

*Supplementary of*

## **Liquid-liquid phase separation in organic particles containing one and two organic species: importance of the average O:C**

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### **S1. Calculation of average O:C of particles containing two organic species**

The average O:C of particles containing two organic compounds was calculated using the following equation (Song et al., 2012):

$$O:C = \frac{\frac{mf_1 \times O_1}{MW_1} + \frac{mf_2 \times O_2}{MW_2}}{\frac{mf_1 \times C_1}{MW_1} + \frac{mf_2 \times C_2}{MW_2}} \quad (1)$$

where  $MW_1$  and  $MW_2$  refer to the molecular weight of organic species 1 and 2, respectively;

$mf_1$  and  $mf_2$  refer to the mass fraction of organic species 1 and 2 in the mixture in the absence of water; and  $O_1$  and  $O_2$  refer to number of oxygen atoms in organic species 1 and 2; and  $C_1$  and  $C_2$  refer to the number of carbon atoms in organic species 1 and 2, respectively.

### **S2. Liquid-liquid phase separation in two organic species**

Humidity cycles were performed with particles containing two organic species. Results for increasing RH and decreasing RH are summarized in Table S1.

Table S1. Summary of studies with particles containing two organic species.  $LLPS_{lower}$  represents the lower boundary of the liquid-liquid phase separation determined from experiments with increasing RH and decreasing RH.  $LLPS_{upper}$  represents the upper boundary of the liquid-liquid phase separation determined from the experiments with increasing RH and

1 decreasing RH. Uncertainties represent  $2\sigma$  of multiple measurements and the uncertainty from  
2 the calibration. “No LLPS” indicates that only one phase was observed for the full range of  
3 relative humidity (RH) explored (approximately 0 to 100 % RH). Average O:C of the mixtures  
4 containing two organic species are also included.

Mixtures	Average O:C	Increasing RH		Decreasing RH	
		LLPS <sub>lower</sub>	LLPS <sub>upper</sub>	LLPS <sub>lower</sub>	LLPS <sub>upper</sub>
		(%)	(%)	(%)	(%)
Diethyl sebacate, Propylene glycol	0.33	92.4±2.1	100±2.0	91.5±2.3	100±2.0
Diethyl sebacate, Glyceryl tributyrate	0.34	96.3±4.3	100±2.0	95.6±3.9	100±2.0
Diethyl sebacate, Suberic acid monomethyl ester	0.36	97.4±3.1	100±2.0	95.5±3.3	100±2.0
Diethyl sebacate, Polyethylene glycol-400	0.39	5.2±3.8	100±2.0	5.2±3.6	100±2.0
Glyceryl tributyrate, Propylene glycol	0.39	93.8±2.3	100±2.0	93.1±2.9	100±2.0
Propylene glycol, Suberic acid monomethyl ester	0.42	97.7±2.3	100±2.0	96.8±2.1	100±2.0
Glyceryl tributyrate, Suberic acid monomethyl ester	0.42	97.6±3.4	100±2.0	96.7±2.4	100±2.0
Propylene glycol, Polyethylene glycol-400	0.47	73.9±2.5	89.9±3.0	74.4±2.4	89.3±2.2
Glyceryl tributyrate, Polyethylene glycol-400	0.47	16.0±2.3	100±2.0	15.9±2.6	100±2.0
Diethyl sebacate, Diethyl L- tartrate	0.48	92.8±3.4	100±2.0	92.8±3.3	100±2.0
Suberic acid monomethyl ester, Polyethylene glycol-400	0.50	93.2±3.2	100±2.0	93.0±2.9	100±2.0
Glyceryl tributyrate, Diethyl L-	0.55	93.9±3.3	100±2.0	94.0±3.3	100±2.0

tartrate					
Polyethylene glycol-400, Diethyl L- tartrate	0.56	No LLPS		No LLPS	
Suberic acid monomethyl ester, Diethyl L- tartrate					
	0.58	97.3±2.7	100±2.0	95.9±2.6	100±2.0
Diethyl L- tartrate, Polyethylene glycol-400	0.68	No LLPS		No LLPS	

## Reference

Song, M., Marcolli, C., Krieger, U. K., Zuend, A., and Peter, T.: Liquid-liquid phase separation in aerosol particles: Dependence on O:C, organic functionalities, and compositional complexity, Geophys. Res. Lett., 39, Artn L19801, Doi 10.1029/2012gl052807, 2012.