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 #3

The review by Peng et al. is an ambitious study in trying to summarize aerosol hygroscopicity measurements in China. The authors efforts are commendable and will certainly guide the future research efforts (at least in China and beyond). The review is well written and easy to follow, so should be acceptable for publication after addressing the comments.

Reply: We would like to thank ref #3 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.

One major comment is arising from the author's efforts to make a fair summary of all the measurements, but without connecting observations with processes and/or sources. As such, reading the large portions of text becomes boring, because it only mentions facts (easily found in individual papers by concerned readers) without linking or extending scientific knowledge. The review is not only meant to provide a summary of observations (that would be rather a report, not scientific study), but most importantly to critically analyse available knowledge and subsequently to identify scientific knowledge gaps. CCN part of the review is written much better, but HTDMA part is lacking interpretation on every page or even more often. Few good and bad examples were noted, but the authors should read their text carefully to recognise the rest.

Reply: This is a very good point. In the revised manuscript, we have made large efforts to link reported aerosol hygroscopicity with aerosol composition, processes and sources. As detailed below, we have addressed specific comments raised by the ref #3 and revised the manuscript accordingly. Furthermore, we have additionally provided explanations/interpretations for other observations reported in previous work, and changes can be found in the revised manuscript (e.g., page 12, page 17, page 19, page 24, page 27, page 32-33).

The second major comment relates to uncertainty analysis and even more importantly taking into account that uncertainty when interpreting the results of various studies. "Smaller" or "larger" is irrelevant on absolute scale, it is only important when the differences are outside the uncertainty range of GF or kappa. When the differences are within the uncertainty range it should be stated accordingly. Therefore, it advised to carefully use the words "different", similar" which carry very little scientific significance.

Reply: We understand and completely agree with this concern, and the following changes have been made in the revised manuscript: 1) we have included error bars for the data shown in figures and included uncertainties for the numerical numbers in the main text, when data uncertainties are available in the original papers; 2) as suggested, when we compare measurement data reported, we are cautious when words such as "different", "similar", and etc. are used.

However, it is not always possible to be statistically rigorous when we compare measurements reported, especially for a review paper. In fact, statistically rigorous comparisons are very rare in the original work covered in this review.

The abstract is currently a very formal structural summary when instead it should be a scientific one, highlighting identified knowledge gaps (perhaps, limiting to the most important ones). It should give a flavour what was uncovered by the review and engage the reader.

Reply: We agree with the referee that our abstract is a structural summary, instead of being a scientific one; indeed it will be very nice if we can highlight some major findings and knowledge

gaps. However, for such a big topic in which many studies have been conducted, we find it very difficult to summarize major findings and knowledge gaps in a few sentences in the abstract. Therefore, we would like to use a structural summary to tell readers what we have done in this review paper, and interested readers can refer to the manuscript and/or individual sections for further information.

Minor comments

Line 164. typo in 0.25, same in next instance.

Reply: Both cases have been corrected in the revised manuscript (page 8).

Figure 1 contains no error bars.

Reply: We have added error bars to Figure 1 in the revised manuscript (page 14).

Line 345. It is important for the review paper to give an in depth explanation of the observed phenomenon, not just acknowledge that differences were observed. Diurnal patterns must come from either dynamics of BL, photochemistry or sources, or interplay of the three.

Reply: We agree that it will be very desirable to explain the observed diurnal variation in aerosol hygroscopicity. However, the diurnal variation in aerosol hygroscopicity, reported by Wang et al. (2019b), was quite complex, and the explanation they provided in the original paper was even much more complex. It is very difficult for a review paper to summarize their major findings and explanations using a few sentences. As a result, in our review paper we only mention this aspect in brief.

Line 413. Same comment about summarizing observations without linking to processes and sources. Observed bimodality typically means different sources like traffic and secondary aerosol formation.

Reply: As suggested, in the revised manuscript (page 21) we have expanded these sentences to provide additional explanation to the observation: "Bimodal hygroscopicity distribution, with a dominant more-hygroscopic mode and a smaller nearly-hydrophobic mode, was observed over the whole period, and the average κ, derived from GF measured at 90% RH, increased from 0.250 at 50 nm to 0.340 at 250 nm, as number fractions of aerosol particles in the more-hygroscopic mode increased with particle size (from 68% for 50 nm to 85% for 250 nm) (Liu et al., 2011). Compared to the nighttime, both the average κ and number fractions of particles in the more-hygroscopic mode were larger during the daytime (Liu et al., 2011), because photochemical processes during the daytime led to enhanced formation of secondary species in aerosol particles and thus increase in their hygroscopicity."

Line 417. It should be specifically reworded: "It was found that secondary inorganic aerosol species increased hygroscopic growth of accumulation mode while organics were decreasing hygroscopicity of the Aitken mode".

Reply: As suggested, in the revised manuscript (page 21) we have rephrased this sentence in order to be more specific: "It was found that secondary inorganic species increased hygroscopicity of the accumulation mode while organics decreased hygroscopicity of the Aitken mode (Liu et al., 2014)."

Line 429. ...suggesting that ISORPIA-II was not capable to reproduce ALWC at low RH.

Reply: As suggested, in the revised manuscript (page 21) we have made the following change: "…but was much larger compared to the second method when ambient RH was <60% (Bian et al., 2014), suggesting that ISORROPIA-II was not capable to predict ALWC at low RH."

Line 437. Same comment on observations versus processes.

Reply: The following explanation has been provided in the revised manuscript (page 22) to explain the observation: "It was also found that for the accumulation mode, κ were larger in the

nighttime than the daytime (Ding et al., 2019), as increase in RH during the nighttime led to enhanced formation of sulfate and nitrate from aqueous oxidations of SO_2 and heterogeneous hydrolysis of N_2O_5 (Wang et al., 2017a)."

Line 488. Again missing comment as to what bimodality and increasing kappa means.

Reply: In the revised manuscript (page 25) we have provided further explanation: "Bimodal aerosol hygroscopicity distribution was always observed (Wang et al., 2017c), indicating that aerosol particles were externally mixed. As larger particles contain higher mass fractions of secondary inorganic species, the average κ were found to increase with particle size, from 0.240 at 30 nm to 0.320 at 250 nm."

Line 588. Good example of trying to explain the observations and link to composition and sources, not just documenting them.

Reply: We would like to thank the referee for his/her kind and positive comment.

Line 596. Good example

Reply: We would like to thank the referee for his/her kind and positive comment.

Line 673. ...or internal/external mixtures of organic and inorganic compounds.

Reply: In the revised manuscript (page 34) we have changed the sentence to "…may underestimate hygroscopicity of aerosol organics or mixed inorganic/organic aerosols."

Line 697. Can the reason be discerned? Well mixed aged aerosol removing differences of various sources of origin?

Reply: The referee is right. In the revised manuscript (page 35) we have expanded this sentence to provide some explanation: "Since aerosol particles arriving at this site were heavily aged and well internally mixed, no obvious dependence of average κ on particle size was found."

Line 702. organic matter, not materials

Reply: Corrected in the revised manuscript (page 35)

Line 726. if evident state the number of higher RH. Was it evident at 90%?

Reply: Here higher RH means 80% and 85%. In the revised manuscript (page 36) we have modified this sentence to be more specific: "…bimodal growth behavior appeared at ~75% RH except 40 nm particles and became more evident at higher RH (80% and 85%) (Wu et al., 2018a)."

Line 758. ...almost all of the...

Reply: Corrected in the revised manuscript (page 37).

Line 766. Not even in summary there is interpretation what multimodal hygroscopicity means in terms of processes and sources.

Reply: In the revised manuscript (page 38) we have made following changes to provide further interpretation: "Bimodal or trimodal hygroscopicity distributions suggested that aerosol particles under investigation were externally mixed. Quasi-unimodal hygroscopicity distributions existed but were quite sparse (Chen et al., 2016; Wang et al., 2016; Wang et al., 2017d; Zhang et al., 2017; Wang et al., 2018b), implying that these aerosols were nearly internally mixed."

Line 775. How different? Opposite?

Reply: The referee is right. In the revised manuscript (page 38) we have made the following changes accordingly: "…though opposite results were also reported in several studies"

Line 781. However, Meier et al. (2009) found that primary particles smaller than about 50nm in diameter exhibited decreasing hygroscopicity. If I interpreted correctly.

Reply: The referee is right. In the revised manuscript (page 39) we have made the following changes: "However, different results were also reported, especially for particles at or below 50 nm (Achtert et al., 2009; Meier et al., 2009; Wang et al., 2018; Wang et al., 2019) for which primary emissions could play an important role."

Line 789. The results should be interpreted in terms of processes and sources.

Reply: We have made the following changes in the revised manuscript (page 39) to interpret the observation: "the overall hygroscopicity, and especially hygroscopicity of 150-350 particles, was highest in summer and lowest in winter at the PKU site (Beijing); one possible reason was that aerosol particles examined by Wang et al. (2018c) were most aged in the summer (and thus contained largest fractions of secondary species with high hygroscopicity) and least aged in the winter."

Line 796. That was already stated numerous times, no need to repeat. The paragraph should start with underlying reasons.

Reply: Indeed this has been stated many times elsewhere in the manuscript. Nevertheless, we feel that it is necessary to summarize diurnal variations reported in previous studies in the summary section, especially different diurnal variations have been reported. In our original manuscript, we have discussed underlying reasons for some of the observed diurnal variations. In the revised manuscript (page 39-40) we have expanded these sentences to provide further explanation: "The underlying reason was that photochemical processes during the daytime led to increased relative contribution of secondary aerosols, which were very hygroscopic. However, there are also exceptions. For example, κ was larger in the nighttime than the daytime for the accumulation mode at the NKU site (Tianjin) in March 2017 (Ding et al., 2019), as high RH in the nighttime may enhance sulfate and nitrate formation from aqueous oxidation of SO_2 and heterogeneous hydrolysis of N_2O_5 (Wang et al., 2017a)."

Line 815. If kappa is considered a robust method, it does not matter at which RH GF was measured at, because lower RH would result in lower GF and kappa should be the same. If it was not the same, then that should be highlighted by proper comparison and stated clearly, because that is very important. Not all species exhibit hysteresis and even fewer when internally mixed.

Reply: In general we agree with the referee's concern. As a matter of fact, we compared κ values derived from GF at different RH, and found that they were not the same. Therefore, we stated clearly. As a result, we have made the following statement clearly in our original manuscript (page 40): "Therefore, it can be concluded that using a constant κ to describe aerosol hygroscopic growth at different RH may not always be proper.'

Line 824. NaCl has the highest deliquescence of 75% among the relevant atmospheric species, so the statement should state that no kappa (HTDMA) should be derived below 75-80%. The following Figure is manifesting that, but needs error bars added to data points.

Reply: In the revised manuscript (page 41) we have added error bars in Figure 5a. As the uncertainties for the data shown in Figure 5b were not provided in the original paper, we are not able to include error bars in Figure 5b, and we have added one sentence in the figure caption to explain how error bars are not displayed in Figure 5b.

We agree with the referee that no κ should be derived from H-TDMA measurements carried out at RH before 75-80%. In Section 5 for the revised manuscript (page 53) we have made the following change to make this statement in specific: "Therefore, measurements of aerosol hygroscopicity at different RH are certainly warranted, and hygroscopic growth factors measured at high RH (at 90% RH or above) are preferably used to calculate κ values."

Figure 5. Uncertainty of the calculated kappa is clearly above 10% based on very basic considerations. If one considers size uncertainty of two independent DMA at 10% each and RH measurement which is inherently drifting during HTDMA operation, one would get ~17% total uncertainty. Therefore, no one can objectively claim kappa differences of ~10%, because those will be within the overlapping error bars.

Reply: In the revised manuscript (page 41) we have included error bars in Figure 5a. We also agree with the referee's comments on uncertainties. The uncertainties shown in Figure 5a have two sources: 1) the uncertainties related to individual measurements; 2) the variation of different measurements, as only the average values from different measurements carried out at a given RH were report. Therefore, without getting access to and analyzing original data, an absolutely solid conclusion cannot be reached.

Line 877. was lower, not became lower. There is more to it. Calculated (chemical) kappa is relying on compound specific kappa values, which have uncertainty and without even mentioning rather arbitrary kappa of organic matter.

Reply: In the revised manuscript (page 44), we have changed "became" to "was", also added one sentence to mention the uncertainties in calculating κ values. After revision, the last two sentences in this paragraphs have become to "This was because hygroscopicity estimated using ASCM-measured composition did not consider the contribution of smaller and less-hygroscopic particles (aerosol hygroscopicity was lower for smaller particles, but ACSM only detected >60 nm particles). In addition, the uncertainties associated with κ values assumed for ammonium sulfate, ammonium nitrates and organics may also contribute to the discrepancies between measurement and calculation." As this is a review paper, we would like to refer readers to the original paper for further information related to κ calculation (e.g., κ values assumed for each individual species).

Line 883...while the increase in aerosol hygroscopicity was much smaller due to the change in chemical composition.

Reply: The increase in observed [CCN], was due to two reasons, i.e. increase in particle size and increased in aerosol hygroscopicity (due to change in aerosol composition). Therefore, our original statement is correct and no changes have been made.

Line 897. Was that outside uncertainty range?

Reply: Considering the uncertainties, some differences were very small. To provide actual κ values (and their uncertainties) and to acknowledge the small difference, in the revised manuscript (page 45) we have made the following changes: "Compared to κ values (increasing from 0.291 ± 0.089 at 50 nm to 0.373 ± 0.092 at 350 nm) derived from concurrent H-TDMA measurements, aerosol hygroscopicity derived from CCN activities were slightly lower for <50 nm particles but higher for >100 nm particles (Ma et al., 2016; Zhang et al., 2016b), but the differences were quite small."

Line 1023...and both consistencies... and discrepancies were reported

Reply: Corrected in the revised manuscript (page 51).

Line 1033...research directions can be proposed.

Reply: We have changed "discussed" to "proposed" in the revised manuscript (page 51).

Line 1042...in eastern regions

Reply: Corrected in the revised manuscript (page 52).

Line 1046...hygroscopicity in the cleaner troposphere. "Pristine" can only possibly apply to remote oceanic regions or Antarctica. Not even Arctic is pristine.

Reply: We have changed "pristine" to "cleaner" in the revised manuscript (page 52).

Line 1069. ...can be easily activated at the lowest supersaturation due to their size.

Reply: In the revised manuscript (page 53) we have made the following change: "…as these particles can be easily activated at low supersaturation due to their size."

Line 1074. It should be stated that kappa (HTDMA) derivation should be limited to RH above 75-80% due to reasons discussed.

Reply: As suggested, in the revised manuscript (page 53) we have made the following change: "Therefore, measurements of aerosol hygroscopicity at different RH are certainly warranted, and hygroscopic growth factors measured at high RH (at 90% RH or above) are preferable used to calculate κ values."

Reference

Liu, H. J., Zhao, C. S., Nekat, B., Ma, N., Wiedensohler, A., van Pinxteren, D., Spindler, G., Mueller, K., and Herrmann, H.: Aerosol hygroscopicity derived from size-segregated chemical composition and its parameterization in the North China Plain, Atmospheric Chemistry and Physics, 14, 2525-2539, 2014.

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Ma, N., Zhao, C., Tao, J., Wu, Z., Kecorius, S., Wang, Z., Groess, J., Liu, H., Bian, Y., Kuang, Y., Teich, M., Spindler, G., Mueller, K., van Pinxteren, D., Herrmann, H., Hu, M., and Wiedensohler, A.: Variation of CCN activity during new particle formation events in the North China Plain, Atmospheric Chemistry and Physics, 16, 8593-8607, 2016.

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Zhang, S. L., Ma, N., Kecorius, S., Wang, P. C., Hu, M., Wang, Z. B., Groess, J., Wu, Z. J., and Wiedensohler, A.: Mixing state of atmospheric particles over the North China Plain, Atmospheric Environment, 125, 152-164, 2016b.