General Response: We thank the Referee for your helpful comments. We have addressed all comments and provided point by point response below. The revised manuscript is presented in below Response

The study by Xu et al. investigates the aging of soot particles during Asian dust event. The authors collected samples at three sites and compared soot aging at single particle basis. They mainly used transmission electron microscopy (TEM) to study morphology of soot particles. Several morphological descriptors such as aspect ratio, fractal dimensions are used to quantify morphology of soot particles and they classified mixing state of soot-bearing particles based on their coating thickness. They found that soot particles are compact with highest fractal dimension of soot collected at coastal site (T3) in southwestern Japan. They suggested that compact morphology is due to condensation of secondary coating material and though phase separation at high humidity during transport. The research topic is certainly relevant, but I have several concerns.

Answer: We appreciated the Referee#2's comments which significantly improve the quality of the manuscript. We carefully answer them one by one as below. The modifications were highlighted in red in the revised manuscript.

1. There is not much discussion about the chemical composition of soot-bearing particles. Did you perform any chemical analysis?

Answer: Yes, we applied chemical analysis using an energy-dispersive X-ray spectrometer (EDS). The sulfur-rich and organics are the main component mixed with soot, this has been mentioned in 3.2 and Figure 3. Because soot have typical morphology and only contain C and minor O, therefore we did not have more discussion on it.

2. The backtrajectory analysis is not properly discussed in the manuscript. However, this is key for the dust event discussion. I didn't follow the cold front arrival. The authors should discuss in more detail.

Answer: (1) More discussion about the backward trajectory is added in the manuscript (Line 195-198): "The transport duration from the BTH to T1 and T2 was about 12 hours and 15 hours, respectively. Thus, we estimated that the interval between T1 and T2 was three hours. After passing over T1 and T2, the air masses kept moving southeastward to Japan. The estimated interval between T2 and T3 was 30 hours."

(2) The cold front arrival times is used to confirm the time when the sampling site starts to be influenced by the dust storm. They were defined based on PM concentration in Figure 2 and meteorological data (mainly pressure) in Figures S3-S5. Here we add more description in caption of Figure 2: "The cold front arrival times indicate the time when the sampling site starts to be influenced by the dust storm."

3. What is the relevance of dust storm here? If soot particles are studied during a dust storm, why the authors didn't observe or discuss about mixing of dust and soot particles. The authors should discuss about number fraction of soot particles that are mixed with dust particles and size distribution of both dust and soot particles.

Answer: We appreciate the referee's comments. Soot particles mostly have smaller size <500 nm but dust particles mostly are in larger size (> 2um). Although we observed several dust particles associated with soot particles, the number faction is too small (Figure 7). That is the reason that we did not describe more about the mixture of dust and soot particles. Here we mainly consider the movement of the strong cold front during the dust storm. Here the dramatic high PM10 concentrations during the dust storm can be helpful to confirm the cold front. In light of the primary purpose of this study, we did not focus on the dust particles like our previous studies such as (Li et al., 2016;Li and Shao, 2009).

The size distribution of soot particles is presented in the next comment.

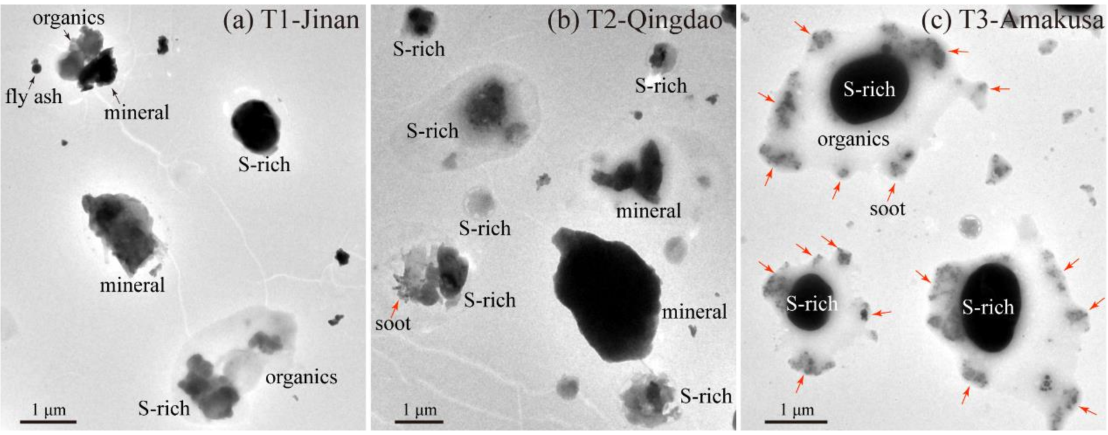


Figure 7. Low-magnification TEM images at T1, T2, and T3.

4. The authors should discuss about the size distribution of three types of soot particles. How did the authors calculate fractal dimension of partially embedded and fully embedded soot particles? For type 3, especially for the fragmented soot ones, it is difficult to measure the required parameters to calculate fractal dimension. They should also provide fractal dimension separately for all three types of soot.

Answer: As the referee's comments, we add more data as below.

(1) The size distribution of soot particles at three sites is provided as follow (Line 273-276): "Size distribution of the soot core indicates a small difference between T1, T2, and T3 during the dust storm period (Figure S9). Thus, the Dp/Dcore increase from T1 to T3 is attributed to the increased coating thickness."

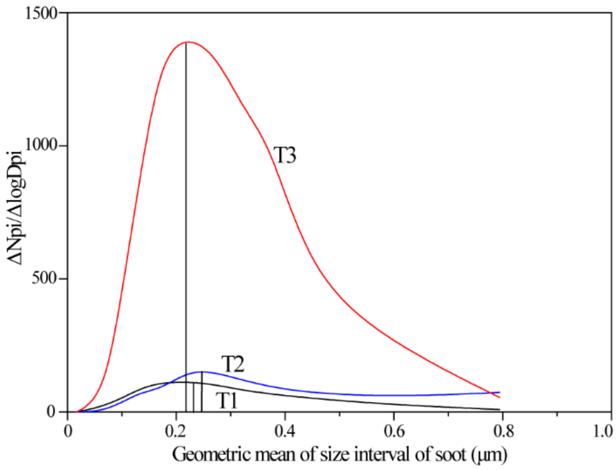
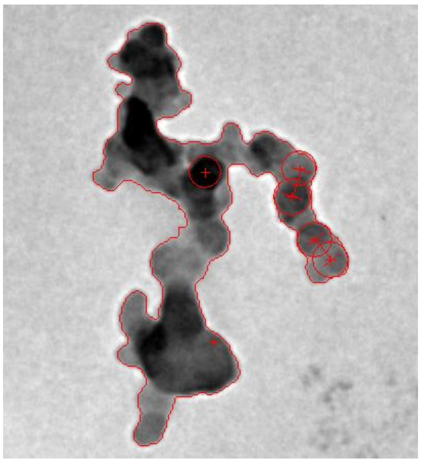
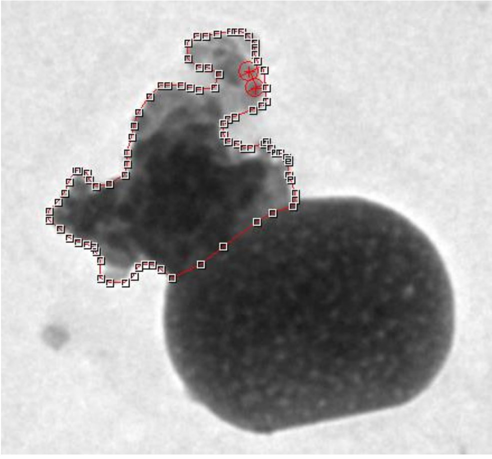


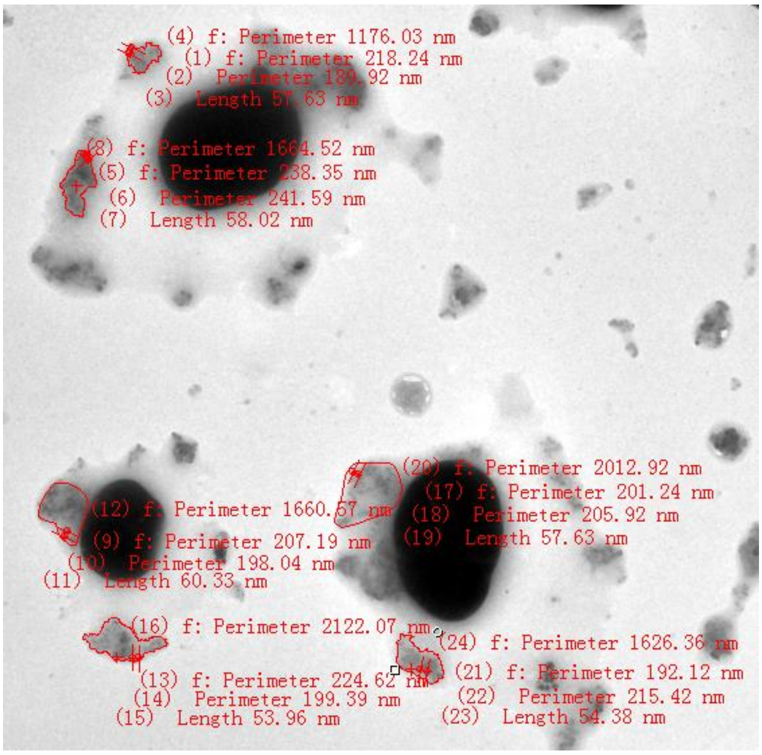
Figure S8. Size distribution of soot core (exclude coating) at T1, T2, and T3.

(2) For soot particles with fractal shape, we manually draw a interpolated polygon to fully cover the edge of soot particles (see figures below). Our iTEM software could obtain the area of this polygon based on our manual drawing, and further convert the area to the equivalent circle diameter (ECD) of soot particles.

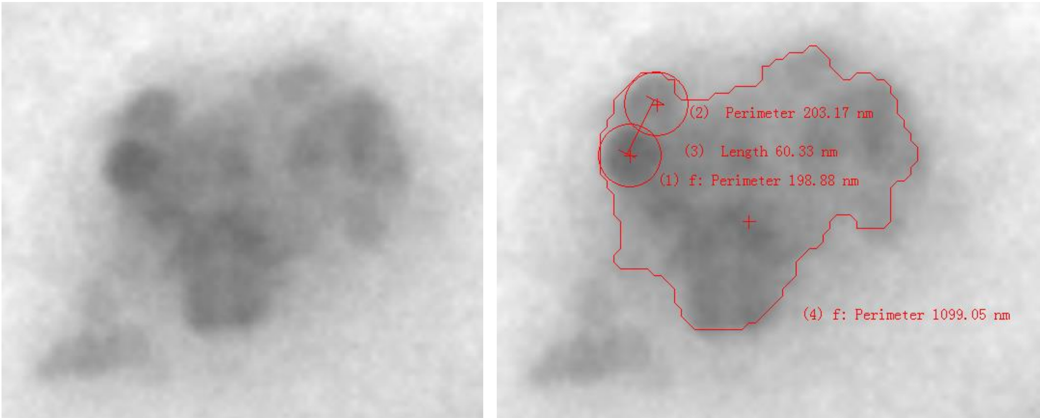




The fractal dimensions and other parameters of the scattered soot particles were acquired same as other soot particles. We manually draw a interpolated polygon to fully cover the edge of soot particles and obtain data from our iTEM software.



It is true that measuring the required parameters to calculate fractal dimension is difficult, especially in low-magnification TEM images. But, we still can acquire the corresponding parameters from high-resolution TEM images.



(3) Thanks to Referee's valuable advice, we realized that there was an issue in the D_f analyses. Thus, we recalculated the D_f in Figure 5. However, there are few soot particles at T1 and T2 that can be used for D_f analyses (34 and 21,respectively). We cannot provide fractal dimension of all three types of soot at T1 and T2. As for T3, we calculate the fractal dimension of partially and fully embeded in the Supporting Information (no fresh soot particle observed at T3).

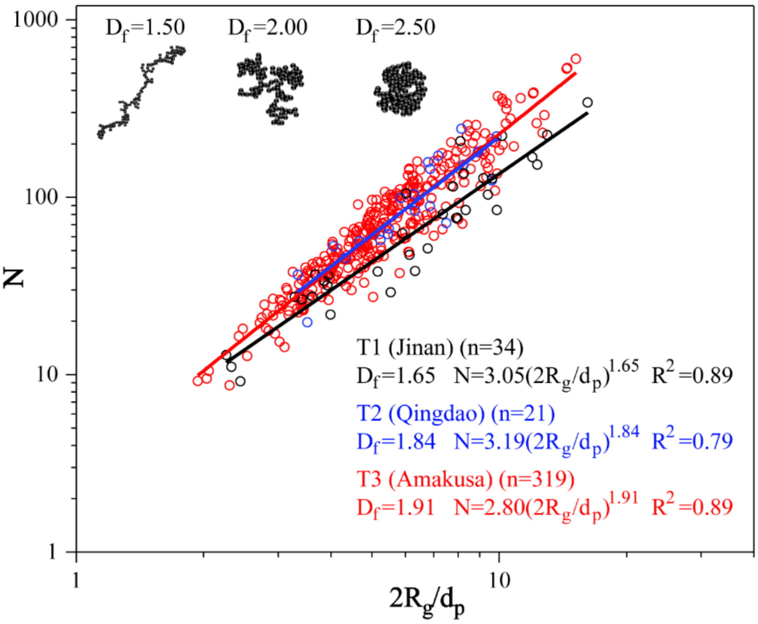


Figure 5. Fractal dimension of soot-bearing particles at the three sampling sites. The parameter n in parentheses represents the total number of soot particles analyzed for each site to calculate Df and kg.

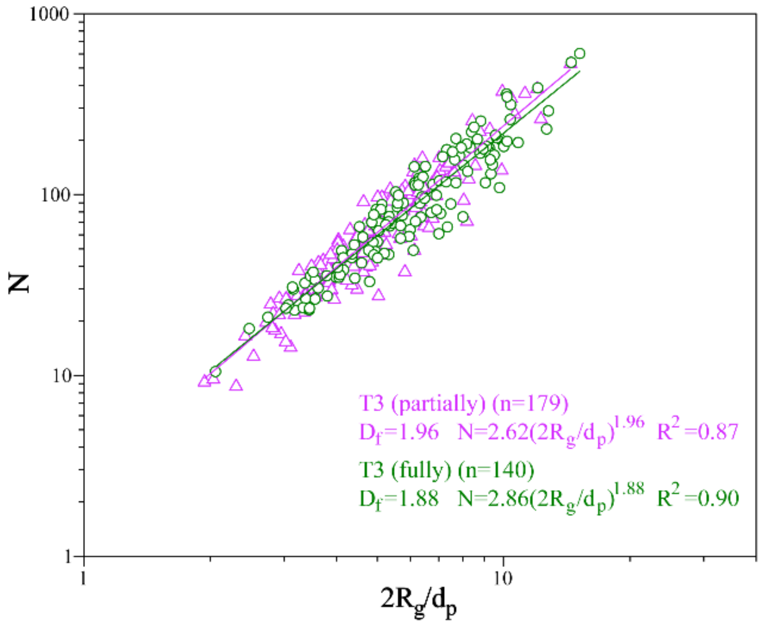


Figure S7. Fractal dimension of partially and fully embedded soot particles at T3 site. It is different to provide fractal dimension of different types of soot at T1 and T2 because of the small number of soot particles at these two sites. The parameter n in parentheses represents the total number of soot particles analyzed for each site to calculate D_f and k_g

5. The discussion about the mixing state configuration needs to be elaborated, like how many soot particles did you observe within individual partially embedded soot particles? How many fragments were observed in type-3 (figure 3d). This information would be useful to understand the aging process and for modeling purposes.

Answer: We provided the frequency of soot fragment number in single soot-bearing particles. More discussion is added in Line 228-229: "Most of partially embedded soot particles include one soot core, only ~ 10% of them contain two soot cores (Figure S7)";

Line 321-323: "More than half of this type of particles contain one or two soot fragments, while 43% of them include more than three soot fragments (Figure S7)."

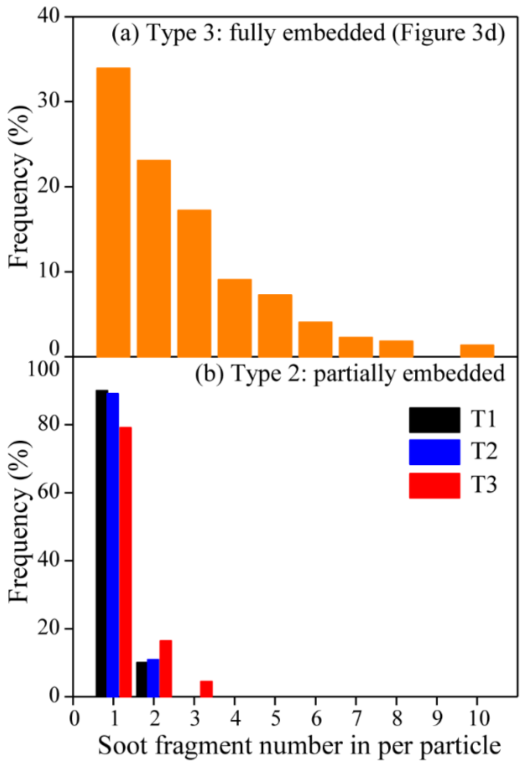


Figure S7. Frequency of soot fragment number in single soot-bearing particles.

6. Need to discuss how many soot particles were studied per sample. How many total samples during event? Overall, particle statistic is poor.

Answer: Totally, we analyzed seven dust samples (Table S1). The number of analyzed soot-bearing particles is shown above the column in Figure 4. The total number of aerosol particles analyzed in this study is presented in the 2.2, which is 412, 486, and 887 for T1, T2, and T3 site, respectively.

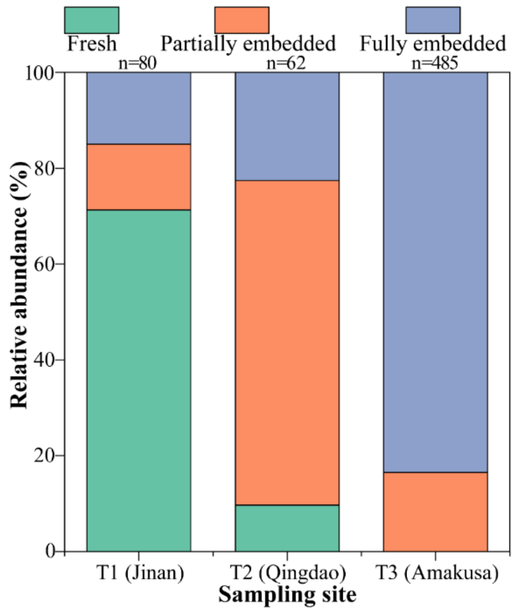


Figure 4. Relative abundance of three types of soot-bearing aerosol particles at the three sampling sites. The number of analyzed soot-bearing particles is shown above the column.

7. If phase separation may be a key mechanism for observation of fully embedded particles, the authors can investigate fraction of fully embedded particles at different RH, not just by sampling site. The RH was high too during certain time at T2 site. The authors should investigate those samples as well.

Answer: There might be some misunderstanding. We did not propose that phase separation may be a key mechanism for fully embedded particles. The high RH in marine air could lead to secondary aerosols change from a solid to liquid phase. During or after sampling, due to the RH decreasing, the phase separation between inorganic and organic components occur. This is a possible cause of the formation of scattered tiny soot in the organic coating (special mixing structure in Figure 3d).

References

Li, W., Sun, J., Xu, L., Shi, Z., Riemer, N., Sun, Y., Fu, P., Zhang, J., Lin, Y., Wang, X., Shao, L., Chen, J., Zhang, X., Wang, Z., and Wang, W.: A conceptual framework for mixing structures in individual aerosol particles, J. Geophys. Res.: Atmos., 121, 13784-13798, https://doi.org/10.1002/2016JD025252, 2016.

Li, W. J., and Shao, L. Y.: Observation of nitrate coatings on atmospheric mineral dust particles, Atmos. Chem. Phys., 9, 1863-1871, https://doi.org/10.5194/acp-9-1863-2009, 2009.