

Referee #2:

condense the introduction to focus more on the study's specific objectives and significance. Reduce background information that isn't directly relevant to your study's aims.

Reply: Thank you for your suggestion, we rewritten the introduction to make it more concise and clearer.

Revision: line 27-82: please see the revised introduction in the manuscript.

Explain the rationale for choosing specific organic salts.

Reply: Thank you for the valuable suggestion. We have added justification for the selected organic-to-inorganic ratios in our introduction and materials and methods sections. In atmospheric aerosols, the organic-to-inorganic ratio and chemical structure can vary by region. Generally, the proportion of organic components is often comparable to that of inorganic components. Therefore, in our study, we used molar ratios of 2:1, 1:1, and 1:2 to represent regions with excess organic matter, a balance between organic and inorganic matter, and excess inorganic matter, respectively.

Revision: line 76-78: “The mole ratio of 1:2, 1:1 and 2:1 was applied to simulate the surplus organic salts, equal, and excessive inorganic slats for mixed aerosols depending on location, aerosol source and season.”

line 90-92: “In the atmosphere, the organic to inorganic ratio varies regionally, but mostly remains at the same order of magnitude. Thus, the selected ratios herein can effectively represent different regions.”

Provide more details about the relative humidity control and measurement, such as the precision and accuracy of RH measurements.

Reply: Thank for your advice. In revised manuscript, the precision and accuracy of RH measurements has been added.

Revision: line 103: “...with a precision of 0.5% and an accuracy of 1%...”

It is suggested to add some summary tables comparing the phase transition behaviors of different organic salts and ammonium salts.

Reply: Thank you for the excellent suggestion, and we have added the summary table to the revised paper.

Revision:

Table 2 The phase transition behaviors of different organic salts and ammonium salts

Mixed aerosols	SP mixed with						(NH ₄) ₂ SO ₄ mixed with	
	NH ₄ Cl			NH ₄ NO ₃	(NH ₄) ₂ SO ₄	SC	ST	
		2:1	1:1	1:2	(1:1)	(2:1)	3:2	1:1
ERH% on	SP	61.7	23.7	×	NaNO ₃ : 35.7–12.7	Na ₂ SO ₄ : 65.7–60.1	Na ₂ SO ₄ : 56.9	×

dehydration	NH ₄ Cl	×	42.5	46.6	SP: ×	SP: ×	SC: ×	
DRH% on	SP	84.3	×	×	NaNO ₃ : 66.1–82.3	Na ₂ SO ₄ : 83.8–90.3	Na ₂ SO ₄ : 70.5–87.2	Na ₂ SO ₄ : 74.8
hydration	NH ₄ Cl	×	71.8	77.7	SP: ×	SP: ×	SC: ×	
ERH% on								
hydration	×	×	×	×	×	×	×	Na ₂ SO ₄ : 43.6

Note : × means no phase transition was observed

Single value represents the onset ERH or DRH

What is the potential impact of temperature on phase transitions? What is the relationship between liquid-liquid phase separation and phase transition behaviors in this study? It is suggested to discuss them in the implication.

Reply: Thanks for reviewer’s suggestion. The potential impact of temperature on phase transitions and the relationship between liquid-liquid phase separation and phase transition behaviors are important for climate effect. However, in our work, there is no LLPS was observed, and phase transition dependence of temperature wasn’t performed. In revised paper, the effects of temperature and LLPS has been discussed based on other work.

Revision: line 452-463: “Except for RH, aerosol phase also depends on other atmospheric conditions, particularly temperature. Solid ammonium sulfate particles with organic coatings was observed under high relative humidity (67 to 98%) at low temperature (−2 to +4 °C), underscoring the key role of temperature in phase transitions (Kirpes et al., 2022). They proposed that solid (NH₄)₂SO₄ formed through contact efflorescence and that temperature induced liquid-liquid phase separation (LLPS). Previous studies have shown that temperature often has a greater impact on LLPS than on efflorescence (Schill et al., 2013). LLPS is crucial for the solidification of ammonium sulfate (Roy et al., 2020) and soot redistribution (Yuan et al., 2023) in aqueous organic-inorganic aerosol droplets. In our work, we observed various aerosol phase states, including crystalline, aqueous, and gel-like states, during dehydration and hydration cycles, and we examined the interplay among aqueous-phase reactions, aerosol hygroscopicity, and phase behavior in detail. However, LLPS was not observed prior to crystallization, either through optical microscopy or ATR-FTIR. Future studies are needed to explore the interplay between various phase transitions, including LLPS, efflorescence, and viscous state formation, considering not only RH but also temperature.”

Ensure all references are up-to-date and relevant. Check if any recent studies on similar topics have been published and could be included. E.g. recent field observation studies have highlighted the significant role of organic acids in modifying the phase behavior of ammonium sulfate aerosols, further complicating the prediction of aerosol phase states (Li et al., EST, 2021, 55, 16339; Yuan et al., 2023, ACP, 23, 9385; Kirpes et al., PNAS, 2022, 119, 14).

Reply: Thanks for the reviewer’s suggestion. The relevant description and references have been added.

Revision: line 41-45: “Recent field observations highlighted the significant role of organic acids in modifying the phase behavior of ammonium sulfate aerosols, further complicating the prediction of aerosol phase states (Li et al., 2021; Zhang et al., 2022; Kirpes et al., 2022) For atmospheric aerosols undergoing liquid–liquid phase separation (LLPS) with an organic shell and an inorganic core, black carbon redistribution can be induced, which leads to a more compact morphology and a reduction in the absorption enhancement effect by 28 %–34% (Zhang et al. 2022).

Revision: line 539-541: “Li, W., Teng, X., Chen, X., Liu, L., Xu, L., Zhang, J., Wang, Y., Zhang, Y. and Shi Z.: Organic Coating Reduces Hygroscopic Growth of Phase-Separated Aerosol Particles, *Environ. Sci. Tech.* 55, 16339–16346, <https://doi.org/10.1021/acs.est.1c05901>, 2021.”

Line 648-655: “Yuan, Q., Wang, Y., Chen, Y., Yue, S., Zhang, J., Zhang, Y., Xu, L., Hu, W., Liu, D., Fu, P., Gao, H., Li W.: Measurement report: New insights into the mixing structures of black carbon on the eastern Tibetan Plateau – soot redistribution and fractal dimension enhancement by liquid–liquid phase separation, *Atmos. Chem. Phys.* 23, 9385–9399, <https://doi.org/10.5194/acp-23-9385-2023>, 2023.
Zhang, J., Wang, Y., Teng, X., Liu, L., Xu, Y., Ren, L., Shi, Z., Zhang, Y., Jiang, J., Liu, D., Hu, M., Shao, L., Chen, J., Martin, S. T., Zhang, X., and Li, W.: Liquid–liquid phase separation reduces radiative absorption by aged black carbon aerosols, *Comm. Earth Environ.*, 3, 128, <https://doi.org/10.1038/s43247-022-00462-1>, 2022.

Line 529-532: “Kirpes, R. M., Lei, Z., Fraund, M., Gunsch, M. J., May, N. W., Barrett, T. E., Moffett, C. E., Schauer, A. J., Alexander, B., Upchurch, L. M., China, S., Quinn, P. K., Moffet, R. C., Laskin, A., Sheesley, R. J., Pratt, K. A., Ault, A. P.: Solid organic-coated ammonium sulfate particles at high relative humidity in the summertime Arctic atmosphere, *PNAS*, 119, e2104496119, <https://doi.org/10.1073/pnas.2104496119>, 2022.

Abstract: The abstract is informative but could be improved by including a brief mention of the methodology (ATR-FTIR spectroscopy) and the key findings related to the RH ranges.

Reply: Thank you. The advice is very helpful. In the abstract, the methodology and key RH have been added.

Line 10-11: “We investigated carboxylate/ammonium salt mixtures using attenuated total reflection fourier transformed infrared spectroscopy (ATR-FTIR).”

Line 15-18: “For SP/ammonium aerosols, NaNO₃ and Na₂SO₄ crystallized from 35.7% to 12.7%, and from 65.7% to 60.1% RH, respectively, lower than pure inorganics (62.5±9–32% RH for NaNO₃ and 82±7–68±5% RH for Na₂SO₄). Upon hydration, the crystalline Na₂SO₄ and NaNO₃ deliquesced at 88.8%–95.2% and 76.5±2–81.9%, higher than those of pure Na₂SO₄ (74±4%–98% RH) and NaNO₃ (65–77.1±3% RH).”

Line 14: A detailed introduction to “aqueous replacement reactions” for better understanding the mechanisms.

Reply: Thank you very much for your suggestion. We have added a definition and examples of “aqueous replacement reactions” in the main text to help readers better understand our work.

Revision: line 53-56: “In aqueous aerosols, the ions between two ionic compounds can exchange to form new compounds through aqueous replacement reactions. For instance, when oxalic acid mixes with nitrates, the H^+ ions can combine with NO_3^- ions to form HNO_3 , which off-gases, leading to the formation of metal oxalate salts in the aerosol (Ma et al. 2019).”

Line 16: 35.7%~12.7%, 64% and 65.5%~60.1% RH?

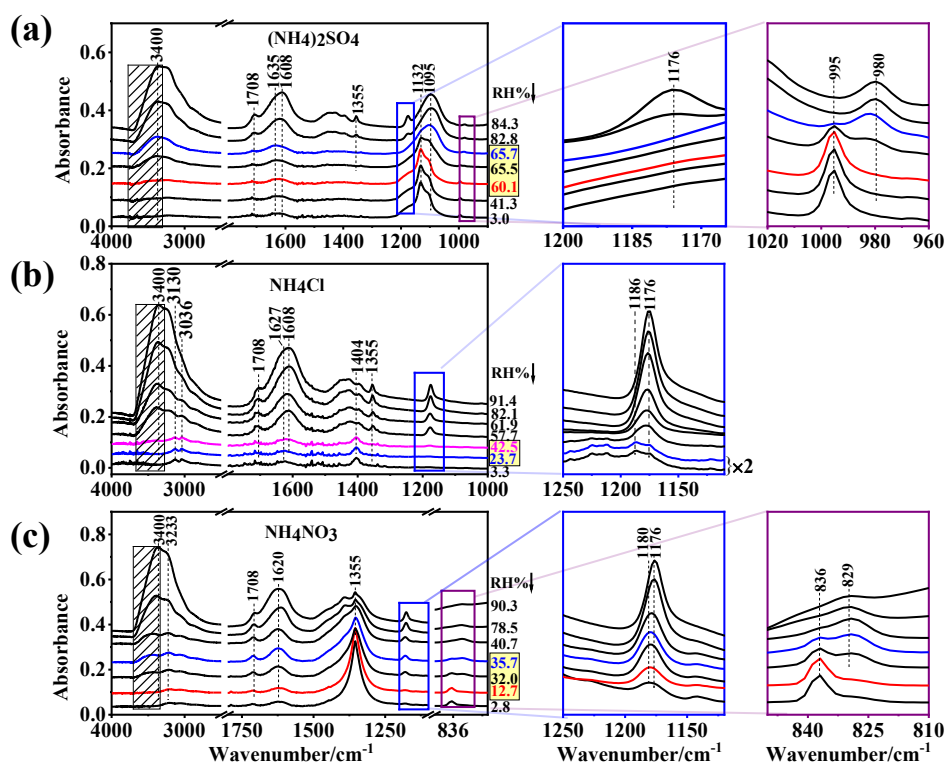
Reply: Thank you very much for your suggestion. We have revised the description in the text to make the sentence clearer.

Revision: line 15-18: “For SP/ammonium aerosols, $NaNO_3$ and Na_2SO_4 crystallized from 35.7% to 12.7%, and from 65.7% to 60.1% RH, respectively, lower than pure inorganics (62.5 ± 9 – 32% RH for $NaNO_3$ and 82 ± 7 – $68 \pm 5\%$ RH for Na_2SO_4). Upon hydration, the crystalline Na_2SO_4 and $NaNO_3$ deliquesced at 88.8% – 95.2% and 76.5 ± 2 – 81.9% , higher than those of pure Na_2SO_4 ($74 \pm 4\%$ – 98% RH) and $NaNO_3$ (65 – $77.1 \pm 3\%$ RH).”

Figure 1: Enhance the figure with annotations indicating key RH ranges where phase changes occur.

Reply: Thank you very much for your suggestion. We have marked the range of phase transition in the Figure 2 (original Figure 1) and added an explanation in the caption.

Revision: Figure 2 (original Figure 1)

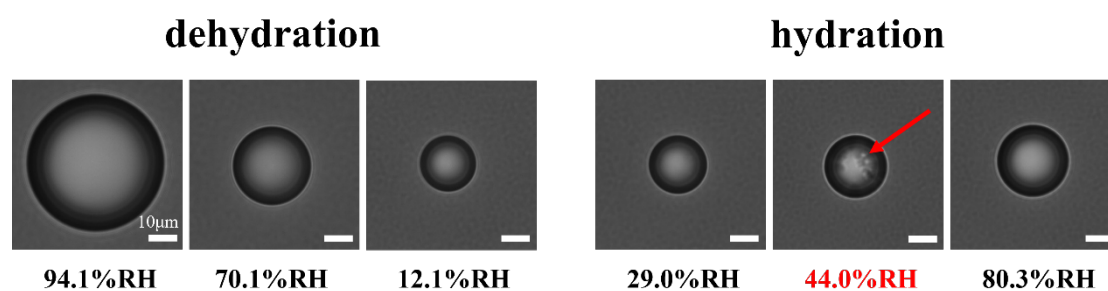


Line 206-208: “Figure 2: ...The yellow frame indicates the RH range when phase transition occurs (ERH of Na₂SO₄: 67.5-60.1%; onsets of ERH for SP and NH₄Cl: 23.7% and 42,5%; ERH of NaNO₃: 35.7-12.7%).”

Figure 7: Include a scale bar in each image to indicate the size of the observed particles. Highlight key features in the images, such as the appearance of solid entities or changes in particle morphology, with arrows or labels.

Reply: Thank you very much for your suggestion. We have added a scale bar to each image and marked the RH and location where phase transitions occur.

Revision: Figure 8 (original Figure 7)



Line 362-364: “Figure 8: ...The red arrow pointed the location where the phase transition occurs. The RH indicated in red marks the occurrence of phase transitions. Scale bar represents 10 micrometers.”