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.

Reply to Ref #4

This paper reports measurements of DRH on bulk samples of some atmospherically-relevant salts. The technique as described is novel and such measurements are of importance in atmospheric chemistry. This paper is suitable for publication in AMT after the authors address the following major issues:

Author reply: We would like to thank Ref #4 for his/her very valuable comments, which have significantly help improve our manuscript. All the comments have been properly addressed in our revised manuscript, as detailed below.

1. Experimental: What is the sample mass needed for this technique? Does this potentially limit its atmospheric applications? If the sample were to be extracted from a filter, what would the mass need to be?

Author reply: In our original manuscript (line 129-131) we have mentioned the mass range of particles used in our work. Indeed this limits the atmospheric application of the technique. In the revised manuscript <u>(line 398-404)</u> we have discussed drawbacks of this technique: "We note that this technique also has a few drawbacks: 1) this technique cannot be used to examine supersaturated droplets or determine efflorescence relative humidities (ERH), due to the contact of particles with the sample pan; 2) substantial amount of particles, typically around or larger than 1 mg, are required by this technique, limiting its application to atmospheric particles even after they are collected (e.g., using a filter or an impactor plate); 3) the experiment is very time-consuming, and a typical experiment can take several hours and even a few days, depending on experimental conditions."

Furthermore, in the revised manuscript <u>(line 136-140)</u> we have included a few sentences to explain how particle samples are prepared and to discuss the effects of sample mass: "Powdered particles are transferred into the sample pan using a small stainless-steel spatula. The mass of the sample would not affect the measured mass ratio of dry particles to associated water under a given condition; however, it would take more time to reach the equilibrium if the sample mass is larger." 2. Results and Discussion: More details on the E-AIM model should be provided: why was it chosen? What were the parameters used?

Author reply: We chose to use the E-AIM model because it is very user-friendly and has been widely accepted and used. We do not provide technical details of this model in our manuscript, but instead refer readers to original papers. In the revised manuscript (line 103-108) we have included a few sentences to explain why the E-AIM model is used in our work: "Detailed description of the E-AIM model can be found elsewhere (Clegg et al., 1998; Friese and Ebel, 2010). Hygroscopic growth factors, calculated using the E-AIM model, has been widely used to compare with experimental measurements to verify the performance of a variety of instruments, techniques and/or methods developed for hygroscopic growth studies (Pope et al., 2010; Lei et al., 2014; Estillore et al., 2016)."

3. Figure 3: The agreement between reference and measured DRH values is excellent for most salts shown in Figure 3. However, (NH4)2SO4 seems to be an outlier. Similarly, the data for (NH4)2SO4 does not fully agree with the model in Figure 4. Can authors comment on this discrepancy? Was there perhaps some contamination in the (NH4)2SO4 sample specifically?

Author reply: The referee raised a very good point. As suggested, in the revised manuscript <u>(line</u> <u>326-340)</u> we have added a few sentences to discuss this issue: "The agreement between measured and calculated growth factors is excellent for NaCl at both temperatures; for (NH4)₂SO₄, the

agreement is not as good as NaCl. This may be caused by two reasons. First, after $(NH_4)_2SO_4$ is deliquesced, mass hygroscopic growth factors increase sharply with RH, and therefore a small difference in RH would lead a relatively large change in measured mass hygroscopic growth factors; if taking into account the uncertainty in RH (±1 %), the difference between our measured and predicted mass hygroscopic growth factors is <15%. Second, inspection of the data in Table 2 reveals that the difference between our measured and previously reported DRH is <1% for all the other compounds except (NH₄)₂SO₄. This may indicate that the purity of (NH₄)₂SO₄ with higher purity and measure its DRH and hygroscopic growth factors. Overall, it can be concluded from the comparison that our measured mass hygroscopic growth factors agree well with theoretical values for (NH₄)₂SO₄ and NaCl at both 5 and 25 oC. This gives us further confidence that the method developed in this work is reliable for hygroscopicity measurements of atmospheric particles."

4. Figure 4: I agree with the other reviewers that more points are needed on these plots to fully appreciate how measurements compare to the model.

Author reply: As requested, we have conducted these experiments with a RH resolution of 10%, and discussion on the comparison between model prediction and our measurement has also been expanded. Please refer to our revised manuscript <u>(line 326-345)</u> for more details.

5. Figure 4 and others: All figures that show experimental measurements need error bars. Similarly, uncertainties should be reported on measured DRH values in Table 1.

Author reply: As requested, information on uncertainties has been provided for all the figures and tables. Please note that in some figures the error bars are too smaller to be clearly visible.

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