We thank the reviewers for carefully reading our manuscript and for their valuable comments. Listed below are our responses in blue font addressing the general and specific comments from the reviewers of our manuscript.

Anonymous Referee #1

Summary: This manuscript reports measurements of the viscosity of internal mixtures of sucrose and ammonium sulfate as a function of relative humidity across a range of organic to inorganic mixing ratios. The results are compared with the predictions from the AIOMFAC-VISC model. Overall this is a nice piece of work applying established methods to quantify viscosity in a previously under-explored chemical system. The observations are clearly presented and the interpretations are well-supported. The manuscript is well-written and the figures are effective in conveying the relevant data.

General Comments of Referee #1

[1] One area that could warrant a deeper discussion and further expand the scope of this work is that of the induced efflorescence of supersaturated mixtures that are rich in AS. The assertion is that the needle allows nucleation of a crystal phase. In particles that also contain sucrose (Figure 3d), the solid phase that nucleates appears to be multiphase. Can the authors discuss the phase of these particles? Are they phase-separated (i.e. AS rich domains and sucrose-rich domains), gel particles (e.g. solid with aqueous / viscous fluid in the void space), well-mixed etc.?

[A1] Thank you for your comment. This is a good point! As we discussed in Sect. 3.1 (lines: 196 – 204), AS/H_2O particles effloresced in the RH range between ~50 and ~40% upon dehydration, which is a well-known ERH range of pure AS particles. At ~50% RH, a population of effloresced particles and non-effloresced particles coexisted on the substrate and when the needle poked the particles, all particles, including non-effloresced particles, cracked as shown in Fig. 1b. Although the act of poking the particles at a RH close to the ERH of AS may induce nucleation of a crystal, similar to the well-known process of contact freezing of supercooled cloud droplets (e.g., Ciobanu et al., 2010; Ladino et al., 2011; Hoose and Möhler,

2012). Based on the observed behaviour, the AS/H_2O particles were determined to be in a solid state for RH ≤ ~50%. To make it clearer, we have modified the paragraph to the following:

"Upon dehydration, AS/H_2O particles effloresced in the RH range between ~50 and ~40% (Fig. 3a), which is a well-known ERH range of pure AS (Winston and Bates, 1960). At ~50% RH, a population of effloresced particles and non-effloresced particles coexisted on the substrate, and when the needle poked the AS/H_2O particles, all particles including non-effloresced particles cracked (Fig. 1b). The act of poking non-effloresced particles at a RH close to the ERH of AS may induce the nucleation of an AS crystal, similar to the well-known process of contact freezing of supercooled cloud droplets (e.g., Ciobanu et al., 2010; Ladino et al., 2011; Hoose and Möhler, 2012). All particles, regardless of whether already effloresced or not, cracked when poked at a RH ≤ ~50%. Moreover, when we tried to poke the particles at ~55 % RH, the $\tau_{(exp,\ flow)}$ of the particles was fast, corresponding to a liquid-like physical state and flow behaviour.. Based on the observed contrasting behaviour at lower RH, the AS/H_2O particles were determined to be in a solid state for RH ≤ ~50%."

Regarding the sucrose/AS particles for an OIR of 1:4 (AS-rich particles), the optical imaging method does not allow us to conclusively determine the composition of each phase in those multiphase particles. Thus, while the presence of a crystalline AS phase is likely (compare Figs. 3a and 3d), it is unclear whether the remaining liquid forms a more structured gel state or an amorphous viscous semisolid or solid state. This ternary system may therefore be of interest for future studies employing other probing techniques and phase composition analysis. Moreover, the sucrose/AS particles did not show liquid-liquid phase separation upon dehydration because of the high O:C ratio of 0.92. Liquid-liquid phase separation in organic/inorganic aerosol particles occurs generally for O:C < 0.80 which is already well-known (Bertram et al., 2011; Song et al., 2012; You et al., 2014).

References:

Bertram, A., Martin, S., Hanna, S., Smith, M., Bodsworth, A., Chen, Q., Kuwata, M., Liu, A., You, Y., and Zorn, S.: Predicting the relative humidities of liquid-liquid phase separation, efflorescence, and deliquescence of mixed particles of ammonium sulfate, organic material, and water using the organic-to-sulfate mass ratio of the particle and the oxygen-to-carbon elemental ratio of the organic component, Atmos. Chem. Phys., 11, 10995-11006, https://doi.org/10.5194/acp-11-10995-2011, 2011.

Ciobanu, V. G., Marcolli, C., Krieger, U. K., Zuend, A., and Peter, T.: Efflorescence of ammonium sulfate and coated ammonium sulfate particles: Evidence for surface nucleation, Phys. Chem. A, 114, 9486-9495, https://doi.org/10.1021/jp103541w, 2010.

Hoose, C.and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, Atmos. Chem. Phys., 12, 9817-9854, https://doi.org/10.5194/acp-12-9817-2012, 2012.

Ladino, L., Stetzer, O., Lüönd, F., Welti, A., and Lohmann, U.: Contact freezing experiments of kaolinite particles with cloud droplets, Geo. Res. Atms, 116, https://doi.org/10.1029/2011JD015727, 2011.

Song, M., Marcolli, C., Krieger, U., Zuend, A., and Peter, T.: Liquid-liquid phase separation and morphology of internally mixed dicarboxylic acids/ammonium sulfate/water particles, Atmos. Chem. Phys., 12, 2691-2712, https://doi.org/10.5194/acp-12-2691-2012, 2012.

You, Y., Smith, M. L., Song, M., Martin, S. T., and Bertram, A. K.: Liquid–liquid phase separation in atmospherically relevant particles consisting of organic species and inorganic salts, Int. Rev. Phys. Chem., 33, 43-77, https://doi.org/10.1080/0144235X.2014.890786, 2014.

Winston, P. W.and Bates, D. H.: Saturated solutions for the control of humidity in biological research, Ecology, 41, 232-237, https://doi.org/10.2307/1931961, 1960.