Response to referee #1

We are grateful for referee #1'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

This manuscript by Yuan et al. reports detailed mixing states and shapes of soot particles mixed with organic matter and sulfate collected at the eastern Tibetan Plateau. They discussed liquid-liquid phase separation and redistribution of soot particles within particles. Mixing states and shapes of soot particles influence the optical properties of internally mixed particles and their radiation. Thus the results are important to the understanding of climate influence. My major concern is that it is probable that the mixing states and shapes that they measured could be influenced by both atmospheric processes and impaction on the substrates when collected. Therefore, I suggest more careful discussion of the influence of the changes on the filter should be provided. I also recommend having some discussion based on chemical and physical processes about liquid-liquid phase separation and soot redistribution.

Major comments

Comment #1: The TEM images show mixing states after the particle collection on the substrates. Thus, changes in shapes and mixing states should be carefully discussed if they had changed in the air or on the substrate. Discussing the two-dimensional mixing states of particles on the substrate is acceptable. However, when discussing their mixing states in the atmosphere, the coating materials should cover the entire surface. When discussing the implication for the climate, the discussion should

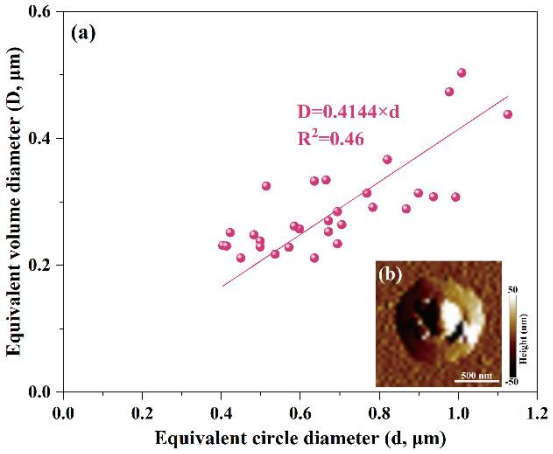
depend on their three-dimensional shape in the atmosphere. At least, the TEM images show that organic coatings cover the perimeter of the spread sulfate, which cannot be a realistic 3D shape in the atmosphere. The coating thickness in the TEM image may be different from that in the atmosphere as they spread over the substrate. Sulfates are also shrinking by losing water on the substrate and have some space with organic coatings (Fig. 3b). There are also some traces around the particles (Fig. 3b). As a result, the shapes and mixing states could have been different from their original or partially the same as the These points need to be clearly discussed in the paper.

Reply: We appreciate the reviewer's comments.

TEM is one of the best technique to determine real mixing state of single particles and has been widely used in large amounts of laboratory studies and field observations (Li et al., 2016). The 3D morphology of the aerosol particles in this study was investigated by using an atomic force microscope (AFM). We provide a typical AFM image of an OM-coating particle in Figure S2. As shown in Fig. S2a, there is a linear relationship between the ECD and EVD of particles and the relation between d and D is D=0.4144×d. The sizes and coating thicknesses of individual particles are calculated based on the EVD and the detailed calculation method can be found in Zhang et al. (2022).

In context, line 122-124: "As shown in Fig. S2a, there is a linear relationship between the ECD and EVD of particles with D=0.4144×d. The sizes and coating thicknesses of individual particles are calculated based on the EVD and the detailed calculation method can be found in Zhang et al. (2022)."

Line 205-206: "Similar to the method employed by Zhang et al. (2022), we calculated the OM-coating thicknesses and the entire particle sizes based on TEM and AFM (Fig. 5)."



"Figure S2. (a) The correlation of equivalent circle diameter (ECD, d) and the equivalent volume diameter (EVD, D) obtained by AFM. (b) An AFM image of an OM-coating particle."

Comment #2: Although liquid-liquid phase separation and soot redistribution is interesting results, I suggest having more discussion based on chemical and physical processes. Why do they have such a process? What are the physical and chemical processes (e.g., the hygroscopicity of soot, surface tension, viscosity, etc.)? When did these processes occur? Some more discussion in Fig. 9 will be helpful in interpreting the results.

Reply: We appreciate the reviewer's comments. We conducted a solid reference survey and added the detailed explanation of liquid-liquid phase separation and soot redistribution phenomenon.

In context, line 165-171: "Optical and fluorescence microscopy analyses revealed that the LLPS could occur in individual ambient aerosols, with the presence of two separate phases: inner ammonium sulfate and outer secondary organic material (You et al., 2012). 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). Furthermore, the LLPS particles have been widely observed in Arctic air (Kirpes et al., 2022;Yu et al., 2019), rural and mountain areas (Zhang et al., 2022), and forest air (Li et al., 2020). Therefore, we concluded that S-soot-OM-coating particles as shown in Figure 2 can be considered as soot particles mixed with the LLPS particles."

In context, line 174-181: “It is well known that soot particles typically contains hydrocarbons, polycyclic aromatic hydrocarbons, and partially oxidized organics generated during combustion (Long et al., 2013;Wang, 2011). Moreover, TEM observations revealed a thin amorphous organic coating on carbon nanospheres of fresh soot particles (Buseck et al., 2014). The combustion processes always produce extremely thin organic layers on each soot monomer (Leskinen et al., 2023;Chen et al., 2016). Freedman (2017) showed that the LLPS process can influence surface and interfacial tensions among different phases in individual particles. Therefore, some studies used the intermolecular forces and interactions between similar chemical bonds to explain the phenomenon of soot redistribution in individual particles (Brunamonti et al., 2015;Zhang et al., 2022).”

Specific comments

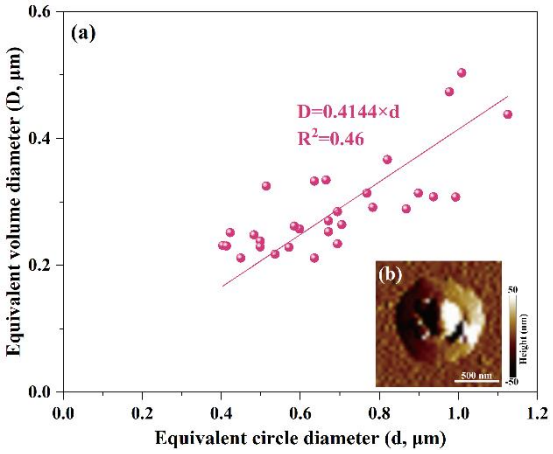
Comment #1: Line 52 “This uncertainty in BC radiative forcing is largely” Are you discussing an uncertainty or “a large difference in several model studies” here? Is this uncertainty caused by only “the lensing effects of the coating”? I assume that different emission inventories are also the cause of large uncertainty.

Reply: We appreciate the reviewer’s comments. We revised this sentence as follow:

In context, line 53-54: “This differences and uncertainties in BC radiative forcing are largely caused by the variability of the actual BC-mixing state in most models and in the ambient air (Hu et al., 2021;Zhai et al., 2022;Fierce et al., 2020;Riemer et al., 2019;Adachi and Buseck, 2013).”

Comment #2: Line 121 “The equivalent circle diameter (ECD, d) and the equivalent volume diameter (EVD, D) were calculated according to the bearing area and bearing volume.” Is soot particle included in the plot? If so, EVD cannot be related to ECD because of its fractal shape.

Reply: We appreciate the reviewer’s comments. Soot particles were not included in the plot. We provide a typical AFM image of an OM-coating particle in Figure S2.



"Figure S2. (a) The correlation of equivalent circle diameter (ECD, d) and the equivalent volume diameter (EVD, D) obtained by AFM. (b) An AFM image of an OM-coating particle."

Comment #3: Line 151 "S4b). The sulfate core and OM-coating in secondary particles were identified as LLPS" Why? Please explain this reason.

Reply: We appreciate the reviewer's comments. We added the result of EDS spectrum and provided some references to confirm the LLPS particles as follow:

In context, line 158-164: "Finally, the third type of soot-containing particle is core-shell particles with soot in either the core or the coating (Fig. 2e, 2g). The EDS spectrum shows that the coating is most likely to be organic matter (OM) with significantly higher carbon and lower sulfur content compared to the core (Fig. 2f, 2h). This similar core-shell particles have been identified as "OM-coating structure" (Li et al., 2016), which were reported in previous field observations and laboratory studies (Adachi et al., 2022;Li et al., 2021;Freedman, 2020;Li et al., 2020;Shi et al., 2008). Consequently, we called soot internally mixed within sulfate core or OM-coating as "S-soot-OM-coating" (Fig. 2e, 2g)."

In context, line 165-171: "Optical and fluorescence microscopy analyses revealed that the LLPS could occur in individual ambient aerosols, with the presence of two separate phases: inner ammonium sulfate and outer secondary organic material (You et al., 2012). 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). Furthermore, the LLPS particles have been widely

observed in Arctic air (Kirpes et al., 2022;Yu et al., 2019), rural and mountain areas (Zhang et al., 2022), and forest air (Li et al., 2020). Therefore, we concluded that S-soot-OM-coating particles as shown in Figure 2 can be considered as soot particles mixed with the LLPS particles."

Comment #4: Line 157 "A laboratory study and field observations have shown that LLPS can drive soot in core–shell particles from inside inorganic aerosols to outer organic aerosols, which is called the soot redistribution phenomenon" Why does it happen? Please explain this soot redistribution phenomenon in more detail.

Reply: We appreciate the reviewer's comments. We provided the detailed explanation of soot redistribution phenomenon as follow:

In context, line 174-181: "It is well known that soot particles typically contains hydrocarbons, polycyclic aromatic hydrocarbons, and partially oxidized organics generated during combustion (Long et al., 2013;Wang, 2011). Moreover, TEM observations revealed a thin amorphous organic coating on carbon nanospheres of fresh soot particles (Buseck et al., 2014). The combustion processes always produce extremely thin organic layers on each soot monomer (Leskinen et al., 2023;Chen et al., 2016). Freedman (2017) showed that the LLPS process can influence surface and interfacial tensions among different phases in individual particles. Therefore, some studies used the intermolecular forces and interactions between similar chemical bonds to explain the phenomenon of soot redistribution in individual particles (Brunamonti et al., 2015;Zhang et al., 2022)."

Comment #5: Line 182 "Therefore, we can conclude that soot redistribution in secondary particles is a common phenomenon on Mt. Emei." The results were obtained only from limited samples and periods. Therefore, it is difficult to have a general conclusion.

Reply: We appreciate the reviewer's comments. We revised this sentence as follow:

In context, line 204: “Therefore, we conclude that soot redistribution in secondary particles is a common occurrence on Mt. Emei during the sampling period.”

Comment #6: Line 188 “Figure 5b shows that the entire particle size and coating thickness exhibited good correlations, suggesting that larger particles along with thicker OM-coatings can drive soot particles into the organics from the sulfate core due to LLPS.” I do not think the correlation suggests the latter sentence. There is a large gap between observation and the discussion.

Reply: We appreciate the reviewer’s comments and we revised the content as follow:

In context, line 206-208: “Figure 5b shows that there is a certain positive correlation between the OM-coating thicknesses and the entire particle sizes, implying that larger S-soot-OM-coating particles tend to contain thicker OM-coating.”

Comment #7: Line 202 “The results suggest that the coarser particles following the thicker Omcoatings captured more soot particles in the OM-coating during the redistribution process” Why can it be concluded that it happened “during the redistribution process”? Can they simply be coagulated in the atmosphere, not “during the redistribution process”?

Reply: We appreciate the reviewer’s comments and we revised the content as follow:

In context, 238-239: “These results suggested that there was a higher tendency for multiple soot particles to distribute in the larger LLPS particles (Fig. 7c-e).”

Comment #8: Line 204 “direct in situ evidence” I do not think it is direct and in situ evidence. They are obtained from the observation of filter samples.

Reply: We appreciate the reviewer’s comments and we revised the content as follow:

In context, line 240: “All of these observations provided evidence for soot redistribution in LLPS particles in the atmosphere over the eastern TP.”

Comment #9: Line 205 "soot size" Is soot size provided? Fig. 4a shows that soot has a narrow size distribution. Which data should I see?

Reply: We appreciate the reviewer's comments. The original statement was deemed controversial and thus has been revised based on previous studies to provide clarity. The revised content is as follows:

In context, line 242: "The soot redistribution is probably governed by the entire particle size and the ratio of OM-coating thickness to soot size."

Comment #10: Line 210 "The average Df of externally mixed soot on Mt. Emei was 1.79±0.09 (Table 1), which was slightly higher than that on the southeastern TP (1.75±0.08) (Yuan et al., 2019), suggesting that the sources of soot particles in the eastern TP atmosphere were more complex" First, I do not understand the interpretation of "more complex." Second, values 1.79±0.09 and 1.75±0.08 essentially have no difference.

Reply: We appreciate the reviewer's comments. We have deleted this sentence.

Comment #11: Line 216 "The sulfate-coated soot and organic-coated soot particles had a higher CV (0.87 and 0.87, respectively), higher RN (0.41 and 0.42, respectively) and lower AR (1.61 and 1.61, respectively) than those of externally mixed soot (avg. CV=0.81, avg. RN=0.38, and avg. AR=1.63)." Interestingly, the sulfate-coated and organic-coated soot particles had nearly the same morphological parameters. Are they contradict the discussion of their fractal dimension in line 220? I do not see "a significant increase in fractal dimension" (line 220) when considering their error range and the plot in Fig 8a. The difference can be within an uncertainty range.

Reply: We appreciate the reviewer's comments. The differences of D_f between sulfate-coated BC and organic-coated BC were really small. We have deleted this sentence and rewrote the content as follow:

In context, line 252-254: "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)."

Comment #12: Figure 7a. Please add a unit for the x-axis (nm).

Reply: We appreciate the reviewer's comments. We modified the Figure 7.

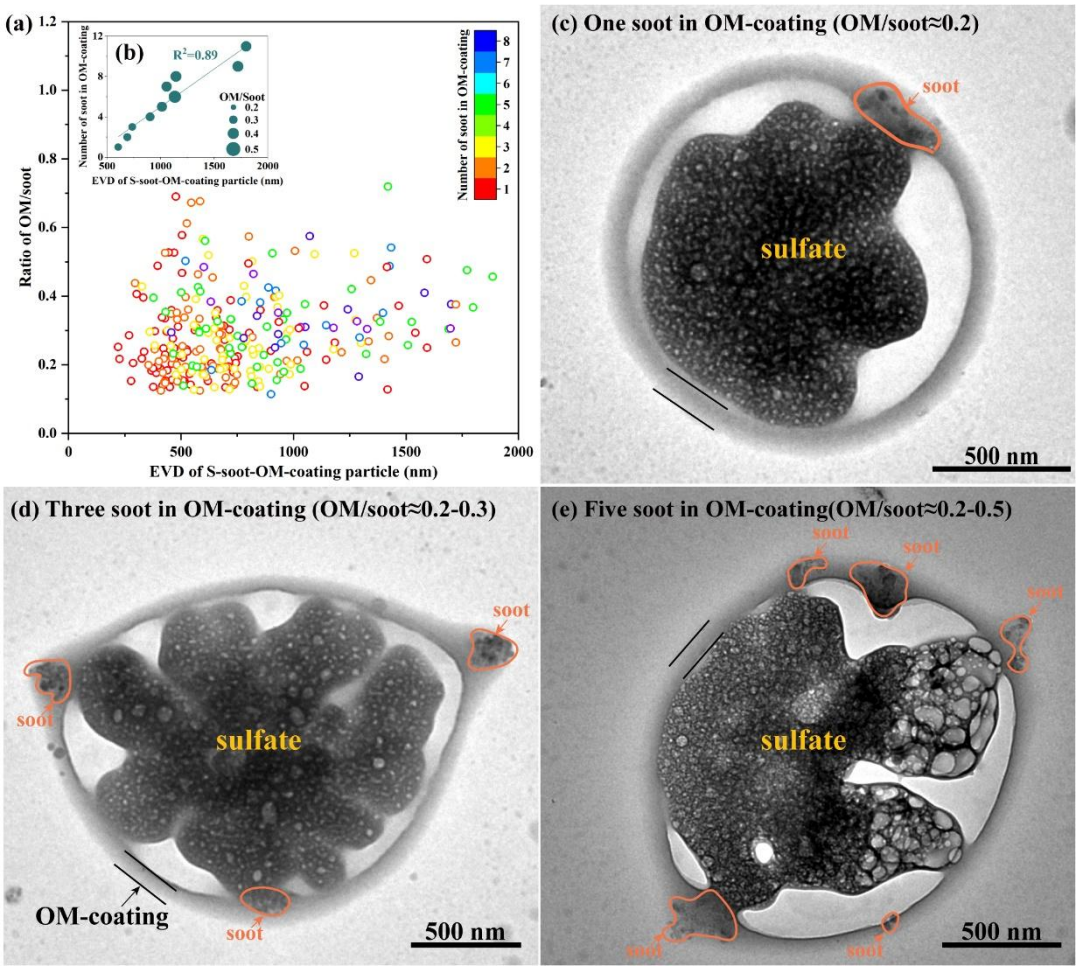


Figure 7. (a) Scatter diagram of OM/soot and the entire particle size of the S-soot-OM-coating particles. Different colours represent the number of soot particles being captured in the OM-coating. (b) Correlation between the average size of the S-soot-OM-coating particle and the average number of soot particles in the OM-coating. The size of the circle point represents the average ratio of OM/soot. (c) A typical TEM image of a S-soot-OM-coating particle with one soot particle in an OM-coating (OM/soot≈0.2, the size of S-soot-OM-coating≈336 nm). (d) A typical TEM image of a S-soot-OM-coating particle with three soot particles in an OM-coating (OM/soot≈0.2-0.3, the size of S-soot-OM-coating≈652 nm). (e) A typical TEM image of a S-soot-OM-coating particle with five soot particles in an OM-coating (OM/soot≈0.2-0.5, the size of S-soot-OM-coating≈582 nm).

Comment #13: Figure 9. I suggest having more discussion in Fig. 9. What do (>90%) and (>70%) mean? At high RH, I guess sulfates deliquesced and had a much larger size. I suggest adding how the liquid-liquid separation and soot redistribution occur in this figure.

Reply: We appreciate the reviewer's comments. We modified Fig. 9 and added some discussion as follow:

In context, line 263: "54% of the total particles were soot-containing particles (Figs. S3 and 9)."

In context, line 271-274: "Once the OM/soot ratio exceeded 0.2, more than 80% of the soot tended to distribute in the organic coating because of the possible intermolecular forces and interactions with increasing coating thickness (Figs. 6b, 9). Conversely, when the OM/soot ratio was less than 0.1, all of the soot in the S-soot-OM-coating particles was found in the sulfate core (Figs. 6b, 9)."

In context, line 275-277: "Theoretical calculations have shown that aged soot particles that were internally mixed with sulfate and organics had higher D_f values (1.84±0.07 for sulfate-coated soot, 1.95±0.06 for organic-coated soot) than those of externally mixed soot (1.79±0.09)."

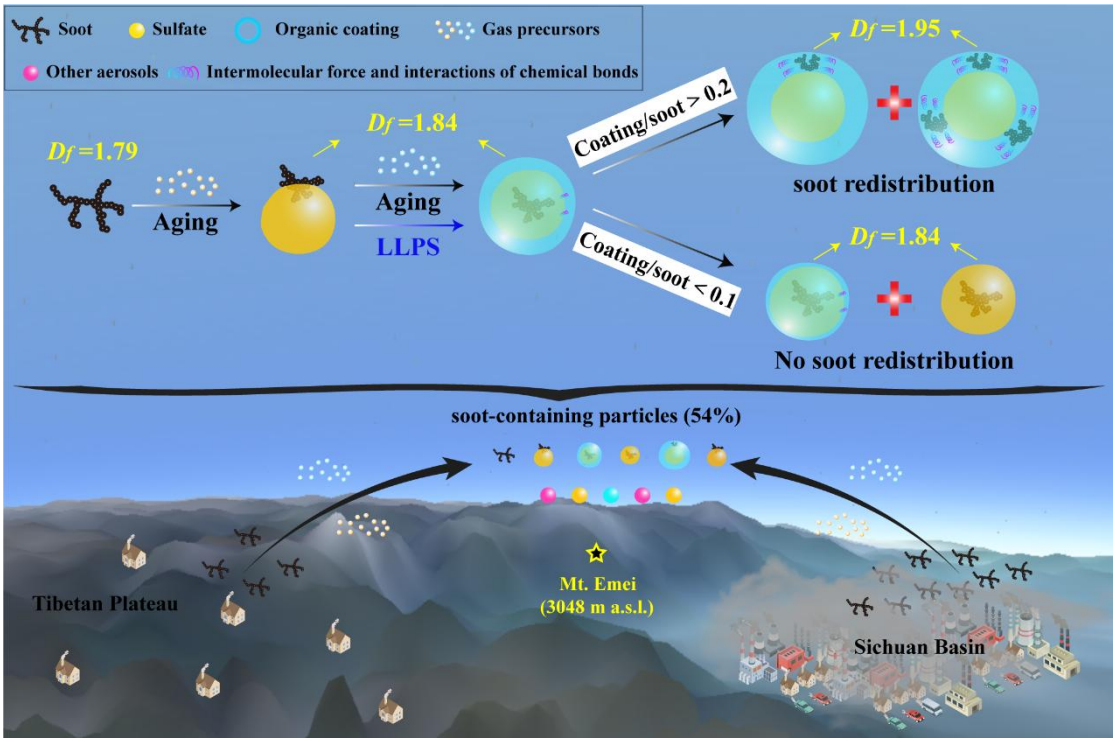


Figure 9. A conceptual model illustrating the atmospheric processes of BC on the eastern rim of the Tibetan Plateau.

Reference:

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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.

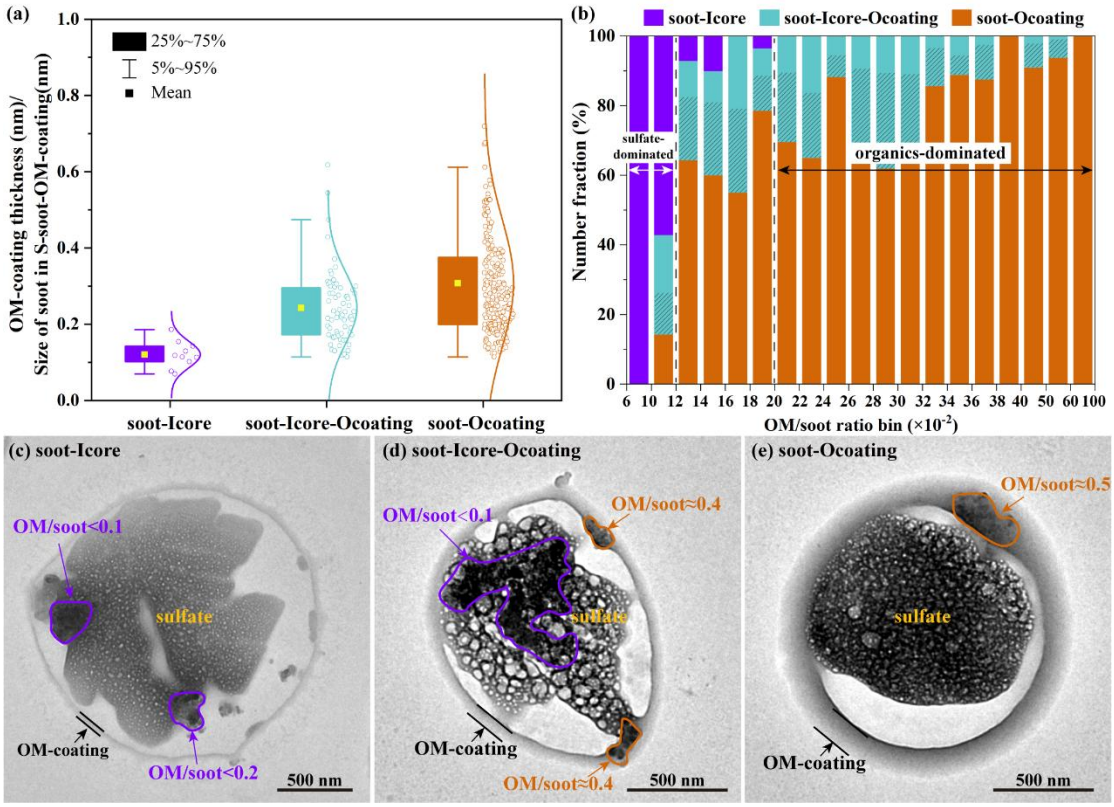
(2022). There, the authors came up with a ratio of 0.24, now they state this ratio is 0.2. My feeling is there is no significant difference between these thresholds (as they are somewhat arbitrary), but the authors need to discuss this.

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).”

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