Response to Anonymous Referee #2.

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

Mikhailov et al. present an instrument characterization of a newly constructed high humidity tandem DMA instrument. The instrument shows improved capabilities compared to previously described setups. The manuscript is well written and I recommend it for publication in AMT.

We thank the Referee #2 for these positive remarks. Responses to individual comments are given below.

The authors might consider adding a section comparing the versatility and accuracy of the system with other techniques. Specifically the Leipzig based LACIS instrument and the filter-based massbased hygroscopicity method used by the same author previously would be interesting to compare in this context.

## The following text has been added:

In addition to the HTDMA methods, other techniques have been used to determine the aerosol hygroscopicity at high RH (Tang et al., 2019). Two of these methods are the Leipzig Aerosol Cloud Interaction Simulator (LACIS; Stratmann et al., 2004) and the inverted streamwise-gradient cloud condensation nuclei counter (Ruehl et al. 2010), which could be operated at RH over the range of 85.8 - 99.1 % and 99.4 - 99.9 %, respectively. Both methods have accurate humidity control, but the optical detectors used to determine the wet particle size distribution are subjected to limitations in accuracy resolution due to uncertainties in refractive index and the conversion from optical to physical diameter. This leads to uncertainty in the measured growth factors of ~ 4% (Wex et al., 2005).

Mikhailov et al. (2011) developed a filter-based differential hygroscopicity analyzer (FDHA), which was employed as an offline method to investigate hygroscopic properties of ambient aerosol particles (Mikhailov et al., 2013, 2015). An updated version of the instrument allows measuring the hygroscopic growth up to 99.6 % with accuracy of  $\pm$  0.1 % RH. The uncertainty in the determination of the mass growth factors was estimated to be ~1 % at 30 % RH and ~10% at 99 % RH. FDHA measures water mass absorbed by aerosol particles deposited on the filters. Due to mass conservation, this method is not influenced by the effects of capillary condensation and restructuring of porous and irregularly shaped particles that usually limit the applicability and precision of mobility diameter-based HTDMA and CCNC (Cloud Condensation Nuclei Counter) experiments. Since FDHA is katharometer-based technique, it takes on average of 2 days, to measure one aerosol sample, which is a drawback of this instrument.

The authors state that activity coefficients can be determined from the data without the need to assume volume additivity by relying only on known (bulk) solution density. The authors should add that solution density is rarely known for metastable solutions and systems of interest to be studied with the HHTDMA.

On line 338 the following text has been added:

For many atmospheric aerosols, the concentration dependence of the aqueous solution density is not well defined. At the same time, for a number of model systems of interest, the aerosol solution density was measured in both unsaturated and supersaturated solutions. In this case  $x_w$  can be obtained without assumption of volume additivity by iteratively solving Eq. (8) with other equation where  $\rho$  and concentration is given explicitly.

The data should be made available in a FAIR aligned repository. Making data "available upon request to the author" is inconsistent with the AMT data policy (<u>https://www.atmospheric-measurement-techniques.net/about/data\_policy.html</u>).

The data are available at <a href="https://osf.io/87526/">https://osf.io/87526/</a>