Viscosity of erythritol and erythritol–water particles as a function of water activity: new results and an intercomparison of techniques for measuring the viscosity of particles

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Abstract. A previous study reported an uncertainty of up to 3 orders of magnitude for the viscosity of erythritol (1,2,3,4-butanetetrol)–water particles. To help reduce the uncertainty in the viscosity of these particles, we measured the diffusion coefficient of a large organic dye (rhodamine B isothiocyanate–dextran, average molecular weight \(\sim 70000\ \text{g mol}^{-1}\)) in an erythritol–water matrix as a function of water activity using rectangular-area fluorescence recovery after photobleaching (rFRAP). The diffusion coefficients were then converted to viscosities of erythritol–water particles using the Stokes–Einstein equation. In addition, we carried out new viscosity measurements of erythritol–water particles using aerosol optical tweezers. Based on the new experimental results and viscosities reported in the literature, we conclude the following: (1) the viscosity of pure erythritol is \(184^{+122}_{-73}\ \text{Pa s}\) (2 standard deviations); (2) the addition of a hydroxyl (OH) functional group to a linear C4 carbon backbone increases the viscosity on average by a factor of \(27^{+6}_{-5}\) (2 standard deviations); and (3) the increase in viscosity from the addition of one OH functional group to a linear C4 carbon backbone is not a strong function of the number of OH functional groups already present in the molecule up to the addition of three OH functional groups, but the increase in viscosity may be larger when the linear C4 carbon backbone already contains three OH functional groups. These results should help improve the understanding of the viscosity of secondary organic aerosol particles in the atmosphere. In addition, these results show that at water activity \(> 0.4\) the rFRAP technique, aerosol optical tweezers technique, and bead-mobility technique give results in reasonable agreement if the uncertainties in the measurements are considered. At water activity \(< 0.4\), the mean viscosity values determined by the optical tweezers technique were higher than those determined by the bead-mobility and rFRAP techniques by 1–2 orders of magnitude. Nevertheless, the disagreement in viscosity measured using multiple techniques reported in this paper is smaller than reported previously.

1 Introduction

Secondary organic aerosol (SOA) is produced by the oxidation of volatile organic compounds followed by the 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., 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
Researchers have investigated the dependence of viscosity on the number of hydroxyl (OH) functional groups on a carbon backbone and found that viscosity increased, on average, by a factor of 22–45 following the addition of an OH functional group to linear C$_4$, linear C$_5$, branched C$_5$, and linear C$_6$ carbon backbones. However, the study by Grayson et al. (2017) revealed a large discrepancy between the viscosity of erythritol (1,2,3,4-butanetetrol) measured with the bead-mobility technique (Grayson et al., 2017) and measured with the aerosol optical tweezers technique (Y. C. Song et al., 2016) at ≤ 25% relative humidity (RH). This led to uncertainties when predicting the effect of adding OH functional groups to a linear C$_4$ carbon backbone on viscosity. This also led to uncertainties regarding the viscosity of tetroils, 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 tetroils is the hydrolysis of isoprene epoxidial (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).

To help reduce the uncertainty in the viscosity of erythritol–water particles, we measured the diffusion coefficients of a large organic dye (rhodamine B isothiocyanate–dextran, referred to as RBID in the following; average molecular weight ~ 70 000 g mol$^{-1}$) in erythritol–water matrices as a function of water activity ($a_w$) using the rectangular-area fluorescence recovery after photobleaching (rFRAP) technique (Deschout et al., 2010). The diffusion coefficients were then converted to viscosities using the Stokes–Einstein equation (Eq. 1). RBID has a hydrodynamic radius that is more than 16 times larger than that of erythritol (Table 1 and Fig. 1). We assume that the viscosity of an erythritol–water particle can be accurately calculated from the diffusion coefficient of RBID and the Stokes–Einstein equation, since the Stokes–Einstein equation accurately predicts diffusion coefficients when the diffusing molecules are large in size relative to the matrix molecules and when the matrix viscosity is comparable to or less than 10$^5$ Pa s (Chenyakin et al., 2017; Price et al., 2016), which is the case for erythritol–water particles (Grayson et al., 2017; Y. C. Song et al., 2016).

In addition to determining viscosities from the rFRAP diffusion measurements, we carried out new viscosity measurements for erythritol–water particles at $a_w < 0.1$ using the aerosol optical tweezers technique. The new viscosity results

Table 1. The molar masses ($M_w$) and hydrodynamic radii ($R_H$) of erythritol and rhodamine B isothiocyanate–dextran (RBID), which are used as the matrix and diffusing fluorescent dye in this work, respectively.

| Compound                        | $M_w$ (g mol$^{-1}$) | $R_H$ (Å) | References                      |
|---------------------------------|----------------------|-----------|---------------------------------|
| Erythritol                      | 122.12               | 3.4 ± 0.3 | Kiyosawa (1991)                 |
|                                 |                      |           | Schultz and Solomon (1961)      |
| Rhodamine B isothiocyanate–dextran (RBID) | 70 000 (on average) | 59 ± 1    | Floury et al. (2015)            |
|                                 |                      |           | Paës et al. (2017)              |

In addition to determining viscosities from the rFRAP diffusion measurements, we carried out new viscosity measurements for erythritol–water particles at $a_w < 0.1$ using the aerosol optical tweezers technique. The new viscosity results
The solubility of erythritol in water at 293 K is \(\sim 38\) wt % (Haynes, 2015), which corresponds to an \(a_w \approx 0.92\), based on Raoult’s law and assuming erythritol does not dissociate in water (Koop et al., 2011). In our experiments, all thin films were conditioned at \(a_w < 0.92\) and were therefore supersaturated with respect to crystalline erythritol. To prepare these supersaturated thin films, a bulk solution containing 20 wt % erythritol in water and 0.056 wt % (0.01 mmol L\(^{-1}\)) RBID was prepared gravimetrically. The prepared bulk solution was then filtered using a 0.45 µm Millex\textsuperscript{®}-HV syringe filter unit (Millipore Sigma Ltd., Etobikoe, ON, Canada) to eliminate solid impurities. Next, the solution was nebulized onto a siliconized hydrophobic glass slide (22 × 22 mm, VWR, Radnor, PA, USA), which had been rinsed with Milli-Q\textsuperscript{®} water (18.2 MΩ cm). This resulted in droplets with radii ranging from 100 to 185 µm on the hydrophobic glass slide. For most of the experiments, the slide holding the droplets was then transferred into a flow cell in an inflatable glove bag (Glas-Col, Terre Haute, IN, USA) for conditioning at a particular \(a_w\). For some of the experiments, the slide holding the droplets was placed in a sealed container above a saturated inorganic salt solution with a known \(a_w\). A handheld hygrometer (OMEGA, Norwalk, CT, USA) with an accuracy of \(\pm 2.5\) % was used to measure the RH at the flow cell outlet and in the glove bag or above the bulk solutions. The \(a_w\) was calculated from the measured RH (\(a_w = \text{RH(\%)/100}\)) (Seinfeld and Pandis, 2006). The time used for conditioning droplets ranged from 21.5 to 96 h. See Sect. S1 and Table S1 in the Supplement for details. After conditioning the droplets to a known \(a_w\) and creating the thin films, the concentration of RBID in the thin films ranged from 0.2 to 0.3 wt %. At this concentration, the fluorescence intensity of the thin films was proportional to the RBID concentration (Sect. S2 and Fig. S2 in the Supplement).
nucleation of organic crystals (Bodsworth et al., 2010; Pant et al., 2004, 2006; Wheeler and Bertram, 2012; Yeung et al., 2009).

2.1.2 rFRAP technique and data extraction

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.

In this work, a Zeiss LSM510 confocal laser scanning microscope with a 10× objective and 0.30 numerical aperture was used. The pinhole was set at 120 µm. Photobleaching was performed using a helium–neon laser with an emission wavelength of 543 nm, at a power of 330 µW. The photobleached area ranged from 3 × 3 to 6 × 6 µm². The bleach parameters, including the number of iterations and scanning speed of the laser, were adjusted to obtain a reduction in fluorescence intensity of ∼ 30 % in the bleached region, as recommended by Deschout et al. (2010). After photobleaching, images were taken using the same laser at a power of 4 µW, at time intervals ranging from 2 to 30 s, depending on the speed of recovery of the fluorescence signal, with longer intervals used when the fluorescence recovery was slower. All experiments were performed at room temperature (292–294 K).

Figure 2 shows an example of a series of images recorded during an rFRAP experiment. All of the images recorded after photobleaching were normalized against an image captured prior to photobleaching, using ImageJ software. After normalization, the images were downsized from 512 × 512 pixels to 128 × 128 pixels by averaging pixels to reduce the level of noise (Chenyakin et al., 2017).

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):

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

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 bleached 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 parameters, including the number of iterations and scanning speed of the laser, were adjusted to obtain a reduction in fluorescence intensity of ∼ 30 % in the bleached region, as recommended by Deschout et al. (2010).
In these experiments, we uniformly photobleached the entire photobleached region, experiments with small droplets (10–30 µm in diameter) containing erythritol, water, and trace amounts of RBID (approximately 0.3 wt %) were carried out. The red line represents a linear fit to the data.

Cross-sectional views of the fluorescence intensity along the x axis for different times after photobleaching are shown in Fig. 4. The cross sections for the measured intensities (blue dots) were generated by averaging the normalized fluorescence intensities \( \frac{F}{F_0} \) over the width of the photobleached region in the y direction, at each position x. Calculated cross sections of the fluorescence intensities (red lines) were generated from fits of Eq. (2) to the experimental data. The close agreement between the measured and calculated cross sections illustrates that Eq. (2) describes our experimental data well.

Equation (2) was derived by assuming no diffusion in the axial direction (i.e., z direction). Deschout et al. (2010) have shown that Eq. (2) gives accurate diffusion coefficients when the numerical aperture of the microscope is low (\( \leq 0.45 \)) and the thickness of the films is small (\( \leq 120 \mu m \)), consistent with the numerical aperture of 0.30 and film thickness of 30–50 µm used in our experiments. Equation (2) also assumes that the only mechanism for recovery in the photobleached region is diffusion. 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; Sinneker 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 amounts of RBID (approximately 0.3 wt %) were carried out. In these experiments, we uniformly photobleached the entire droplet, resulting in \( \sim 30 \% \) 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 Fig. S3 in the Supplement. The fluorescence intensity remained constant within the uncertainty of the measurements, indicating that reversible photobleaching was not an important mechanism in our rFRAP experiments.

2.2 Aerosol optical tweezers

The application of the aerosol optical tweezers technique to measure the viscosity of aerosol particles has been discussed in detail in previous publications (Bzek et al., 2016; Y. C. Song et al., 2016) and will only be briefly reviewed here. Two optical traps are formed using a holographic optical tweezers instrument equipped with a laser at 532 nm (Laser Quantum Opus 3W). The holographic arrangement uses a spatial light modulator (Hamamatsu, X10468) to encode phase information into the expanded laser-light wavefront, creating an interference pattern in the trapping plane that resembles two tightly focused beams. Aerosol droplets are captured from a cloud of aerosol generated from a medical nebulizer and introduced into a RH-controlled trapping cell with the RH recorded by a capacitance probe (Honeywell, HIH-4202A). Typical particle diameters are 9–16 µm. Experiments were performed at room temperature of 293 K. Droplet sizes and refractive indices are inferred from the discrete wavelengths commensurate with whispering gallery modes (WGMs) that are observed in the Raman scattering fingerprints recorded from the two droplets. Particle size and refractive index are estimated from comparison with calculated WGM wavelengths using Mie scattering theory and can be determined with an accuracy of \( \leq \pm 2 \) nm and \( \leq \pm 0.0005 \), respectively (Preston and Reid, 2013).

Following a conditioning period of many hours, identified by a steady droplet size over a period of tens of minutes, the particles are coalesced by manipulating the optical trap positions. Once brought into contact, the shape of the composite particle relaxes over a timescale of microseconds to hours, dependent on the viscosity. One of three methods is then chosen to infer particle viscosity from the shape relaxation based on the relaxation timescale:

1. For relaxation timescales of \( < 1 \) ms (equivalent to viscosities \( < 10 \) Pa s) (Power and Reid, 2014), the time dependence of the backscattered light intensity can be used to monitor the change in shape using a silicon photodetector (Thorlabs, DET 110) and oscilloscope (LeCroy, HDO 6034-MS). At timescales longer than this, the movement of the trapped particle relative to the laser beam focus (i.e., the relaxation in trapped position) contributes to the change in light-scattering signal and becomes convoluted with the change arising from the relaxation in shape. Thus, light-scattering measure-
Figure 4. Cross-sectional view of the normalized fluorescence intensities ($F/F_0$) along the $x$ axis during an rFRAP experiment. Blue dots correspond to the measured data, while the red lines represent fits to the experimental data using Eq. (2). The sample films were conditioned at $a_w = 0.023 \pm 0.023$. RBID concentration in the conditioned films was approximately 0.3 wt %. Panel (a) shows the cross section prior to photobleaching. $F/F_0$ equals 1 because the image was normalized against itself. Panels (b–f) show the cross sections at 0, 360, 720, 1080, and 1440 s after photobleaching, corresponding to frames (b–f) in Fig. 2.

3 Results and discussion

3.1 Diffusion coefficients in and viscosities of erythritol–water particles as functions of $a_w$ measured by the rFRAP technique

Shown in Fig. 5a and listed in Table S2 (Supplement) are the measured diffusion coefficients of RBID in erythritol–water matrices as a function of $a_w$. The diffusion coefficient decreased by 2–3 orders of magnitude as $a_w$ decreased from approximately 0.5 to 0. This decrease in the diffusion coefficients with a decrease in $a_w$ is due to the plasticizing effect of water (Koop et al., 2011; Power et al., 2013).

The Stokes–Einstein equation and measured diffusion coefficients were used to calculate the viscosity of erythritol–water particles. It has been found that the Stokes–Einstein equation significantly underestimates the diffusion coefficients of small molecules such as water and ozone within a matrix containing larger molecules (Bastelberger et al., 2017; Davies and Wilson, 2016; Li et al., 2015; Marshall et al., 2016; Price et al., 2014; Shiraiwa et al., 2011). On the other hand, as discussed in Sect. 1, the Stokes–Einstein equation gives accurate diffusion coefficients when the diffusing species is similar in size to or larger than the matrix molecules and when the viscosity of the matrix is comparable to or lower than $10^4$ Pa s (Chenyakin et al., 2017; Price et al., 2016). Hence, in this study, we assume that the viscosity of erythritol–water particles can be accurately calculated using the measured RBID diffusion coefficient and the
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 panels. Horizontal error bars indicate the upper and lower limits of $a_w$. Vertical error bars correspond to 2 standard deviations of diffusion coefficient (a) and log_{10}(viscosity / Pa s) (b).

Stokes–Einstein equation, because RBID is much larger than the matrix molecules (Table 1) and the highest reported viscosity of erythritol in the literature is on the order of $10^4$ Pa s (Grayson et al., 2017; Y. C. Song et al., 2016).

Figure 5b and Table S2 show the viscosity of erythritol–water particles (calculated using diffusion coefficients from Fig. 5a and the Stokes–Einstein equation) as a function of $a_w$. As $a_w$ decreased from approximately 0.5 to 0, the viscosity increased from approximately $1 \times 10^{-1}$ to $5 \times 10^1$ Pa s. The symbols in Fig. 5 are color-coded by the time allowed to condition the samples to a particular $a_w$ value before measuring the diffusion coefficient. The color scale in the top left corner applies to both panels (a, b). No clear trend is observed between the conditioning time and the diffusion coefficient or particle viscosity.

To further investigate the effect of the time used to condition the samples to a particular $a_w$ value, in Fig. 6 the measured RBID diffusion coefficients in erythritol–water matrices are plotted as a function of conditioning time at $a_w$ ≤ 0.105. The data shown in Fig. 6 were taken from the data shown in Fig. 5a. Included as a secondary $x$ axis is the sample conditioning time in multiples of $\tau_w$ (characteristic time for water diffusion within the sample droplets used in the conditioning experiments (see Sect. S1 in the Supplement for details). Consistent with Fig. 5a, Fig. 6 illustrates that there is no clear trend between the diffusion coefficient and the time allowed for conditioning the samples prior to the diffusion measurements. Figure 6 also suggests that a sample conditioning time of $\geq 21.5 \ h$, or $\geq 6.5 \tau_w$, was sufficient to reach near equilibrium between the RH used for conditioning and the $a_w$ in particles.

3.2 Viscosity of erythritol–water particles as a function of $a_w$ measured by the aerosol optical tweezers technique

Erythritol viscosity measurements using the aerosol optical tweezers technique are shown in Fig. 7. The viscosity of pure water at 293 K (Korson et al., 1969) is also included for comparison. The red circles represent the new aerosol optical tweezers measurements obtained in this work (also listed in Table S3 in the Supplement), based solely on bright-field images. The gray circles represent the viscosities reported in Y. C. Song et al. (2016). The new averaged viscosities reported here based on the aerosol optical tweezers technique are lower than those reported by Y. C. Song et al. (2016)
Sect. 2.2). The viscosity at $a_w$ $\geq$ 0.43 were based on backscattered light intensity (where viscosities were ages alone, and those at $a_w$ = 0.22 $\pm$ 0.02 reported by Y. C. Song et al. (2016) was based on backscattered images alone, and those at $a_w$ $\geq$ 0.43 were based on backscattered light intensity (where viscosities were

3.3 Update on the viscosity of erythritol–water particles as a function of $a_w$ and an intercomparison of techniques for measuring the viscosity of particles

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 Y. C. Song et al. (2016) (open circles) and this study (solid circles). The previous measurements at $a_w$ $\leq$ 0.1 by Y. C. Song et al. (2016) 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 (2 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.

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 $\pm$ 0.22 (2 standard deviations), corresponding to a viscosity of pure erythritol of 184$^{+122}_{-73}$ Pa s.

3.4 Effect of the addition of OH functional groups to a linear C$_4$ carbon backbone

Grayson et al. (2017) previously estimated the effect of adding OH functional groups to the viscosity of a linear C$_4$ compound. Here we repeat this analysis (Fig. 10) based on the updated viscosity of pure erythritol (184$^{+122}_{-73}$ 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; Y. C. Song et al., 2016). Table S4 (in the Supplement) 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 $\pm$ 0.08 (2 standard deviations), which indicates that the viscosity of a linear C$_4$ molecule increases on average by a factor of 27$^{+6}_{-5}$ per ad-
The viscosity increase from the addition of OH functional groups to a carbon backbone may depend on the level of prior functionalization. To investigate this aspect further, we calculated the viscosity sensitivity parameter \( S_\eta \) for a linear C\(_4\) carbon backbone using the viscosity data presented in Fig. 10 and the following equation (Rothfuss and Petters, 2017):

\[
S_\eta = \Delta \log_{10}(\eta/\text{Pa s})/\Delta N,
\]

where \( \Delta \log_{10}(\eta/\text{Pa s}) \) is the change in viscosity on a \( \log_{10} \) scale and \( \Delta N \) is the change in the number of OH functional groups. \( S_\eta \) was estimated based on the addition of one OH functional group (\( \Delta N = 1 \)), starting from \( n \)-butane. The relationship between \( S_\eta \) and \( N \) is shown in Fig. 11 for a linear C\(_4\) carbon backbone. \( S_\eta \) is between 0.7 and 1.9 for \( N = 1 \text{–} 3 \). On the other hand, \( S_\eta \) is between 1.6 and 2.5 for \( N = 4 \), suggesting \( S_\eta \) likely 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.

4 Summary and conclusion

In this work, viscosities of erythritol–water particles as a function of \( a_w \) at 292–295 K were measured using the rFRAP and aerosol optical tweezers techniques. In the rFRAP measurements, a trace amount of RBID (0.2–0.3 wt %) was
The viscosity sensitivity parameter at 292–295 K is estimated at 4, to the addition of three OH functional groups. On the other hand, the degree of increase in viscosity is likely larger when the linear C₄ carbon backbone already contains three OH groups. These results should help improve the understanding of the viscosity of SOA particles in the atmosphere.

Data availability. Data for this paper are available in the Supplement.

The Supplement related to this article is available online at https://doi.org/10.5194/amt-11-4809-2018-supplement.

Author contributions. AKB designed and oversaw this study. YC, EE, and YCS carried out the experiments. SK provided technical support. YC and EE wrote the manuscript with contributions from all authors.

Competing interests. The authors declare that they have no conflict of interest.

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