Dr. Mingjin Tang
Associate Editor of Atmospheric Measurement Techniques

Dear Mingjin,

Listed below are our responses to the comments from the reviewers of our manuscript. For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. ***[1]***). Authors' responses are in blue below each referee statement with matching numbers (e.g. ***[A1]***). The revised text according to referees' comments is in green below each response made with line numbers in the original manuscript (e.g. ***[R1, lines 310-322]***). We thank the reviewers for carefully reading our manuscript and for their helpful suggestions!

Sincerely,

Allan Bertram
Professor of Chemistry
University of British Columbia

Anonymous Referee #1

Summary

The authors report on new measurement of the viscosity of aqueous erythtritol particles at different RH. One set of data is obtained by utilizing the rFRAP technique yielding the diffusivity of a large dye molecule and using the Stokes Einstein relationship to estimate viscosity; the other data are obtained by analyzing the shape relaxation of two particles coalescing in an optical tweezer setup. The two methods as well as previous bead mobility data agree within error. The new data are used to update a previous parametrization on how the addition of an OH functional group to a linear C4 carbon backbone effects viscosity.

This is a paper well suited for publication in AMT as we need more intercomparison between different measurement techniques to obtain particle viscosity data in the high viscosity range to get a better understanding on the limitations of the different techniques. The paper is well written, the figures illustrate the results adequately and the discussion is based on the experimental findings. I recommend publishing the paper, but ask the authors to consider the following comments.

General comments

[1] In section 3.2. the authors explain why they consider previous tweezers measurements compromised. They write this is due to the limited time resolution of the Raman spectroscopy method (being about 1 s). Since this being a technical paper, the reader would greatly benefit from seeing the corresponding "raw" data together with the raw brightfield imaging data versus time. I assume one would then appreciate that the transition to a spherical particle after coalescence is happing on a timescale to fast to be resolved with the Raman method.

[A1] In section 3.2, we have added the brightfield images as a function of time during the coalescence of two erythritol particles at $a_w = 0.04 \pm 0.02$. It demonstrates that the transition to a spherical particle occurred within 56 milliseconds, a timescale that is too short to be resolved by Raman spectral measurements (time resolution of 1 s). This information has been added to the text. ***[R1, lines 310-322]*** In the previous aerosol optical tweezers measurements at $a_w < 0.1$ (Song et al., 2016b), the timescale for relaxation to a sphere was estimated from two methods: the change in coalesced particle shape as recorded by the brightfield images and the reappearance of WGMs in the Raman spectrum. Figure 8 shows an example of captured brightfield images as a function of time after the coalescence of two erythritol particles at $a_w = 0.04 \pm 0.02$. The relaxation to a spherical particle occurred within 56 milliseconds, a timescale that is too short to be resolved by Raman spectral measurements (time resolution of 1 s, see Sect. 2.2). Therefore, previous erythritol viscosity measurements under dry conditions using the Raman spectral measurements (Song et al., 2016b) were compromised by the limited time resolution (1 s, equivalent to ~ 10^4 Pa s) and higher than those estimated from brightfield imaging, yielding an overestimate of the viscosity. Since the new aerosol optical tweezers measurements in this work are based solely on the brightfield images, they are more accurate than the previous results at $a_w < 0.1$ as a consequence of the higher time resolution of the brightfield imaging measurement compared to the Raman spectroscopy measurement. The viscosity at $a_w = 0.22 \pm 0.02$ reported by Song et al. (2016b) was based on brightfield images alone and those at $a_w \geq 0.43$ were based on back-scattered light intensity (where viscosities were < 10 Pa s, see Sect. 2.2).

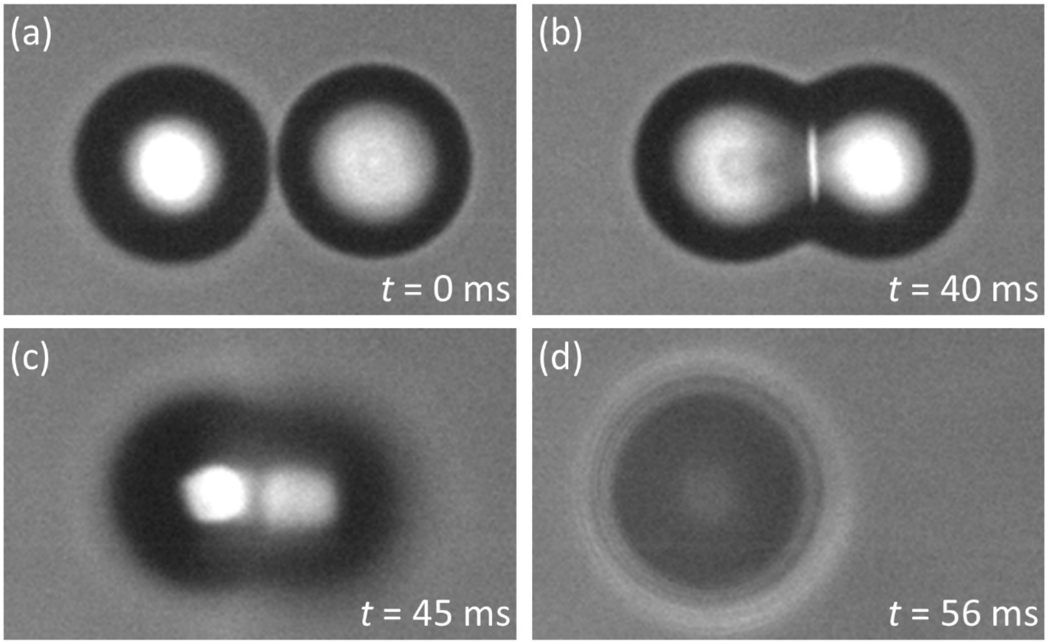

Figure 8. An example of the captured brightfield images as a function of time after the coalescence of two erythritol particles in optical tweezers at $a_w = 0.04 \pm 0.02$. The relaxation to a spherical particle occurred within 56 milliseconds.

[2] In the introduction, it is written that viscosity has implications for predicting size and mass distribution of SOA particles as well as implications for long-range transport of pollutants. I feel it should made more clear, that it is the diffusivity of a certain molecule in the viscous matrix which is the primary parameter needed for prediction not the viscosity as such. As the authors point out correctly, the Stokes-Einstein relation may fail for small molecules, but of course helps to estimate diffusivities.

[A2] We have revised the introduction to clarify that the diffusivity of molecules in particles plays a key role in predicting the size and mass distribution of SOA, affecting the reaction rates and long-range transport of pollutants.

[R2, lines 40-58] Secondary organic aerosol (SOA) is produced by the oxidation of volatile organic compounds followed by condensation of oxidation products (Hallquist et al., 2009). SOA contributes approximately 20 to 70% to the mass of fine aerosol particles, depending on location (Hallquist et al., 2009; Jimenez et al., 2009; Kanakidou et al., 2005; Zhang et al., 2007). Despite the abundance of SOA in the atmosphere, some physical and chemical properties of SOA remain poorly understood. An example is the diffusion of organic molecules within SOA particles (Cappa and Wilson, 2011; Mikhailov et al., 2009; Perraud et al., 2012; Reid et al., 2018; Vaden et al., 2011). Diffusion rates of organic molecules in SOA have implications for predicting the size and mass distribution of SOA particles (Lu et al., 2014; Saleh et al., 2013; Shiraiwa and Seinfeld, 2012; Zaveri et al., 2014, 2018). Diffusion rates of molecules in SOA also influence reaction rates (Berkemeier et al., 2016; Chu and Chan, 2017a, 2017b; Gatzsche et al., 2017; Hinks et al., 2016; Houle et al., 2015; Kuwata and Martin, 2012; Li et al., 2015; Lignell et al., 2014; Liu et al., 2018; Steimer et al., 2015; Wang et al., 2015; Wong et al., 2015; Zhou et al., 2012), the long-range transport of pollutants (Bastelberger et al., 2017; Shrivastava et al., 2017; Zelenyuk et al., 2012) and optical properties of SOA particles (Adler et al., 2013; Robinson et al., 2014). Diffusion may also have implications for the ice nucleating ability of SOA (Bodsworth et al., 2010; Ignatius et al., 2016; Ladino et al., 2014; Murray and Bertram, 2008; Schill et al., 2014; Wilson et al., 2012). Diffusion limitations in SOA have also been investigated through aerosol population mixing experiments, evaporation studies, and other approaches (Cappa and Wilson, 2011; Gorkowski et al., 2017; Liu et al., 2016; Perraud et al., 2012; Ye et al., 2016, 2018; Zaveri et al., 2018). In addition, researchers have used measurements of SOA viscosity together with the Stokes-Einstein equation to estimate diffusion rates of organics within SOA. The Stokes-Einstein equation is as follows:

$$D = k_\mathrm{B}T \,/\, (6\pi\eta R_\mathrm{H}), \qquad (1)$$

where D is the diffusion coefficient (m^2 s^{-1}), k_B is the Boltzmann constant (1.38×10^{-23} J K^{-1}), T is the temperature (K), η is the matrix viscosity (Pa s), and R_H is the hydrodynamic radius (m) of the diffusing species.

Technical comments

[3] Line 184: It might help to put the normalization factor into eq. (2) to see what is exactly meant by normalization.

[A3] We added a "*B*" parameter to the right-hand side of equation, where *B* represents the normalization factor. During the image fitting using a Matlab script, *B* was left as a free parameter and returned a value close to 1.

[R3, lines 175-190] The fluorescence intensity at position (x,y) and time t after photobleaching a rectangular area in a thin film can be described by the following equation (Deschout et al., 2010):

$$\frac{F(x,y,t)}{F_0(x,y,t)} = B\left\{1 - \frac{K_0}{4}\left[\mathrm{erf}\left(\frac{x+\frac{l_x}{2}}{\sqrt{r^2+4Dt}}\right) - \mathrm{erf}\left(\frac{x-\frac{l_x}{2}}{\sqrt{r^2+4Dt}}\right)\right] \times \left[\mathrm{erf}\left(\frac{y+\frac{l_y}{2}}{\sqrt{r^2+4Dt}}\right) - \mathrm{erf}\left(\frac{y-\frac{l_y}{2}}{\sqrt{r^2+4Dt}}\right)\right]\right\}, \qquad (2)$$

where $F(x,y,t)$ is the fluorescence intensity at position (x,y) and time t after photobleaching, $F_0(x,y)$ is the fluorescence intensity at position (x,y) prior to photobleaching, l_x and l_y are the lengths of the rectangular photobleached area, K_0 is related to the fraction of molecules photobleached in the bleach region, r is the resolution of the microscope, t is the time after photobleaching, and D is the diffusion coefficient of the fluorescent dye. B is a normalization constant, and "erf" is the error function.

Following the rFRAP experiments, individual images were fit to Eq. (2) using a Matlab script, with terms B and K_0 left as free parameters. The combined term of $r^2 + 4Dt$ was also left as a free parameter. Due to the normalization of images to a pre-bleached image, B returned a value close to 1, as expected. From Eq. (2), a value for the combined term $r^2 + 4Dt$ was obtained for each image taken after photobleaching. Next, $r^2 + 4Dt$ was plotted as a function of time after photobleaching, and a straight line was fit to the plotted data. An example plot of $r^2 + 4Dt$ versus t and a linear fit to the data are shown in Fig. 3. Diffusion coefficients were determined from the slope of the fitted line. The diffusion coefficient at each a_w reported in Sect. 3 is the average of at least four measurements.

[4] Line 337 onwards: I do not understand this regression. As the viscosity of pure water is precisely known and an exponential dependence of viscosity with viscosity is assumed for the regression there is only one free parameter, namely the slope in Fig.8. The intercept follows from slope and the value of pure water. Of course, this leads to an uncertainty for the viscosity for the pure erythritol.

[A4] The linear fit in Fig. 9 (which was Fig. 8 in the original manuscript) was based on the orthogonal distance regression (or total least squares) fitting algorithm using IGOR Pro 6, which was weighted based on the x and y uncertainties of each data point. The algorithm assigns a greater weight to the viscosity of water ($a_w = 1$) than to the other data points ($a_w < 1$) in Fig. 9 since the uncertainty in the viscosity of pure water is small. Both the slope and y-axis intercept were left as free parameters. To address the referee's comment, more information has been added regarding the regression.

[R4, lines 337-340] To determine the viscosity of pure erythritol under dry conditions (at $a_w = 0$), a straight line was fit to the data in Fig. 9 based on the orthogonal distance regression-fitting algorithm using IGOR Pro 6 and then extrapolated to $a_w = 0$. This algorithm weighted the fit based on the x and y uncertainties of each data point. The viscosity of pure water ($a_w = 1$) is well constrained (Korson et al., 1969), giving it a larger weighting than data points at $a_w < 1$. The intercept on the y-axis was 2.27 ± 0.22 (two standard deviations), corresponding to a viscosity of pure erythritol of 184^{+122}_{-111} Pa s.

Anonymous Referee #2

Summary

A new method for determining the viscosity of bulk solutions using rectangular area fluorescence recovery after photobleaching (rFRAP) is presented. This is a useful and relatively simple way to determine viscosity, which has been a major topic regarding the viscosity of organic atmospheric aerosol particles. We do need more and more reliable techniques to determine viscosity (and really diffusivity), so this is a valuable contribution to the atmospheric chemistry community. The authors focus on erythritol as it is a proxy for polyalcohols that form from the hydrolysis of IEPOX (an isoprene oxidation product) in the atmosphere, and due to a large disagreement in its viscosity previously reported using very different techniques (bead mobility, and optical tweezers). I appreciate their honesty in re-evaluating their prior optical tweezers experiments and acknowledging unrecognized issues in the viscosity measurements under dry conditions due to limited time resolution of the measured signal. I recommend the manuscript for publication in AMT, but would like to see the following questions and comments addressed first to further improve the clarity and quality of the manuscript.

General comments

[5] I do agree with the other referee that it is really important to clarify that it is diffusivity and not viscosity that is the key property that governs all the important processes they discuss such as vapor uptake, evaporation, reactive uptake, etc. Viscosity and diffusivity are related, but the relationship breaks down in highly viscous systems, as the authors discuss. Please discuss the key role of diffusivity instead of focusing on viscosity.

[A5] Please see our response ***[A2]*** and the revised text ***[R2, lines 40-58]***.

[6] The concept of photobleaching needs a more thorough explanation, since it is key to this technique, and very new to the atmospheric science community. It appears to be an irreversible process? This is important to explain. If the fluorophore can regenerate that would require a different approach using this technique, since the fluorescent recovery is entirely attributed to diffusional transport of new fluorophores into the optical volume. Regeneration of the fluorophore does not seem to be the case however. So what is it that causes the photobleaching? Photolysis at this long laser wavelength seems unlikely, unless there is extensive vibrational overtone excitement taking place (unlikely). Does the laser produce a reactant that reacts with the fluorophore? Even if the mechanism isn't entirely understood more explanation of the photobleaching process is required.

[A6] Photobleaching of the fluorescent dye (i.e., RBID) in our rFRAP experiments was an irreversible process, possibly due to the reaction between molecular oxygen and RBID molecules in their excited singlet or triplet states causing permanent destruction of the fluorophore (Song et al., 1995; Widengren and Rigler, 1996). The excited singlet state is achieved via the absorption of photon energy (from the laser) by RBID molecules in the ground state. The triplet state is reached via intersystem crossing of RBID molecules in their excited singlet state. In section 2.1.2 we have added discussion regarding the photobleaching mechanism.

[R6, lines 151-156] In the rFRAP experiments, a confocal laser scanning microscope was used to photobleach RBID molecules in a small volume of the thin film. The photobleaching process occurs as follows. First, RBID molecules in the ground state absorb photons generated by the 543 nm laser and undergo an electronic transition from the ground state to the excited singlet state. Next, these excited molecules can either relax to the ground state via the emission of a photon (this process is fluorescence, which does not result in photobleaching), or undergo intersystem crossing from the excited singlet state to the excited triplet state. In the excited singlet or triplet state, RBID molecules may react with molecular oxygen, resulting in permanent destruction of the fluorophore (Song et al., 1995; Widengren and Rigler, 1996). After photobleaching, a gradual recovery of fluorescence within the photobleached region occurred due to the diffusion of unbleached fluorescent molecules from outside the bleached region into the bleached region. The diffusion coefficient of the fluorescent dye was determined by monitoring the time-dependent recovery of the fluorescence intensity using the same confocal laser scanning microscope used for photobleaching.

Specific comments

[7] Line 204-206: Related to this, the concept of "reversible photobleaching" needs to be explained. Do you have any direct evidence that this does /not/ occur in your experiments?

[A7] To address the referee's comment the discussion about reversible photobleaching in our experiments (originally in the Supporting Information) has been moved to the main text. Figure S3 (Supporting Information) shows that reversible photobleaching is not important in our experiments.

[R7, lines 205-208] An additional possible mechanism is reversible photobleaching (or photoswitching), where the fluorescent molecules convert between a fluorescent and a non-fluorescent state without being permanently photobleached (Fukaminato, 2011; Long et al., 2011; Sinnecker et al., 2005). To determine if reversible photobleaching was responsible for the recovery of fluorescence in the photobleached region, experiments with small droplets (10–30 µm in diameter) containing erythritol, water, and trace amount of RBID (approximately 0.3 weight percent) were carried out. In these experiments, we uniformly photobleached the entire droplet, resulting in ~ 30% reduction in fluorescence intensity. Uniform bleaching ensures that the diffusion of fluorescent RBID molecules will not result in a change in fluorescence intensity. After bleaching, the average fluorescence intensity of the entire droplet was monitored over time, as shown in Figure S3 (Supporting Information). The fluorescence intensity remained constant within the uncertainty of the measurements, indicating that reversible photobleaching was not an important mechanism in our rFRAP experiments.

[8] Line 60 on: Another important way that viscosity and the diffusion limitations this may create in organic aerosols has been assessed is through aerosol population mixing experiments, optical tweezers, and other related methods. These experiments test for diffusion limitations that would prevent prompt mixing of organic components through evaporation and then re-dissolution into the aerosol phase. Please cite and discuss these approaches as well. (Gorkowski et al., 2017; Liu et al., 2016; Ye et al., 2016, 2018).

[A8] To address the referee's comment, discussions regarding aerosol population mixing experiments, aerosol size change and other related methods has been added to the manuscript. Please see the revised text ***[R2, lines 40-58]***.

[9] Like 77: Should state that tetrols are important components of atmospheric aerosols as they are hydrolysis products of IEPOX, which is a major oxidation product of isoprene that has been extensively studied recently. This will help to better motivate this study and focus on erythritol.

[A9] The formation of tetrols via IEPOX hydrolysis has been added. Please see the revised text below.

[R9, lines 76-79] This also led to uncertainties regarding the viscosity of tetrols, which have been observed in ambient SOA particles and SOA particles generated in environmental chambers (Claeys, 2004; Edney et al., 2005; Surratt et al., 2006, 2010). An important formation pathway for tetrols is the hydrolysis of isoprene epoxidiol (IEPOX). IEPOX has been identified as a key intermediate during the oxidation of isoprene, an SOA precursor (Guenther et al., 2006; Surratt et al., 2010).

[10] Eqn 2: Is "erf" the error function used to describe a lognormal population? Please define so this is clear.

[A10] Yes. We have added a definition of "erf" in the text. Please see the revised text ***[R3, lines 175-190]***, right below the response ***[A3]***.

[11] Line 184: If r is the microscope's resolution, why is r^2 a free parameter? Shouldn't the microscope's resolution be fixed and known? Or is just combined as one free term as r^2 + 4Dt?

[A11] During the image fitting process using equation (2), $r^2 + 4Dt$ was combined as one term and left as a free parameter. Please see the revised text ***[R3, lines 175-190]***, right below the response ***[A3]***.

[12] Line 210: Is it aerosol optical tweezers or tweezer? I am used to seeing "tweezers" in the literature.

[A12] We agree that "tweezers" should be consistently used in the manuscript rather than its singular form. We have changed the terminology throughout the manuscript.

[13] Line 260: Much more discussion of why adding an –OH group to the carbon backbone would increase viscosity is needed. This will be unclear to non-experts. Please discuss in terms of the intermolecular interactions the alcohol groups create, as intermolecular interactions are what determine viscosity, and to a large extent diffusivity as well. Please also discuss/speculate, again using a structure-activity relationship perspective, why the effect might diminish after the 3rd -OH group is added. What could explain these diminishing returns? A nice concise summary of what is known regarding how structure influences viscosity, especially for -OH and similar functional groups, would be a really helpful addition here.

[A13] For the first part of the comment, more discussion on the viscosity increase due to addition of an –OH group has been added to the text. Specifically, we focus on the role of hydrogen bonding between alcohol and polyol molecules in affecting the intermolecular force and thus viscosity. This effect of functionality on viscosity has also been observed for carboxyl groups (–COOH) (Rothfuss and Petters, 2017; Song et al., 2016). For the second part of the comment, the effect of –OH group addition on viscosity slightly strengthens rather than diminishes for the fourth –OH group addition, as the viscosity sensitivity parameter (S_η) at $N = 4$ is slightly higher than those at $N = 1 – 3$ (Fig. 11). The reason for this small increase for the fourth –OH group is unclear to us, and we would prefer to not speculate at this point.

[R13, lines 343-351] Grayson et al. (2017) previously estimated the effect of adding OH functional groups on the viscosity of a linear C_4 compound. Here we repeat this analysis (Fig. 10) based on the updated viscosity of pure erythritol (184^{+122}_{-111} Pa s) determined above. For those compounds with the same number but different positions of OH functional groups, the average of their viscosities was taken from the literature (Grayson et al., 2017; Rothfuss and Petters, 2017; Song et al., 2016b). Table S4 (Supporting Information) lists the values and sources of literature data used. The data in Fig. 10 were fit to a linear equation, resulting in a slope of 1.43 ± 0.08 (two standard deviations), which indicates that the viscosity of a linear C_4 molecule increases on average by a factor of 27^{+6}_{-5} per addition of an OH functional group. The increase in viscosity with the addition of an OH functional group to a linear C_4 backbone is attributed to the increased number of hydrogen bonds (H–O···H) formed between adjacent molecules, as discussed previously (Rothfuss and Petters, 2017). Similarly, the addition of carboxyl groups (–COOH) leads to an increase in viscosity due to enhanced formation of intermolecular hydrogen bonds (Rothfuss and Petters, 2017).

[R13, lines 359-363] The relationship between S_η and N is shown in Fig. 11 for a linear C_4 carbon backbone. S_η is between 0.7 and 1.9 for $N = 1 – 3$. On the other hand, S_η is between 1.6 and 2.5 for $N = 4$, suggesting S_η increases with the addition of the fourth OH functional group to the linear C_4 carbon backbone. However, additional studies are needed in order to reduce the uncertainties of the measurements and make stronger conclusions.

[14] Fig. 5: Why weren't more a_w values studied, especially at the higher range to see how consistently the trend holds? Measurements in the apparent transition region of ~0.05-0.2 a_w would be really useful.

[A14] We have performed additional experiments at $a_w = 0.153 \pm 0.025$ using the rFRAP technique. The additional measurement was consistent with the trend of RBID diffusion coefficients and erythritol-water particle viscosity. The revised Fig. 5 is shown below. Related contents (e.g. updated Figs. 9–11) in the manuscript and SI have also been revised.

[R14, lines 744-751]

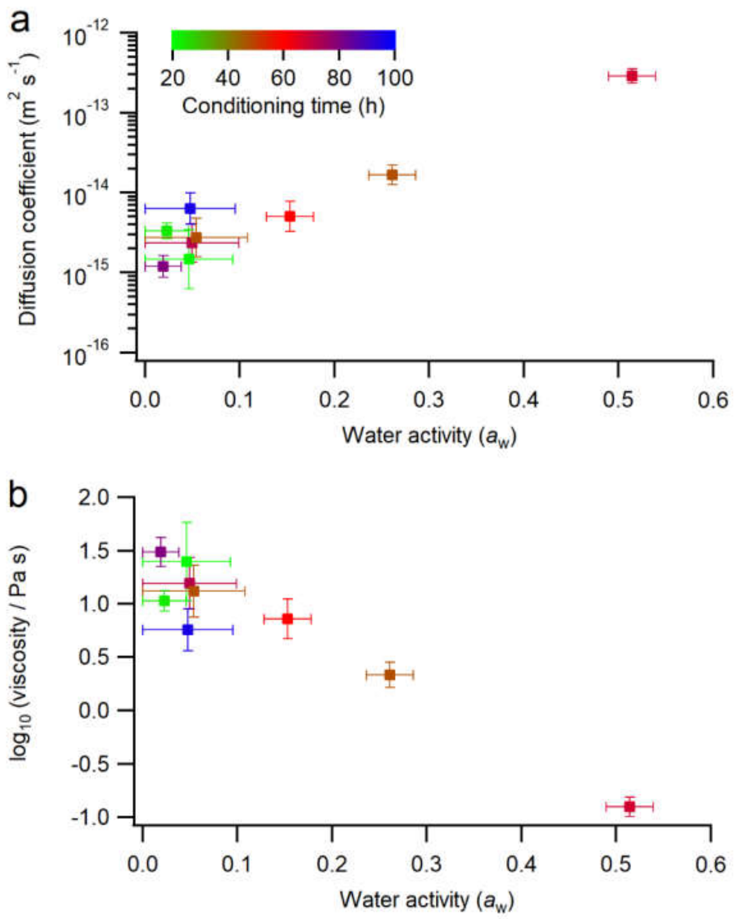

Figure 5. (a) The measured diffusion coefficients of RBID as a function of a_w. (b) The viscosity of erythritol-water particles as a function of a_w based on the measured RBID diffusion coefficients and the Stokes-Einstein equation. Results from rFRAP measurements are color-coded by the sample conditioning time prior to the rFRAP experiments. The color scale applies to both panel (a) and (b). Horizontal error bars indicate the upper and lower limits of a_w. Vertical error bars correspond to two standard deviations of diffusion coefficient (in panel a) and $\log_{10}$ (viscosity / Pa s) (in panel b).

[15] Fig. 8: The aerosol optical tweezers results seem consistently biased higher compared to the bead-mobility and rFRAP results presented here. Please discuss. I don't see the "reasonable agreement" (line 335) between all three methods (excluding the old optical tweezers data) that is stated.

[A15] The mean values from aerosol optical tweezers measurements were higher than those from rFRAP and bead-mobility measurements, but when considering the error bars (which equal two standard deviations) the results from three different techniques differed from one another only slightly at $a_w < 0.4$, and agreed with one another at $a_w > 0.4$. The text has been revised accordingly. Note that the original Fig. 8 is Fig. 9 in the revised manuscript.

[R15, lines 326-335] In Fig. 9, we have summarized the previous and current measurements of the viscosity of erythritol-water particles as a function of a_w. The black triangles represent measurements by Grayson et al. (2017) using the bead-mobility technique. The blue squares represent the rFRAP results from this work, where experimental data at similar a_w have been binned together so as not to give extra weight to the rFRAP data. The red circles indicate aerosol optical tweezers measurements from Song et al. (2016b) (open circles) and this study (solid circles). The previous measurements at $a_w \leq 0.1$ by Song et al. (2016b) were excluded from Fig. 9, because the new aerosol optical tweezers measurements reported in this study at $a_w \leq 0.1$ are thought to be more accurate. At $a_w > 0.4$, the viscosity measurements from the bead-mobility, rFRAP, and optical tweezers techniques are in reasonable agreement, if the experimental uncertainties are considered. At $a_w < 0.4$, the mean viscosity values determined using optical tweezers are higher than those from rFRAP and bead-mobility measurements by 1 – 2 orders of magnitude. The error bars (two standard deviations) overlap in some, but not all, cases. Nevertheless, the disagreement in viscosity measured using multiple techniques seen here is smaller than reported previously.

Works cited:

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