Thomas Koop,
Editor of Atmospheric Chemistry and Physics
Professor of Chemistry
Bielefeld University

Dear Thomas,

Listed below are our responses to the comments from reviewer 1 and 2. The reviewer's comments are in bold type and our responses are in normal text. We thank the referees for carefully reading our manuscript and for excellent comments.

Sincerely,
Allan Bertram
Professor of Chemistry
University of British Columbia

**Referee #1**

**There is one point that should be added to the discussion of LLPS of the investigated systems, namely, which of the investigated organic substances show LLPS when mixed with water even in the absence of an inorganic salt, i.e. for an OIR of infinity. This would help to better judge the salting-out effects of the investigated salts.**

This is a very good question, but one that is not easy to answer since we didn't measure the relative humidity at which LLPS occurs in the absence of salts. We do know from preparing solutions for particle production that the solubility of liquid diethyl sebacate and liquid suberic acid monomethyl ester is less than 1 % by weight in water. Hence, for these two systems, liquid-liquid phase separation is expected even without the salt at greater than or equal to roughly 99% RH. For the other systems, the solubility of organic species in water was greater than or equal to 1 % by weight. To address the referee's comments, we have added this discussion to the experimental section.

**As a minor technical point: the authors should try to improve the contrast of the optical images shown in Fig. 1.**

We have tried to improve the contrast of the optical images in Fig.1, but were unsuccessful. The movies included in the Supplemental Information show more clearly the phase separation, and we hope the referee is satisfied with the combination of optical images and the movies.

**Referee #2**

**Measurements are only done at an organic-to-inorganic mass ratio of 2:1. The authors should some broader justification for this and discuss how this compares with recent work by other authors. They should also provide some discussion of how the trends observed might be expected to depend on the organic-to-inorganic mass ratio.**

This is a good suggestion. To address the referee's comments, in the Experimental Section we have added a discussion on previous studies of SRH as a function of OIR. In addition, in Section 3.1 (second to last paragraph) we added discussion on how the trends observed might be expected to depend on the organic-inorganic mass ratio.

**- On line 19 of page 20086, the sentence "Shown in Fig. 4a are the SRH results of the organics (20 out of the 23 investigated) that followed the trend $(NH_4)_2SO_4$> $NH_4HSO_4$> NaCl> $NH_4NO_3$." This sentence needs clarifying (do they mean the SRHs are in this order?) and the authors should talk through more explicitly how this relates to the Hofmeister series. I felt this paragraph was perhaps not as clear/unambiguous as it could be.**

We thank the referee for the feedback. We re-wrote this paragraph to make it clearer (we hope!). The following is the re-written paragraph.

"SRH results shown in Figure 3 do not vary drastically with the type of inorganic salt. However, out of the 23 organics investigated, the SRH of 20 organics followed the SRH trend: $(NH_4)_2SO_4 \geq NH_4HSO_4 \geq NaCl \geq NH_4NO_3$ (see Figure 4a). In other words, the SRH-value measured with $(NH_4)_2SO_4$ was greater than or equal to the SRH-value measured with $NH_4HSO_4$, and so on. Early in the last century, Randall and Failey showed the following trends for the salting out efficiencies of ions relevant to our work: $Na^+ > NH_4^+ > H^+$ and $SO_4^{2-} > Cl^- > NO_3^-$ (Randall and Failey, 1927). In addition, the Hofmeister series, which consists of a ranking of cations and anions in terms of their ability to salt-out proteins follows the same trend (i.e. $Na^+ > NH_4^+$ and $SO_4^{2-} > Cl^- > NO_3^-$) (Kunz et al., 2004; Hofmeister, 1887; Hofmeister, 1888). These trends allow one to compare the salting out efficiency (or SRH) of two salts if they have a common cation or anion. Based on these early salting out studies, we would expect $(NH_4)_2SO_4$ to have a greater salting out efficiency (or higher SRH) compared to $NH_4HSO_4$ since the salting out efficiency follows the trend $NH_4^+ > H^+$. In addition, we would expect $(NH_4)_2SO_4$ to have a greater salting out efficiency (or higher SRH) than $NH_4NO_3$, since the salting out efficiency follows the trend $SO_4^{2-} > NO_3^-$. These expectations are consistent with the trends observed for 20 out of the 23 organics investigated (Figure 4a)."

**- Fig. 3: It is not why Sigmoidal-Boltzmann fits are chosen, particularly as these are intended only to guide the eye. Most of the systems in which phase separation occurs lie above the fit line with the systems for which phase separation was not observed receiving undue weighting in the fitting. Are these fits related in anyway (or how do they compare) with the expected behaviour from the previous parameterizations by the authors (recognising that these were only for ammonium sulphate)?**

The referee asks a very good question, and we thank him/her for bringing it up. Following Song et al [GRL, 2012], the Sigmoidal-Boltzmann function was chosen to avoid unphysical values at both low and high O:C values. Many of the systems in which phase separation was observed lie above the fit line since the curve was fit to both zero and non-zero SRH values. Alternatively we could have fit only the non-zero SRH values, but this would give extra weight to cases where phase separation was observed.

To address the referee's comments we have added the discussion above to the manuscript. We have also added a new table (Table 3) to the document where we report the parameters from the fit. We also point out that the fit for ammonium sulfate is qualitatively consistent with fits previously reported in the literature.