Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-539-RC1, 2020

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Tracing the evolution of morphology and mixing state of soot particles along with the movement of an Asian dust storm" *by* Liang Xu et al.

Anonymous Referee #1

Received and published: 20 July 2020

This study investigates soot mixing states mainly using a transmission electron microscope (TEM) in China and Japan. They collected samples by following the aging of an air plume during a dust event and found changes of soot mixing states as the plume aged. Their findings will be an interesting case study to follow the soot aging in an ambient atmosphere and a heavily polluted area. As the soot aging process is of interest for an understanding of their climate influences through the accurate estimates of soot lifetime and optical properties, their result will have contributions to the climate prediction. However, I have a list of concerns regarding their discussion and interpretations of their results. A major concern is that the many discussion regarding T3 samples (and Type 3 particle) was based only on a particle shown in Fig. 3 d. The authors also

try to have a general model based on the one-particle observation; a better statistic is necessary here. Please see the specific comments for more suggestions.

1. Line 108-110: from 1 min to 10 min: I see only 1 to 2 min in Table S1.

2. Line 113 (and 86): TEM has been defined as "transmission electron microscopy" in line 86, whereas it is defined as "transmission electron microscope" here. Please be consistent.

3. Section 2.5 Line 160-162: It is unclear how the Eq. 3 is used to obtain α and ka here.

4. Line 180-183: How were the cold front arrival times defined? I do not see a clear change at the time of the arrival in Figures S3-5. As the timing is used to evaluate soot aging time, please make them specific. Why did the authors use Beijing time instead of UTC? As the local time in T3 site is not Beijing time, UTC will be better.

5. Line 196-198: The dust distribution and back-trajectory model in Fig. 1 indicates that the Dust samples at T3 site are not from the dust plume. On 8:00 18 March in Fig. 1, the edge of the dust plume was 18 and 24 hours ahead for the Dust 1 and 2 sampling at T3 site, respectively. Thus, the dust should arrive between 2:00-8:00 19 March at T3 site, whereas the samplings were done at 23:16, 18 March and 6:36, 19 March. At least, the sentence "together verified that the dust storm event" is not valid, unless I miss something. Figure S2 may be helpful, but I cannot see details in the figure. I recommend rechecking the modeling data and showing robust data to prove the dust arrival before the sampling at T3 site. TEM data of dust particles from the T3 site samples may be useful.

6. Line 205-208: It is surprising that the dust samples have so many soot particles. Are there so many dust particles as well together with soot particles? TEM images with low-magnification showing more particles, including soot and dust, will be helpful to have an idea of how such a large number of soot particles occur in the dust samples.

7. Line 223-224: Do coagulations between soot particles and others contribute to the "partially embedded soot" formation?

8. Line 237-242: When the soot particles are fully coated, such as aged samples at T3, the deposition efficiency of fully embedded soot-bearing particles may not differ from those without soot. I assume that the increase of the number fraction of soot-bearing particles is simply due to an increase of mixing state index as aging (Riemer and West, 2013; Healy et al., 2014), but more discussion will be useful here.

9. Line 247-254 and 258-259: I suggest adding TEM images, including many soot particles having high or low fractal dimensions, so that readers visually see the compaction of soot particles as age.

10. Line 263-265: It is unclear how the Dcore was obtained. As soot particles have a fractal structure, Dp and Dcore should have different relations in Fig. S6. Please explain the methodology.

11. Line 307-327: The discussion in this paragraph based on only one particle (Fig. 3d), and it is difficult to have a general conclusion. I suggest showing more particle images or adding statistics of the fraction having particles with phase separation or tiny soot particles. I also question how the fractal dimensions or other parameters were obtained from the scattered soot particles.

12. Line 326-330: I do not see any evidence of cloud-aqueous process discussed here. Are there any could in the dust plume? How do "the cloud-aqueous process and the phase separation of organic and sulfate components in the soot-bearing particles likely break the chain-like soot" happen? Do the "the cloud-aqueous process and the phase separation" make soot compact or scatter? Careful discussions should be provided here.

13. Line 363-366: The conclusion was obtained only one soot particle. Please show more data or revise the conclusion. It sounds that the conclusion “that the compli-

cated aging processes of individual particles can break the chain-like soot formation” contradicts with soot compaction process. Please have a comment on this.

14. Figure 2: Dust 1 in T2 was collected ahead of the samples in T1. Dust 2 in the T2 was collected only 3 hour ahead of the T3 dust 1 sample. Ideally, if someone wants to compare the aging process, the samples should be collected within the same plume, following the airmass movement. The sampling strategy may cause an influence, and some discussion about the sampling strategy may be helpful.

15. Figure 3: I recommend having more particle images, including low-magnification images from T1, T2, and T3 sites.

16. Figures 4 and 5a. The number of analyzed soot particles does not agree between Fig. 4 and 5a (e.g., 80 vs 36). Did the author select soot particles for the analysis in Fig. 5a? If so, how and does the method cause any bias?

17. Fig. 5b: The error bars are too high to compare the data. What do the bars indicate?

18. Fig. 6: If there are soot and other emissions at T2, the aging process will be rather complicated, i.e., they should be the mixtures of aged soot and freshly emitted soot. The influence of the emission at T2 should be discussed.

19. Table S1. The wind directions for the samples within the same sampling site vary largely (almost opposite directions (e.g., 121 vs. 358 at T3). The wind direction in Fig S1 at T3 is also complicated. I question if the samples were collected from dust plume. Again, TEM images of dust particles may be helpful.

20. Figure S2. It is difficult to see the details.

21. Figure S6. Is the plot include soot particles?

Healy, R. M., Riemer, N., Wenger, J. C., Murphy, M., West, M., Poulain, L., Wiedensohler, A., O’Connor, I. P., McGillicuddy, E., Sodeau, J. R., and Evans, G. J.: Single particle diversity and mixing state measurements, Atmos. Chem. Phys., 14, 6289–6299, https://doi.org/10.5194/acp-14-6289-2014, 2014.

Riemer, N. and West, M.: Quantifying aerosol mixing state with entropy and diversity measures, Atmos. Chem. Phys., 13, 11423–11439, https://doi.org/10.5194/acp-13-11423-2013, 2013.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-539, 2020.