obvious phase transitions was observed for methyl-, ethyl- and octyl-OS aerosol particles in H-TDMA measurements, suggesting that these aerosol particles which were produced by drying aqueous droplets to <5% RH may exist in amorphous state.”

5. Figure 6, at the same RH the GFs of methyl-, ethyl- and octyl-OS derived from VSA measurements were found to be consistently smaller than those measured using H-TDMA. Can the authors elaborate this observation?

Reply: To address this comment, we have added one sentence in the revised manuscript (Line 303-305) to explain our observation: "The small but systematical differences between VSA and H-TDMA results, as evident from Figure 6, could stem from volume additivity assumption used to convert mass growth to diameter growth, uncertainties in OS densities, and DMA sizing errors."

6. Line 351, "Such underestimation at 70% and 75% RH is likely to due to that inorganic compounds (AS, in our work) may dissolve partially in the organics/water solution (which can be formed at much lower RH due to continuous water uptake of organics) before the mixed particle is completely deliquesced (Svenningsson et al., 2006; Zardini et al., 2008; Wu et al., 2011);" Can the authors comment if the amount of AS partially dissolved could be estimated based on their assumption and the hygroscopic data?

Reply: The difference between predicted and measured GF at a given RH (70% or 75%) was due to water associated with partially dissolved AS, and subsequently we can estimate the amount of AS partially dissolved. In the revised manuscript (line 365-367) we have added one sentence to state the amount of partially dissolved AS at 70% RH: "For example, the ratios of partially dissolved AS to total AS at 70% RH were estimated to be 0.95, 0.85 and 0.49 for methyl-OS/AS, ethyl-OS/AS, and octyl-OS/AS mixtures with a mass ratio of 1:1."

7. Line 392, "No significant difference was observed between κ_{gf} and κ_{ccn} for five types of aerosol particles, and the relative differences between κ_{ccn} and κ_{gf} values do not exceed 25%. However, octyl-OS appears to be an exception, and the average κ_{ccn} value (0.206) was ~1.4 times larger than the average κ_{gf} value (0.086)." Given the differences between κ_{gf} and κ_{ccn} were larger than the error bars (Figure 8), can the authors discuss why they would like to claim there is no significant difference between κ_{gf} and κ_{ccn}.

Reply: We agree with the referee, and have revised this paragraph (Line 404-411) in the revised manuscript accordingly: "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."

8. Line 403, “Petters and Kreidenweis. (2008) demonstrated that cloud droplet activation was highly sensitive to the solubility for sparingly soluble compounds in the range of 5×10^{-4}-2×10^{-1}, expressed as volume of solute per unit volume of water (Petters and Kreidenweis, 2008). Compared to the highly soluble methyl- and ethyl-OS, the solubility of octyl-OS (8.43×10^{-4}-4.26×10^{-2}) (Chemistry Dashboard, 2021) is rather limited.” What are the estimated water solubility for methyl- OS and ethyl-OS? Are they highly soluble or sparingly soluble?

Reply: The two compounds are highly soluble, and in the revised manuscript (Line 422) we have provided their solubilities: “Compared to the highly soluble methyl- and ethyl-OS (their solubilities are 0.127-0.219 and 0.075-0.151), the solubility of octyl-OS (8.43×10^{-4}-4.26×10^{-2}) (Chemistry Dashboard, 2021) is rather limited...”

9. Line 408, “incomplete dissolution at subsaturated condition in H-TDMA measurements may lead to underestimation of κ_{gf} values for octyl-OS; as a result, the solubility limit may explain the observed difference between κ_{gf} and κ_{ccn} for octyl-OS.” As mentioned by the authors, shall the authors estimate the κ_{gf} values for octyl-OS with solubility limit and surface tension correction?

Reply: As pointed out correctly by the referee, it will be very nice to assess the contribution of solubility limit and surface tension reduction to the observed difference between κ_{gf} and κ_{ccn}. Nevertheless, this needs sophisticated numerical models and may be beyond the scope of our manuscript. We have added one sentence in the revised manuscript (line 434-436) to acknowledge this caveat: “We note that some numerical models (Petters and Kreidenweis, 2008; Petters and Kreidenweis, 2013; Riipinen et al., 2015) are available to quantitatively assess contribution of solubility limit and surface tension reduction to the discrepancy between κ_{gf} and κ_{ccn}.”