Comments by referees are in blue.

Our replies are in black.

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

The work is important and of relevance to the readership. The authors present a review of hygroscopicity measurements as it pertains to the atmosphere and also orthogonal scientific fields (surface science, heterogeneous catalysis, geochemistry/astrochemistry, pharmaceutical and food science, etc). Thus the publication will be of interest to the ACP readership and other fields. The authors have done a good job to describe "non-conventional atmospheric" hygroscopicity measurements. That is they provide an overview of comprehensive laboratory techniques that due to time or spatial resolution is not necessarily applied to field studies. For instance, the spectroscopy section is quite thorough and provides information on the numerous spectroscopy techniques that have been applied in controlled laboratory settings. The review is useful in that it provides a current overview of the current state of technology for hygroscopicity. However, the paper does not include a review of the theoretical hygroscopicity equations (although, I do not think that this is the purpose of the manuscript). Furthermore, I was quite disappointed to find that CCN and IN techniques were not discussed at all and perhaps this omission should be reflected in the title. E.g. "A review of experimental techniques for unsaturated aerosol hygroscopicity studies". Regardless of this disappointment, I highly recommend the work for publication in ACP. The work will be cited heavily in the future. The following are a few concepts and ideas that may strengthen and or clarify ideas in the manuscript. I sincerely encourage the authors to consider addressing these comments before eventual publication.

Reply: We would like to thank Ref #1 for his/her insightful comments as well as recommending our manuscript for final publication. We do not review theories related to aerosol hygroscopicity because our manuscript is focused on techniques for aerosol hygroscopicity measurements; similarly, we do not review techniques used for CCN and IN measurements. However, in the original manuscript we have referred readers to literature where aerosol hygroscopicity theories and techniques for CCN and IN measurements are reviewed. We feel that the title we use is proper, although we understand what the referee means. This is because hygroscopicity usually means interaction of water vapor with particles at <100% RH. We have also addressed all the other comments adequately in the revised manuscript, as detailed below.

Comments.

1. The fluorescence spectroscopy section seems tangential to the hygroscopicity discussion. Much of this sections suggest that EDB is the actual technique and then fluorescence is used to measures the particle properties.

Reply: It is true that fluorescence spectroscopy does not measure aerosol hygroscopicity; however, it provides information (for example, the ratio of solvated water to free water) closely related to hygroscopicity. Therefore, we chose to discuss this technique using two paragraphs.

2. Time resolution of hygroscopicity measurements should also be discussed as a recommendation to improve measurements. The DASH-SP and HFIMS are the only fast resolution hygroscopicity measurements techniques currently used. Recent work by Wang et al (HFIM) should also be discussed.

Reply: Indeed time resolution is an important aspect, and this has been discussed in the original manuscript (future direction #1, page 92). We would like to thank the referee for bringing the work by Wang et al. (2019) to our attention, and in the revised manuscript (page 95) the work have been cited in addition to that by Langridge et al. (2011) and Pinterich et al. (2017b) as examples of high time resolution instruments for aerosol hygroscopicity measurements.

3. The authors may consider discussing how advances in orthogonal fields may be of future importance to atmospheric measurements. For example, although not currently relevant to aerosol, the production of highly sensitive humidity sensors should be considered (e.g., Liang et al, 2018). The Dash-P and HFIMS, use faster sizing instrumentation however faster RH technology may also advance studies.

Reply: The advancement in RH measurements will definitely be very valuable. In the revised manuscript (page 95, future direction #2) we have added the following sentences to discuss this aspect: "Furthermore, currently RH measurements typically have an absolute uncertainty of 1% or larger, and uncertainties in RH measurement would affect hygroscopic growth factors reported at a given RH, especially for high RH at which growth factors are more sensitive to RH; therefore, advancement in RH measurements (Liang et al., 2018) will contribute to the improvement in aerosol hygroscopicity measurement techniques."

Minor Corrections

Reply: We would like to thank the referee for carefully reading our manuscript and pointing out those typos, as detailed below.

L83, aqueous particle becomes supersaturated?

Reply: It means that the aqueous particle becomes a supersaturated solution. In the revised manuscript (page 4) we have expanded this sentence to provide further clarification: "the aqueous particle would become supersaturated (i.e. the aqueous particle becomes a supersaturated solution)."

L85. efflorescence is also kinetically controlled? How? Not clear how this statement is made.

Reply: Here we mean that efflorescence not only depends on thermodynamics but also is kinetically controlled. In the revised manuscript (page 4) we have expanded this sentence to provide further explanation: "Therefore, efflorescence is also kinetically controlled (in addition to being thermodynamically controlled) and…"

L70 – Remove or define "and etc"

Reply: As suggested, in the revised manuscript we have deleted "and etc".

L81 – spelling "dehumification"

Reply: In the revised manuscript this typo has been corrected.

L95 - , insert word - leading to the formation OF two coexisting liquid"

Reply: Corrected.

L95 – change to : in one particle"

Reply: Corrected.

L120 – Remove "in specific,"

Reply: Removed.

L189 – Change recently to recent

Reply: We have checked the manuscript and here "recently" should be used.

L385 – Change isotherm to isothermal

Reply: Corrected.

L1480 – "aerosol size is measured as" to "aerosol size is measured AT"

Reply: Corrected.

Additional References to consider

Wang, Yang, et al. "Retrieval of High Time Resolution Growth Factor Probability Density Function from a Humidity-controlled Fast Integrated Mobility Spectrometer." Aerosol Science and Technology just-accepted (2019): 1-18.

Liang, Jun-Ge, et al. "Thickness effects of aerosol deposited hygroscopic films on ultrasensitive

humidity sensors." Sensors and Actuators B: Chemical 265 (2018): 632-643.

Reply: As suggested, in the revised manuscript we have discussed these two studies.