Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information for:

Relative humidity-dependent viscosity of single quasi aerosol particle and possible implications for atmospheric aerosol chemistry

Yu-Kai Tong,[†] Yaoyao Liu,[†] Xiangxinyue Meng,[‡] Jie Wang,[†] Dongping Zhao,[†] Zhijun Wu,[‡] and Anpei Ye[*,†]

[†]*Key Laboratory for the Physics and Chemistry of Nanodevices, School of Electronics, Peking University, Beijing 100871, China*

[‡]*State Key Joint Laboratory of Environmental Simulation and Pollution Control, College of Environmental Sciences and Engineering, Peking University, Beijing 100871, China*

E-mail: yap@pku.edu.cn

S.1 Aqueous-Sucrose Viscosity

Table S1: Experimental data at varying RH reported in this study. The uncertainties in the reported viscosities are the standard deviations associated with an average of multiple measurements at the reported RH value.

RH / %	log(Viscosity / Pa s)	Temperature / ^{o}C
30	8.14±0.5	23.00±0.29
40	6.62±0.12	23.33±0.32
50	4.02±0.18	23.32±0.30
60	1.77±0.29	23.65±0.44
70	0.39±0.20	23.91±0.41
80	-0.59±0.35	23.32±0.30

Table S2: Parameters used in the parabolic fitting of experimental data where RH is expressed as a percentage ($\log \eta = A + B \cdot RH + C \cdot RH^2$).

Parameters	Value	Error
A	16.88	0.897
B	-0.314	0.033
C	0.001	3.03×10^{-4}

S.2 Aqueous-Sulfate/Nitrate Viscosity

Table S3: Experimental data of aqueous $(NH_4)_2SO_4$ at varying RH reported in this study. The uncertainties in the reported viscosities are the standard deviations associated with an average of multiple measurements at the reported RH value.

RH / %	log(Viscosity / mPa s)	Temperature / ^{o}C
40	2.72±0.058	23.64±0.87
50	2.21±0.063	23.64±0.87
60	1.73±0.061	22.53±0.47
70	1.29±0.059	23.30±0.42
80	0.56±0.091	23.30±0.42
82	0.37±0.134	23.30±0.42

Table S4: Experimental data of aqueous NH_4NO_3 at varying RH reported in this study. The uncertainties in the reported viscosities are the standard deviations associated with an average of multiple measurements at the reported RH value.

RH / %	log(Viscosity / mPa s)	Temperature / ^{o}C
30	0.98±0.065	24.73±0.31
35	0.87±0.022	24.73±0.31
40	0.80±0.049	24.73±0.31
50	0.71±0.058	24.73±0.31
60	0.54±0.057	24.73±0.31
70	0.36±0.065	25.21±0.54
80	0.18±0.076	24.57±0.20

Table S5: Experimental data of aqueous $(NH_4)_2SO_4$ / NH_4NO_3 mixture at varying RH reported in this study. The uncertainties in the reported viscosities are the standard deviations associated with an average of multiple measurements at the reported RH value.

RH / %	log(Viscosity / mPa s)	Temperature / ^{o}C
40	2.26±0.039	23.44±0.73
50	1.85±0.063	24.63±0.53
55	1.69±0.034	23.44±0.73
60	1.45±0.096	24.63±0.53
65	1.39±0.095	23.44±0.73
68	1.37±0.061	24.63±0.53
70	1.08±0.079	24.63±0.53
78	0.73±0.037	23.44±0.73
80	0.58±0.069	24.63±0.53

Table S6: Experimental data of aqueous Na_2SO_4 at varying RH reported in this study. The experiment was conducted at 22.55 ± 0.67 ^{o}C.

RH / %	log(Viscosity/mPa s)	RH / %	log(Viscosity/mPa s)
60	2.43±0.039	80	1.37±0.038
70	1.73±0.031	88	0.86±0.034

Table S7: Experimental data of aqueous $NaNO_3$ at varying RH reported in this study.

RH / %	log(Viscosity / mPa s)	Temperature / ^{o}C
40	2.30±0.021	22.48±0.33
50	1.89±0.016	22.48±0.33
60	1.47±0.017	22.57±0.41
70	1.09±0.039	22.57±0.41
80	0.76±0.035	22.57±0.41

Table S8: Experimental data of aqueous $CuSO_4$ at varying RH reported in this study.

RH / %	log(Viscosity / mPa s)	Temperature / ^{o}C
40	2.55±0.036	22.22±0.44
50	2.33±0.035	22.22±0.44
60	1.89±0.018	22.22±0.44
70	1.40±0.057	21.65±0.21
80	0.89±0.044	22.60±0.38

Table S9: Experimental data of aqueous $MgSO_4$ at varying RH reported in this study. The experiment was conducted at 21.67 ± 0.64 ^{o}C.

RH / %	log(Viscosity/mPa s)	RH / %	log(Viscosity/mPa s)
40	3.87±0.071	70	1.44±0.013
50	2.26±0.030	80	1.11±0.043
60	1.69±0.031		

Table S10: Experimental data of aqueous $Mg(NO_3)_2$ at varying RH reported in this study. The experiment was conducted at 21.43 ± 0.42 ^{o}C.

RH / %	log(Viscosity/mPa s)	RH / %	log(Viscosity/mPa s)
40	0.79±0.023	70	0.24±0.022
50	0.64±0.021	80	0.09±0.013
60	0.37±0.046		

Table S11: Parameters used in the parabolic fitting of experimental data ($\log \eta = A + B \cdot RH + C \cdot RH^2$, unit: mPa s).

Solute	A	B	C
$(NH_4)_2SO_4$	3.17±0.71	0.60±0.48	-4.83±2.09
NH_4NO_3	0.96±0.19	0.19±0.09	-1.48±0.58
$(NH_4)_2SO_4$ / NH_4NO_3	3.11±0.53	-1.29±1.82	-2.24±1.51
Na_2SO_4	7.08±2.11	-9.54±5.17	2.86±2.47
$NaNO_3$	4.27±0.35	-5.55±1.25	1.44±1.08
$MgSO_4$	10.24±3.35	-23.13±11.12	15.00±9.09
$Mg(NO_3)_2$	1.70±0.31	-2.45±1.06	0.55±0.86
$CuSO_4$	2.81±0.36	1.21±1.23	-4.55±1.04

S.3 Aqueous-Organics/Inorganics Viscosity

Figure S1 Experimental Data - red, 50/50 wt% $(NH_4)_2SO_4$ / sucrose mixture; blue, 50/50 wt% NH_4NO_3 / sucrose mixture.

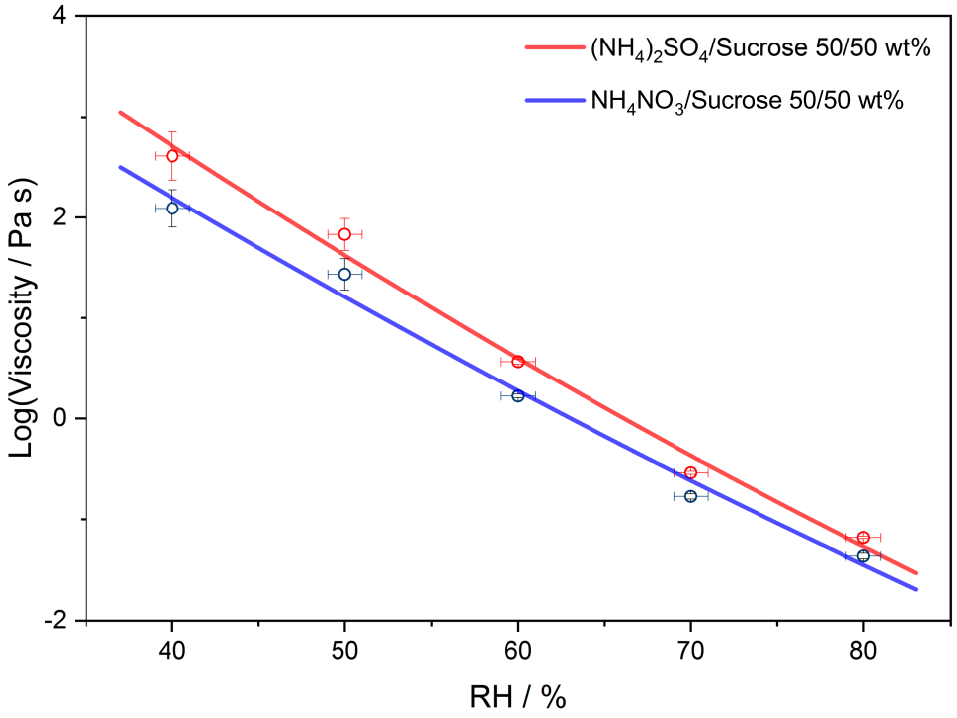


Table S12: Experimental data of aqueous $(NH_4)_2SO_4$ / sucrose mixture at varying RH reported in this study.

RH / %	log(Viscosity / Pa s)	Temperature / ^{o}C
40	2.61±0.239	24.14±0.54
50	1.83±0.158	24.14±0.54
60	0.57±0.025	24.15±0.37
70	-0.54±0.015	24.15±0.37
80	-1.18±0.01	24.15±0.37

Table S13: Experimental data of aqueous NH_4NO_3 / sucrose mixture at varying RH reported in this study.

RH / %	log(Viscosity / Pa s)	Temperature / ^{o}C
40	2.09±0.186	24.44±0.46
50	1.43±0.156	24.44±0.46
60	0.22±0.018	24.44±0.46
70	-0.77±0.026	24.44±0.46
80	-1.36±0.028	24.14±0.54

Table S14: Parameters used in the parabolic fitting of experimental data ($\log \eta = A + B \cdot RH + C \cdot RH^2$).

Parameters	$(NH_4)_2SO_4$ / Sucrose	NH_4NO_3 / Sucrose
A	7.67±1.97	6.63±2.05
B	-13.63±6.84	-12.07±7.09
C	3.07±5.67	2.46±5.88

S.4 Atmospheric Aerosol Proxy Viscosity

Figure S2 Experimental Data - of atmospheric aerosol proxies of 2015 and 2020 based on the research on aerosol composition of Lu et al.[1] Red, 2015 Proxy of 16.8/20.3/62.9 wt% $(NH_4)_2SO_4$ / NH_4NO_3 / sucrose mixture; blue, 2020 Proxy of 19.8/37.4/42.8 wt% $(NH_4)_2SO_4$ / NH_4NO_3 / sucrose mixture.

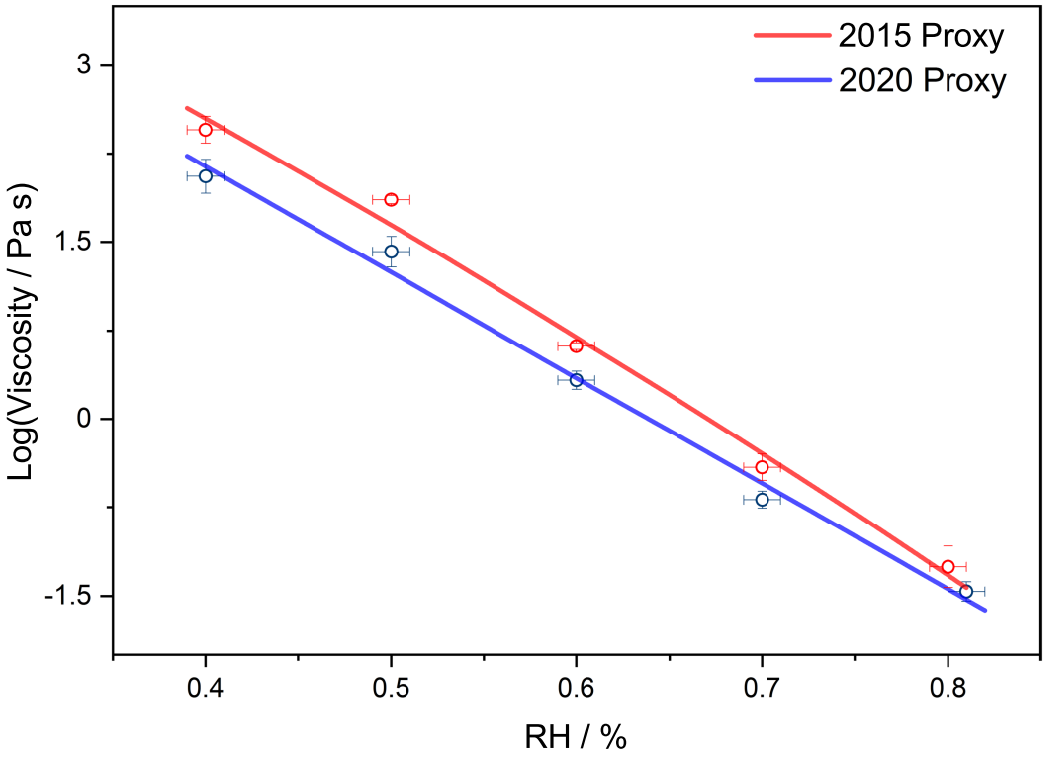


Table S15: Experimental data of 2015 proxy at varying RH reported in this study. The uncertainties in the reported viscosities are the standard deviations associated with an average of multiple measurements at the reported RH value.

RH / %	log(Viscosity / Pa s)	Temperature / ^{o}C
40	2.45±0.114	20.20±1.18
50	1.87±0.035	20.20±1.18
60	0.62±0.029	22.57±1.24
70	-0.41±0.113	22.57±1.24
80	-1.25±0.173	22.57±1.24

Table S16: Experimental data of 2020 proxy at varying RH reported in this study. The uncertainties in the reported viscosities are the standard deviations associated with an average of multiple measurements at the reported RH value.

RH / %	log(Viscosity / Pa s)	Temperature / ^{o}C
40	2.06±0.144	21.76±0.65
50	1.42±0.129	21.76±0.65
60	0.33±0.078	22.69±0.70
70	-0.68±0.072	20.20±1.18
81	-1.46±0.081	20.20±1.18

Table S17: Parameters used in the parabolic fitting of experimental data ($\log \eta = A + B \cdot RH + C \cdot RH^2$).

Parameters	2015 Proxy	2020 Proxy
A	5.76±1.86	5.75±1.57
B	-7.21±6.46	-9.05±5.39
C	-2.06±5.35	0.07±4.34

S.5 Mass Transfer

Table S18: Ratio of half-time for response in size of aqueous $(NH_4)_2SO_4$ / sucrose mixture droplet to half-time for RH probe response.

initial RH / %	final RH / %	Response time ratio
80	70	2.77
80	60	5.02
80	50	6.13
80	40	3.33
70	80	2.15
70	60	1.82
70	50	8.71
70	40	4.21
60	80	6.35
60	70	8.79
60	50	12.11
60	40	3.37
50	80	5.02
50	70	4.04
50	60	15.13
50	40	13.43
40	80	15.35
40	70	8.06
40	60	4.65
40	50	33.89

Table S19: Ratio of half-time for response in size of aqueous NH_4NO_3 / sucrose mixture droplet to half-time for RH probe response.

initial RH / %	final RH / %	Response time ratio
80	70	1.23
80	60	2.97
80	50	2.52
80	40	2.5
70	80	1.60
70	60	2.82
70	50	7.59
70	40	2.96
60	80	2.46
60	70	1.83
60	50	5.15
60	40	2.71
50	80	7.62
50	70	3.24
50	60	10.57
50	40	8.56
40	80	6
40	60	11.11
40	50	12.70

Table S20: Ratio of half-time for response in size of aqueous 2015 proxy droplet to half-time for RH probe response.

initial RH / %	final RH / %	Response time ratio
80	40	2.33
70	60	5
70	50	6.67
70	40	7.88
60	70	1.63
60	50	1.55
60	40	15.86
50	70	1.98
50	60	2.52
50	40	18.33
40	80	5.89
40	70	4.04
40	60	4.31
40	50	16.24

Table S21: Ratio of half-time for response in size of aqueous 2020 proxy droplet to half-time for RH probe response.

initial RH / %	final RH / %	Response time ratio
80	60	1.63
80	40	4.84
70	60	1.42
70	50	3.10
70	40	3.94
60	70	2.35
60	50	4.42
60	40	3.18
50	70	2.74
50	60	3.95
50	40	6.32
40	80	9.67
40	70	6.21
40	60	8.91
40	50	9.53

Figure S3 - (a) Time-dependent radius of the droplet of 2015 proxy during the process of water evaporation when RH changes from 40% to 37%. (b) The linear-fitting (red dashed line) of $r^3 - r_0^3$ and $\sqrt{t}$ for calculating the water diffusion coefficient using the differential step isothermal method, where r is the time-dependent radius, r_0 is the initial radius prior to RH steps, and t is time.

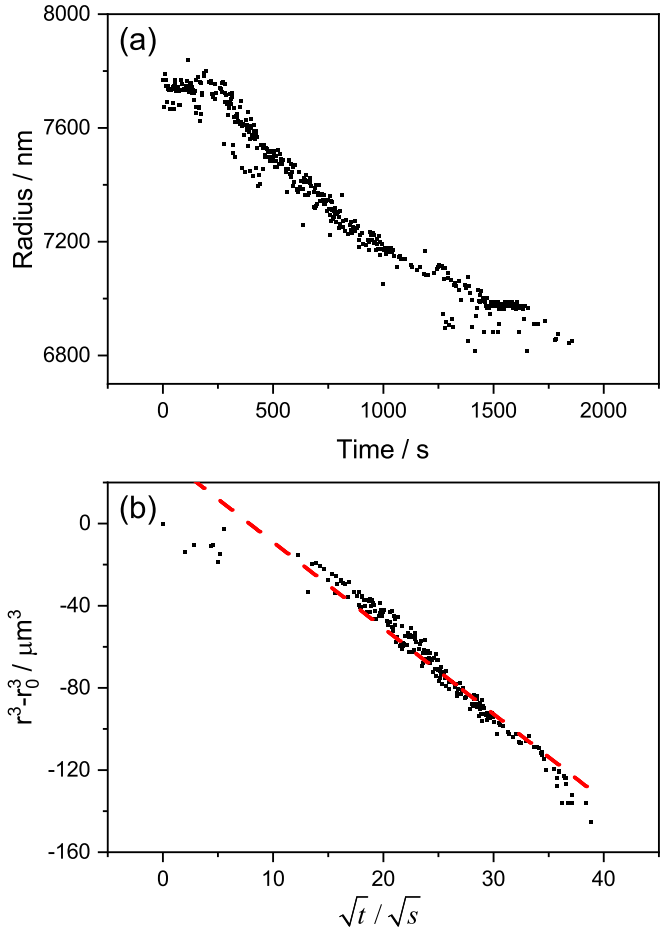


Figure S4 - Ratio of half-time for response in size of aqueous 2015 proxy (a1 and a2) and 2020 proxy (b1 and b2) droplet to half-time for RH probe response, mapped over range in initial and final RHs. Data points above the diagonals in (a1) and (b1) mean the process of water condensation and those below the diagonals mean the process of water evaporation. (a2) and (b2) are contours plotted by smoothing the data points in (a1) and (a2). The experiments were conducted at $22.61 \pm 0.37\,^{o}$C for 2015 proxy and $22.25 \pm 0.54\,^{o}$C for 2020 proxy, respectively.

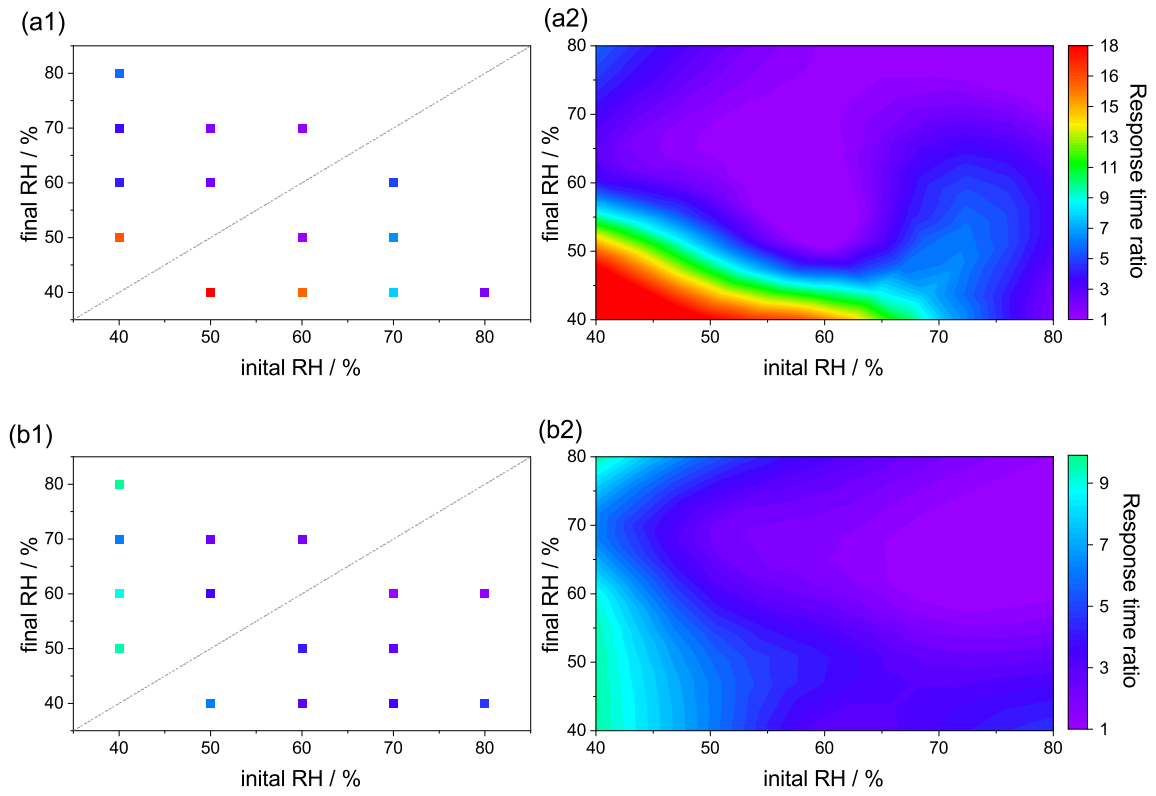
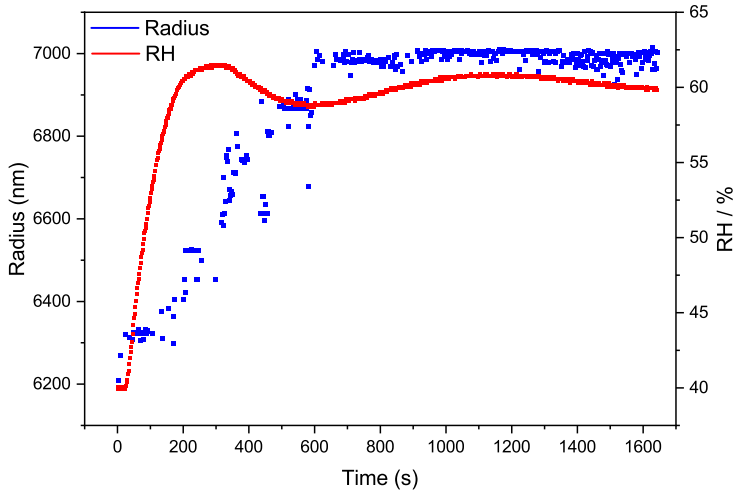


Figure S5 - Example of the hysteresis of response in size of the droplet of 2015 proxy when RH changes from 40% to 60%.



S.6 Influence of Temperature on Viscosity

Figure S6 - Temperature-dependent viscosity of bulk solutions of atmospheric proxies and sucrose (53%, mass fraction). The solid lines are fitting curves of the experimental data based on the Andrade equation. The viscosity and temperature was measured using a cone/plate viscometer (Brookfield, DV3TLV). Each measurement lasts for 1 minute and the data point is recorded every 2 seconds. The uncertainties in the reported viscosities are the standard deviations of the data points recorded in one measurement. The temperature was controlled by a thermostatic bath. Previous studies[2 3] on aqueous sucrose solution are presented in the figure for comparison as well.

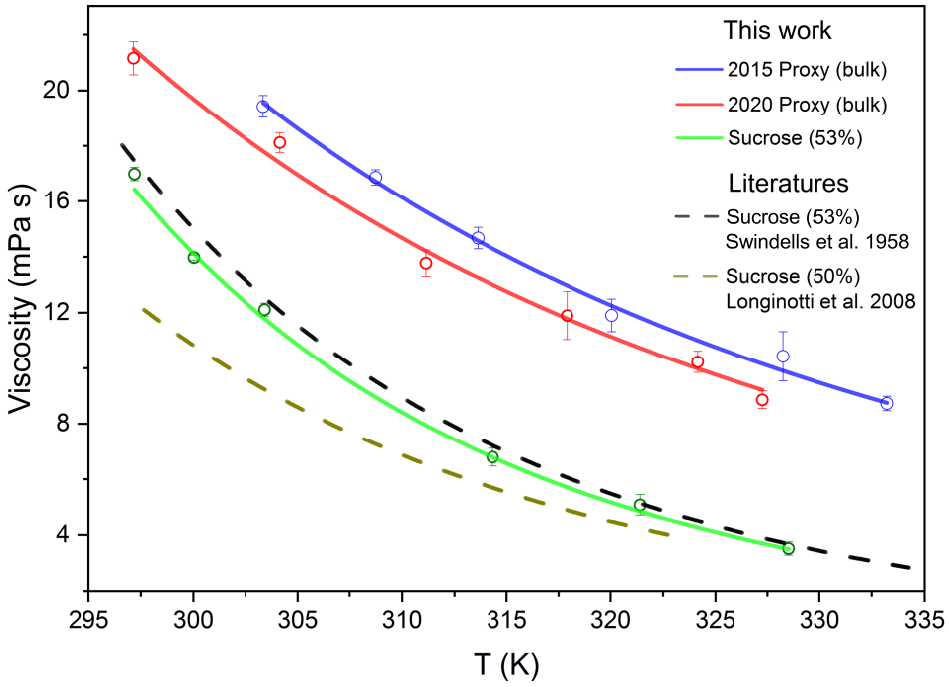


Table S22: Experimental data of temperature-dependent viscosity of the 2015 proxy bulk solution.

Temperature/K	Viscosity/mPa s	Temperature/K	Viscosity/mPa s
303.35	19.43±0.39	320.05	11.91±0.59
308.75	16.86±0.27	328.25	10.43±0.88
313.63	14.67±0.38	333.23	8.73±0.26

Table S23: Experimental data of temperature-dependent viscosity of the 2020 proxy bulk solution.

Temperature/K	Viscosity/mPa s	Temperature/K	Viscosity/mPa s
297.16	21.15±0.59	317.92	11.90±0.86
304.15	18.12±0.36	324.15	10.23±0.38
311.15	13.77±0.48	327.26	8.86±0.32

Table S24: Experimental data of temperature-dependent viscosity of the aqueous sucrose bulk solution (53%, mass fraction).

Temperature/K	Viscosity/mPa s	Temperature/K	Viscosity/mPa s
297.20	16.98±0.24	314.33	6.79±0.32
300.03	13.97±0.09	321.40	5.08±0.36
303.42	12.11±0.23	328.51	3.52±0.24

Table S25: Parameters used in the fitting of experimental data according to the Andrade equation ($\eta = A \exp B/T$), where η (mPa s) is the viscosity of flow and T is temperature.

Solute	A	B
2015 Proxy	$(2.51 \pm 0.48) \times 10^{-3}$	2718.17±59.77
2020 Proxy	$(2.18 \pm 0.96) \times 10^{-3}$	2732.55±134.90
Sucrose	$(1.47 \pm 0.16) \times 10^{-6}$	4823.41±237.27

S.7 Example of Droplets Coalescence

Figure S7 - Example of inferring the relaxation time from measurements of elastically backscattered light collected in a droplets coalescence event using PMT. The example shown is from the coalescence of aqueous-2015 aerosol proxy droplets at 70% RH.

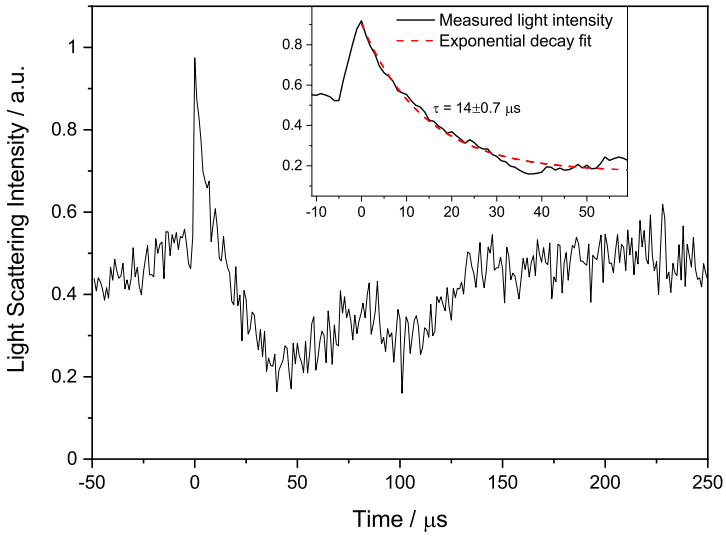
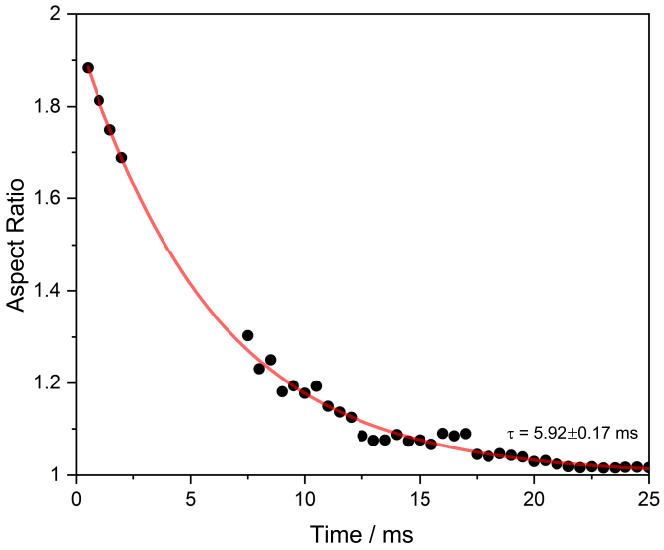
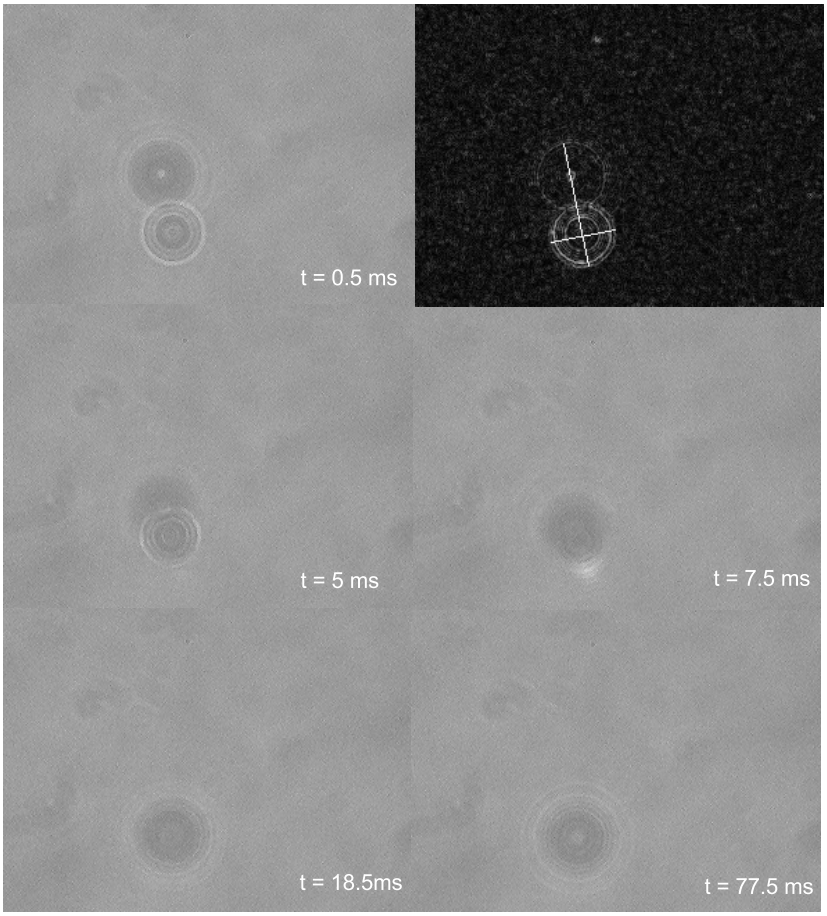


Figure S8 - Example of inferring the relaxation time from measurements of the droplet aspect ratio through brightfield imaging with high-speed camera. The time-series images correspond to different periods during droplets coalescence process. The top right image shows an example of edge crispening for inferring aspect ratio. The example shown is from the coalescence of aqueous-$(NH_4)_2SO_4$ / sucrose mixture droplets at 50% RH.





References

(1) Lei, L.; Zhou, W.; Chen, C.; He, Y.; Li, Z.; Sun, J.; Tang, X.; Fu, P.; Wang, Z.; Sun, Y. Long-term characterization of aerosol chemistry in cold season from 2013 to 2020 in Beijing, China. *Environ. Pollut.* **2021**, *268*, 115952.

(2) Swindells, J. F.; Snyder, C. F.; Hardy, R. C.; Golden, P. E. *Viscosities of Sucrose Solutions at Various Temperatures: Tables of Recalculated Values*, 1st ed.; U. S. Government Printing Office: Washington D. C., 1958.

(3) Longinotti, M. P.; Corti, H. R. Viscosity of concentrated sucrose and trehalose aqueous solutions including the supercooled regime. *J. Phys. Chem. Ref. Data* **2008**, *37*, 1503–1515.