

We want to thank the reviewers for their further comments. Below, we list each reviewer's comment (regular font), followed by our response (indented, **bold font**), followed by corresponding changes in the revised manuscript (**indented, blue font**). RL represents the line number in the revised version.

### **Anonymous Referee #1**

This manuscript by Cheng et al. has been improved and been revised accordingly based on the reviewers' comments. I support the publication of this manuscript on this journal. I have two technical comments.

1. Abbreviations are defined several times in the manuscript (e.g., SEM, STXM-NEXAFS, and FRXPART). Please check them.
2. Figure S7. The last panel should be “l” instead of “i”

**We appreciate the reviewer for providing us with these comments. We have revised the manuscript and SI based on these comments.**

### **Anonymous Referee #2**

In general, the author's answer and revised sentences are much clearer than before. However, I still need some clarification regarding the authors' responses to resolve some of the possible confusion. I have several more comments that the authors should address before publication of this manuscript. Please see below.

**We appreciate the constructive feedback from the reviewer. We attempted to clarify further and noted the study's limitations. Below are our responses to each comment:**

#### **Comments:**

1. I confused on the definition of the phase state of the field samples. It must be contain various phases inside the particles as shown in the figures. Do you mean the particle is overall solid, semisolid or liquid? A clear definition of the concepts for the real aerosol particles in the introduction should be added.

**We thank the reviewer for pointing this out and agree that it should be clearly defined in the manuscript. As the reviewer noted, we study the overall phase state of individual (internally mixed) FT particles. Based on our knowledge, only a limited number of studies investigated ambient particle phase state (e.g., Bateman et al., 2016; Liu et al., 2017, 2021; Pajunoja et al., 2016; Slade et al., 2019), and all of them investigated the phase state of entire particles. We agree that the phase state inside the particles (e.g., inorganic inclusions) can be different, causing phase separation. We added the following sentence in our introduction to specify that we are looking at the overall phase state of particles:**

RL117-119: “In this study, we present an overview of the overall phase state of individual FT atmospheric aerosol particles (internally mixed) collected at OMP over three different years, which are July 2014 (Pico 2014), June and July 2015 (Pico 2015), and 2017 (Pico 2017).”

2. The authors mentioned that the samples were analyzed less than one year after collection. One year maybe enough time to change the chemical compositions and related phase states of the collected samples. Can you provide some previous work to solve this issue?

**In the previous response, we noted the potential modification of particles due to storage (comment 4 in the previous response). It would be fantastic if we could analyze our samples right after collecting them. However, that is often not feasible to accomplish for offline measurements of samples collected at the remote site since these sites are usually difficult to access, sample delivery time can be long, and there are always limited instruments and labor time to analyze them, which makes these type of research typically publish their results few years after the sample collection (see (Allen et al., 2021; China et al., 2017; Cozic et al., 2008; Dzepina et al., 2015; Lata et al., 2021; Moffet et al., 2010; Schum et al., 2018; Zhang et al., 2013). As the reviewer noted, we cannot exclude with certainty that some transformations might have taken place between the sampling and the analysis times. This is a limitation of any offline analysis of field-collected samples. We have tried to minimize oxidation and photolysis by wrapping the sample with Al foil and kept in zip lock bags immediately after collection, which is a typical method for storing field-collected aerosol samples (e.g., Adachi and Buseck, 2011; Kirillova et al., 2016; Marsh et al., 2017; Stockwell et al., 2016). Although we cannot avoid potential particle modification due to storage, we believe our results still provide essential scientific findings for aerosol research in the phase state of the FT particles. Future studies should focus on sample storage strategy for offline measurements. To make these clearer, we revised the previous revised sentence as below:**

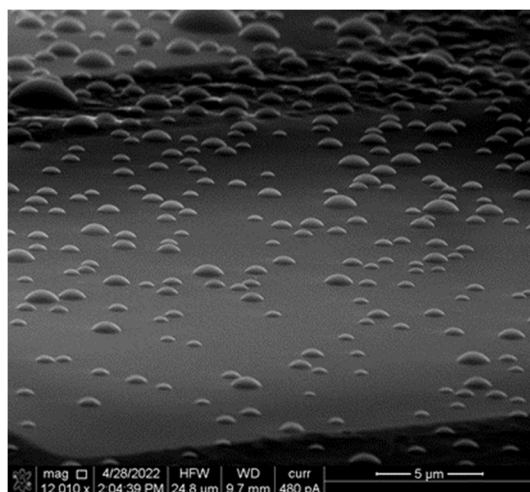
RL140-147: “Samples were placed in dedicated storage boxes wrapped in Al foil and kept in zip lock bags immediately after collection to avoid exposure to light and outside air, which is a typical sample storage strategy for field-collected aerosol samples (e.g., Adachi and Buseck, 2011; Kirillova et al., 2016; Marsh et al., 2017; Stockwell et al., 2016). The samples were then stored at ambient conditions to reduce the chances of particle modifications and oxidation that might have partially intercurrent. However, we cannot exclude with certainty that some of such transformations might have taken place between the sampling and the analysis times. This is a limitation of any offline analysis of field-collected samples. We underline that the site is quite difficult to access; therefore, samples were delivered and analyzed as soon as it was feasible (less than one year after collection).”

3. Still I am confusing how you described/concluded the phases for the filter samples collected from FT even the experimental condition was different. During the experiment, the experimental conditions of RH and temperature were significantly different compared to the environment conditions of FT (higher RH and lower temperature). This should be stated clearer.

**Thanks for this comment. As we explained in our response to the reviewer’s comment 1 in the first reviewer comments document, we agree there might be some potential modification of the particle phase inside the ESEM chamber due to vacuum and dry conditions, which is a common caveat of this method. Based on that, we also revised the manuscript in our revision. We agree that the temperature and RH were different at the site compared to those during our offline experiments, in which the temperature is usually about 10 K higher, and RH is usually about 6-**

67% lower. The higher temperature can reduce viscosity during experiments, while lower RH can increase viscosity as a competition effect. However, we mentioned this caveat in the previous response, *“our inference of the particle’s phase state at the time of collection is based on the shape the particle acquires at impaction on the substrate, which unlikely would change significantly within the ESEM chamber due to adhesion forces between the particle and the substrate.”* We confirmed this by collecting sucrose and ammonium sulfate mixture (50/50 wt%) at 83% RH. Based to (Tong et al., 2022), sucrose and ammonium sulfate mixture should be liquid at 83% RH and semisolid at 0% RH. As shown in Fig. R1, these sucrose and ammonium sulfate mixture particles have an average aspect ratio of about  $3.15 \pm 0.27$ , indicating they are still in the liquid state under vacuum conditions, which validates that the shape of organic particles after impact on the substrate would unlikely change significantly within the ESEM chamber. However, to make the potential change in particle phase state clearer, we revised the following sentence:

RL169-175: “Ambient particle samples were analyzed with ESEM at 293 K, under vacuum conditions ( $\sim 2 \times 10^{-6}$  Torr) and therefore at RH values near zero, which might lead to losses of volatile and semivolatile materials. Moreover, the temperature and RH inside the ESEM chamber differed from those at the OMP during sample collections (about 10 K higher and 6-67% lower, respectively, see Fig. S2). RH and T affect the phase state of airborne particles; however, our inference of the particle’s phase state at the time of collection is based on the shape the particle acquires at impaction on the substrate, which unlikely would change significantly within the ESEM chamber due to adhesion forces between the particle and the substrate. These limitations need to be considered when interpreting our results.”



**Figure R1. ESEM tiled image of 50/50 wt% sucrose and ammonium sulfate mixture particles collected on Carbon Type-B TEM grids at 83%RH.**

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