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. overview aerosol hygroscopicity data in China in terms of hygroscopic parameter and CCN activity. This review paper seems to simply list the results reported by earlier work without much interpretation based on atmospheric chemistry. For example, the hygroscopic parameters varied with particle size, chemical composition (primary vs secondary species), seasonal effect, and so on. This paper lacks the discussions on how the variable hygroscopic parameters are related to many factors mentioned above. I recommend the authors to provide more underlying mechanisms about the hygroscopicity characteristic in China and otherwise the paper would be just collection of the results. This current manuscript is not ready for publication and requires major revisions.

Reply: We would like to thank ref #2 for reviewing our manuscript. We agree that in our review we should further discuss factors related to aerosol hygroscopicity. In the revised manuscript, we have made large efforts to explain/interpret underlying mechanisms for aerosol hygroscopicity in China. For more information about changes we have made, we kindly refer ref #2 to the revised manuscript as well as our responses to specific comments raised by the two referees.

The introduction needs improvements. The authors describe their motivation to report the current study with fairly rational information. However, it still lacks why one needs to review aerosol hygroscopicity in China now under what circumstances. Elaborating such points would put this study in better context.

Reply: In response to this comment, in the revised manuscript we have added a few sentences in the fourth paragraph in the introduction Section (page 5) to explain why we would like to write this review paper: "In the last few decades, a number of field studies have investigated tropospheric aerosol hygroscopicity in China. However, a general overview of spatial and temporal variation of aerosol hygroscopicity in China is yet to be provided, and the dependence of aerosol hygroscopicity on aerosol composition, mixing state, and etc. has not been fully elucidated. In this paper we provide a comprehensive review of hygroscopic properties of ambient aerosols measured using H-TDMA in China; in addition, CCN activities of tropospheric aerosols measured in China are also reviewed and discussed."

The title would be misleading. It seems to me that this study focuses on aerosol hygroscopicity in China, but not its measurements.

Reply: The referee is right. We have changed the title to "Tropospheric aerosol hygroscopicity in China".

I am aware of the recent review paper on aerosol hygroscopicity (Tang et al., 2019). It is not convincing at all if the authors do not provide any elaboration on how the current study is distinguishable by Tang et al.

Reply: The recent review paper by Tang et al. (2019) is focused on the experimental techniques for aerosol hygroscopicity measurements. In the revised manuscript (page 5), we have expanded the sentence to further clarify the difference between our current review and the review paper by Tang et al. (2019): "In addition, a recent paper (Tang et al. 2019) has reviewed aerosol hygroscopicity measurement techniques, but it only discussed several exemplary studies to

illustrate how specific techniques can help us better understand tropospheric aerosol hygroscopicity."

Line 97: I do not agree that the single particle studies are too limited for the overall aerosol hygroscopicity. This type of lab studies has proven powerful to establish aerosol thermodynamic models that can provide useful information on the overall aerosol hygroscopicity. It is understood that this manuscript focuses on field measurements of ambient aerosol hygroscopicity, but it needs to rephrase the argument.

Reply: The referee is right. In the revised manuscript (page 5) we have rephrased our argument to avoid misleading implications: "Single particles techniques (Krieger et al., 2012; Li et al., 2016) have provided physiochemical data which are very valuable to test aerosol thermodynamic models, largely helping us better understand tropospheric aerosol hygroscopicity. However, as numbers of particles examined in single particle studies are very limited, these studies usually do not provide direct information of overall aerosol hygroscopicity in the ambient air and thus are not discussed herein."

Line 200: Can you explain why is there no obvious difference in aerosol hygroscopicity between summer and winter?

Reply: As suggested, we have added one sentence in the revised manuscript (page 12) to explain the observation: "In addition, no obvious difference in aerosol hygroscopicity was found between summer and winter, because constantly high mass fractions (~50% wt) of carbonaceous materials (nearly hydrophobic or less hygroscopic), related to extensive usage of fossil fuel, were observed in both seasons for submicrometer particles (Massling et al., 2009)."

In Figure 1, the hygroscopic parameter was always highest in summer and lowest in winter. Photochemical processes and secondary products play a role in this trend. Can you also explain the seasonal difference in the hygroscopic parameter in terms of chemical composition difference? For instance, what is the major inorganic and organic species between two seasons and O/C ratios?

Reply: We agree with the referee that photochemical processes and enhanced formation of secondary species play a role in enhanced aerosol hygroscopicity in summer, compared to winter; however, the original paper provided no information on seasonal variation in aerosol composition. To address this comment, in the revised manuscript (page 15) we have added one sentence to explain the possible reason for the observed seasonal variation in aerosol hygroscopicity: "The difference in aerosol hygroscopicity between summer and winter may be caused by enhanced photochemical processes in the summer and as a result increased fractions of secondary species."

In Figure 2, it is related to the questions above. The hygroscopic parameter increased with particle size, which was attributed to enhanced contribution of secondary species. Can you provide more information on how chemical composition of PM changes with particle size increase?

Reply: In response to this comment, we have added one sentence in the revised manuscript (page 16-17) to explain the variation of aerosol hygroscopicity with particle size: "Figure 2 shows that aerosol hygroscopicity increased with particle size and pollution level (Wang et al., 2017d), because mass fractions of hydrophilic species, such as sulfate, nitrate and oxidized organics increased with particle size, especially under highly polluted conditions."

Lines 319-322: How did the size-resolved mass fractions of secondary inorganic species lead to slight decrease in the hygroscopic parameter in winter as particle size increases?

Reply: From the size-resolved H-TDMA and HR-TOF-AMS measurements, the average κ slightly decreased from 0.271 at 110 nm to 0.260 at 200 nm, while the mass fractions of secondary inorganic species decreased from 61.8% at 110 nm to 59.3% at 200 nm. In the revised manuscript (page 18) we have made the following changes to provide further explanation: "The average κ

increased from 0.158 at 40 nm to 0.271 at 110 nm in winter, and further increase in particle size (to 200 nm) led to slight decrease in κ, because mass fractions of secondary inorganic species decreased slightly from 61.8% at 110 nm to 59.3% at 200 nm (Fan et al., 2020). For comparison, the average κ increased with particles size in summer, from 0.211 at 40 nm to 0.267 at 200 nm, as mass fractions of secondary inorganic species increased from 56.7% at 80 nm to 63.0% at 200 nm (Wang et al., 2019b; Fan et al., 2020)."

Line 856: Why there was no size dependence of the hygroscopic parameters of activated particles despite the large values?

Reply: Aerosol particles in Xinzhou was highly aged and well internally mixed, and the variation of chemical compositions with particle size was negligible; therefore, κ were larger compared to other sites in the NCP and showed no obvious size dependence. We have made following changes in the revised manuscript (page 45) to provide further explanation: "This is because aerosols observed at this site were highly aged and well internally mixed after undergoing regional transport for a long time, and thus the variation of chemical compositions with particle size was negligible."

Lines 952-955: The logic is not clear. The authors mentioned that If measurement sites were affected by primary emissions, CCN activities could be reduced. However, if so, the averaged hygroscopic parameters can be reduced too. I imagine that CCN activities would be essentially attributable to the hygroscopic parameters. How can the contribution of soot and organics reduce CCN activities while the hygroscopic parameters remain high?

Reply: Indeed if significantly affected by primary emissions, both hygroscopic properties (RH <100%) and CCN activities (RH>100%) should be reduced, as suggested by several studies (Rose et al., 2010; Gunthe et al., 2011; Zhang et al., 2014; Wu et al., 2017) mentioned in this paragraph. The work by Zhang et al. (2017) found high aerosol hygroscopicity, but aerosols investigated by this work were heavily aged, instead of being affected by primary emissions. In order to reduce confusion, in the revised manuscript (page 49) we have made the following changes: "…due to enhanced contribution of soot and organics. We note that a few recent studies (Atwood et al., 2017; Zhang et al., 2017; Cai et al., 2020) also reported higher aerosol hygroscopicity. For example, the average κ observed at the Xinzhou site appeared to be larger than those reported at other continental site (Zhang et al., 2017)…"

Lines 979-980: What are potential reasons for the consistence and discrepancies?

Reply: Ideally, they should be consistent, while discrepancies were not usual due to several reasons, including solution non-ideality of aerosol droplets, limited solubility of some components contained by aerosol particles, surface tension effects, and etc. We have added one sentence in the revised manuscript (page 51) to provide further explanation: "The discrepancies could be caused by several factors (Petters and Kreidenweis, 2008; Wex et al., 2009; Petters and Kreidenweis, 2013; Liu et al., 2018), such as solution non-ideality of aerosol droplets, limited solubility of some components contained by aerosol particles, surface tension effects, and etc."

Minor points:

Line 217: A typo of "exntensive".

Reply: Corrected in the revised manuscript (page 13).

Line 544: One possible reason for what?

Reply: In the revised manuscript (page 30) we have provided further information to clarify it: "One possible reason for difference in aerosol hygroscopicity observed in the two periods at the same site was that in April-May 2014 organic species made a large contribution to submicrometer aerosols…"

Line 619: A typo of "he".

Reply: Corrected in the revised manuscript (page 33).

Line 760: You mean "exceptions"?

Reply: That is right, and we have corrected it in the revised manuscript (page 40).

Line 823: Please add "respectively". Were the hygroscopic parameters for organics assumed to be 0?

Reply: The hygroscopicity parameter was indeed assumed to be 0 for organics. We have rephrased this sentence in the revised manuscript (page 43) to be clearer: "…if κ were assumed to be 0.53 for inorganics and 0 for organics, respectively."

Reference

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