

Response to referee #1

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

In this manuscript, the authors derived water activity and surface tension from hygroscopic growth of aerosol particles smaller than 100 nm composed of aqueous ammonium sulfate, D-glucose, and their mixtures. The hygroscopic growth of particles was measured over a wide range of RH from 2.0% to 99.6% using a high humidity tandem differential mobility analyzer (HHTDMA). The derived water activity and surface tension were compared with those from electrodynamic balance and bulk measurements and thermodynamic model predictions (E-AIM and UNIFAC). Overall, the manuscript is well written, and the topic fits the scope of Atmospheric Chemistry and Physics very nicely. I recommend the manuscript for publication after the authors address the following comments.

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

Major comment:

The authors derived water activity and surface tension using differential Kohler analysis, i.e., fitting parameters for the $s(D_s)$ dependence with the same g_b (i.e., Eq. 3). This approach assumes that water activity and surface tension depend on g_b only (i.e., independent of D_s). However, due to surface-bulk partitioning, both water activity and surface tension also depend on D_s (i.e., surface area to volume ratio). I would suggest that the authors include relevant discussions on how such dependence affects the accuracy of derived water activity and surface tension and conclusion of this study.

Line 122 and below, the following text has been added:

According to Eq.(3), DKA assumes that water activity and surface tension depend on g_b only (i.e. concentration). However, in case of bulk-surface partitioning, both water activity and surface tension may also depend on D_s . To minimize this effect, we used surface inactive compounds. Previous studies on the example of NaCl and $(\text{NH}_4)_2\text{SO}_4$ nanoparticles have shown that the size effect is negligible for such compounds (Bahadur and Russell, 2008; Cheng et al., 2015, Supplement)

New reference: Bahadur, R. and Russell, L. M.: Effect of surface tension from MD simulations on size-dependent deliquescence of NaCl nanoparticles, *Aerosol Sci. Technol.*, 42(5), 369-376, <https://doi.org/10.1080/02786820802104965>, 2008.

Line 326 and below, new text has been inserted instead of old one:

As mentioned above, DKA assumes that thermodynamic parameters depend only on concentration and are independent of particle size. Good agreement of DKA-derived a_w and σ with the literature data, showed that for particles in the size range of 20-100 nm, this approach is acceptable for single-component solutions of AS and GI and their mixtures, at least at moderate concentrations. Additional HHTDMA-DKA studies are needed to evaluate the accuracy of this approximation for nanoparticles containing surface-active molecules. We plan to conduct such studies to compare the DKA-derived a_w and σ with available experimental data and models that account for size-dependent bulk-surface partitioning.

Minor comments:

Line 188-189: The growth factor is a monotonic function of RH (first derivative is consistently above zero).

The text was modified as following:

The measurement data in Fig. 3b and Fig. 3d also demonstrate that for D-glucose and AS/GI (1:1) particles in the vicinity of $RH \approx 48\%$ the $g_b(RH)$ dependence has an inflection point, (derivative dg_b/dRH has extremum, see insert in Fig. 3b and Fig. 3c) indicating a distinct water sorption mechanism, and hence a different phase state of the particles before and after this RH point.

Line 205-210: Please provide more details on how the uncertainties are estimated.

Line 89, the following sentence has been added: A detailed calculation of growth factor uncertainty is described in Mikhailov et al., 2020 (Section 2.5).