Anonymous Referee #2

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The authors describe the physical phase states of 2-Methylbutane-1,2,3,4-tetraol and mixtures of tetrol which can be a marker for isoprene-derived SOA and alpha-pinene-derived SOA. The results provide new/additional insight for the phase state of SOA particles which is still unknown in the atmospheric chemistry community. This manuscript is clear, concise, and well-written. I recommend this manuscript for publication in ACP. I have several comments that the authors should consider prior to publication.

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

1. Page 2, line 19 – page 3, line 6; Page 15, line 8 – page 16, line 4: Recently, many research groups have focused on determination of phase states of isoprene-derived SOA and α-pinene-derived SOA particles using different techniques (i.e. measurements of viscosity, diffusion rate, evaporation rate, reactivity, and etc.) besides bounce experiments. Below are the relevant references. Maclean et al., and Song et al. showed that SOA particles produced from biogenic VOC are to be liquid even at dry condition at 290 K. Please add these references even more and compared the results in detail.

Maclean, A. M., Butenhoff, C. L., Grayson, J. W., Barsanti, K., Jimenez, J. L., and Bertram, A. K.: Mixing times of organic molecules within secondary organic aerosol particles: a global planetary boundary layer perspective, Atmos Chem Phys, 17, 13037-13048, 10.5194/acp-17-13037-2017, 2017.

Song, M., Liu, P. F., Hanna, S. J., Li, Y. J., Martin, S. T., and Bertram, A. K.: Relative humidity-dependent viscosities of isoprene-derived secondary organic material and atmospheric implications for isoprene-dominant forests, Atmos Chem Phys, 15, 5145-5159, 2015.

We thank the referee for pointing out to us these two references, which are now referred to at the appropriate places, i.e. on new page 2 and new page 15 in marked version.

2. Please provide the fitting parameters for Fig. 4. It would be useful for readers.

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

solutions at different concentrations and heating rates. (Sorbitol is a hexane-hexol and thus structurally similar to tetraol.) From these measurements we concluded a correction value of 0.93 K that is subtracted from the measured tetraol solution ice melting points. The water activity at the heating rate-corrected ice melting points was then calculated using equation S2.

The water activities at 25°C and those at the melting points for the different concentrations was then fitted using equation S3 (Zobrist et al., 2008), whereby the fit parameters *a, b* and *c* obtained from fitting equation S1 were kept fixed

$$a_w(w_2,T) = \frac{(1 + a \cdot w_2)}{(1 + b \cdot w_2 + c \cdot w_2^2)} + (T - T^\theta) \cdot (d \cdot w_2 + e \cdot w_2^2 + f \cdot w_2^3 + g \cdot w_2^4) \qquad \text{S3}$$

The last bracket in the equation is a fourth order polynomial function that is meant to describe the concentration dependence of the slope of the linear fit.

With these equations our measured glass transition temperatures at different mass fractions as well as the corresponding Gordon-Taylor fit can be converted from a mass fraction dependence to a water activity dependence. We note here that this transformation is not exact for several reasons. First we derived the temperature dependence of the water activity by fitting only two points for each concentration. Second we had to correct our ice melting points for a high heating rate which is a potential source for uncertainty. Third our data could not be fitted very well for the entire concentration range with the original fourth-order polynomial equation from Zobrist et al.: while the fourth order polynomial in equations S3 fitted the low tetraol concentration range very well, we obtained a better fit at medium tetraol concentrations with a second order polynomial (i.e. by setting parameters f and g to zero, see fitting parameter values in table S1). To take these uncertainties into account we took the following measures for the data transformation from the mass fraction dependence to the water activity dependence.

We did not transfer the actual Gordon-Taylor fit itself into the water activity regime but the 3σ range of the fit. Furthermore, we did the transformation with both the second order as well as the fourth order polynomial function and then overlapped the resulting ranges. With this procedure we derived a relatively broad glass transition range rather than an actual glass transition line. While this procedure reduces the precision of the glass transition temperature as a function of water activity, it enhances the certainty that the glass transition will take place within the range of glass transition temperatures depicted in figure 4.

Table S1. Fitting parameters of equation S3 used for figure 4 in the main paper.

rac-(2R,3R)-tetraol							
fitting parameter	a	b	c	d	e	f	g
4th order polynomial	-0.9996	-0.86678	0.06027	0.00973	-0.01623	-0.00169	0.00819
2nd order polynomial	-0.9996	-0.86678	0.06027	0.0056	-0.0056	0	0
rac-(2R,3S)-tetraol							
fitting parameter	a	b	c	d	e	f	g
4th order polynomial	-0.9997	-0.86188	0.04763	0.01384	-0.03261	0.02025	-0.00148
2nd order polynomial	-0.9997	-0.86188	0.04763	0.00631	-0.00631	0	0

Syntheses

General information

Materials used

The following chemicals were obtained from commercial suppliers and were used as received. The quality specified by the supplier is given in parentheses: (*S*)-1,1'-bi-2-naphthol (99%), *tert*-butanol (100%), calcium chloride (85%), citric acid monohydrate (> 99.5%), 2,2-dimethoxypropane (98%), Dowex® 50 WX4 (100 - 200 mesh, Sigma-Aldrich Chemie GmbH), lipase A *Candida antarctica* immobilised on Immobead 150, recombinant from *Aspergillus oryzae* (CAL-A, ≥ 500 U/g, product nr.:41658, Sigma-Aldrich Chemie GmbH), lithium aluminium hydride (for synthesis), maleic acid (99.99%), magnesium sulfate (> 99%), methanol (100%), (*E*)-2-methylbut-2-enedioic acid (99%), (*Z*)-2-methylbut-2-enedioic acid (99+%), *N*-methylmorpholine-*N*-oxide (97%), potassium carbonate (> 99.0%), potassium osmate dihydrate (for synthesis), potassium permanganate (99%), sodium hydroxide (99%), sodium sulphite (98%), concentrated sulfuric acid (95%), toluene (99.9%), vinyl butanoate (> 98.0%.).

Tetrahydrofuran (99.7%) was distilled from sodium/benzophenone prior its use. Solvents used for work-up and chromatography were of technical grade and were distilled prior their use. Deionized water was used for the syntheses.