Supplementary Information

Technical Note: New methodology for measuring viscosities in small volumes characteristic of environmental chamber particle samples

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Viscosity vs. RH relationships for glycerol-water solutions at 20 ^oC.

Viscosity vs. concentration data was taken from Sheely (1931) and Segur and Oberstar (1951) where the viscosities reported by Segur and Oberstar were corrected to the viscosity of water at 20 ^oC (1.002 x 10^{-3} Pa s) (Kestin et al., 1978;Swindells et al., 1952). The concentrations used in these experiments were converted to relative humidity using the parameterization shown in Table S1. The resulting viscosity vs. RH data was then fit to a 3rd degree polynomial. The results of the fit are shown in Table S2.

Viscosity vs. RH relationships for Glycerol-water solutions at 0 ^oC.

Viscosity vs. concentration data was taken from Trejo González et al. (2011). Concentrations used in these experiments were converted to RH using the parameterizations shown in Table S1. The resulting viscosity vs. RH data was then fit to a 3rd degree polynomial (see Table S2).

Viscosity vs. RH relationships for sucrose-water solutions at 20 ^oC.

Viscosities measured at different concentration was taken from Swindells et al. (1958), Quintas et al. (2006) and Perry and Green (2008). The concentrations used in these references were converted to RH using the parameterization of Zobrist et al. (2011). The resulting viscosity vs. RH data was fit to a 3rd degree polynomial (see Table S2).

Viscosity of olive oil samples at 20 C.

A Haake RotoVisco 550 viscometer (sensor MV-1P) was used to determine the viscosity of the same olive oil sample used in the bead mobility experiments. The resulting shear stress vs. shear rate data are shown in Fig. S2. From the slope of the line in Fig. S2, the viscosity of the solution was determined to be 0.097 (± 0.006) Pa s.

Surface tension values for glycerol-water mixtures.

For glycerol-water solutions, the concentration of glycerol at a given RH was determined using the parameterization in Table S2 and the corresponding surface tension was determined from a 3rd degree polynomial fit to the data of Ernst et al. (1936) (see Table S3).

Surface tension values for other organic and organic-water mixtures.

Other surface tension values used to construct Figure 7c (main text) are given Table S4. Both 25 ^{o}C and 20 ^{o}C surface tension data from the literature were used to construct Fig. 7c. It is assumed that the surface tension does not depend strongly on temperature over 20-25 ^{o}C, a good assumption for the types of compounds used herein. For example, the surface tension of pure glycerol is 63.3-63.5 mN/m at 20 ^{o}C (Jańczuk et al., 1993;Panzer, 1973), and 62.5 mN/m at 25^{o}C. (Ernst et al., 1936)

Table S1: Best fit parameters for concentration vs. relative humidity data for glycerol-water mixtures. $RH = 100 + Ax + Bx^2 + Cx^3 + Dx^4 + Ex^5$ where x= mass fraction glycerol. Data used to generate this equation taken from Marcolli and Peter (2005), Ninni et al. (2000), and Chenlo et al. (2004).The corresponding R^2 from the best fit function shows the quality of the fit.

A	B	C	D	E	R^2
-15.08	-30.62	-21.12	7.406	-36.17	0.9996

Table S2: Best fit parameters for viscosity vs. relative humidity data where log(viscosity) = A + B(RH) + C(RH)^2 + D(RH)^3 and the corresponding R^2 from the best fit function shows the quality of the fit.

Aqueous solution	A	B	C	D	R^2
Glycerol, 20°C	3.407×10^{-1}	-3.778×10^{-2}	1.914×10^{-4}	-1.409×10^{-6}	0.9997
Glycerol, 0°C	1.276	-4.352×10^{-2}	1.746×10^{-4}	-1.350×10^{-6}	0.9999
Sucrose, 20°C	44.33	-1.400	1.566×10^{-2}	-6.387×10^{-5}	0.9988

Table S3: Best fit parameters for surface tension vs. mass fraction of glycerol. The data of Ernst et al. (1936) was fit to a 3rd degree polynomial where surface tension (mN/m) $=A-Bx+Cx^2+Dx^3$ where x= mass fraction of glycerol and the corresponding R^2 from the best fit function shows the quality of the fit.

Aqueous solution	A	B	C	D	R^2
Glycerol	71.97	-15.05	12.16	16.32	0.9997

Table S4: Surface tensions from the literature used to construct Fig. 7c in the main text.

Droplet material	Range of concentrations studied (wt% organic)	Molecular formula of the organic	Surface tension (mN/m)	References
Glycerol + Water	35 - ~100	$C_3H_8O_3$	62.5-68.2	(Ernst et al., 1936)
1,2,6-hexanetriol	~100	$C_6H_{14}O_3$	50	(Aldrich Chemical Company, 1996)
Tetraethylene glycol	~100	$C_8H_{15}O_5$	46	(Dow Chemical Company, 2007;Moumen et al., 2006)
Oleic Acid	~100	$C_{18}H_{34}O_2$	32.79	(Chumpitaz et al., 1999)
PEG-300	~100	$H(OCH_2CH_2)_nOH$, n=6-7	34.3	(Mazandarani et al., 2007)
Sucrose	55	$C_{12}H_{22}O_{11}$	76	(Docoslis et al., 2000;MacDonald et al., 1996)
PEG-400	~100	$H(OCH_2CH_2)_nOH$, n=8-9	44.5	(Ciobanu et al., 2009;Marcolli and Krieger, 2006)
Tergitol™ NP-7	~100	$C_{29}H_{52}O_8$	32[a]	(Dow Chemical Company, 2004)

[a] Upper limit on the surface tension of pure Tergitol NP-7 because literature value is for 1 wt% Tergitol™ NP-7 in water. The critical micelle concentration is < 1 wt% (Dow Chemical Company, 2004). Surface tension vs. concentration data for structurally similar Tergitol™ NP-10 shows a minimal decrease in surface tension at concentrations greater than the critical micelle concentration (Park and Bielefeldt, 2003) so surface tension of ~100 wt% Tergitol™ NP-7 used herein is likely close to the value reported here.

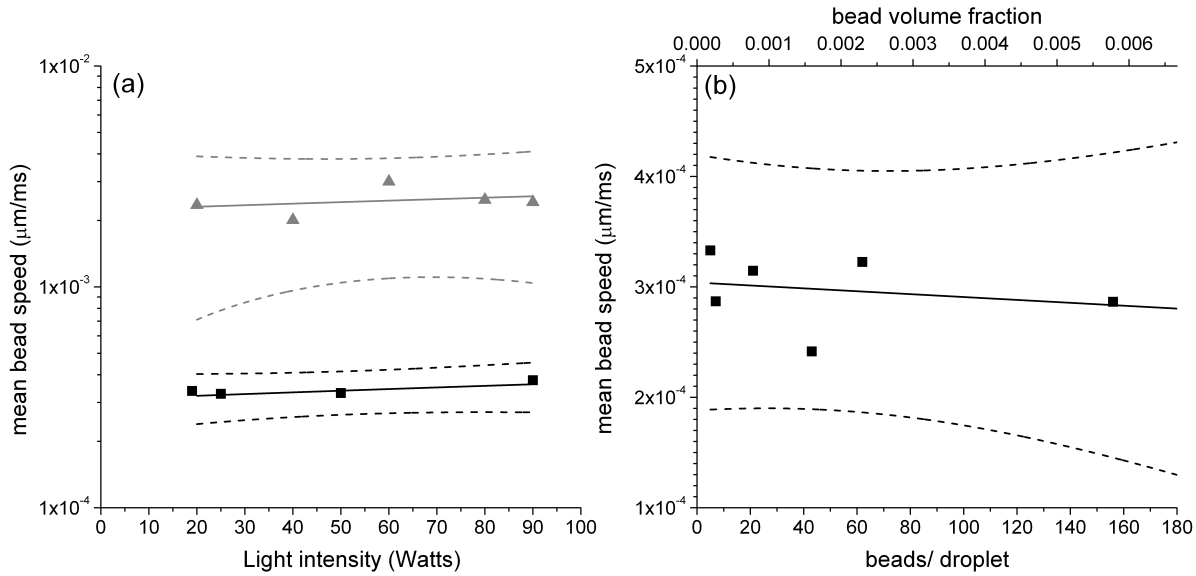

Figure S1: a) Mean melamine bead speed vs. light intensity of halogen light source on microscope in droplets composed of ~100 wt% glycerol (black squares) and ~100 wt% olive oil (grey triangles) at room temperature, b) Mean melamine bead speed vs. number of beads/droplet or bead volume fraction in droplets composed of ~100 wt% glycerol. In (a) and (b), the solid lines represent the best fit functions and the dashed lines represent the upper and lower 95% prediction intervals.

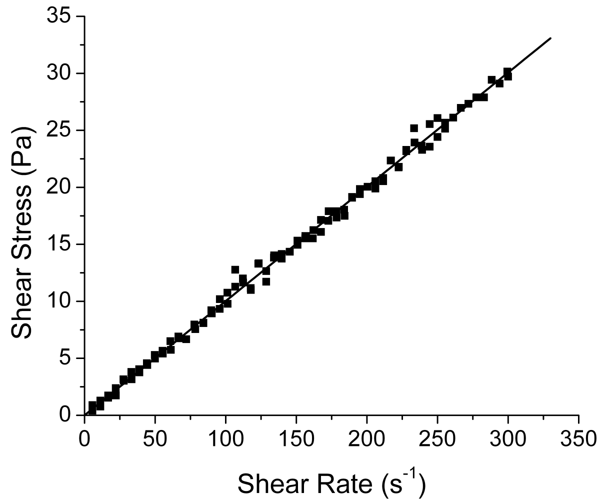

Figure S2. Shear stress v. shear rate data obtained with the Haake RotoVisco 550 viscometer using the virgin olive oil that was used in the bead mobility experiments. The slope of the best fit line (0.097 (± 0.006) Pa s) gives the measured viscosity.

Movies

Movie S1: Sample movie showing beads moving through a droplet matrix. The droplet shown here is an aqueous glycerol droplet at 85% RH. The first three frames correspond to Figs. 3 a-c in the main text. Frame rate = 5 frames/second and frame interval = 200 ms. (real time). The white rings around the perimeter of the droplet are due to light diffraction and are an experimental artifact.

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