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Effects of molecular weight and temperature on liquid–liquid phase separation in particles containing organic species and ammonium sulfate

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Abstract

Atmospheric particles containing organic species and inorganic salts may undergo liquid–liquid phase separation when the relative humidity varies between high and low values. To better understand the parameters that affect liquid–liquid phase separation in atmospheric particles, we studied the effects of molecular weight and temperature on liquid–liquid phase separation in particles containing one organic species mixed with ammonium sulfate. In the molecular weight dependent studies, we measured liquid–liquid phase separation relative humidity (SRH) in particles containing ammonium sulfate and organic species with large molecular weights (up to 1153 Da). These results were combined with recent studies of liquid–liquid phase separation in the literature to assess if molecular weight is a useful parameter for predicting SRH. The combined results, which include results from 33 different particle types, illustrate that SRH does not depend strongly on molecular weight (i.e. a clear relationship between molecular weight and SRH was not observed). In the temperature dependent studies, we measured liquid–liquid phase separation in 20 particle types at 244 ± 1 K, 263 ± 1 K, and 278 ± 1 K, as well as 290 ± 1 K for a few of these particle types. These new results were combined with previous measurements of the same particle types at 290 ± 1 K. The combined SRH data illustrate that for the particle types studied the SRH does not depend strongly on temperature. At most the SRH varied by 9.7 % as the temperature varied from 290 to 244 K. In addition, for all the particle types studied and at all the temperatures studied, liquid–liquid phase separation was always observed when the $O:C < 0.57$, frequently observed when $0.57 \leq O:C < 0.8$, and never observed when $O:C \geq 0.8$. These combined results suggest that liquid–liquid phase separation is likely a common occurrence in the atmospheric particles at temperatures from 244–290 K. Additional studies at temperatures < 244 K and with other organic species are still needed.

1 Introduction

A large fraction of particles in the atmosphere contains both organic species and inorganic salts (Murphy and Thomson, 1997; Murphy et al., 1998, 2006; Middlebrook et al., 1998; Buzorius et al., 2002; Murphy, 2005; Tolocka et al., 2005; Chen et al., 2009; Pratt and Prather, 2010). Ammonium sulfate is one of the most common inorganic salts in these particles (Dibb et al., 1996, 2000; Huebert et al., 1998; Talbot et al., 1998; Lee et al., 2003). The types of organic species in these particles are numerous with only around 10 % by mass of these organic species identified at the molecular level (Hamilton et al., 2004; Goldstein and Galbally, 2007; Decesari et al., 2006; Hallquist et al., 2009). Organic functional groups found in the mixed organic-inorganic salt particles include carboxylic acids, alcohols, oxidized aromatic compounds, ethers, and esters (Finlayson-Pitts and Pitts, 2000; Seinfeld and Pandis, 2006; Decesari et al., 2006; Hallquist et al., 2009; Takahama et al., 2011; Rogge et al., 1993; Saxena and Hildemann, 1996; Finlayson-Pitts and Pitts, 1997; Day et al., 2009; Gilardoni et al., 2009; Liu et al., 2009; Russell et al., 2009, 2011; Fu et al., 2011; Fuzzi et al., 2001). The molecular weight of organic molecules in these mixed particles is thought to range from less than 100 Da to as high as 1000 Da (Gao et al., 2004; Tolocka et al., 2004; Kalberer et al., 2004; Nguyen et al., 2010). In addition, the average oxygen-to-carbon elemental ratios (O : C) of the organic material in ambient particles generally range from 0.1–1.0 (Chen et al., 2009; Aiken et al., 2008; DeCarlo et al., 2008; Jimenez et al., 2009; Hawkins et al., 2010; Heald et al., 2010; Ng et al., 2010; Takahama et al., 2011), while the average organic-to-inorganic mass ratios (OIR) of atmospheric particles generally range from 0.2–3.5 (Chen et al., 2009; Zhang et al., 2007; Jimenez et al., 2009). When the relative humidity varies in the atmosphere, mixed organic-inorganic salt particles can undergo different phase transitions, including efflorescence, deliquescence and liquid–liquid phase separation (Martin, 2000; Clegg et al., 2001; Brooks et al., 2002, 2003; Choi and Chan, 2002; Chan and Chan, 2003; Pankow, 2003; Wise et al., 2003; Braban and Abbatt, 2004; Pant et al., 2004; Parsons et al., 2004a, 2006; Badger et al., 2006;

Chang and Pankow, 2006; Erdakos et al., 2006; Marcolli and Krieger, 2006; Salcedo, 2006; Anttila et al., 2007; Buajarern et al., 2007; Ling and Chan, 2008; Zuend et al., 2008, 2010, 2011; Ciobanu et al., 2009; Treuel et al., 2009; Bodsworth et al., 2010; Kwamena et al., 2010; Prisle et al., 2010; Bertram et al., 2011; Reid et al., 2011; Smith
5 et al., 2011; Krieger et al., 2012; Pöhlker et al., 2012; Smith et al., 2012; Song et al., 2012a, b; You et al., 2012, 2013; Zuend and Seinfeld, 2012, 2013; Drozd et al., 2013; Shiraiwa et al., 2013; Song et al., 2013; Veghte et al., 2013, 2014). Of these phase transitions, recent laboratory measurements and calculations have focused on liquid–liquid phase separation. This phase transition can influence the partitioning of organic
10 molecules between the gas and the particle phase (Seinfeld et al., 2001; Chang and Pankow, 2006; Zuend et al., 2010; Shiraiwa et al., 2013) and influence the reactive uptake of important gas-phase molecules, such as N_2O_5, into atmospheric particles (Anttila et al., 2006; Folkers et al., 2003; Escoreia et al., 2010; Cosman and Bertram, 2008; Park et al., 2007; Thornton and Abbatt, 2005; McNeill et al., 2006; Gaston et al.,
15 2014). Liquid-liquid phase separation can also modify the deliquescence and efflorescence relative humidity of the inorganic salt in the mixed organic-inorganic salt particles (Bertram et al., 2011; Smith et al., 2012; Song et al., 2013). In addition, liquid–liquid phase separation can alter the ice nucleation properties of particles (Schill and Tolbert, 2013).

20 Recent work has investigated different parameters that influence liquid–liquid phase separation in particles containing mixtures of organic species and inorganic salts (You et al., 2014). Understanding the parameters that affect these transitions is necessary for predicting these phase transitions in atmospheric particles. Studies have shown that O : C is an important parameter for predicting liquid–liquid phase separation in these
25 particles, with this phase transition always observed for O : C less than 0.5, frequently observed for O : C values between 0.5 and 0.8, and never observed for O : C values greater than 0.8 (Bertram et al., 2011; Song et al., 2012a, b; You et al., 2013). Studies have also shown that the type of functional groups can also influence the relative humidity required for liquid–liquid phase separation when the O : C of the organic ma-

terial is in the range of 0.56–0.8 (Song et al., 2012b). On the other hand, previous studies have suggested that the occurrence of liquid–liquid phase separation is not a strong function of the OIR, the number of organic species, or the type of inorganic salt in mixed organic-inorganic salt particles (Marcolli and Krieger, 2006; Ciobanu et al., 2009; Bertram et al., 2011; Song et al., 2012a, b; You et al., 2013; Schill and Tolbert, 2013).

In the following paper, we investigate if molecular weight of the organic material and temperature of the particles influence liquid–liquid phase separation in particles containing organic species and inorganic salts. The effect of molecular weight of the organic material on the occurrence of liquid–liquid phase separation in mixed organic-inorganic salt particles has not been explored. However, recent studies have shown that molecular weight of the organic material is important for predicting glass transitions in organic particles (Koop et al., 2011; Saukko et al., 2012; Zobrist et al., 2008). Here we carried out a systematic study of the effect of molecular weight of the organic material on liquid–liquid phase separation. We first studied liquid–liquid phase separation in particles containing ammonium sulfate mixed with one of ten organic species, with molecular weights up to 1153 Da. The data from these studies were then combined with recent studies of liquid–liquid phase separation in particles reported in the literature (You et al., 2013) to assess if molecular weight is a useful parameter to predict the occurrence of liquid–liquid phase separation and the liquid–liquid phase separation relative humidity (SRH).

In the troposphere, the temperature ranges from approximately 220–300 K. However, only two studies have investigated liquid–liquid phase separation in mixed organic-inorganic salt particles at temperatures below 290 K. Bertram et al. reported that SRH results were similar at 273 K and 290 K for particles containing ammonium sulfate and 1,2,6-hexanetriol (Bertram et al., 2011). Schill and Tolbert reported that SRH results were similar for temperatures from 240 to 265 K for particles containing ammonium sulfate and 1,2,6-hexanetriol and particles containing ammonium sulfate, 1,2,6-hexanetriol, and 2,2,6,6-tetrakis(hydroxymethyl)cyclohexanol (Schill and Tolbert,

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2013). To gain a better understanding of the effect of temperature on liquid–liquid phase separation in mixed organic-inorganic salt particles, we investigated liquid–liquid phase separation in particles containing ammonium sulfate mixed with one of twenty organic species at $244 \pm 1\,K$, $263 \pm 1\,K$, and $278 \pm 1\,K$, respectively. Some of these particle types were also studied at $290 \pm 1\,K$. These new data were combined with previous measurements of liquid–liquid phase separation at $290 \pm 1\,K$ by You et al. (You et al., 2013) to assess the effect of temperature on liquid–liquid phase separation in mixed organic-inorganic salt particles.

2 Experimental

2.1 Sample preparation and apparatus

Solutions of ammonium sulfate and one organic species were prepared in high purity water (Millipore, $18.2\,M\Omega\,cm$) with OIR of 2.0 ± 0.1. The solutions were then nebulized to produce submicron particles, which impacted onto a hydrophobic glass slide (Hampton Research) and coagulated into super-micron droplets. Water was then evaporated to generate mixed organic-ammonium sulfate particles with lateral dimensions ranging from 10 to 35 µm.

The glass slide supporting the organic-ammonium sulfate particles was mounted to a temperature and relative humidity controlled flow cell, which was coupled to an optical reflectance microscope (Zeiss Axiotech; 50 × objective) (Koop et al., 2000; Parsons et al., 2004b; Pant et al., 2006; Bodsworth et al., 2010). To control the relative humidity in the flow cell, dry and humidified nitrogen gas flows were combined and continuously passed through the cell. The total flow rate was approximately $1.5\,L\,min^{-1}$. While the RH was decreased, liquid–liquid phase separation was identified by monitoring the change in morphology of the particles. The relative humidity of the gas was determined with a chilled mirror hygrometer (General Eastern, Model 1311DR), which

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was calibrated by measuring the deliquescence relative humidity of ammonium sulfate particles.

2.2 Molecular weight dependent studies

Particles containing ammonium sulfate mixed with one of ten organic species were studied at 290 ± 1 K (see Table 1). Most of previous laboratory studies of liquid–liquid phase separation in particles containing organic species mixed with ammonium sulfate used organic species with molecular weight less than 200 Da (Bertram et al., 2011; Song et al., 2012a, b; You et al., 2013). To complement these previous studies, in the current study we studied particles containing ammonium sulfate and organic species with molecular weight ranging from 180 to 1153 Da (see Table 1). The specific organic species selected for these studies (Table 1) also had a relatively wide range of O : C values. The organic-to-inorganic mass ratio (OIR) was 2.0 ± 0.1 in all the studies.

In a typical experiment, the RH in the cell was ramped down at a rate of 0.4–0.6 % RH min^{-1}, while the temperature of the cell was held at 290 ± 1 K. At the same time, images of the particles were captured continuously until one of the following conditions occurred: liquid–liquid phase separation was observed, the particles effloresced, or the RH reached ≤ 0.5 %. For each type of particles, experiments were repeated at least three times. All of the organic species were purchased from Sigma-Aldrich and had purities ≥ 95 %, except for maltoheptaose, which had a purity ≥ 90 %.

2.3 Temperature dependent studies

Particles consisting of ammonium sulfate mixed with one of twenty organic species were studied (see Table 2) at 244 ± 1 K, 263 ± 1 K, and 278 ± 1 K, as well as 290 ± 1 K for a few of these particle types. The organic species studied (see Table 2) were selected because they had a wide range of O : C values and included functional groups found in atmospheric particles. In addition, most of these organic species were studied previously at 290 K (You et al., 2013), so comparisons could be made between the

current studies and the previous studies by You et al. (2013). The OIR was 2.0 ± 0.1 in all the studies. The organic species studied were purchased from Sigma-Aldrich with purities $\geq 98\,\%$. All organics were used without further purification. In a typical experiment, the RH was ramped down at a rate of around $0.1–0.5\,\%\,\mathrm{RH\,min^{-1}}$. Images were recorded in the same way as the molecular weight dependent studies.

3 Results and discussion

3.1 Effect of molecular weight on liquid–liquid phase separation

Particles containing ammonium sulfate mixed with one of ten organic species were studied at $290 \pm 1\,\mathrm{K}$. In these studies organic species with large molecular weights (180 to 1153 Da) were used. The organic-to-inorganic mass ratio (OIR) was 2.0 ± 0.1 in all the studies. The results from these studies are listed in Table 3 as well as the data from You et al., who also studied particles with an OIR of 2.0 ± 0.1 at $290 \pm 1\,\mathrm{K}$ (You et al., 2013). The combined data set in Table 3, which includes results on liquid–liquid phase separation for 33 different particle types and includes results for organic species with molecular weights ranging from 92 to 1153 Da, was used to determine the importance of molecular weight of the organic species on liquid–liquid phase separation.

In Fig. 1, the data from Table 3 were plotted as a function of O : C and molecular weight. Open circles indicate liquid–liquid phase separation was observed and stars indicate liquid–liquid phase separation was not observed. No clear relationship between molecular weight and the occurrence of liquid–liquid phase separation was observed, however a relationship between occurrence of liquid–liquid phase separation and O : C was clear: liquid–liquid phase separation was always observed when O : C < 0.57 (orange hatched region), was never observed when O : C > 0.83 (green hatched region), and was frequently observed when O : C ranged from 0.57–0.83.

In Fig. 2, the SRH data from Table 3 were plotted as a function of molecular weight of the organic species. The colors of the symbols indicate the O : C of organic species

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in the mixed particles. Data at SRH = 0 % indicate liquid–liquid phase separation was not observed even at the lowest relative humidity studied. Similar to Fig. 1, no correlation with molecular weight was apparent. For contrast, in Fig. 3, we show the same SRH data plotted as a function of O : C of organic species with the color of the symbols representing the molecular weight of organic species. The solid curve is a Sigmoidal–Boltzmann fit to all the SRH data. Following Song et al. (Song et al., 2012b), a Sigmoidal–Boltzmann function was chosen to avoid unphysical values at high and low SRH values (negative values or values greater than 100 %). The fit to the current SRH data set is qualitatively consistent with previous fits for SRH values of mixed organic-ammonium sulfate particles (Bertram et al., 2011; Song et al., 2012b; You et al., 2013). Consistent with previous results (Bertram et al., 2011; Song et al., 2012b), a correlation between SRH and O : C is apparent. These results suggest that O : C is more important for predicting the occurrence of liquid–liquid phase separation in atmospherically relevant mixed organic-ammonium sulfate particles compared with molecular weight.

3.2 Effect of temperature on liquid–liquid phase separation

In the studies that explored the effect of temperature on liquid–liquid phase separation, particles containing ammonium sulfate mixed with one of twenty organic species were studied at 244 ± 1 K, 263 ± 1 K, and 278 ± 1 K. Some of these particle types were also studied at 290 ± 1 K. These new results were included in Table 4 as well as results from You et al., who studied most of the same types of particles but only at 290 ± 1 K (You et al., 2013). The combined SRH data from Table 4, which cover the temperature range of 290–244 K, were plotted in Fig. 4a as a function of O : C of the organic species. Data at SRH = 0 % indicate liquid–liquid phase separation was not observed even at the lowest relative humidity studied. The results show that for all the particle types studied and at all the temperatures studied liquid–liquid phase separation was always observed when O : C < 0.57, frequently observed when $0.57 \leq$ O : C < 0.8, and never observed when O : C $\geq$ 0.8. We conclude that the O : C range at which liquid–liquid

phase separation was observed at 290–293 K in previous studies (Ciobanu et al., 2009; Bertram et al., 2011; Song et al., 2012b; You et al., 2013), is consistent with the range observed at temperatures down to 244 ± 1 K.

Figure 5 shows the same data as in Fig. 4a, but displayed in a slightly different way. The SRH results for the twelve types of mixed organic-ammonium sulfate particles that underwent liquid–liquid phase separation in Fig. 4a, are shown as a function of temperature, with the colors of the symbols representing the O : C values of the organic species in the particles. For all the particle types included in Fig. 5, the SRH varied by less than 9.7 % RH as the temperature varied from 244 to 290 K. Figures 4a and 5 illustrate that SRH is not a strong function of temperature for the particle types investigated. These results are consistent with earlier studies by Bertram et al. and Schill and Tolbert discussed in the Introduction.

The average O : C of organic material in atmospheric particles has been measured at many locations in the Northern Hemisphere and in the Amazon and has been shown to range from 0.1 to 1.0 (Chen et al., 2009; Aiken et al., 2008; DeCarlo et al., 2008; Jimenez et al., 2009; Hawkins et al., 2010; Heald et al., 2010; Ng et al., 2010; Takahama et al., 2011). This range of O : C values is indicated in Fig. 4b. The range of average O : C values measured in the atmosphere overlaps with the range of O : C values where liquid–liquid phase separation was observed at temperatures ranging from 244 to 290 K. This overlap suggests that liquid–liquid phase separation is likely a common occurrence in the atmosphere over this temperature range.

Although SRH does not appear to be a strong function of the temperature for the particle types investigated over the temperature range of 244 to 290 K, SRH may be a strong function of temperature at temperatures lower than 244 K. Some mixtures of organic species and inorganic salts can become highly viscous at lower temperatures and low relative humidities (Tong et al., 2011; Zobrist et al., 2008, 2011; Koop et al., 2011; Murray, 2008; Mikhailov et al., 2009; Saukko et al., 2012). At these low temperatures and relative humidities, liquid–liquid phase separation may not occur on typical atmospheric time scales due to diffusion limitations. Related, previous research has

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shown that efflorescence is inhibited in particles containing organic species and in-organic salts at low temperatures due to diffusion limitations (Bodsworth et al., 2010; Schill and Tolbert, 2013). Additional studies of SRH at temperatures lower than 244 K and for other particle types are still needed. Possible particle types that may show a stronger dependence on temperature are particles containing organic species with high molecular weights and particles that undergo liquid–liquid phase separation at low relative humidities, since viscosity is a strong function of relative humidity (Koop et al., 2011; Renbaum-Wolff et al., 2013; Power et al., 2013; Kidd et al., 2014).

4 Conclusions

Studies with particles containing ammonium sulfate mixed with one organic species illustrated that the occurrence of liquid–liquid phase separation and SRH do not de-pend strongly on the molecular weight of the organic species, at least for the particle types studied. These current studies also illustrate that the occurrence of liquid–liquid phase separation and SRH do not depend strongly on temperature over the range of 290–244 K. For all the particle types studied and at all the temperatures studied (290 ± 1 K to 244 ± 1 K) liquid–liquid phase separation was always observed when the $O:C < 0.57$, frequently observed when $0.57 \le O:C < 0.8$, and never observed when $O:C \ge 0.8$. The SRH varied by at most 9.7 % RH as the temperature varied from 290 K to 244 K for the particle types studied. The combined results suggest that liquid–liquid phase separation is likely a common occurrence in the atmospheric particles at 244–290 K. At the same time, additional studies of SRH at temperatures lower than 244 K and for other particle types are still needed.

Appendix A

Parameterizations of SRH results at 290 ± 1 K as a function of the oxygen-to-carbon elemental ratio ($O:C$) of the organic species. The range of $O:C$ in this study is from

0.29 to 1.33.

$$\text{SRH (\%)} = \frac{100}{1 + \exp\left(\frac{(\text{O:C}) - 0.69}{0.10}\right)} \times 100\%$$

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Table 1. List of the ten organic species used in molecular weight dependent measurements. Each organic species was separately mixed with ammonium sulfate to make particles, and liquid–liquid phase separation was studied in these particles at 290 ± 1 K.

Compounds	Formula	Molecular weight (Da)	O : C
Glucose	$C_6H_{12}O_6$	180.2	1
Poly (ethylene glycol) bis (carboxymethyl) ether	$C_{2n+4}H_{4n+6}O_{n+5}$	250	0.83
Sucrose	$C_{12}H_{22}O_{11}$	342.3	0.92
Poly (ethylene glycol)	$C_{2n}H_{4n+2}O_{n+1}$	400	0.56
Ouabain	$C_{29}H_{44}O_{12}$	584	0.41
Raffinose	$C_{18}H_{32}O_{16}$	594.5	0.89
Poly (ethylene glycol)	$C_{2n}H_{4n+2}O_{n+1}$	600	0.54
Maltopentaose	$C_{30}H_{52}O_{26}$	829	0.87
Poly (ethylene glycol)	$C_{2n}H_{4n+2}O_{n+1}$	900	0.53
Maltoheptaose	$C_{42}H_{72}O_{36}$	1153	0.86

Table 2. Summary of the twenty organic species used in temperature dependent experiments. Each organic species was separately mixed with ammonium sulfate to make particles, and liquid–liquid phase separation was studied in these particles at 244 ± 1 K, 263 ± 1 K and 278 ± 1 K. Two of the organic species (poly (ethylene glycol) diacrylate and raffinose) mixed with ammonium sulfate were also studied at 290 ± 1 K.

Compounds	Formula	O : C	Molecular weight (Da)	Functional group (s)
2,5-hexanediol	$C_6H_{14}O_2$	0.33	118.2	alcohol
Poly (propylene glycol)	$C_{3n}H_{6n+2}O_{n+1}$	0.38	425	alcohol, ether
Poly (ethylene glycol) diacrylate	$C_{2n+6}H_{4n+6}O_{n+3}$	0.5	575	ester, ether, C-C double bond
Poly (ethylene glycol) 900	$C_{2n}H_{4n+2}O_{n+1}$	0.53	900	alcohol, ether
α,4-dihydroxy-3-methoxybenzeneacetic acid	$C_9H_{10}O_5$	0.56	198.2	alcohol, aromatic, carboxylic acid, ether
Diethylmalonic acid	$C_7H_{12}O_4$	0.57	160.2	carboxylic acid
3,3-dimethylglutaric acid	$C_7H_{12}O_4$	0.57	160.2	carboxylic acid
2,5-hydroxybenzoic acid	$C_7H_6O_4$	0.57	154.2	carboxylic acid, aromatic
Poly (ethylene glycol) 300	$C_{2n}H_{4n+2}O_{n+1}$	0.58	300	alcohol, ether
Poly (ethylene glycol) 200	$C_{2n}H_{4n+2}O_{n+1}$	0.63	200	alcohol, ether
Poly (ethylene glycol) bis (carboxymethyl) ether	$C_{2n+4}H_{4n+6}O_{n+5}$	0.63	600	alcohol, ether
2-methylglutaric acid	$C_6H_{10}O_4$	0.67	146.1	carboxylic acid
2,2-dimethylsuccinic acid	$C_6H_{10}O_4$	0.67	146.2	carboxylic acid
Diethyl-L-tartrate	$C_8H_{14}O_6$	0.75	206.2	alcohol, ether
Glycerol	$C_3H_8O_3$	1.00	92.1	alcohol
Glutaric acid	$C_5H_8O_4$	0.8	132.1	carboxylic acid
Levoglucosan	$C_6H_{10}O_5$	0.83	162.1	alcohol, ether
Raffinose	$C_{18}H_{32}O_{16}$	0.89	594.5	alcohol, ether
Citric acid	$C_6H_8O_7$	1.17	192.1	carboxylic acid
Malonic acid	$C_3H_4O_4$	1.33	104.1	carboxylic acid

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Table 3. Combined data set used to assess the effect of molecular weight on SRH in mixed organic-ammonium sulfate particles. This data set includes the ten types of particles studied here (see Table 1) and the SRH results of twenty-three types of particles containing single organic species and ammonium sulfate studied by You et al. (2013). OIR = 2.0 ± 0.1 in all the experiments. Uncertainties include 2σ of multiple SRH measurements and the uncertainty from the calibration.

Compounds	Formula	Molecular weight (Da)	O : C	SRH (%)	Reference
Glycerol	$C_3H_8O_3$	92.1	1	Not observed	You et al. (2013)
Malonic acid	$C_3H_4O_4$	104.1	1.33	Not observed	You et al. (2013)
Maleic acid	$C_4H_4O_4$	116.1	1	Not observed	You et al. (2013)
2,5-hexanediol	$C_6H_{14}O_2$	118.2	0.33	88.8 ± 3.7	You et al. (2013)
Glutaric acid	$C_5H_8O_4$	132.1	0.8	Not observed	You et al. (2013)
Malic acid	$C_4H_6O_5$	134.1	1.25	Not observed	You et al. (2013)
1,2,6-hexanetriol	$C_6H_{14}O_3$	134.2	0.5	76.7 ± 2.5	You et al. (2013)
2-methylglutaric acid	$C_6H_{10}O_4$	146.1	0.67	75.3 ± 2.8	You et al. (2013)
2,2-dimethylsuccinic acid	$C_6H_{10}O_4$	146.2	0.67	Not observed	You et al. (2013)
2,5-hydroxybenzoic acid	$C_7H_6O_4$	154.2	0.57	Not observed	You et al. (2013)
Diethylmalonic acid	$C_7H_{12}O_4$	160.2	0.57	89.2 ± 3.0	You et al. (2013)
3,3-dimethylglutaric acid	$C_7H_{12}O_4$	160.2	0.57	89.1 ± 3.4	You et al. (2013)
Levoglucosan	$C_6H_{10}O_5$	162.1	0.83	Not observed	You et al. (2013)
Glucose	$C_6H_{12}O_6$	180.2	1	Not observed	Current study
Suberic acid monomethyl ester	$C_9H_{16}O_4$	188.2	0.44	100 ± 2.5	You et al. (2013)
Citric acid	$C_6H_8O_7$	192.1	1.17	Not observed	You et al. (2013)
α,4-dihydroxy-3-methoxybenzeneacetic acid	$C_9H_{10}O_5$	198.2	0.56	72.6 ± 2.6	You et al. (2013)
Poly (ethylene glycol)	$C_{2n}H_{4n+2}O_{n+1}$	200	0.63	79.8 ± 4.1	You et al. (2013)
Diethyl-L-tartrate	$C_8H_{14}O_6$	206.2	0.75	90.2 ± 3.0	You et al. (2013)
Poly (ethylene glycol) bis (carboxymethyl) ether	$C_{2n+4}H_{4n+6}O_{n+5}$	250	0.83	67.6 ± 2.5	Current study
Diethyl sabacate	$C_{14}H_{26}O_4$	258.4	0.29	100 ± 2.5	You et al. (2013)
Poly (ethylene glycol)	$C_{2n}H_{4n+2}O_{n+1}$	300	0.58	86.7 ± 2.8	You et al. (2013)
Sucrose	$C_{12}H_{22}O_{11}$	342.3	0.92	Not observed	Current study
Poly (ethylene glycol)	$C_{2n}H_{4n+2}O_{n+1}$	400	0.56	88.3 ± 3.0	Current study
Poly (propylene glycol)	$C_{3n}H_{6n+2}O_{n+1}$	425	0.38	94.1 ± 3.2	You et al. (2013)
Poly (ethylene glycol) diacrylate	$C_{2n+6}H_{4n+6}O_{n+3}$	575	0.5	94.7 ± 2.5	You et al. (2013)
Ouabain	$C_{29}H_{44}O_{12}$	584	0.41	90.1 ± 3.3	Current study
Raffinose	$C_{18}H_{32}O_{16}$	594.5	0.89	Not observed	Current study
Poly (ethylene glycol)	$C_{2n}H_{4n+2}O_{n+1}$	600	0.54	89.5 ± 2.7	Current study
Poly (ethylene glycol) bis (carboxymethyl) ether	$C_{2n+4}H_{4n+6}O_{n+5}$	600	0.63	92.0 ± 2.7	You et al. (2013)
Maltopentaose	$C_{30}H_{52}O_{26}$	829	0.87	Not observed	Current study
Poly (ethylene glycol)	$C_{2n}H_{4n+2}O_{n+1}$	900	0.53	92.9 ± 2.8	Current study
Maltoheptaose	$C_{42}H_{72}O_{36}$	1153	0.86	Not observed	Current study

Table 4. Combined data set used to determine the effect of temperature on SRH. This includes the measurements at $244 \pm 1\,K$, $263 \pm 1\,K$ and $278 \pm 1\,K$ in the current studies (Table 2) and results from You et al. (2013) at $290 \pm 1\,K$. OIR $= 2.0 \pm 0.1$ in all the experiments. Uncertainties include 2σ of multiple SRH measurements and the uncertainty from the calibration.

Compound	O : C	SRH (%) at different temperatures			
		$244 \pm 1\,K$	$263 \pm 1\,K$	$278 \pm 1\,K$	$290 \pm 1\,K$
2,5-hexanediol	0.33	84.0 ± 5.1	84.6 ± 2.9	88.0 ± 2.5	88.8 ± 3.7[a]
Poly (propylene glycol)	0.38	87.7 ± 3.0	89.4 ± 3.2	92.5 ± 2.6	94.1 ± 3.2[a]
Poly (ethylene glycol) diacrylate	0.5	88.5 ± 3.6	90.4 ± 3.8	95.0 ± 2.5	94.7 ± 2.5
Poly (ethylene glycol) 900	0.53	89.2 ± 6.0	88.7 ± 2.5	91.8 ± 2.7	92.9 ± 2.8[a]
α,4-dihydroxy-3-methoxybenzeneacetic acid	0.56	76.0 ± 2.7	67.3 ± 10.1	72.3 ± 2.9	72.6 ± 2.6[a]
Diethyl malonic acid	0.57	89.8 ± 13.8	87.0 ± 3.3	88.6 ± 2.7	89.2 ± 3.0[a]
3,3-dimethylglutaric acid	0.57	98.2 ± 3.1	88.5 ± 2.7	88.5 ± 2.7	89.1 ± 3.4[a]
2,5-hydroxybenzoic acid	0.57	Not observed	Not observed	Not observed	Not observed[a]
Poly (ethylene glycol) 300	0.58	85.6 ± 6.5	83.6 ± 2.9	85.6 ± 2.8	87.6 ± 2.8[a]
Poly (ethylene glycol) 200	0.63	71.3 ± 2.5	77.2 ± 2.7	79.7 ± 2.5	79.8 ± 4.1[a]
Poly (ethylene glycol) bis (carboxymethyl) ether 600	0.63	87.8 ± 3.8	90.4 ± 2.5	90.4 ± 3.1	92.0 ± 2.7[a]
2-methylglutaric acid	0.67	76.7 ± 3.1	76.2 ± 2.6	76.6 ± 2.5	75.3 ± 2.8[a]
2,2-dimethylsuccinic acid	0.67	Not observed	Not observed	Not observed	Not observed[a]
Diethyl-L-tartrate	0.75	86.9 ± 3.9	85.0 ± 2.9	87.4 ± 2.9	90.2 ± 3.0[a]
Glutaric acid	0.8	Not observed	Not observed	Not observed	Not observed[a]
Levoglucosan	0.83	Not observed	Not observed	Not observed	Not observed[a]
Raffinose	0.89	Not observed	Not observed	Not observed	Not observed
Glycerol	1	Not observed	Not observed	Not observed	Not observed[a]
Citric acid	1.17	Not observed	Not observed	Not observed	Not observed[a]
Malonic acid	1.33	Not observed	Not observed	Not observed	Not observed[a]

[a] Data taken from You et al. (2013). All other data are from this study.

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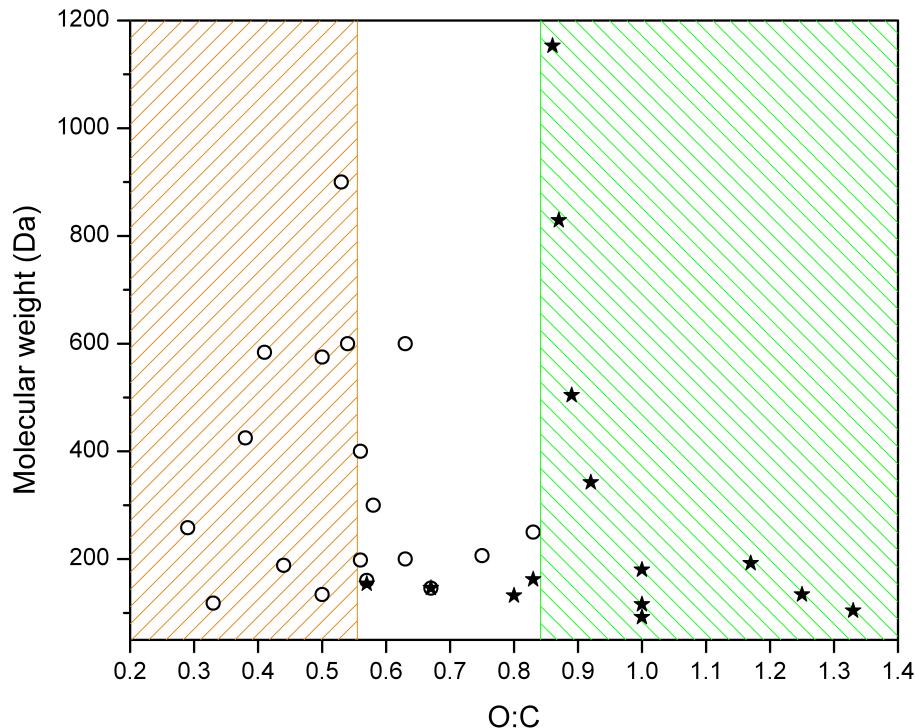
Figure 1. The effect of molecular weight and O : C of the organic species on the occurrence of liquid–liquid phase separation in mixed organic-ammonium sulfate particles (OIR = 2.0 ± 0.1). Data plotted are from the current study and You et al. (2013), and are summarized in Table 3. Open circles indicate liquid–liquid phase separation was observed, while stars indicate liquid–liquid phase separation was not observed. The orange hatched region corresponds to the molecular weight and O : C of the organic species when liquid–liquid phase separation was always observed, and the green hatched region corresponds to the molecular weight and O : C of the organic species when liquid–liquid phase separation was never observed.
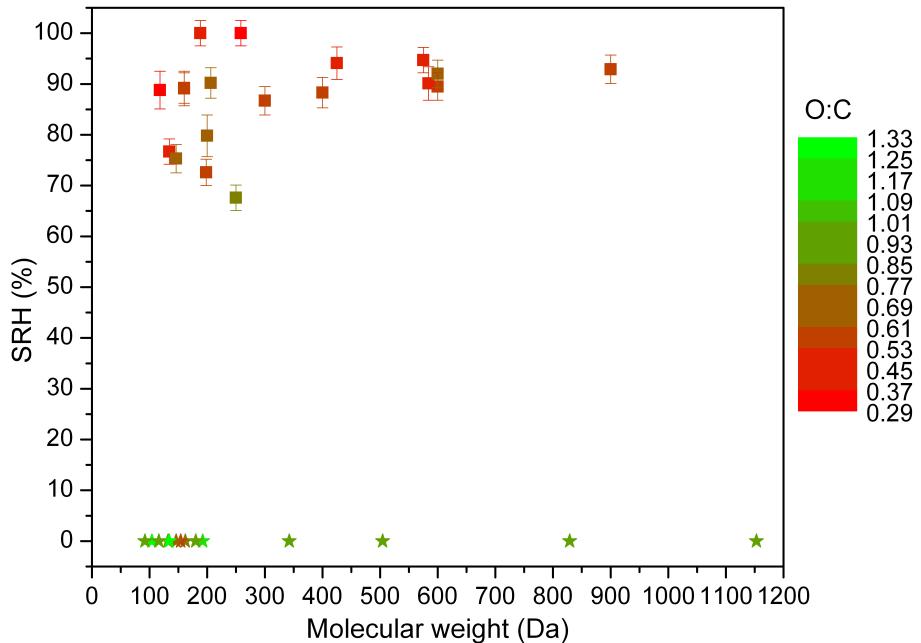
Figure 2. SRH as a function of molecular weight of organic species in the particles at 290 ± 1 K. The SRH results are from the current study and You et al. (2013) (see Table 3). The colors represent the O : C of different organic species. Squares represent SRH of particles in which liquid–liquid phase separation was observed. Bars for the squares include 2σ of multiple measurements and the uncertainty from the calibration. Stars indicate that liquid–liquid phase separation was not observed. OIR = 2.0 ± 0.1 in all the experiments.
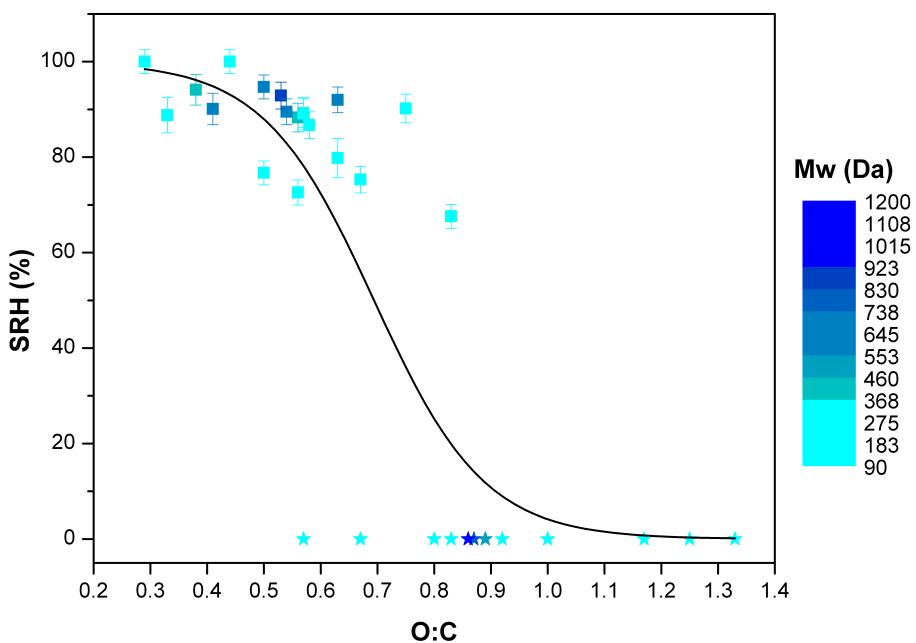
Figure 3. SRH as a function of O : C of the organic species at 290 ± 1 K. The SRH results are from the current study and You et al. (2013) (see Table 3). The colors represent the molecular weight of the different organic species. Squares represent the SRH of particles in which liquid–liquid phase separation was observed. Bars for the squares include 2σ of multiple measurements and the uncertainty from the calibration. Stars indicate liquid–liquid phase separation was not observed. OIR = 2.0 ± 0.1 in all the experiments. The black curve is a Sigmoidal–Boltzmann fit to the data. The equation of the fit is included in the Appendix A.
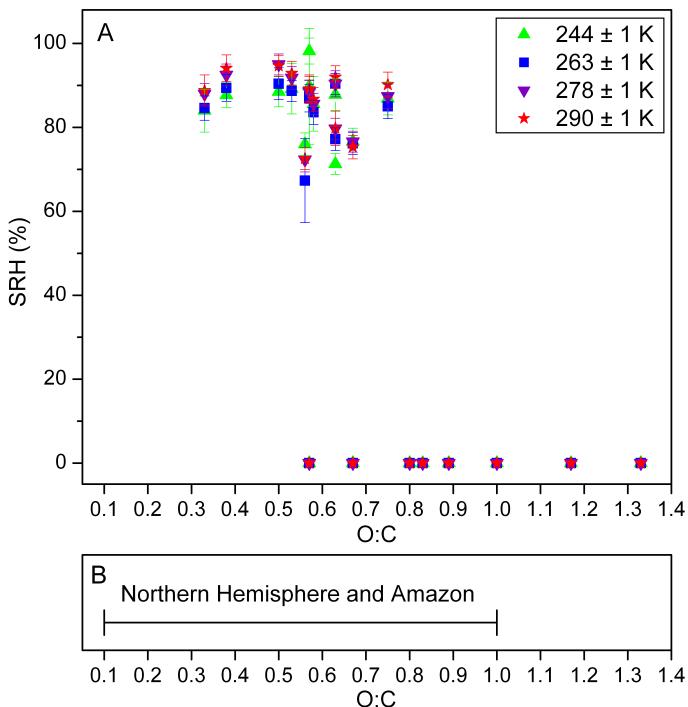
Figure 4. (A) SRH of mixed organic-ammonium sulfate particles as a function of O : C measured at four different temperatures. Different symbols represent the different temperatures. Bars for the data include 2σ of multiple measurements and the uncertainty from the calibration. Data at SRH = 0 % indicate liquid–liquid phase separation was not observed. Data plotted are summarized in Table 4. The OIR = 2.0 ± 0.1 in all the experiments. **(B)** Range of the average O : C of organic material in particles from measurements at many locations in the Northern Hemisphere and the Amazon.
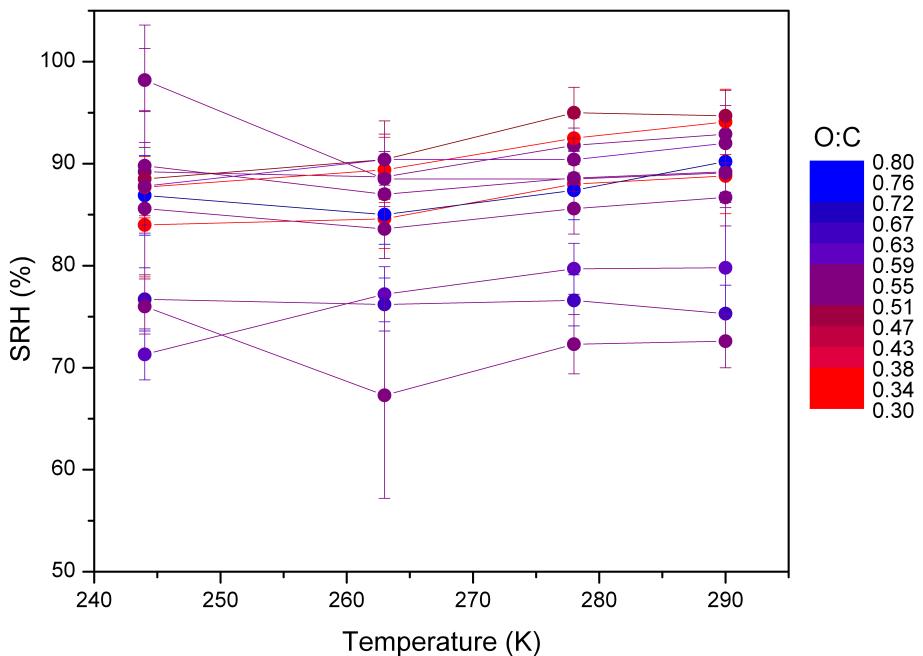
Figure 5. Summary of SRH as a function of temperature for mixed organic-ammonium sulfate particles that underwent liquid–liquid phase separation. Data plotted were taken from Table 4. Bars for the data include 2σ of multiple measurements and the uncertainty from the calibration. Colors represent the O : C values of different organic species in the particles. OIR = 2.0 ± 0.1 in all the experiments.

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