Responses to the reviewer

Thanks for the reviewer's helpful comments and suggestions again. We made every effort to respond to the reviewer's questions point to point, and improved our manuscript and supplementary information (SI) accordingly. For clarity, the reviewer's comments are shown in *black italic font*. The response is shown in black normal font. The revised text in the manuscript and/or the SI is shown in **bold blue font**.

General comments: Thank you for your work on these revisions. You have addressed many of my concerns, but there are a few notes I would like to follow up on. I gently recommend that more effort be placed in making changes in the text of the manuscript or in the supplemental to address comments or questions that a reviewer raises.

Specific comments:

1. I requested information on freezing for these samples and this was provided in the response, but no changes were made in the document. Please add some notes either to the manuscript or the supplement to provide context for the possible effect of freezing on the samples.

[Response]: Thanks for your comment! We searched for more research on freezing effects on field samples during storage. The relevant descriptions were added as follows.

[Revise]: Line 130-136 in the manuscript: "Previous results indicate that the chemical composition of organic aerosols (especially secondary organic aerosols, SOA) and the mass concentrations of black carbon (BC) both remained stable for several weeks under low-temperature storage conditions (–20°C and –80°C for organic aerosols, and 2°C, 4°C, and 5°C for BC); while significant changes occurred over time when samples were stored at room temperature even just for a few days (Mori et al., 2016; Mori et al., 2019; Resch et al., 2023; Ueda et al., 2025; Wendl et al., 2014)."

2. I requested information about the possible beam damage in the samples. I am happy to see that minimal signal changes were observed, please add this information either to the manuscript or the supplement as it will be helpful for future studies in this area.

[Response]: Thanks for your suggestion! We added information about beam damage in the manuscript and the SI. Please see as follows.

[Revise]: Line 177-179 in the manuscript: "Optical density detected over the same spot at different photon energies at the carbon and nitrogen K-edges was displayed in Fig. S1, and less beam damage during the experiment was confirmed."

Line 107-112 in the SI:

“

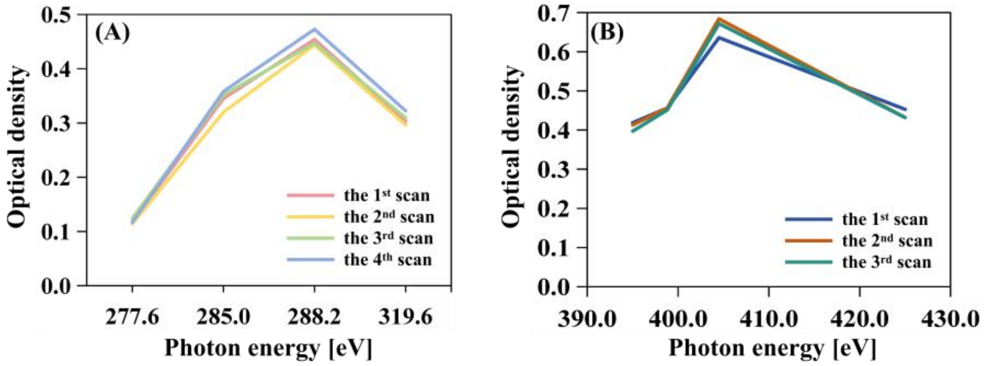

Figure S 1: Spectra collected over the same spot at different photon energies at the (A) carbon and (B) nitrogen K-edges.

As displayed in Fig. S1, optical density at different photon energies of several scans resembles each other at both the carbon and nitrogen K-edges, which indicates no obvious beam damage occurred.”

3. In a comment I requested information on the absorption enhancements and soot location. The response to reviewer comments makes some good statements, but the underlying question is not addressed, and no changes were made to the manuscript. Please consider revising this portion of the manuscript. To clarify my concerns, I will pose the following problem:

if a particle has the black carbon on the edge of the particle, but the particle lands such that the soot is centrally located in the impacted/collected particle, you cannot tell that this is a particle with soot on the edge vs. soot that is at the center. This problem adds some uncertainty in all estimates of morphology from STXM data, and it should be clearly stated so that this and future data sets are not over-interpreted in terms of absorption enhancements based on organic/BC morphology.

[Response]: Thanks for your valuable comments! We added some descriptions in the manuscript. Please see as follows.

[Revise]: Line 294-298 in the manuscript: “While, it should be noted that particle deformation may occur during the particle collection process due to the high particle impaction velocity of the impactor (O’Brien et al., 2014). Therefore, the distribution of chemical components within individual particles displayed in the images may differ from that of aerosol particles in ambient atmospheres.”

4. I asked for a comparison of the STXM O/C with the AMS O/C. In the response, several reasons for why these might be different are provided and it is stated that they are not comparable. I understand the limitations, but please provide the O/C values from the AMS data sets for these time periods either in the main text or the supplemental. You are welcome to clarify the limitations of the comparisons, but *future studies may want to make those comparisons, and the data should be provided here to enable that. For this comparison, I would also argue that your third statement in the response does not make sense compared to what is stated in the manuscript. In the manuscript, the O/C from STXM for the organic dominated particles is used to argue that LLPS is possible. Why would the AMS data not be comparable to this if you are focusing on organic dominated particles for the STXM O/C calculations?*

[Response]: Thanks for your comments! We added data set in the SI and the relevant descriptions in the manuscript. Please see as follows.

[Revise]: Line 313-319 in the manuscript: "For comparison, the value of the O:C ratio by AMS during the individual particle collection period is also calculated (0.60 ± 0.01), and the data set was displayed in the SI. The difference between the O:C ratio results by STXM and LToF-AMS may be due to the reasons as follows: (1) STXM measures individual particles, while AMS targets bulk aerosols; (2) Particles collected onto the last stage using a four-stage cascade impactor with the 50% cut-point aerodynamic diameter of 250-nm were used for STXM analysis, while AMS measured the non-refractory chemical composition of submicron particles."

Line 92-94 in the SI:

"Table S 2 The data set of the oxygen-to-carbon (O:C) ratio based on AMS data using "Improved-Ambient" Methods (Canagaratna et al., 2015) during the individual particle collection period."

Date and Time	O:C
2019/10/1 17:05	0.61
2019/10/1 17:06	0.61
2019/10/1 17:07	0.60
2019/10/1 17:08	0.59
2019/10/1 17:09	0.58

5. I asked about the note that is made in the manuscript that the peak at 288.6 eV is always found at the outer shell of the particles. This statement is now on line 375 of the document with track changes. The response to reviewer comments note that this can be observed in the particles, but no corrections were made to the text in the manuscript. Please correct the text in this sentence.

[Response]: Thanks for your comment! We added the relevant descriptions in the manuscript. Please see as follows.

[Revise]: Line 391-397 in the manuscript: "The peak appearing at 288.6 eV represents the characteristic transition of carboxylic carbonyl functional groups ($\mathrm{C\ 1s} \rightarrow \pi^*_{\mathrm{R(C^*=O)OH}}$), which refers to organic matter and is generally dominant in the outer shell of particles (Moffet et al., 2016; Prather et al., 2013). One should note that there could be extra components in both core and shell in a

phase-separated particle with an inorganic-rich core and an organic-rich shell, for example, organic in core or inorganic in shell (Gaikwad et al., 2022). Therefore, the 288.6-eV peak may also be observed in a particle core with relatively low peak intensity.”