Response to referee #2

We are grateful for referee #2's comments. Those comments are all valuable and helpful for improving our paper and English writing. We answered the comments carefully and have made corrections in the submitted manuscript. The corrections and the responses are as following:

In the revised manuscript, the red color was marked as the revised places.

General comments

The authors provide a measurement report about how black carbon (BC) is distributed within aged mixed organic/inorganic aerosol particles collected on the eastern Tibetan Plateau mountain site in July 2016. The used ground based collection on TEM grids and TEM and AFM to obtain size, mixing state and morphology. Basically, they confirm their previous result, Zhang et al. (2022), that liquid-liquid phase separation redistributes BC to the organic coatings for a wide range of relative humidities. In addition to their previous work, they deduced the fractal dimension (Df) of the BC and see a ranking with decreasing Df from externally mixed BC to sulfate coated BC to organic coated BC.

As the morphology of BC in internally mixed aged aerosol is clearly important for analyzing its radiative impact, I feel this measurement report should be published as it reconfirms previous work measured at different sites. However, I ask the authors to take the following comments/suggestions into account for a revised manuscript.

Major comments

Comment #1: The reader would benefit, if the connection to their previous work (Zhang et al., 2022) would be made stronger throughout the whole manuscript. For example, it remains unclear to me whether there is a significant difference in the ratio between organic coating thickness and BC size as a threshold above which the BC redistributes to the organic coating between the present study and that of Zhang et al.

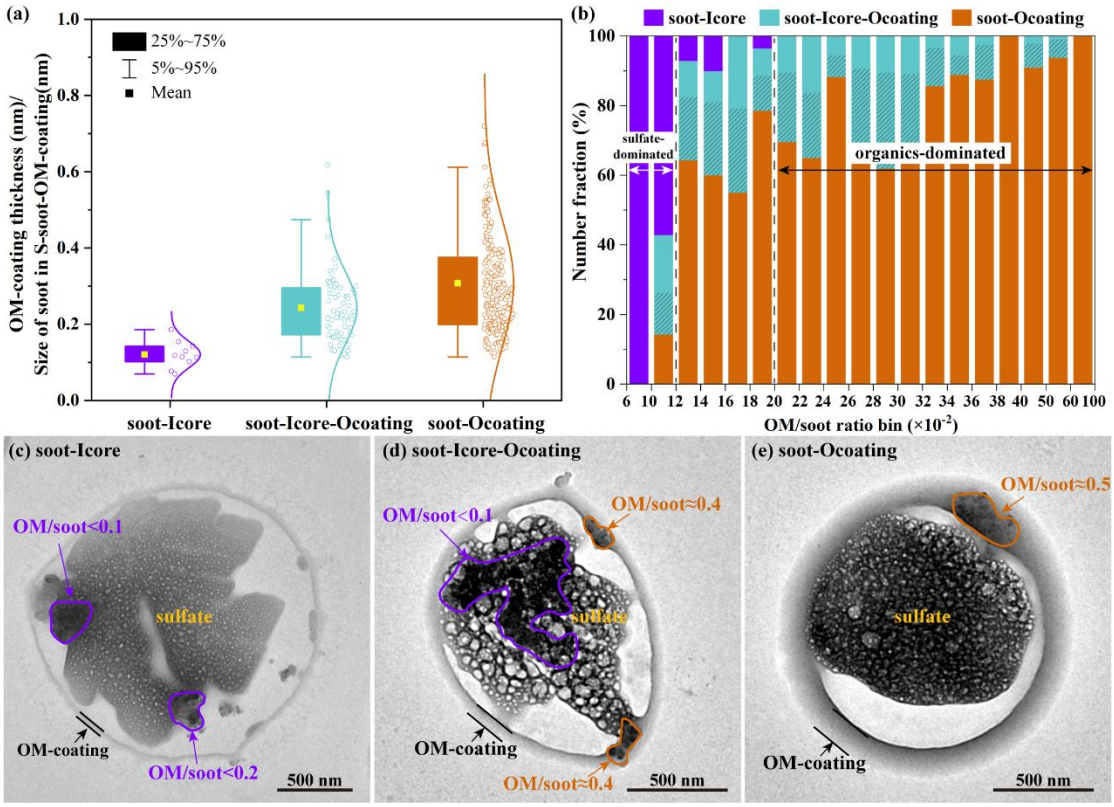
(2022). There, the authors came up with a ratio of 0.24, now they state this ratio is 0.2. My feeling is there is no significant difference between these thresholds (as they are somewhat arbitrary), but the authors need to discuss this.

Reply: We appreciate the reviewer's comments. We have improved the data analysis of the OM/soot ratio and compared our results with that reported by Zhang et al. (2022). We also modified the Figure 6b. The detailed revision was shown as below.

In context, line 215-222: "To further explore this trend accurately, we divided the ratios into 15 bins between 0.1 and 0.4, which collectively accounted for over 80% of the total OM/soot ratios (Fig. 6b). We observed that when the OM/soot ratio was less than 0.1, all of S-soot-OM-coating particles were soot-Icore particles (Fig. 6b). As the ratio increased beyond 0.2, none of soot-Icore particles was observed, and nearly 60% of the total S-soot-OM-coating particles were identified as soot-Ocoating particles (Fig. 6b). When the ratio exceeded 0.32, more than 80% of the S-soot-OM-coating particles were identified as soot-Ocoating particles. Nearly all soot particles occurred in the OM-coating when the ratio of OM/soot was larger than 0.6 (Fig. 6b). These results suggest that soot tended to distribute into the organic coating instead of the inorganic core following an increasing ratio of OM/soot (Fig. 6b)."

In context, line 223-233: "Zhang et al. (2022) reported that the dominant type of the laboratory-generated soot-containing particles shifts from soot-Icore particles to soot-Ocoating particles when the OM/soot ratio increased from 0.04 to 0.34. Their field-observed soot-containing particles were almost soot-Ocoating particles when the OM/soot ratio exceeded 0.24. Our study at 0.32 of the OM/soot was close to their laboratory results, suggesting the reliability of our research outcomes. However, our field observation was slightly higher than the previous reported 0.24, and this discrepancy could be attributed to the considerable presence of soot-Icore-Ocoating particles in our study, which was rarely observed in Zhang et al. (2022). Over 50% of soot particles were distributed within OM-coating in all the soot-Icore-Ocoating particles (as shown in the oblique bar in Fig. 6b). Consequently, combining the soot-Ocoating (brown bar in Fig. 6b) and soot in OM-coating of soot-Icore-Ocoating

particles (oblique bar in Fig. 6b), we can infer that when the OM/soot ratio exceeds 0.2, most of soot (>80%) tend to distribute in organic phase in the atmosphere of Mt. Emei during sampling period (as indicated by the organics-dominated region in Fig. 6b)."



"Figure 6. Variations of OM/soot ratios with different distribution positions of soot in S-soot-OM-coating particles. (a) The different ratios of OM/soot in the S-soot-OM-coating particles. (b) Number fractions of soot-Icore, soot-Icore-Ocoating, and soot-Ocoating particles in all S-soot-OM-coating particles in different ratios of OM/soot. Oblique bar represents that soot were distributed within OM-coating in all of the soot-Icore-Ocoating particles. (c) A typical TEM image of a soot-Icore particle with two soot particles in a sulfate core (OM/soot<0.2). (d) A typical TEM image of a soot-Icore-Ocoating particle with a soot particle in a sulfate core (OM/soot<0.1) and two soot particles in an OM-coating (OM/soot≈0.4). (e) A typical TEM image of a soot-Ocoating particle with a soot particle in the OM-coating (OM/soot≈0.5)."

Comment #2: My other concern is the significance of the differences they observe in the fractal dimension between the different morphologies. I can see that the difference between externally mixed BC and internally mixed BC in Df is significant. I doubt that

the small differences the authors see between sulfate coated BC and organic coated BC are significant. The authors need to explain in detail their uncertainty analysis for the values they provide in Table 1. While they state "The standard error for Df was calculated from the uncertainty in the mean-square fit considering the uncertainty in N and dp.", the details remain unclear to the reader. In addition, they do not comment on that Df is higher at elevated RH for sulfate coated BC compared to organic coated BC while it is the opposite at lower RH.

Reply: We appreciate the reviewer's comments.

1. The differences of D_f between sulfate-coated BC and organic-coated BC were really small. We have deleted the sentence and revised the content as follow:

In context, line 251-253: "The conclusion derived from all these morphological parameters was consistent with the compacted soot particles enclosed by sulfate and organics. Indeed, several field and laboratory studies found that soot embedded with sulfate and organics could increase its compactness after coating (Wang et al., 2021;Xue et al., 2009;Saathoff et al., 2003)."

2. As we used the ensemble method, the uncertainty of the D_f of black carbon mainly comes from the uncertainties in the total number (N) and the average diameter (d_p) of soot monomers. N can be calculated using the equation 1 as below:

$$N = k_a\left(\frac{A_a}{A_p}\right)^{\alpha} \qquad (1)$$

A_a and A_p can be obtained directly by analyzing TEM images. α and k_a in this equation are determined by the overlap parameter (δ). Therefore, the uncertainty of N is mainly from δ of soot monomers. δ is calculated by equation 2 as below:

$$\delta = \frac{2a}{l} \qquad (2)$$

a is the monomer radius and l is the monomer spacing. Note that the monomers overlap in the three-dimensional structure which can cause darkened color from gray to dark on the projection of soot particles in TEM images. We cannot figure out the lattice spacing between every pair of monomers in individual soot aggregate. We also can't obtain the diameter of every soot monomer through our manual efforts and

usually use the average diameter of several soot monomers for calculation. The quantification of this uncertainty is represented by the standard error of the slope given by the mean-square fit. Several previous studies have pointed out the uncertainty of D_f (China et al., 2013;Pang et al., 2022) and used the same quantification method (China et al., 2013;Yuan et al., 2019). The uncertainties of convexity (CV), roundness (RN), and aspect ratio (AR) were calculated by standard error of all individual soot particles. We added the content as follow:

In context, line 131-135: "In this study, we employed the ensemble method to obtain a mean D_f of soot particles with different mixing states (Wang et al., 2017). The uncertainty of the D_f was attributed to the uncertainties in the numbers and diameters of soot monomers, which were mainly manually determined (Pang et al., 2022). The quantification of this uncertainty was expressed by the standard error of the slope given by the mean-square fit (China et al., 2013;Yuan et al., 2019)."

Line 140-141: "These morphological parameters can be calculated using the methods in China et al. (2013) and Yuan et al. (2019). The uncertainties of *CV*, *RN*, and *AR* were expressed by standard errors of these values in all individual soot particles."

3. Given the really small variations of D_f values of organic-coated soot between different RH and the complicated mechanism of soot aging process under high RH, we deleted the discussion about the comparisons of D_f values of soot between different RHs.

Specific comments

Comment #1: Line 149: I suggest citing here some of the relevant lab studies, in particular also the cryo TEM work of the Freedman group as well. In particular, she showed that there is a size dependence on LLPS (e.g. Altlaf et al., 2016).

Reply: We appreciate the reviewer's comments. We cited the lab study in the revised manuscript.

In context, line 166-168: “Cryo-TEM measurements further confirmed that the LLPS formed the distinct core-shell structures with sulfate core and OM-coating in ambient aerosols (Altaf et al., 2016;Li et al., 2021).”

Line 178-179: “Freedman (2017) showed that the LLPS process can influence surface and interfacial tensions among different phases in individual particles.”

Line 193-195: “The result is similar to the previous reports that particle size plays a crucial role to influence the LLPS of individual particles (Altaf et al., 2016;Li et al., 2021).”

Reference:

Altaf, M. B., Zuend, A., and Freedman, M. A.: Role of nucleation mechanism on the size dependent morphology of organic aerosol, Chemical Communications, 52, 9220-9223, 2016.

China, S., Mazzoleni, C., Gorkowski, K., Aiken, A. C., and Dubey, M. K.: Morphology and mixing state of individual freshly emitted wildfire carbonaceous particles, Nature Communications, 4, 2122, 2013.

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Li, W., Liu, L., Zhang, J., Xu, L., Wang, Y., Sun, Y., and Shi, Z.: Microscopic Evidence for Phase Separation of Organic Species and Inorganic Salts in Fine Ambient Aerosol Particles, Environ Sci Technol, 55, 2234-2242, 2021.

Pang, Y., Wang, Y., Wang, Z., Zhang, Y., Liu, L., Kong, S., Liu, F., Shi, Z., and Li, W.: Quantifying the Fractal Dimension and Morphology of Individual Atmospheric Soot Aggregates, Journal of Geophysical Research: Atmospheres, 127, e2021JD036055, 2022.

Saathoff, H., Naumann, K. H., Schnaiter, M., Schock, W., Mohler, O., Schurath, U., Weingartner, E., Gysel, M., and Baltensperger, U.: Coating of soot and $(NH_4)_2SO_4$ particles by ozonolysis products of alpha-pinene, Journal of Aerosol Science, 34, 1297-1321, 2003.

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Wang, Y. Y., Liu, F. S., He, C. L., Bi, L., Cheng, T. H., Wang, Z. L., Zhang, H., Zhang, X. Y., Shi, Z. B., and Li, W. J.: Fractal dimensions and mixing structures of soot particles during atmospheric processing, Environmental Science & Technology Letters, 4, 487-493, 2017.

Xue, H. X., Khalizov, A. F., Wang, L., Zheng, J., and Zhang, R. Y.: Effects of coating of dicarboxylic acids on the mass-mobility relationship of soot particles, Environmental Science & Technology, 43, 2787-2792, 2009.

Yuan, Q., Xu, J., Wang, Y., Zhang, X., Pang, Y., Liu, L., Bi, L., Kang, S., and Li, W.: Mixing State and Fractal Dimension of Soot Particles at a Remote Site in the Southeastern Tibetan Plateau, Environmental Science & Technology, 53, 8227-8234, 2019.

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, Communications Earth & Environment, 3, 128, 2022.