Anonymous Referee #3

Received and published: 8 September 2018

Review of "Physical state of 2-methylbutane-1,2,3,4-tetraol in pure and internally mixed aerosols" by Lessmeier et al. In this paper the authors 1) synthesized 2-methylbutane-1,2,3,4-tetraol, 2) measured the glass transition temperature of the tetraol, 3) determined a phase diagram for water/tetraol mixtures over the whole tropospheric temperature and RH range, 4) investigated water diffusion in the tetraol, and 5) measured the glass transition temperatures of mixtures of the tetraol with 3-methylbutante-1,2,3-tricarboxylic acid. This combined information was used to argue that isoprene-derived SOA will be a liquid in the lower troposphere and at high RH values, but a semisolid or even glassy state in the upper troposphere. The results have important implications for the growth and reactivity of atmospheric secondary organic aerosols. The paper combines first-rate physical chemistry and important atmospheric insight, and hence I strongly support this manuscript for publication in ACP. Due to the high quality of the manuscript, I have very few comments.
Below are a few comments I have that the authors may want to consider before publication. Since I am not an expert on synthesis, I do not have any comments on the synthesis part of the manuscript.

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

Minor comments:

1) Page 5, line 3. Can the authors confirm that there was no loss of material after the DSC sample pans were sealed? In other words, did the seal prevent evaporation or water uptake? I assume this can be checked from repeated DSC measurements.

The DSC sample pans are closed with an aluminum lid and then sealed with a crimper press, which is why they are called "hermetic pans". This sealing indeed allows no water evaporation or uptake which was confirmed by repeated DSC measurements and by checking the sample mass before and after the measurements via weighing. We have added the term "hermetically sealed" to that sentence.

2) Figure 5. The shell of the second sphere from the left looks aqua, not light green. Should the shell be light green to be consistent with the figure caption?

Yes, the referee is correct, thanks for pointing this out to us. We have revised the figure accordingly and also slightly modified the figure caption.

3) Figure 7a. Should the annotation on the arrow be changed to "diffusion into the material (without further uptake)" or "diffusion into the particles (without further uptake)"?

Yes, this is a very good and valid point: "into the material" is indeed a better description and we have changed the text accordingly, see new Fig.7a.

4) From the water uptake experiments (e.g. Figure 7) would it be possible to extract a diffusion coefficient. This is, obviously, beyond the scope of the current paper, but I am interested in the authors' response.

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 the referee – beyond the scope for this paper and is a topic for future studies.

5) Page 20, line 10 and Page 21, line 19-21. Can the authors comment on how the kgt they determined compares with the kgt assumed in the recent modelling study by Shiraiwa et al. 2017. Shiraiwa, M., Li, Y., Tsimpidi, A. P., Karydis, V. A., Berkemeier, T., Pandis, S. N., Lelieveld, J., Koop, T., and Poschl, U.: Global distribution of particle phase state in atmospheric secondary organic aerosols, Nature Communications, 8, 10.1038/ncomms15002, 2017.

We have added a short paragraph discussing this issue on new p. 22/23 in marked version:

"We note that the value of k_{GT} = 3.79 ±0.15 of the binary tetraol/3-MBTCA mixture determined here differs significantly from the value of k_{GT} = 1 assumed for multicomponent SOA mixtures in a recent modeling study (Shiraiwa et al., 2017). We also note that to our knowledge this tetraol/3-MBTCA mixture is the second binary mixture of individual SOA compounds investigated so far, the other being pinonic acid/3-MBTCA (Dette et al., 2014). Hence, it would be premature to conclude that these two binary systems are representative for the entire variety of atmospheric binary mixtures, which may also exhibit k_{GT}-values smaller than one. Therefore, as the different k_{GT} values may compensate each other in multicomponent mixtures a mean k_{GT} of one is not implausible, although our present study may be a first hint that k_{GT} may be slightly larger than 1."

6) Supplement, Page 4, line 13. Delete "we".
Thanks, "we" has been deleted.