05 October, 2020

Dear Professor Jingkun Jiang,

Thank you very much for handling our manuscript (MS No.: acp-2020-386) submitted to Atmospheric Chemistry and Physics.

Our revised manuscript, submitted on 02 September, was reviewed by three referees. One referee suggested that the manuscript can be accepted after technical correction, and minor revision was required by the other two referees. We have adequately addressed all the comments raised, and revised our manuscript accordingly again. For more information, please refer to our revised manuscript and replies to referees.

I would like to thank you and referees for all your efforts, which have largely help us improve our manuscript.

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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

Peng et al. have adequately responded to most comments I raised. Nonetheless, some parts need further clarification and improvement. Overall, I recommend its publication in ACP with minor revisions noted below.

Reply: We would like to thank ref #2 for reviewing our manuscript again and recommending it for final publication after minor revision. All the comments have been addressed in our revised manuscript, as detailed below.

Line 251 in the revised text: It still lacks the in-depth interpretation of the no obvious difference in aerosol hygroscopicity between summer and winter. The authors claimed that high mass fractions of carbonaceous materials were responsible for such no seasonal trend. More elaboration on how such high mass fractions caused the no seasonality deserves to appear here. Or, at least, potential mechanisms need to be mentioned here.

Reply: In response to this comment, in the revised manuscript (page 12) we have modified this sentence to make the explanation concise and clear: "In addition, no obvious difference in aerosol hygroscopicity was found between summer and winter, because mass fractions of soluble inorganic species were similar in the two seasons at each individual particle size (Massling et al., 2009)."

It seems to me that the authors directly answered the question in their rebuttal on "How can the contribution of soot and organics reduce CCN activities while the hygroscopic parameters remain high?". The authors claimed in line 1042 that if measurement sites were affected by primary emissions (less hygroscopic), CCN activities could be significantly reduced. However, at the same time, the hygroscopicity in the sites was found to be high (κ>0.3). How can you reconcile the discrepancy? Or did I miss something?

Reply: Here ref #2 may misunderstand our statement. In general, CCN activities were quite high (κ>0.3) in these sites. However, when these sites were significantly affected by primary emissions, CCN activities would be largely reduced, and in such cases the measured κ values would be significantly smaller than 0.3. The work at Backgarden, Guangzhou (Rose et al., 2010, Rose et al., 2011) gave a very good examples to illustrate the effects of primary emissions (to be more specific, biomass burning) on CCN activities (see Table S5 for more details).

Line 1090: This is a relatively minor point, but it would be better if the authors can give some examples of locations that should be examined as a representative of a clean environment compared to eastern regions.

Reply: As suggested, in the revised manuscript (page 52) we have given a few examples for these locations: "…measurements in areas far from by human activities (e.g., Mt. Gongga in Sichuan Province, Mt. Waliguan in Qinghai Province, and Xianggelila in Yunnan Province) will be especially important…"

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Our replies are in black.
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Reply to referee #4

General Comments: The authors have put many efforts into reviewing aerosol hygroscopicity measurements in China, which is helpful for researchers to get a quick grasp of what has been done so far regarding this topic and may provide guidance for future research in this area. Most comments raised by the two reviewers have been addressed adequately by the authors, and the manuscript is ready for publication after the following specific comments are addressed.

Reply: We would like to thank ref #4 for reviewing our manuscript and recommending it for publication after minor revision. All the comments have been properly addressed in our revised manuscript, as detailed below.

Specific Comments: Line 208, Please be more specific, otherwise it could be misleading. Does "κ_t describe the overall aerosol properties" mean that κt describes the overall aerosol hygroscopicity? If this was meant, then this statement is not correct. Assuming MAF to be 1 will certainly influence the hydrophobic part of aerosol particles, however, measured CCN activities certainly cannot reflect variations of aerosol hygroscopicity of particles larger than ~ 300 nm. Thus, κ_t might only represent overall hygroscopicity of particles that within CCN relevant diameter ranges. Overall, this is not an accurate description and should be altered.

Reply: We agree with the referee, and in the revised manuscript (page 10) we have rephrased this sentence to be more accurate: "if it is forced to be 1 (two-parameter fit), the derived activation diameter (d_t) and single hygroscopicity parameter (κ_t) describe the overall properties of aerosol particles whose diameters did not exceed the maximum diameter scanned (Rose et al., 2010)."

L258 explore -> explored

Reply: This has been corrected in our revised manuscript (page 12).

Line 286, To be more precise, Wu et al., 2016 derived a linear relationship between organic aerosol hygroscopicity and O:C, which does not mean that derived κ of organics depended linearly on their O:C ratios. Also, one can see from Fig.8 in Wu et al., 2016 that κOA did not exhibit significant a linear dependence on O:C. Please rephrase this sentence.

Reply: As suggested, in the revised manuscript (page 13) we have rephrased this sentence to be more accurate: "The measured κ could be well predicted using the AMS data, and a linear relationship was found between the derived κ of organics and their O:C ratios (Wu et al., 2016)"

Line 340-341: Add references to support this clarification.

Reply: The work by Wang et al. (2017d) has been cited in the revised manuscript.

Line 361: What's the difference?

Reply: As suggested, we have added several sentences in the revised manuscript (page 17-18) to further clarify the difference: "To be more specific, the average κ of 40 nm particles increased in daytime during clean periods due to strong photochemical reactions, while showed a reverse pattern during polluted periods due to dominant contribution by primary emissions. For 150 nm particles, average κ showed similar diurnal variations for clean and polluted periods, reaching maximum values at noon."

Line 466: The closure results from only one site during specific periods proves nothing. Please change to "contribution of organics to aerosol hygroscopic growth was quite limited during that campaign". For example, results of Kuang et al. (2020) show that variations of organic aerosol can dominate the diurnal variations of overall aerosol hygroscopicity due to the dominant contribution of organic aerosol to aerosol mass and strong photochemical processes during daytime, which

resulted in quick daytime SOA formation. Results from Jin et al. (2020) and (Li et al., 2019) also demonstrated that organic aerosol can contribute substantially to aerosol liquid water content.

Reply: We agree with the referee. In the revised manuscript (page 22) we have modified our statement to be more accurate: "…implying that the contribution of organics to aerosol hygroscopic growth was quite limited during their campaign."

Line 473: Both CCN and HTDMA measurements are not precise down to 0.001, please change 0.364 to 0.36 and also revise similar cases throughout the manuscript and the supplement materials.

Reply: We agree with the referee that κ values cannot be measured with a precision down to 0.001. Nevertheless, this is a review paper and we would like to keep our values consistent with those reported in literature.

Line 716: similar issue as in comment for Line 286

Reply: As suggested, in the revised manuscript (page 34) we have rephrased this sentence: "A linear relationship was found between GF and O:C ratios for aerosol organics…"

L916-919, The explanation for the discrepancy between ACSM calculation and CCN or HTDMA measurements needs to be improved. ACSM measures the bulk compositions of PM2.5 or PM1, so the kappa derived from ACSM measurements using volume mixing rule should be understood as the average of kappa hygroscopicity of different diameters of PM2.5 or PM1 with aerosol volume as the weight, therefore represents the overall hygroscopicity of entire aerosol population of PM1 or PM2.5. However, the HTDMA or CCN measurements only represents aerosol hygroscopicity of specified diameter or diameter range. Thus, the closure between Kappa calculated using ACSM measurements between HTDMA measurements or CCN measurements is not physically appropriate, while their variation trends may be comparable, they should not be compared against each other in closure studies. If all measurements (including aerosol chemical compositions measurements, HTDMA measurements and organic aerosol hygroscopicity) were accurate, large discrepancies can still be expected from their comparison due to their intrinsic difference in their representations of distinct aerosol populations. Volume contributions of particles with diameter < 60 nm are generally below 3% and has almost negligible impacts on Kappa calculations based on ACSM measurements, thus the inconsistency should be dominantly determined by their diameter discrepancy.

Reply: We entirely agree with the referee on this point. In response, in the Section 4.5 of the revised manuscript (page 51), we have added a few sentences to further discuss the hygroscopicity closure analysis: "In hygroscopicity closure studies (either hygroscopic growth or CCN activity), average aerosol compositions are usually used to calculate hygroscopicity, and thus the calculated hygroscopicity represents the volume-weighted hygroscopicity of the entire aerosol population; on the other hand, H-TDMA and CCN measurements only provide hygroscopicity of aerosols of specific diameters or diameter ranges. As a result, although variation trends between measured and calculated hygroscopicity may be comparable, strictly speaking direct comparison is not physically appropriate. It would be more proper to compare measured hygroscopicity with that calculated using size-resolved chemical composition, as demonstrated by a closure study carried out by a campaign in central Germany (Wu et al., 2013)."

Line 1046, Give concrete values, like "larger than (range1 versus range2)……

Line 1048, Give concrete values.

Reply: The two comments point to the same issue and are thus addressed together. It would be nice to provide concrete values here, as suggested by the referee. However, reported κ values depended on particle size, and therefore it is difficult to use a few numbers to provide concrete values. Instead, in the revised manuscript (page 49) we have expanded a sentence to refer readers

to Table S5 for these numbers: "We note that a few recent studies (Atwood et al., 2017; Zhang et al., 2017; Cai et al., 2020) also reported higher aerosol hygroscopicity, as shown in Table S5. For example, the average κ observed at the Xinzhou site (Zhang et al., 2017) appeared to be larger than those reported at other continental sites…"

Line 1127, It might be better to include most recent results on organic aerosol hygroscopicity in the North China Plain (Kuang et al., 2020) in this part.

Reply: The work by Kuang et al. (Atmos. Chem. Phys., 20, 865–880, 2020) has been cited in the revised manuscript (page 54).

Line 1073, Please include Liquid-Liquid phase separation

Reply: In the revised manuscript (page 51) liquid-liquid phase separation has been included: "…such as solution non-ideality of aerosol droplets, limited solubility of some components contained by aerosol particles, surface tension effects, liquid-liquid phase separation, and etc."