Anonymous Referee #1

Received and published: 23 August 2018

This manuscript investigated the phase state of tetraol, which is an important secondary organic aerosol component. The authors synthesized teraol and studied the glass transition temperatures (Tg) of tetroal in pure and mixed particles with another important oxidation product, 3-MBTCA. This manuscript derived the phase diagram of water/tetraol mixtures at the atmospheric relevant conditions and determined the water activity of water/tetraol mixtures as a function of temperature and solute mass concentration. This study provides a set of valuable data for the phase state important SOA at different temperature and humidity. Due to the lack of experience on the synthesis, I was not able to provide assessment on this part. Besides that, the experimental methods are valid and the scientific approach and discussion are sound. The paper is well written and organized. I recommend it for publication for a minor revision. Please see the following comments which the authors may want to consider in the revision.

We thank the referee for her/his positive evaluation and the comments to improve our manuscript.

Minor comments:

1. P11, L17-22, It is not trivial to understand the Tg’ with currently short description. It will be useful to provide details how the Tg’ was determined.

We have added a paragraph describing what Tg’ is and how it can be measured (new p. 11 in marked version):

“The T_g’ of an aqueous mixture can be measured, when in samples with high to medium water mass fractions ice crystallization is the first phase transition that occurs upon cooling. In these cases, some of the water nucleates and forms small ice crystals. Because of this ice formation the remaining solution is depleted in water and becomes enriched in tetraol until the system reaches the ice/solution equilibrium, i.e. where the current temperature matches the freezing point of the remaining solution. When the temperature is decreased further, as is done in the experiments described above, the ice crystals continue to grow and the solution becomes more and more concentrated in tetraol thus increasing in viscosity. At some point the temperature is so low and the tetraol concentration becomes so large that the remaining solution forms a glass and no further ice crystal growth occurs due to the hindered macroscopic diffusion of water molecules in the now glassy phase. This temperature is termed T_g’ and marks the intersection of the ice melting point curve with the glass transition curve.”

It could be also useful to provide measured average values instead of Tg’ is about 200 K.

We added the exact mean Tg’ values for both stereoisomers to the caption of Figure 2:

“… is depicted as the mean Tg’ shifted towards the Gordon-Taylor fit (blue star) with a value of 197 K for *rac*-(2*R*,3*R*)-tetraol and 196 K for *rac*-(2*R*,3*S*)-tetraol.”

2. P13, L2-11, How does the Tg look like when using Tg(dry) from the spray-dried samples? It is suggested to derived Tg using both Tg(dry) from vacuum-dried and spray-dried samples, or provide uncertainties in Figure 2. Does the uncertainty in kGT cover this variation?

When the T_g(dry) is derived from spray-dried samples the T_g-curves change only slightly. For the *R,S*-racemate the k_{GT} value changes from 1.53 ± 0.08 to 1.68 ± 0.13, so both values agree within their respective uncertainty. For the *R,R*-racemate the k_{GT} value changes from 1.58 ± 0.05 to 1.84 ± 0.15. Even though this difference is slightly outside of the respective uncertainty range, it is within the uncertainty of the atmospherically more relevant representation of the Tg curve in water activity, see Figure 4. Nevertheless, following the suggestion of the referee we now add the k_{GT}-values derived when using the T_g(dry) from the spray-dried samples in the text (new p. 13 in marked version):

"For that reason, we used the T_g values of the vacuum-dried tetraols for the Gordon-Taylor analysis shown in Figure 2 to allow for a better comparability. Using the T_g derived from spray-dried samples of pure tetraol changes the derived Gordon-Taylor-curve only slightly yields a k_{GT} of 1.68 ± 0.13.
The partial phase diagram of *rac*-(2*R*,3*S*)-tetraol (Figure 2b) is very similar to that of the diastereomeric racemate. The glass transition curve fitting results in a k_{GT} of 1.58 ± 0.05 (1.84 ± 0.15 when using the T_g derived from a spray-dried sample of pure tetraol)."

L5-6, the maximum error in the water mass fraction is estimated to be 0.03 (a) and 0.05(b), do you mean in figure 2(a) and 2(b)?

Yes. This has been corrected (new p. 13 in marked version).

3. P14, L8-13, It will be great to provide the fitting parameters that could be directly used by the readers.

The fitting parameters are now provided in the supporting information where the fitting procedure is explained in detail (new Table S1 and corresponding text).

4. P16 section 3.4, is it possible to estimate the diffusion coefficient of water vapor using this water uptake/DSC experiment?

The kinetics of the observed diffusion process of water into the pure tetraol phase depends primarily on the diffusion coefficients of water and of tetraol, both of which depend upon temperature and composition of the tetraol/water mixture. Because the DSC-measurement was done in a sealed sample pan with no further water uptake, the spatial distribution of the components, i.e. the tetraol concentration and the thickness of the shell and the core all change during the course of the experiment, and so does temperature. For these reasons the system is underdetermined for extracting a back-of-the-envelope estimate of the diffusion coefficients from this single experiment. The latter would require a composition and space-resolved model that includes a parameterization of the composition- and temperature-dependent diffusion coefficients for the full range of mixing ratios of the two components, which in turn would require several experiments at different temperatures and starting compositions, which was – as already noted by referee 3 – beyond the scope for this paper and is a topic for future studies.