

# Supplementary Information

for

## *A predictive group-contribution model for the viscosity of aqueous organic aerosol*

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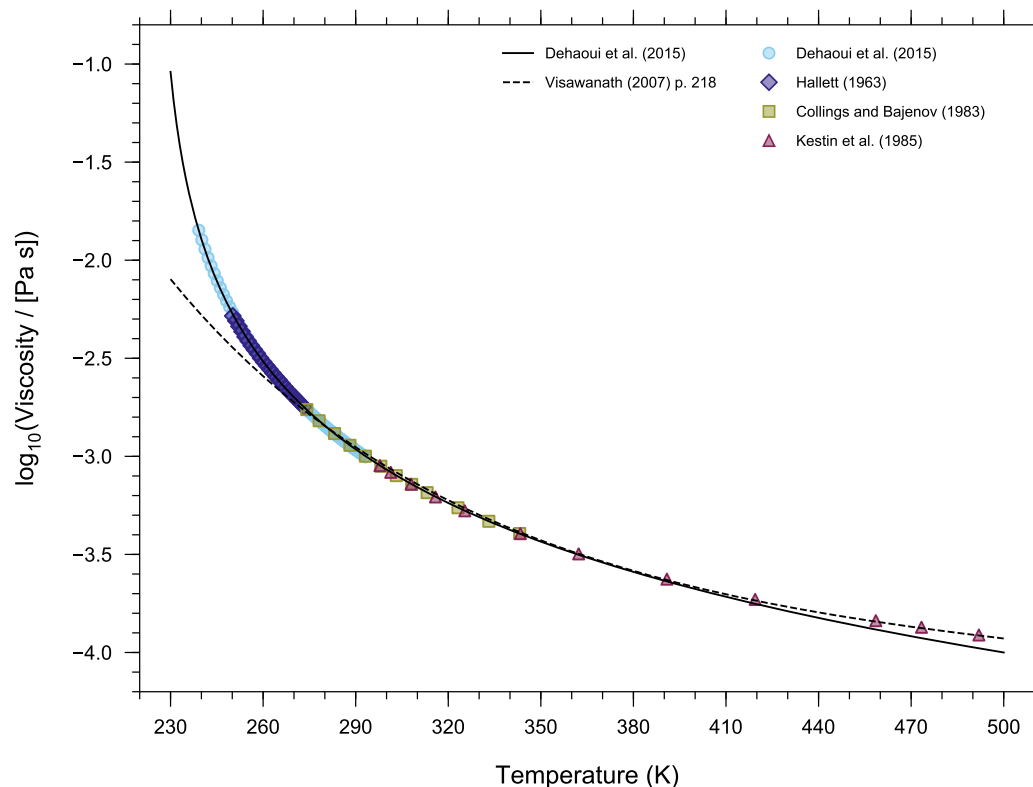
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### **S1 Estimation of the pure component viscosity of water**

The pure component viscosity of water was estimated using the parameterization developed by Dehaoui et al. (2015) for all model simulations in this work; see Eq. (10) of main text. The experimental data used for developing the Dehaoui et al. (2015) parameterization extends from 239.15 K to 491.95 K. The parameterization is in excellent agreement with the data when  
5 temperatures are below  $\sim 400$  K. In Fig. S1, we compare the Dehaoui et al. (2015) parameterization with a parameterization by Viswanath et al. (2007) and with experimental data. The parameterization by Viswanath et al. (2007) is in better agreement with experimental data above  $\sim 400$  K when compared to the Dehaoui et al. (2015) parameterization. The Viswanath et al. (2007) parameterization is also in excellent agreement with the experimental data down to  $\sim 270$  K, below which it begins to deviate substantially from the available experimental data. Between 270 K and  $\sim 380$  K the two parameterizations are  
10 almost indistinguishable. Here we choose to use the Dehaoui et al. (2015) parameterization given that it is the more robust parameterization at lower temperatures of relevance in the troposphere.

### **S2 Exploration of the relationship between pure component vapour pressure and viscosity**

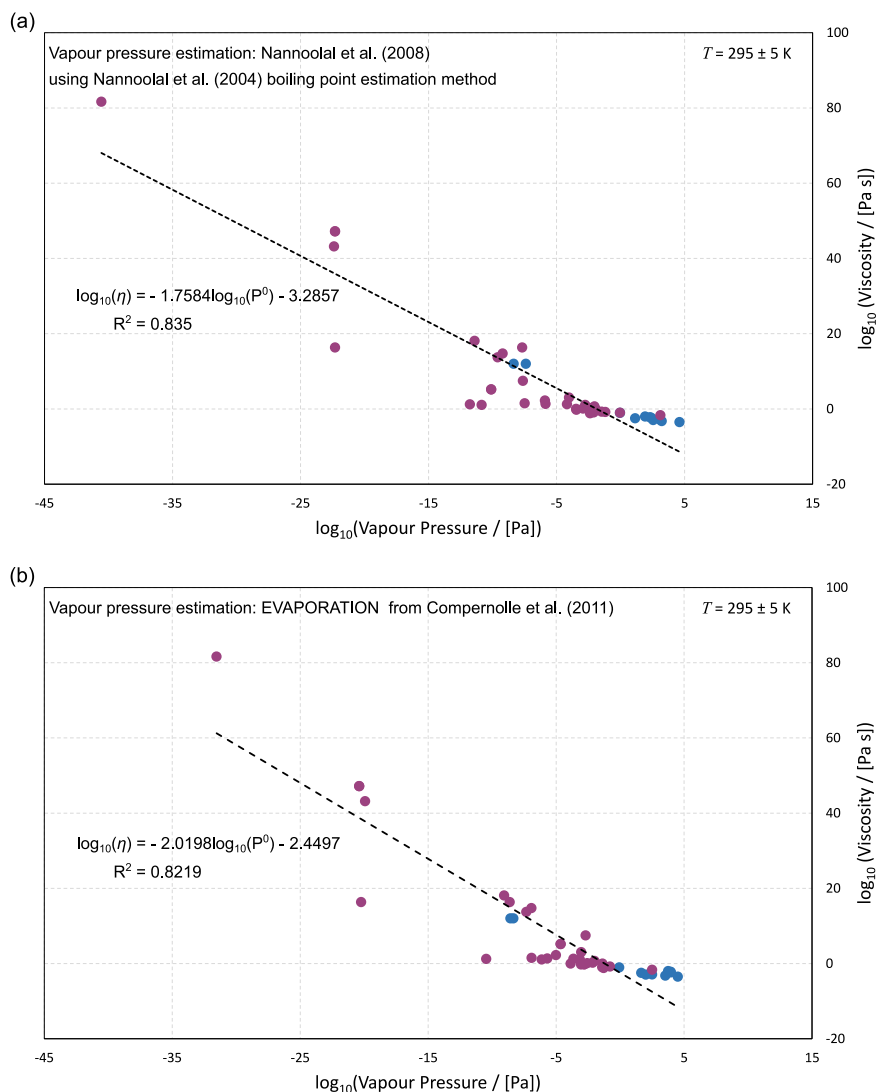
In this study, initially an attempt was made to estimate the pure component viscosity of organic compounds from their pure component vapour pressures. The pure component viscosity is shown as a function of pure component vapour pressure in  
15 double logarithm space in Fig. S2. There is only a weak linear relationship between viscosity and vapour pressure when considering the range of viscosity from liquid to glassy for both the Nannoolal et al. (2008) and EVAPORATION model vapour pressure predictions. A stronger linear relationship exists in the liquid range, but below a vapour pressure of  $10^{-5}$  Pa, the relationship between viscosity and vapour pressure becomes less clear and reliable data are scarce. We still hypothesize a relationship to exist between the two pure-component properties even in the semi-solid and glassy regimes. Although, it is likely



**Figure S1.** Parameterizations of the pure-component viscosity of water from Dehaoui et al. (2015) (solid line) and Viswanath et al. (2007) (dashed line). Markers represent experimental data where error bars have been omitted for clarity. The Dehaoui et al. (2015) parameterization is supported by measurements from  $\sim 230$  to  $400$  K and the Viswanath et al. (2007) parameterization is supported by measurements from  $\sim 270$  to  $500$  K.

this relationship is not resolved with the vapour pressure and viscosity estimation tools used here, given these tools have been trained with compounds that have higher vapour pressure and liquid viscosity only. Just as direct measurements of ultra-high pure-component viscosities are challenging to make, so too are measurements of ultra-low pure component vapour pressures. In order to fully elucidate the relationship between the two material properties, more precise experimental measurements are

5 needed to better constrain pure-component property estimation tools.



**Figure S2.** Reference or modelled pure component viscosity as a function of modelled pure component vapour pressure. Vapour pressures have been estimated using (a) the online tool UManSysProp (<http://umansysprop.seaes.manchester.ac.uk>) with the Nannoolal et al. (2008) vapour pressure model and the Nannoolal et al. (2004) boiling point estimation method and (b) the EVAPORATION model (Compennolle et al., 2011). Purple markers indicate values where the viscosity has been modelled using the method by Nannoolal et al. (2009). Blue markers indicate reference viscosity values either from direct experimental measurements or from an extrapolation with the Vogel–Tamman–Fulcher equation to  $T = 293.15 \text{ K}$  using pure-component viscosity values measured at higher temperatures. All model values have been calculated at  $293.15 \text{ K}$ . Reference viscosity values are taken at a range of temperatures ( $295 \pm 5 \text{ K}$ ). Dashed black lines indicate linear regressions (in logarithm space) to the combined reference and model data.

**Table S1.** Measured, parameterized, or modelled values of the glass transition temperature ( $T_g$ ) from the literature. Uncertainty values are listed when they are provided from their source.

Compound	$T_g$ (K)	Reference	Compound	$T_g$ (K)	Reference
1,2,4-Butanetriol	200.7	Nakanishi and Nozaki (2011)	Glycerol	187	Angell (1997)
1,2,6-Hexanetriol	202	Böhmer et al. (1993)		193	Angell (1997)
	206.4 $\pm$ 0.5	Dorfmueller et al. (1979)		190	Böhmer et al. (1993)
	201.9	Nakanishi and Nozaki (2011)		191 $\pm$ 0.9	Lienhard et al. (2012)
	192 $\pm$ 2	Zhang et al. (2018)		191.7	Nakanishi and Nozaki (2011)
	193.3 $\pm$ 1.3	Zobrist et al. (2008)		196	Seidl et al. (2013)
1,4-Butanediol	158.4 $\pm$ 1.1	Zobrist et al. (2008)		192 $\pm$ 2	Zhang et al. (2018)
Citric Acid	281 $\pm$ 5	Bodsworth et al. (2010)	Raffinose	377.9 $\pm$ 0.9	Lienhard et al. (2012)
	286 $\pm$ 1.5	Dette et al. (2014)		395.7 $\pm$ 21.6	Zobrist et al. (2008)
	273.25 $\pm$ 3.4	Hoppu et al. (2009)	Sorbitol	266	Angell (1997)
	281.9 $\pm$ 0.9	Lienhard et al. (2012)		274	Böhmer et al. (1993)
	284.15 $\pm$ 0.2	Lu and Zografi (1997)		268.3	Nakanishi and Nozaki (2011)
	286 $\pm$ 10	Marsh et al. (2018)		276 <sup>mid</sup>	Simatos et al. (1996)
	260 $\pm$ 10	Murray (2008)	Sucrose	323	Angell (1997)
	283.35 <sup>in situ</sup>	Timko and Lordi (1979)		331 $\pm$ 2	Dette et al. (2014)
	286.65 <sup>bulk</sup>	Timko and Lordi (1979)		350 $\pm$ 3.5	Hancock et al. (1995)
Fructose	307 $\pm$ 5	Zhang et al. (2018)		341	Kawai et al. (2005)
	283.15	Ablett et al. (1993)		341	Rothfuss and Petters (2017)
	286	Angell (1997)		333 <sup>mid</sup>	Simperler et al. (2006)
	283	Ollet and Parker (1990)		347 <sup>calculated</sup>	Simperler et al. (2006)
	289 <sup>mid</sup>	Simatos et al. (1996)		335.7 $\pm$ 3.6	Zobrist et al. (2008)
Glucose	306	Angell (1997)	Trehalose	388	Angell (1997)
	297 $\pm$ 2	Dette et al. (2014)		369 $\pm$ 1.5	Dette et al. (2014)
	309	Kawai et al. (2005)		386	Kawai et al. (2005)
	293.2 $\pm$ 0.9	Lienhard et al. (2012)		380 <sup>mid</sup>	Simperler et al. (2006)
	304	Ollet and Parker (1990)		392 <sup>calculated</sup>	Simperler et al. (2006)
	296 <sup>mid</sup>	Simperler et al. (2006)			
	325 <sup>calculated</sup>	Simperler et al. (2006)			
	296.1 $\pm$ 3.1	Zobrist et al. (2008)			

### S3 Estimation of AIOMFAC-VISC sensitivity

We calculated the sensitivity of AIOMFAC-VISC as a proxy for the uncertainty in the mixture viscosity prediction. We chose to prescribe the AIOMFAC-VISC sensitivity as the response of the mixture viscosity prediction to a small change in mixture composition. A small change in mixture composition is meant to represent the uncertainty in the composition measurement in a laboratory setting, which would be typical of all experiments. Therefore, the AIOMFAC-VISC sensitivity of mixture viscosity,  $s_\eta$ , is calculated using a molar partial derivative:

$$s_\eta = x^{\text{tol}} \left[ \frac{\partial \ln(\eta_{\text{mix}})}{\partial n_{\text{H}_2\text{O}}} \right] \quad (\text{S1})$$

where  $x^{\text{tol}}$  is the molar tolerance (the prescribed uncertainty) in the mixture composition. To retrieve  $x^{\text{tol}}$  we first perturb the mass of water by  $\delta_m = 2\%$  of the mass of the total system,

$$m_{\text{H}_2\text{O}} = m_{\text{H}_2\text{O},\text{init}} + \delta_m, \quad (\text{S2})$$

where  $m_{\text{H}_2\text{O},\text{init}}$  is the initial mass of water in the mixture (e.g.  $m_{\text{H}_2\text{O},\text{init}} = w_{\text{H}_2\text{O},\text{init}}$  for 1 kg of total mass of the mixture) and  $m_{\text{H}_2\text{O}}$  is the perturbed mass. Next, the mass fractions of all components are normalized to account for the mass addition via

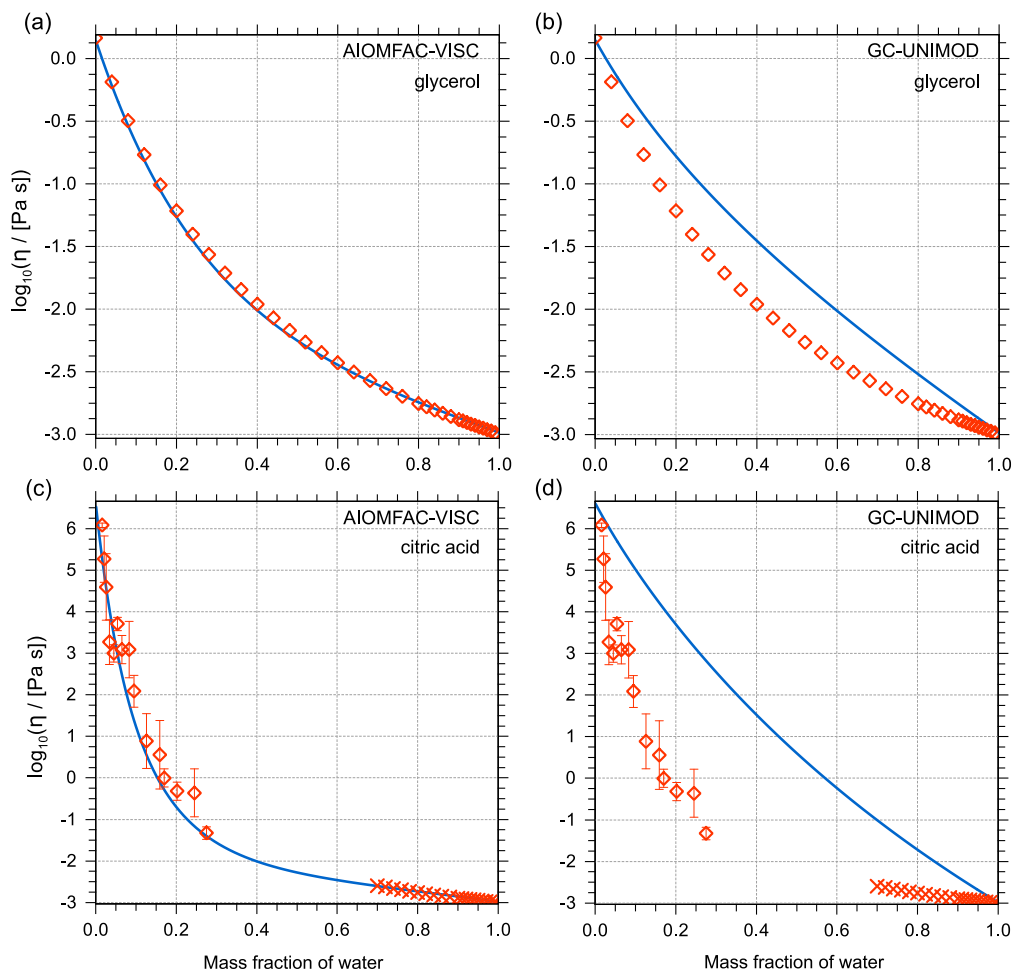
$$w_i = \frac{w_{i,\text{init}}}{1 + \delta_m}, \quad (\text{S3})$$

where  $w_i$  represents the normalized mass fraction of a given component  $i$  given the initial mass fraction  $w_{i,\text{init}}$ . By doing this, we prescribe the model sensitivity as strictly a change in water content of the mixture, where the mixing ratio of organic constituents remains fixed. The normalized mass fractions are then converted to mole fractions ( $x_i$ ) and finally,  $x^{\text{tol}}$  is calculated as the difference between the mole fractions of the perturbed system and the unperturbed system.

$$x^{\text{tol}} = x_{\text{H}_2\text{O}} - x_{\text{H}_2\text{O},\text{init}}. \quad (\text{S4})$$

### S4 Comparison of AIOMFAC-VISC and GC-UNIMOD

Here we compare the performance of the mixture viscosity prediction of AIOMFAC-VISC with the original Cao et al. (1993), GC-UNIMOD model. To compare the mixture viscosity prediction absent of uncertainty introduced by the pure-component viscosity prediction, we have fixed the pure-component viscosity of citric acid to a fitted value at the temperature of interest here (as described in the main text) and we have used the experimental pure-component viscosity of glycerol. As seen in Fig. S3 the AIOMFAC-VISC mixture viscosity prediction is greatly improved from that by the GC-UNIMOD model. The same behaviour was observed for the other binary aqueous mixtures investigated in this work.



**Figure S3.** A comparison of predicted mixture viscosity as a function of mass fraction of water (blue curves) shown for glycerol (top two panels) and citric acid (bottom two panels). The AIOMFAC-VISC mixture viscosity prediction for glycerol (a) and citric acid (c) is in significantly better agreement with the experimental data (red markers) as compared to the GC-UNIMOD mixture viscosity prediction for both glycerol (b) and citric acid (d).

## S5 Binary aqueous mixture viscosity predictions for all training data

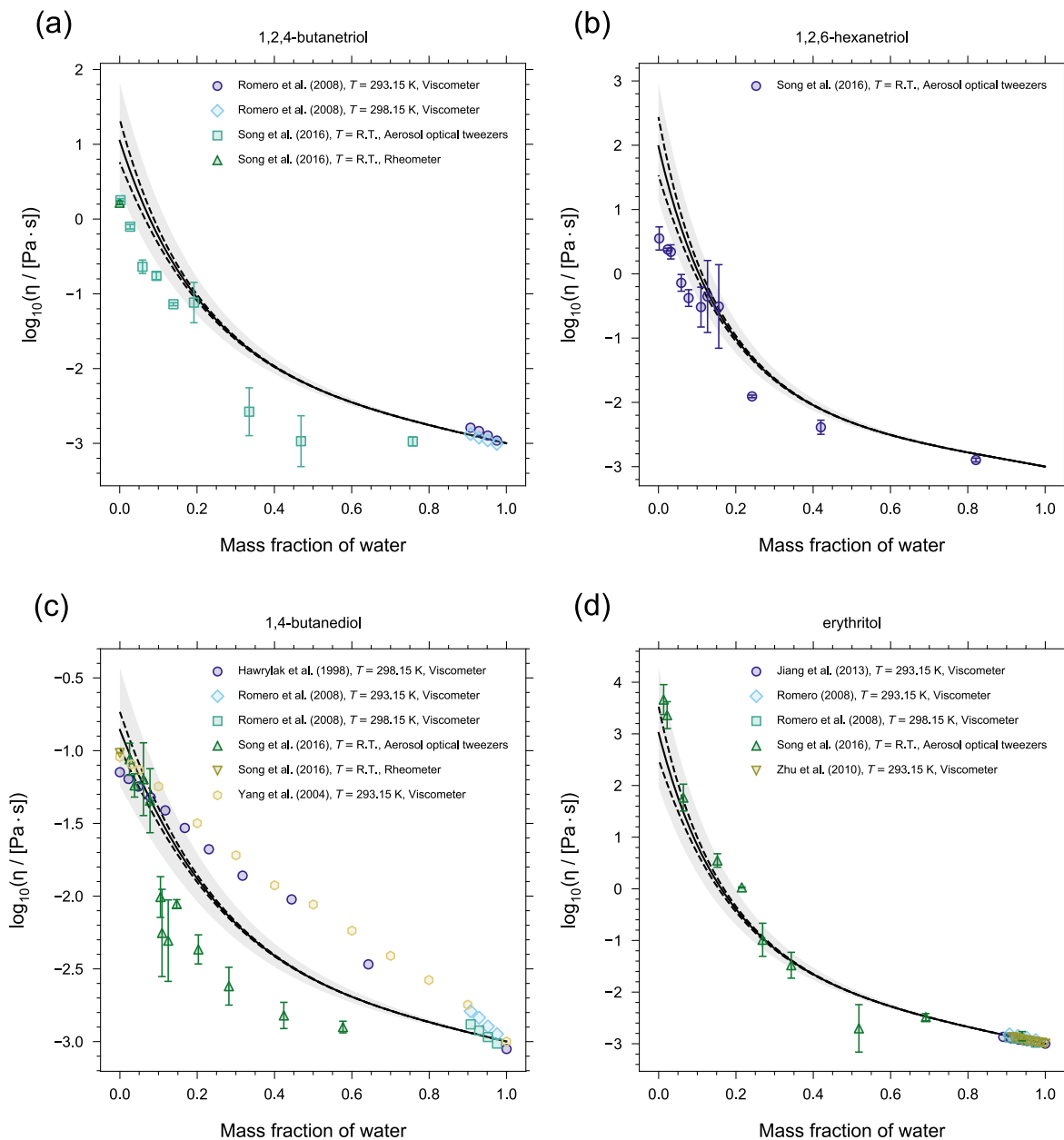
To optimize the mixing model of AIOMFAC-VISC, we attempted to simultaneously fit the mixing model prediction to experimental viscosity data for the binary aqueous mixtures shown in Figs. S4, S5, and S6. The fit is captured by an adjustable parameter multiplied by the residual component of the mixture viscosity model. The determination of an optimal fit parameter is a global minimization problem, ideally approached by using a set of global optimization methods. For this, we used the optimization approach described by Zuend et al. (2011). The optimal fit parameter was determined to be  $\sim 1.0$ , therefore no further adjustments were made to the mixture viscosity model aside from those adjustments made to the original Cao et al. (1993) formulation described in the main text.

## S6 Determination of SOA systems

For all three SOA systems simulated in this work, each surrogate compound was assigned a fixed molar concentration in the particulate matter (PM). These fixed molar concentrations in  $\text{mol m}^{-3}$  (of air) are listed in Tables S2, S3, and S4 for  $\alpha$ -pinene-, toluene-, and isoprene-derived SOA, respectively. To determine those molar concentration of constituents for the  $\alpha$ -pinene and isoprene SOA systems, we begin by calculating the equilibrium gas-particle partitioning of the surrogate species in each SOA system using the MCM-EVAPORATION-AIOMFAC approach (Zuend et al., 2011) where the initial total molar concentrations (PM plus gas phase) for  $\alpha$ -pinene and isoprene SOA were taken from Zuend and Seinfeld (2012) and Rastak et al. (2017), respectively. We extract the molar concentration of each constituent in the PM phase for a relative humidity of 40 %. When relative humidity is held at 40%, the average O : C ratio of the SOA produced via our gas-particle partitioning prediction is representative of known O : C ratios from experiments. We then hold the molar concentrations of organics in the PM constant during calculations of mixture viscosity. In the case of  $\alpha$ -pinene SOA, we have made one additional adjustment by scaling the molar amount of surrogate compound C108OOH in the PM phase by a factor of 30. This is done to better match the curvature of the experimental viscosity data at high relative humidity. In the case of toluene SOA, we have selected several constituents from the MCM-derived list of surrogate components from toluene photo-oxidation by OH radicals. To determine the molar concentrations of a given constituent ( $n_i$ ) in the PM phase we use the following formula:

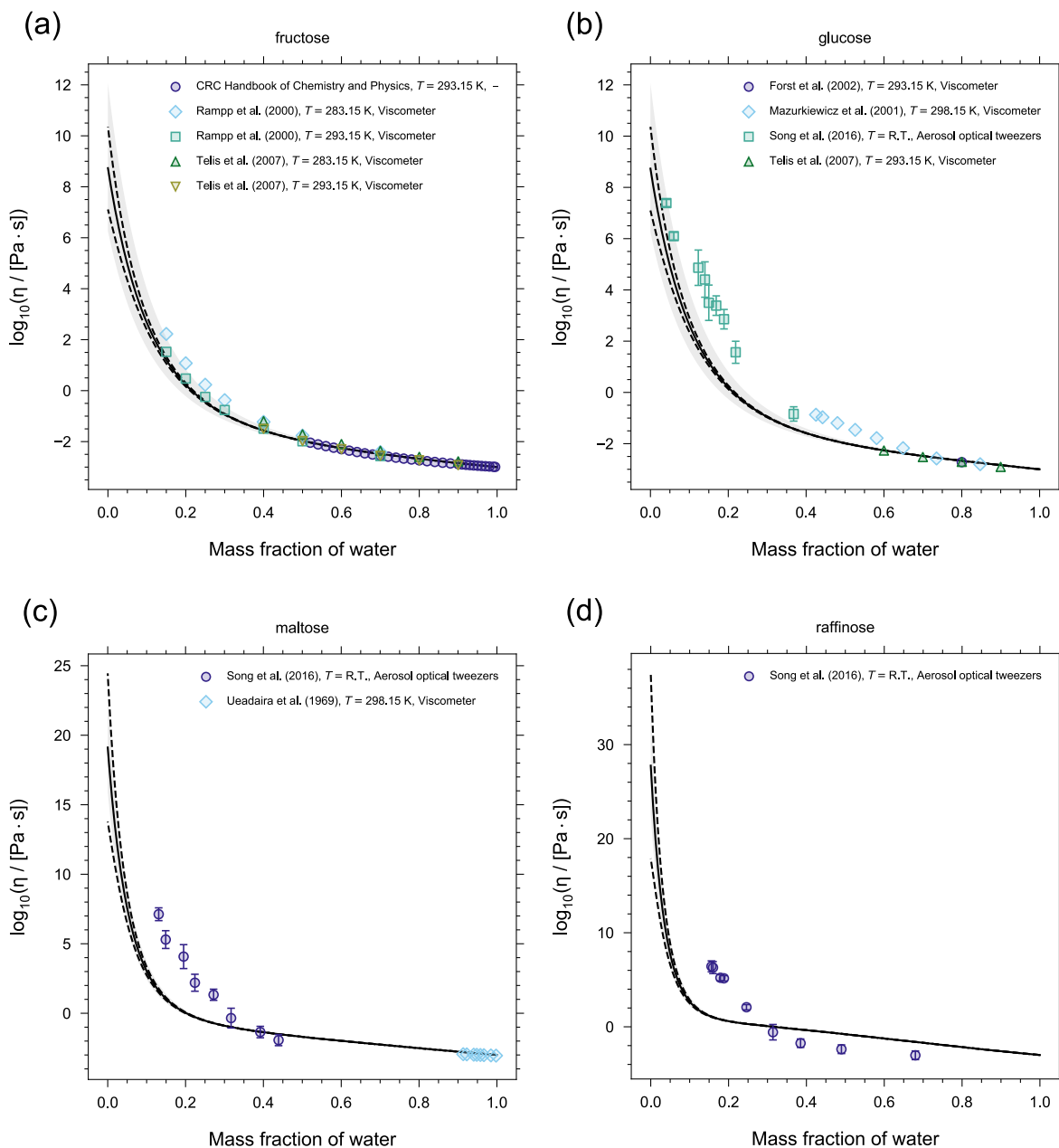
$$n_i = \text{O : C} \times T_g \times 10^{-10}. \quad (\text{S5})$$

Using this scaling results in the O : C of the SOA produced to be similar to what is expected from laboratory chamber experiments. We note here that we have increased the concentration of compound C535OOH by a factor of 5 to increase the average mixture O:C from 0.96 to 1.12.

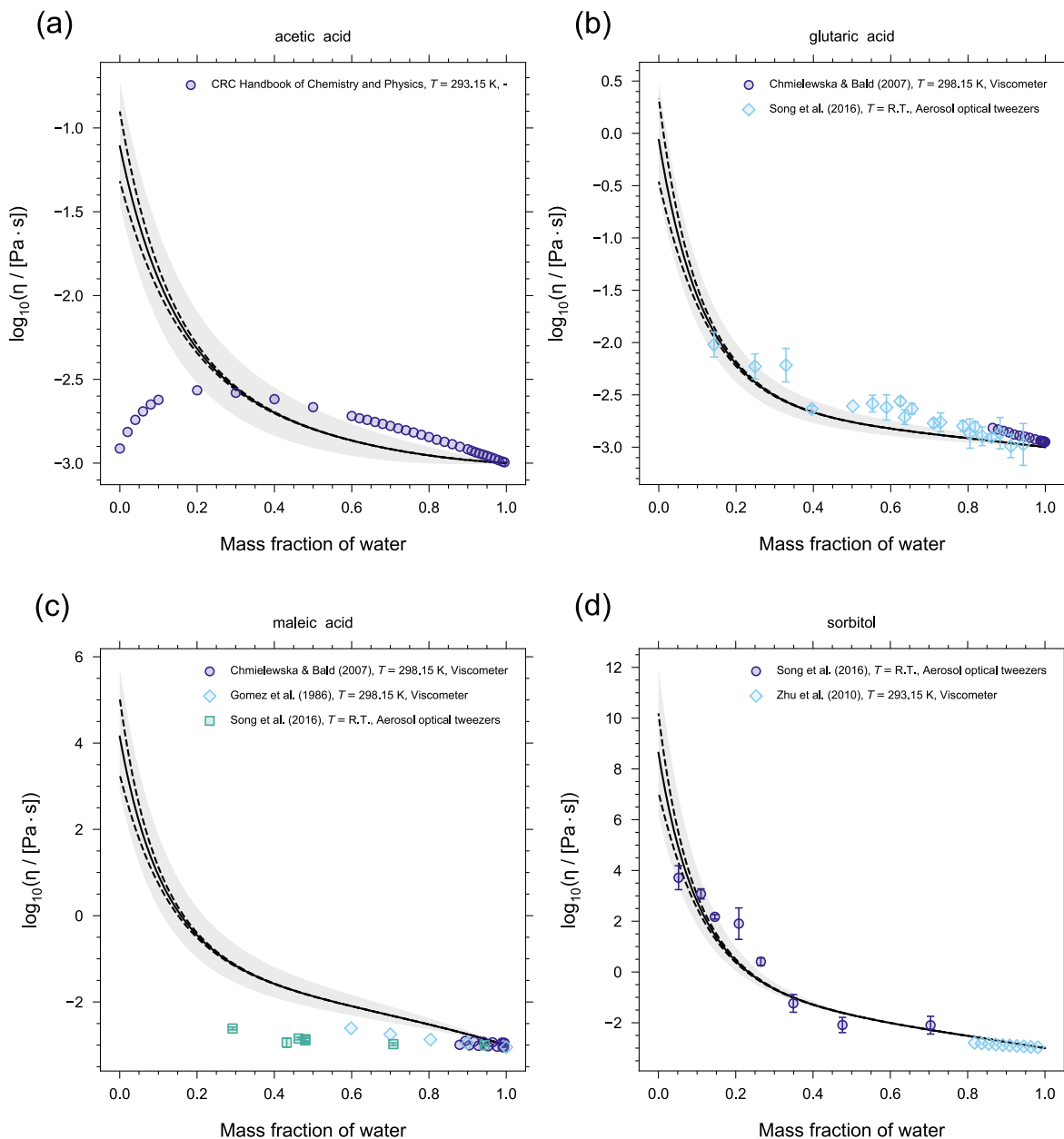


**Figure S4.** AIOMFAC-VISC mixture viscosity predictions as a function of mass fraction of water at 293.15 K for (a) 1,2,4-butanetriol, (b) 1,2,6-hexanetriol, (c) 1,4-butanediol, and (d) erythritol. The solid black line is the AIOMFAC-VISC mixture viscosity prediction. The dashed black lines show the AIOMFAC-VISC sensitivity. The sensitivity is assessed by calculating the response of the model to a small change in mixture composition. The grey shaded region denotes a 5 % uncertainty in the prediction of  $T_g$ . Markers show experimental data. Error bars have been omitted when the length of the error bar does not exceed the width of the marker.





**Figure S5.** AIOMFAC-VISC mixture viscosity predictions as a function of mass fraction of water at 293.15 K for (a) fructose, (b) glucose, (c) maltose, and (d) raffinose. The solid black line is the AIOMFAC-VISC mixture viscosity prediction. The dashed black lines show the AIOMFAC-VISC sensitivity. The sensitivity is assessed by calculating the response of the model to a small change in mixture composition. The grey shaded region denotes a 5 % uncertainty in the prediction of  $T_g$ . Markers show experimental data. Error bars have been omitted when the length of the error bar does not exceed the width of the marker.



**Figure S6.** AIOMFAC-VISC mixture viscosity predictions as a function of mass fraction of water at 293.15 K for (a) acetic acid, (b) glutaric acid, (c) maleic acid, and (d) sorbitol. The solid black line is the AIOMFAC-VISC mixture viscosity prediction. The dashed black lines show the AIOMFAC-VISC sensitivity. The sensitivity is assessed by calculating the response of the model to a small change in mixture composition. The grey shaded region denotes a 5 % uncertainty in the prediction of  $T_g$ . Markers show experimental data. Error bars have been omitted when the length of the error bar does not exceed the width of the marker.

**Table S2.** MCM-derived surrogate components for alpha-pinene oxidation by ozone and their fixed amounts in  $\text{mol m}^{-3}$  in the particulate matter (PM) phase.

Name (MCM)	O:C	$M$ ( $\text{g mol}^{-1}$ )	$\text{mol m}^{-3}$ in PM phase
C107OOH	0.4	200.231	$2.1860 \times 10^{-10}$
PINONIC	0.3	184.232	$1.2356 \times 10^{-10}$
C97OOH	0.44	188.22	$2.5175 \times 10^{-9}$
C108OOH	0.5	216.231	$8.4010 \times 10^{-8}$
C89CO2H	0.33	170.206	$2.010 \times 10^{-11}$
PINIC	0.444	186.205	$8.0263 \times 10^{-9}$
C921OOH	0.56	204.220	$9.2106 \times 10^{-9}$
C109OOH	0.4	200.231	$1.5748 \times 10^{-11}$
C812OOH	0.625	190.194	$8.4291 \times 10^{-9}$
HOPINONIC	0.4	200.232	$2.3266 \times 10^{-9}$
C811OH	0.375	158.094	$8.9370 \times 10^{-11}$
C813OOH	0.75	206.193	$3.2969 \times 10^{-9}$
ALDOL dimer	0.375	368.421	$5.9996 \times 10^{-10}$
ESTER dimer	0.375	368.421	$2.3998 \times 10^{-9}$

The ALDOL dimer and ESTER dimer are not predicted by MCM. Justification for including the dimers can be found in Zuend and Seinfeld (2012).

The average O:C ratio of the predicted  $\alpha$ -pinene SOA mixture is 0.507 (for  $27.248 \mu\text{g m}^{-3}$  of SOA formed at  $T = 293.15 \text{ K}$ ).

**Table S3.** MCM-derived surrogate components for toluene oxidation by OH and their fixed amounts in  $\text{mol m}^{-3}$  in the particulate matter (PM) phase.

Name (MCM)	O:C	$M$ ( $\text{g mol}^{-1}$ )	$\text{mol m}^{-3}$ in PM phase
C5134CO2OH	0.8	130.099	$1.9868 \times 10^{-8}$
C5CO234	0.6	114.099	$1.3525 \times 10^{-8}$
PMALNHY2OH	0.714	174.151	$1.9267 \times 10^{-8}$
C6H5CH2OOH	0.286	124.137	$5.8337 \times 10^{-9}$
CRESOOH	0.857	190.151	$2.4241 \times 10^{-8}$
TLEPOXMUC	0.429	140.137	$1.9987 \times 10^{-8}$
MALANHY	0.75	98.057	$1.6884 \times 10^{-8}$
C3DIALOOH	1.333	104.062	$3.0168 \times 10^{-8}$
C33CO	1.0	86.046	$2.2626 \times 10^{-8}$
C23O3CCHO	0.8	130.099	$1.9868 \times 10^{-8}$
C535OOH	1.4	180.113	$2.0366 \times 10^{-7}$
C534OOH	1.4	180.113	$4.0863 \times 10^{-8}$

The average O:C ratio of the predicted toluene SOA mixture is 1.12 (for  $301 \mu\text{g m}^{-3}$  of SOA formed at  $T = 295.15 \text{ K}$ ).

**Table S4.** MCM-derived surrogate components for isoprene photo-oxidation and their fixed amounts in  $\text{mol m}^{-3}$  in the particulate matter (PM) phase.

Name (MCM)	O:C	$M$ ( $\text{g mol}^{-1}$ )	$\text{mol m}^{-3}$ in PM phase
IEB1OOH	1.0	150.1120	$2.1859 \times 10^{-9}$
IEB2OOH	1.0	150.1120	$3.8058 \times 10^{-11}$
C59OOH	1.0	150.0940	$6.4468 \times 10^{-9}$
IEC1OOH	1.0	150.0940	$2.2503 \times 10^{-9}$
C58OOH	1.0	150.1120	$2.2710 \times 10^{-10}$
IEPOXA	0.6	118.1308	$1.6303 \times 10^{-31}$
C57OOH	1.0	150.1120	$1.8452 \times 10^{-10}$
IEPOXC	0.6	118.1308	$3.7912 \times 10^{-21}$
HIEB1OOH	1.2	166.1120	$2.3492 \times 10^{-9}$
INDOOH	1.4	197.1380	$1.6072 \times 10^{-9}$
IEACO3H	1.0	148.0960	$1.8935 \times 10^{-19}$
C525OOH	1.2	166.0940	$1.7850 \times 10^{-9}$
HIEB2OOH	1.2	166.1120	$1.0495 \times 10^{-9}$
IEC2OOH	1.0	148.0600	$2.0814 \times 10^{-17}$
INAOOH	1.4	197.1380	$7.2618 \times 10^{-10}$
C510OOH	1.4	195.1040	$5.5325 \times 10^{-13}$
INB1OOH	1.4	197.1380	$4.6077 \times 10^{-10}$
IECCO3H	1.0	148.1148	$1.2558 \times 10^{-17}$
INCOOH	1.4	197.1380	$8.7075 \times 10^{-11}$
INB2OOH	1.4	197.1380	$1.8653 \times 10^{-10}$
Tetrol dimer	1.43	254.2768	$3.9110 \times 10^{-18}$

The average O:C ratio of the predicted isoprene SOA mixture is 1.12 (for  $3.406 \mu\text{g m}^{-3}$  of SOA formed at  $T = 295.15 \text{ K}$ ). See the SI of Rastak et al. (2017) for chemical formulas and justification for the tetrol dimer.

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