Reply to comments on "In−situ observation for RH−dependent mixing states of submicron particles containing organic surfactants and inorganic salts" by Xiong et al.

Reply to Anonymous Referee #1

1) The research provides direct observation and analysis of the dynamic mixing state and phase transitions of submicron particles containing inorganic and surface-active organic constituents in response to relative humidity (RH) cycling. The research also conducted on the interaction between phase transitions of aerosol particles and related hygroscopicity, and CCN activity which could make a significant contribution to the field. This approach allows for a comprehensive understanding of the dynamic evolution of inorganic-organic particles under fluctuating atmospheric conditions. This study fits within the scope of the journal Atmospheric Chemistry and Physics. However, after carefully examining and revising this article in light of the comments below, it could be considered for publishing in ACP.

Response:

We truly appreciate the constructive comments and suggestions raised by the reviewer. Those comments are valuable and very helpful for improving our paper. Below we provide a point-by-point response to individual comment. The responses are shown in brown and bold fonts, and the added/rewritten parts are presented in blue and bold fonts.

Major comments:

1) In experimental section, it's important to ensure the ESEM reproducibility. It's unclear from the conclusion if the experiments were repeated and the results were consistent. Further verification in this aspect might be needed.

Also, how did you determine the RH step from (a) to (c) of Figure 3? This progress occurred a really narrow range within 0.8% RH. Does it make sense to determine the decimal point of RH range because the authors used 2-3% RH/min condition for RH changes?

Furthermore, in Figure 4, if you change the brightness and contrast of the images, do you still obtain same results?

Response:

1) Each image in our study contained at least 5 particles (or droplets) to ensure the ESEM reproducibility and decrease the uncertainty. We have added the error bar in Figure 1 in the manuscript. In addition, we have reproduced some of the experiment for validation, e.g., in the RH decreasing period, and the results showed good reproducibility (Fig. R1 below).
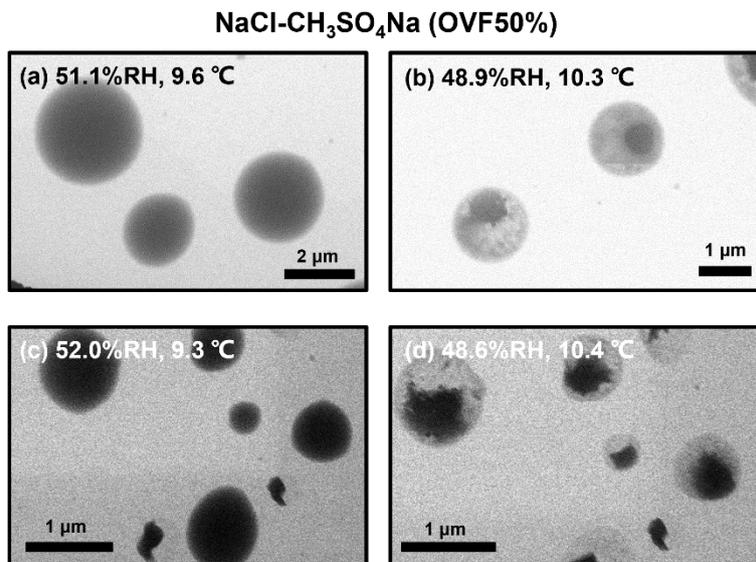
NaCl-CH₃SO₄Na (OVF50%)

(a) 51.1%RH, 9.6 ℃   2 μm

(b) 48.9%RH, 10.3 ℃   1 μm

(c) 52.0%RH, 9.3 ℃   1 μm

(d) 48.6%RH, 10.4 ℃   1 μm

Figure R1. ESEM images of solid phase state of NaCl−CH₃SO₄Na with 50% OVF in two repeated experiments (Experiment 1: a and b, Experiment 2: c and d). Phase separated RH showed good consistency.

2) By adjusting the scanning rates, RH varying rate and video recording parameters, the varying range of RH value between two consecutive pictures were mostly 0.2~0.4%RH (very narrow), in order to capture the possible quick transitions of mixing states. Meanwhile, we think it is necessary to determine the decimal point of RH range, since some transitions occurred in very short time (e.g., some Ostwald ripening process occurred within 10 seconds and 1% RH).

3) We have adjusted the contrast, brightness, as well as the magnification in Figure 4 to make it clear (Fig. R2a), and we could not see distinct separation edge (thus homogeneous) in Fig. 4a, but it was clearly identified in Fig. R2b (thus phase-separated).
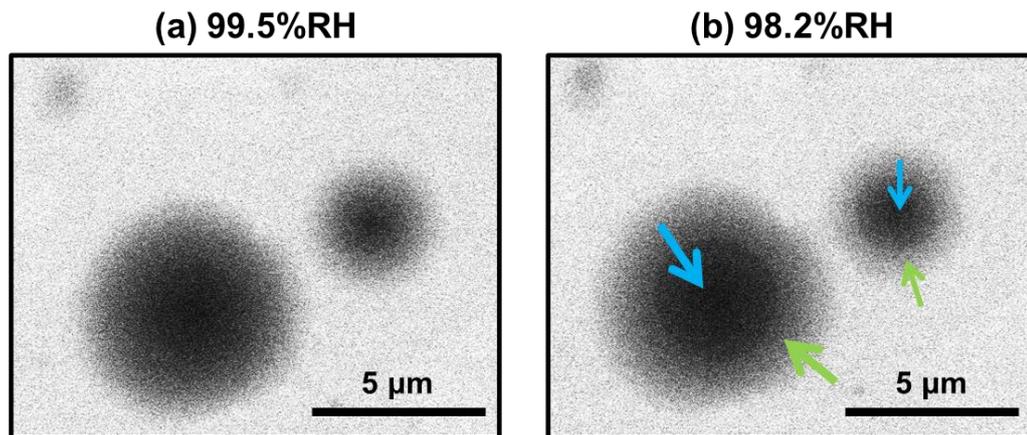
| (a) 99.5%RH | (b) 98.2%RH |

5 µm   5 µm

Figure R2. ESEM images of (a) homogeneous AS−C$_8$H$_{17}$SO$_4$Na particles (70% OVF) underwent (b) LLPS

Revision/addition:

1.1) SI: Fig. R1 was added in the SI (Fig. S1).

1.2) Line 129: "possible beam damage (O'brien et al., 2015). The varying range of RH value between two consecutive pictures were mostly 0.2~0.4%RH (very narrow), in order to capture the possible quick transitions of mixing states. Each image in our study contained at least 5 particles (or droplets) to ensure the ESEM reproducibility and decrease the uncertainty. In addition, we have repeated some of the experiment (e.g., in the RH decreasing period) for reproducibility validation, and the results showed good consistence (Fig. S1)"

2) Please provide the size range of the aerosol particles investigated during the experiments and discuss/compare the size effect with previous study of Freedman et al. in the result section. A figure would be helpful.

Response: Thank you for the suggestion. In our experiment, the initial selected inorganic AS–organic were with lateral dimensions of ~ 1 µm. Therefore, size effect

was not discussed in our study. In the recent years, size-dependent morphology was uncovered by Freedman (2020), who used cryo-electron microsocopy (Cryo-TEM) and observed larger particles were mostly phase–separated but smaller were homogeneous. Ohno et al. (2021) also found that LLPS occurred at lower RH in smaller droplet (70 – 190 nm) than in larger droplet (260 – 370 nm), which was explained by kinetic factors.

Revision/addition:

Line 152: "… form a new phase (Altaf and Freedman, 2017; Altaf et al., 2018; Freedman, 2020; Ott and Freedman, 2021). According to result in Freedman (2020), morphology of most systems were found size-dependent, where large particles were phase-separated and small particles were homogeneous. Furthermore, in their study all systems (e.g, AS–PA and AS–succinic acid systems) with dry diameters larger than 0.7 μm were observed phase-separated no matter the occurrence of size dependence (Altaf and Freedman, 2017). Freedman (2020) expected that phase-separation could be attributed by nucleation and growth, therefore larger particles tended to be phase-separated morphology. In another study, Ohno et al. (2021) also found that LLPS occurred at lower RH in smaller droplet (70 – 190 nm) than in larger droplet (260 – 370 nm)"

3) The authors used different temperature range of 0.1 – 25 ∘C in the experiments because temperature has negligible influence on the LLPS of AS-organic and NaCl-organic particles. Is this statement still valid for the submicron particles? Please discuss it.

Response: Thanks for the suggestion. To our knowledge, no research comprehensively discussed the effect of temperature on LLPS for submicron mixed

droplet. We think this is a very interesting question and further experiments are needed. But in our study, wet diameter of the investigated droplets at high RH was several micrometers before LLPS experiment, though their initial dry lateral dimensions were ranged in 0.7 ~ 1 μm.

According to You and Bertram (2015) and Roy et al. (2020), temperature did not much affect separation RH of micrometer AS–organic and NaCl–organic and mixed droplet (dozens of micrometers), respectively.

O'Brien et al. (2015) used ESEM to observe LLPS of AS–organic particles with smaller droplets (several micrometers) under 275−278 K, and found consistency of SRH with other studies in spite of different temperature and droplet size range, indicating weak dependences of size (from several micrometers to dozens of micrometers) and temperature on LLPS.

Revision/addition:

1) Line 118: "In each experiment, particles with lateral dimensions of ~ 1 μm were selected first (based on the deposition, volume-equivalent size was smaller than 1 μm). Then the RH raised from low…"

2) Line 121: "dissolution (O'Brien et al., 2015). With increased RH, most selected particles grew larger to several micrometers (lateral dimension) before subsequent LLPS experiment. Then, RH decreased to dry condition at similar change rate. Negligible influence on the LLPS of AS−organic (O'Brien et al., 2015; You and Bertram, 2015) and NaCl−organic systems (Roy et al., 2020) in micrometer scale (from several micrometers to dozens of micrometers)."

4) The authors showed that LLPS always occurs when the O:C ratio is 0.4 or below, and never when the O:C ratio is higher than ~0.57. This is inconsistent with previous studies (Bertram et al., 2011 ACP; You et al., 2013 ACP; Song et al., 2012 GRL). Thus, the authors should make a careful comparison with the literature and make a clear conclusion. I recommend that all experimental data points including the previous studies should be shown in Figure 5.

Response: Thank you for the suggestions. We have revised Fig. 5 (Fig. R3) and added all the available experimental data for NaCl–organic and AS–organic binary systems in the literature (Bertram et al., 2011; You et al., 2013; You and Bertram, 2015). In our study, LLPS always occurs when the O:C ratio is below 0.43 (yellow dashed line in Fig. R3) for NaCl–organic and AS–organic droplets. This value was close to the reported value in You et al. (2013) (~ 0.5). However, LLPS was reported never observed when O:C was above ~ 0.8 (grey dashed line in Fig. R3) (Bertram et al., 2011; Song et al., 2012; You et al., 2013),which was higher than that in our experiment (0.57). We ascribe this to the insufficient chemical systems (10 systems) in our experiment, which was notably smaller than those in previous studies (over 30 systems). As a result, the reported O:C ratio determining LLPS were not changed if our results were added in previous studies, e.g., You et al. (2013) and Song et al. (2012), as shown in the revised picture (Fig. R3).
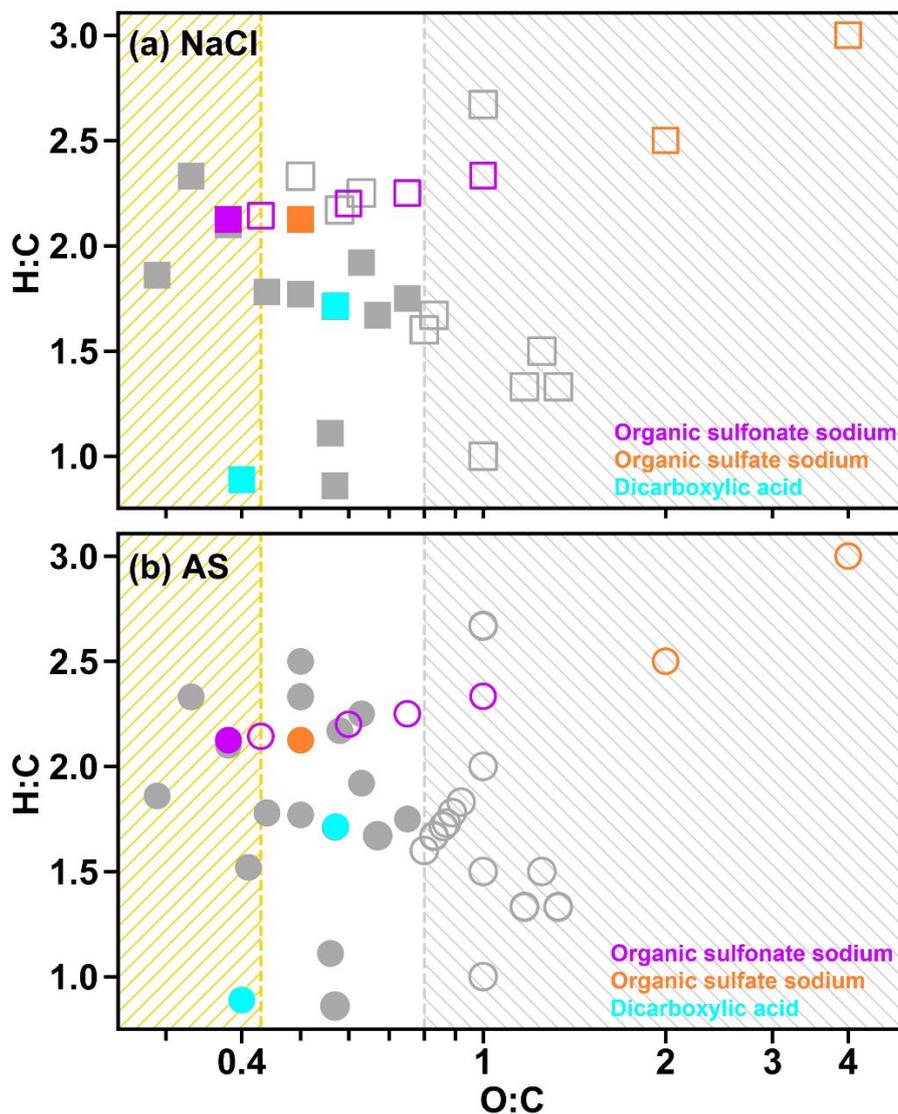
Figure R3. Van Krevelen Diagram for the mixed inorganic−surfactants particles in the current study (symbols in red, orange and cyan): (a) NaCl−surfactants and (b) AS−surfactants systems. Solid symbols indicate that LLPS was observed for particles with at least one OVF, while hollow symbols indicate that LLPS was not observed for particles with all tested OVFs. Symbols in grey were results obtained from Bertram et al. (2011), You et al. (2013) and You and Bertram (2015). Yellow-hatched region (O:C < 0.43) means that LLPS observed in all investigated systems, while grey-hatched region (O:C > 0.8) means no LLPS detected in any of the investigated systems.

Revision/addition:

1) Fig. R3 has been added in the main text (Fig. 5).

2) Line 228: "NaCl−organic surfactant systems. Fig. 5 illustrates the relationship between LLPS occurrence and molar ratios (O:C and H:C) of the surface−active organics, as well as reported results of other binary inorganic salt−organic systems in literature (Bertram et al., 2011; You et al., 2013; You and Bertram, 2015). In our study, LLPS always occurs when the O:C ratio is below 0.43 (yellow dashed line in Fig. 5) for NaCl–organic and AS–organic droplets. This value was close to the reported values in You et al. (2013) (~ 0.5). However, in their results, LLPS was never observed when O:C was above ~ 0.8 (grey dashed line in Fig. 5) (Bertram et al., 2011; Song et al., 2012; You et al., 2013),which was higher than that in our experiment (0.57). We ascribe this to the insufficient chemical systems in our experiment (10 systems), which was notable smaller than in previous studies (over 30). As a result, the reported O:C ratio determining LLPS were not changed if our results were added in previous studies, e.g., You et al. (2013) and Song et al. (2012)"

5) The study also observed that inorganic salt-surfactant systems without LLPS undergo solid phase separation after efflorescence, demonstrating distinct separated phases (pg. 14). This is interesting and pretty new. Thus, I suggest to show the result in the main text and expand the discussion. Figure S2 should be clearer.

Response: Thank you for the suggestion, we have expanded the discussion and gave clearer images (Fig. R4).
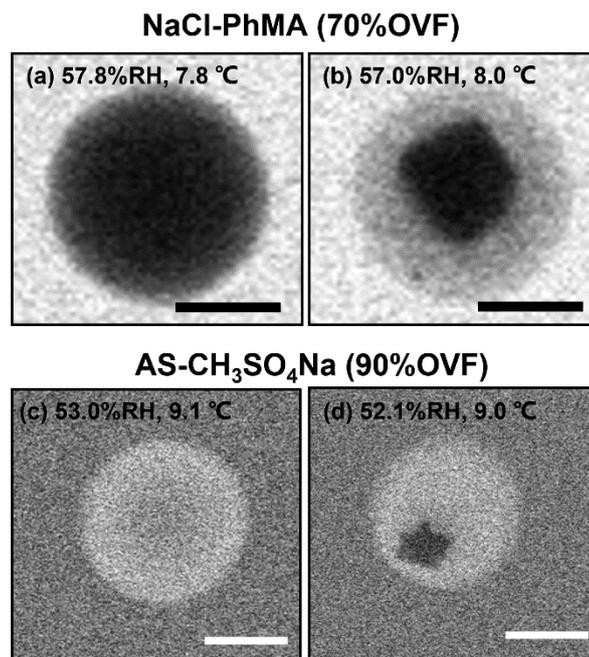
NaCl-PhMA (70%OVF)

(a) 57.8%RH, 7.8 ℃ (b) 57.0%RH, 8.0 ℃

AS-CH₃SO₄Na (90%OVF)

(c) 53.0%RH, 9.1 ℃ (d) 52.1%RH, 9.0 ℃

Figure R4. ESEM images of efflorescence of (a−b) NaCl−PhMA and (c−d) AS−CH$_3$SO$_4$Na systems.

Revision/addition:

1) Fig. R4 has been added in the main text (Fig. 7)

2) Line 265: "For mixed systems, we also found they were directly separated with distinct core−shell phases from homogeneous morphology at low RH. However, this phase transition was different from LLPS, since the inner phase was with irregular shape (LLPS occurred with rounded inner liquid phase), which was attributed to the efflorescence progress of inorganic salt (Fig. 7). We termed it solid phase separation. The efflorescence RH (ERH) of inner inorganic salt, therefore, was the solid phase separation RH.

In Fig. 8a, ERH of NaCl−organic surfactant particles with 50%, 70% and 90% OVF were ranged in 47.0−61.8%, which was higher than the measured ERH (48.3 ± 0.4%) and reported ERH range of pure NaCl (41−51%) (Peng et al., 2022). This could

be explained by the interaction between organic and inorganic matters. For example, Ghorai et al. (2014) found an acid displacement reaction in NaCl−glutaric acid systems, which was driven by gaseous HCl liberation and causing chloride depletion. Such interactions of chloride depletion may facilitate efflorescence transitions, resulting in efflorescence at ∼ 68% RH and ∼ 60% RH, respectively, for internally mixed NaCl−glutaric acid particles with molar ratios of 1:3 and 1:1. Higher ERH could also be attributed to heterogeneous nucleation initiated by chemical purities (Choi and Chan, 2002). Choi and Chan (2002) observed 54.4% ERH for a 1:1mixed NaCl−glutaric acid, and they explained that insoluble additives crystallized and formed nuclei for the heterogeneous efflorescence of inorganic salts, leading to their higher ERH values.

As for AS−organic surfactant systems (Fig. 8b), efflorescence was observed for 27 among 30 aerosol samples. We did not observe distinct occurrence of efflorescence for the rest 3 samples, and 2 samples among 3 were with 90% OVF, which could be explained by the possible loss of AS when it was persistently exposed to electronic beam (Posfai et al., 2013; O'Brien et al., 2015), especially for particles in which inorganic fractions were small (i.e., high OVF). ERH values of AS−organic surfactant particles with 50%, 70%, and 90% OVF ranged in 31.2−46.6%, showing a close result to the reported ERH of pure AS (30−48%) (Peng et al., 2022), but higher than the measured ERH (30.7 ± 0.9%). The potential cause may be the heterogeneous crystallization of AS on organic salts (Wang et al., 2019; Yang et al., 2019; Ma et al., 2021). For example, Wang et al. (2019) investigated the efflorescence of AS in AS−sodium oxalate and found SRH values were 48.9% and 55.3% with organic-inorganic mole ratios of 1:1 and 3:1, respectively, which were higher than that of pure

AS (47.5%). Likely, Yang et al. (2019) also observed that the initial ERH of AS rose to 47.7% and 62% for inorganic mole ratios 1:3 and 1:1 AS−sodium pyruvate mixtures, respectively.”

Minor comments:

1) The manuscript has several instances where grammar and language use could be improved for clarity. This includes sentence structure, punctuation, and the use of certain phrases.

Response: We have checked and revised the manuscript.

2) **Lines 132 – 133: The sentence needs to be clearer. Please expand the discussion.**

Response: We have expanded the discussion in the paper.

Revision/addition:

Line 152: “… form a new phase (Altaf and Freedman, 2017; Altaf et al., 2018; Freedman, 2020; Ott and Freedman, 2021). According to result in Freedman (2020), morphology of most systems were found size-dependent, where large particles were phase-separated and small particles were homogeneous. Furthermore, in their study all systems (e.g, AS–PA and AS–succinic acid systems) with dry diameters larger than 0.7 μm were observed phase-separated no matter the occurrence of size dependence (Altaf and Freedman, 2017). Freedman (2020) expected that phase-separation could be attributed by nucleation and growth, therefore larger particles**

tended to be phase-separated morphology, as was observed in the present study. In another study, Ohno et al. (2021) also found that LLPS occurred at lower RH in smaller droplet (70 – 190 nm) than in larger droplet (260 – 370 nm)"

3) Ensure that the authors used consistent terminology throughout the paper. For example, the terms "inorganic salts-surfactant systems" and "inorganic−surfactant particles" appeared to refer to the same thing, and using them interchangeably may cause confusion.

Response: We have revised the inconsistent terminology to inorganic salts−organic surfactant through the paper.

4) Implication: Strengthen the conclusion by making a clear connection between the results and the larger implications for the field of atmospheric chemistry and physics. Providing a more thorough discussion on how your findings could help reduce the uncertainty of model estimation on the global radiative effect could be beneficial.

Response: We revised our implication as suggested.

Revision/addition:

Implication: "Dicarboxylic acids, organosulfates, and organosulfonates are important surface−active organic constituents in secondary organic aerosol. Few studies comprehensively studied their mixing state upon fluctuating RH cycling, which is a simulate of real atmospheric condition. In this work, we concluded that mixing state affected interactions of inorganic salt with water. Since common assumptions in chemical transport models (including ISORROPIA-II (Fountoukis and Nenes, 2007), EQSAM (Metzger et al., 2002a; Metzger et al., 2002b), and

MOSAIC (Zaveri et al., 2008)) are that water uptake is determined separately by the inorganic compounds and organics (i.e., the effect of mixing state was ignored) (Myhre et al., 2007; Nandy et al., 2021), thereby our results implied further effect of mixing states on estimations of aerosol hygroscopicity (e.g., growth factor), optical properties, and radiative forcing.

During dehydration, we investigated phase−separated before and after efflorescence for inorganic salts−organic surfactant particles. Compared with homogeneous particles, phase−separated particles could decrease trace gas uptake (You et al., 2012), resulting in reduction of the formation of secondary organic aerosols (SOAs) (Zhang et al., 2018). In addition, organic phase was enriched in "outer shell", which can potentially alter aerosol water activity and lower aerosol surface tension, hence affecting aerosol−cloud interactions because water uptake of organic matter in current models (e.g. MPMPO (Griffin et al., 2003) and SOA treat-ment in CMAQ v5.2 (Pye et al., 2017)) is estimated by highly parameterized relationships assuming ideal solutions, e.g., using the kappa hygroscopicity parameter with water surface tension (Petters and Kreidenweis, 2007; Nandy et al., 2021).

Our results provide comprehensive information of mixing states between inorganic salts and organic surfactant in nanoscale perspective, which could help the establish of incorporation atmospheric modeling, to improve predictions on indirect effects of aerosol−climate interactions. We should note that in the atmosphere most particles are smaller (e.g., 0.1 to 0.3 μm) than sample particles and the chemical characteristics of ambient aerosol are not as simple as binary chemical systems in the

current study. Therefore. the water kinetic inhibition should be further investigated for smaller particles containing more complex systems in the future."

Reference

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