Response to Anonymous Referee #1

The referee's comments are in italics, our responses in plain font.

This paper presents a description of a HHTDMA and related method developed for investigating the hygroscopicity of aerosols in RH range of 2-99.6% with the uncertainty of growth factors within 0.9%, which will help explore the interaction between water and aerosols at RH close to 100%. By combining the restructuring modes with hydration/dehydration modes, the GFs can be measured in high precision after the microstructural rearrangement effect is considered. The manuscript is well written and presents a valuable contribution in the field of aerosol measurement techniques. I recommend this manuscript to be published after the following issues to be addressed and modified.

We thank the Referee #1 for the suggestions for improvement that were taken into account upon manuscript revision. Responses to individual comments are given below.

Specific comments:
Section 2.1: The manuscript gives a general description about the design and the components in constructing the HHTDMA. However, temperature and humidity control should be critical issues in operation, for example, maybe a PID control program was used with the input of RH4 and RH5 probe to control the RH in DMA2 precisely.

The temperature and RH control is discussed in Sect. 2.1 and Sect 2.4, respectively. PID control program was not used. The temperature **gradient**, i.e. the temperature profile along the DMA2 column (dT/dL) was not directly measured. The temperature difference between sheath and excess flow was used to estimate temperature variation inside DMA2 as indicated in Sect. 2.1.

How did the GORE-TEX membrane work?

The following clarifying text has been added in Sect. 2.3 to explain how Gore-Tex membrane was used:
The humidity of the aerosol flow (RH3) and sheath air (RH4) in DMA2, is controlled by mixing water saturated and dry air flows in a ratio produced the desired RH. Saturated air is obtained by passing dry air through a Gore-Tex membrane tube submerged inside a temperature controlled water bath (27.0 ± 0.1 ºC). Two separate 6 mm (ID) Gore-Tex tubes, 0.5-m and 2-m long are used for aerosol and sheath flows conditioning, respectively (Humidifier, Fig. 1). For the H1 Nafion exchanger the humid air is prepared by bubbling air directly through water and then mixing with dry air to the required humidity (not shown in Fig.1).

How to adjust the rotation speed of the fans in the DMA2 box?

The fan speed can be changed by varying the applied voltage (manually), but this was not necessary. In Supp. 2.1 we have demonstrated a simple way to compensate for the temperature difference between sheath and excess flows, if it needed.
The text in Suppl. 2.1 was modified as following:
The test measurements showed that the temperature difference between the sheath and excess flows can be changed within ±0.3 ºC by adjusting the rotation speed of the fans. The speed of each fan is affected by applied AC voltage.

Page 13, Line 358-360: This sentence is obscure and should be rewritten.

The sentence is updated:

The FHH (Frenkel, Halsey and Hiil) model is the frequently used to relate surface coverage to a water activity:

$$a_w = exp(-A_{FHH}/\Theta^{B_{FHH}}), \tag{28}$$

where A_{FHH} and B_{FHH} are empirical fit parameters that describe the intermolecular interactions governing the adsorption potential. A_{FHH} characterizes interactions between the surface and first adsorbed water layer as well as interactions between adjacent molecules. B_{FHH} describes the interactions between the surface and subsequent adsorbate layers.

Page 16, Line 470-480: I recommend providing more proofs (e.g. SEM images of particles) or reference(s) to support this claim.

The conclusion about the difference in the porous structure of ammonium sulfate and glucose particles is based on the results of HHTDMA measurements in the h&d mode. To our knowledge, there are no direct methods for measuring the pore network of 100 nm particles. SEM is a useful technique for extracting two-dimensional (2D) images of the microstructures but does not provide the third spatial component of the sample, which is important to find interconnected regions and pore volumes, shapes and sizes.
We have added the SEM images of initial ammonium sulfate and glucose aerosol particles (Fig. 5) in order to strengthen the argument in favor of the discussed aerosol particles morphology and the calculated values of particle shape (χ, β) and porosity (δ, f) presented in Table 1.

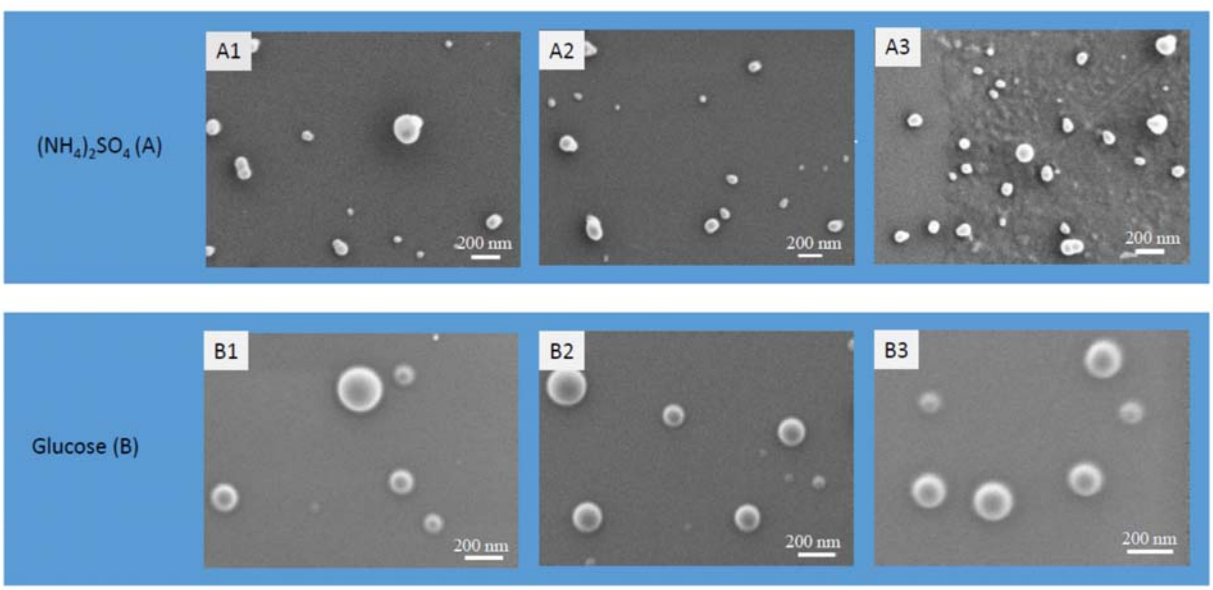

Fig. 5 SEM images of initial ammonium sulfate (A) and glucose (B) aerosol particles. The samples were investigated with a high-resolution SEM (ZEISS Merlin). Operation conditions: 0.4 kV accelerating voltage, 1.5 kV ESB grid voltage, 1.8 mm working distance. Particle samples were collected directly onto a 3mm TEM copper 300 mesh grids, coated with a 30–60 nm thick Formvar film.