General comments:

Based on the HTDMA and VSA measurements, this work has reported a large dataset of hygroscopicity of Ca- and Mg-containing samples, and discussed the temperaturedependence of DRH values. The calculated κ results were comparable to those of previous hygroscopic studies. This can enrich the knowledge of water uptake by Caand Mg-containing mineral dust and sea salts in the troposphere. The quality of this manuscript could be largely improved, providing a better organization of the main contents with clearer and more concise descriptions. A native or professional speaker is recommended for the language revision including a thorough grammar check. The presented work has its scientific importance, while major revision is suggested before the final publication.

Specific comments:

- 1. Page 8, line 175: The authors have suggested a "*baseline drift*" for the determination of the mass growth factor, or rather the DRH value. However, it is confusing to readers that how "*a significant increase in sample mass*" was defined. What is the corresponding reference value for the investigated salts? Does the baseline drift in sample mass always correspond to a same value for different types of Ca- and Mg-containing samples?
- 2. Page 8, line 176: Experiments are repeatable, but how "each measurement" could be repeated?
- 3. Page 8, line 180: It is unclear that when did "*an equilibrium*" or "*a new equilibrium*" state actually reach during the experiments, at least it is difficult to tell from the corresponding figures, e.g., it is still able to expect some increase in the sample mass as displayed in Fig.5.

In addition, the determination of an equilibrium condition will directly influence the accuracy of water-to-solute ratio, mass growth factor, and DRH results reported in this work. In this sense, how to quantify the influence of this "equilibrium" state on measured change in m/m_0 or in normalized sample mass with varying RH, and further on DRH and comparison of different hygroscopic results?

4. Page 9, line 188: Following the comment #3, there is no definition of "normalized sample mass", which is also important for the determination of DRH.

I feel lost in the **authors' response to Specific comment #15 of Ref #3**, which explained that "When plotting these three figures (i.e., Figs.1, 3, and 5), we did not

always to normalize the sample mass to the dry particle mass. This is why the right y-axis in these three figures is labelled as normalized sample mass instead of m/m₀." How to quantify the change in sample mass accurately, if the "normalized sample mass" was not always normalized by the initial dry mass? Accordingly, which kinds of mass were used for the normalization if "not always" to normalize, and how should the readers evaluate the comparability among these normalized sample mass results with increasing RH if without a constant comparison standard/baseline in the same experiment?

- 5. Page 9, line 193: The authors have stated that "*such a large mass increase cannot be solely caused by water adsorption*". Why? Can you provide some clues or evidence to clarify this point, instead of making an assertion without any proof?
- 6. Page 12, line 227: Is the corresponding RH determined to be 57±5%? Accordingly, can you really tell that they are "*in broad consistence with*" the results reported in this work?
- 7. Page 12, line 242: In the expression of "*could have unknown systematical errors*", I was confused by the "unknown" here, as this sentence was supposed to provide the specific reasons. Besides, why can you assume that it is due to systematical errors if it's even unknown?
- 8. Page 14, line 275

"Small changes in m/m_0 (typically <2%) were observed for some compounds (as shown in Tables 2 and 6) when RH was below corresponding DRH values, mainly due to water adsorption/desorption and baseline drift."

This sentence sounds somewhat ambiguous. What does the "small changes" refer to, the increase or decrease in mass change? How to relate the small mass change with the corresponding causes, i.e., water adsorption, water desorption, and baseline drift?

9. Page 16, line 296

"Water activities of $Ca(NO_3)_2$ solutions at 25 °C were measured to be 0.904, 0.812 and 0.712 when the concentrations were 2.0, 3.5 and 5.0 mol kg⁻¹, respectively".

The results might be wrongly sequenced in either RH values or molality concentrations, as which are opposite to the corresponding details included in the following sentence. To avoid such mistakes, please also check for the $Mg(NO_3)_2$ sample and data results elsewhere carefully.

10. Page 16, line 297: Please confirm the physical meaning of "water activity" and "RH of air".

- 11. Page 18, line 330: How was the mass increase (i.e., 48±7%) determined here, which seemed not reflected in Fig.3? What is the baseline drift or reference mass value used for the normalization of sample mass?
- 12. Page 24, line 430: Try to rephrase the whole sentence starting with "For comparison" into a more concise one, so does the beginning sentence in the following paragraph. This will help the readers to catch the point more easily.
- 13. Page 24, line 443: In the section of "Comparison between hygroscopic growth with CCN activities", equations used for the κ_{gf} calculation and relevant details belong to theoretical methodologies other than observed results. It would make more sense to reorganize the corresponding contents, e.g., into the Experimental section.
- 14. Table 5: Corresponding references are needed for "previous studies".
- 15. Page 28, line 519: How much does the "*significant increase*" refer to? Line 521: Why is the DRH determined to be 90.5±1.0% instead of ±0.5%? A similar issue existed in Line 533: "giving a measured DRH of 71.5±1.0%". Please also have a check on the corresponding values such as in the Conclusion section.
- 16. Page 32, line 585: The last sentence is really confusing. What is the connection between "GF is typically expected to increase with RH" and "small decrease in GF (~0.03) for RH between 50 and 70% may reflect uncertainties in GF measurements"? What kinds of uncertainties in GF measurements are you trying to suggest? What is the possible influence of particle phase state on measured growth factors?

Besides, what does it mean that "*HTDMA measurements cannot resolve a GF difference as small as 0.03*", since the presented GF results (i.e., 1.03 ± 0.01 and 1.00 ± 0.01) were actually measured with HTDMA and the uncertainties in the corresponding GF values are as small as ±0.01 ? A more scientific/convincing explanation for this observed decrease in GF will need to be provided.

17. Page 34, line 635: This Sect.3.3.1 was designed to compare the hygroscopicity results between HTDMA and VSA measurements. However, the organization of the whole section was a bit misleading. It seems that only one sentence is directly related to the topic (i.e., "To directly link the mass change (measured using VSA) with diameter change (measured using H-TDMA), solution densities, which also vary with RH, are needed."). I didn't see more details about how these two types of growth factors were actually reconciled.

Although the authors have tried to explore the atmospheric relevance of their experimental results (e.g., applicability to the conditions when RH is below or exceeds DRH), it's important to distinguish the difference in hygroscopicity for both bulk powder samples and individual particles. For clearer clarification, such information might be necessary before detailing the corresponding water uptake results.

Line 662-663: This sentence is confusing. Whether is it suggesting a possible way for the future investigations of the particle phase state with changing RH, or is it trying to answer the open question with the results obtained from previous studies? Then what is the conclusion or possible answers to the question according to previous work?

A similar issue also exists in the last two sentences of this section. Haven't you already measured the water-to-solute ratios and mass growth factors in this study? Why not trying to address the question using your own experimental data?