Reply to Reviewer #2

The manuscript by Zhang et al. investigates two different soot aerosol aging processes by comparing the properties of particles that are transported over land and over the ocean. The authors utilize measurements of particle morphology, number of soot cores and coating thickness, and report differences in optical properties. Overall, it was found that particles that were transported via the sea pathway, and likely underwent aqueous-phase or cloud processing, had an increase in soot core number and a decrease in absorption enhancement compared to particles that were transported over the land, where heterogenous oxidation dominated.

Overall, the study is well conducted, clear, and of interest to the ACP community. I would recommend publication after addressing a few minor comments.

We are grateful for this reviewer's comments. These comments are all valuable and helpful for improving our paper. We added a figure to show the presence of clouds during the transport of haze masses through the sea pathway (Figure S7). We added the *P* value calculation to show the significant difference in coating thicknesses of soot-containing particles between the NCP and the YRD (Figure 7). We added labels (Inland and Sea) in Figure 2, 4, 5, 6, 7, 8, 9. 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 and supplementary information, the red color was marked as the revised places.

1. The authors primarily discuss environmental conditions (cloud processing, RH, etc) as what is causing the differences between the two aging pathways. Although this evidence is convincing, there are likely also significantly different emissions mixing with the haze plume during transport over land than over the ocean. This is briefly mentioned as a possibility at line 95 but not referred back to in the results section. Can the authors comment on if these differences and if they have implications on the observations or results.

   Reply: We appreciated the reviewer's comments and added some discussion

on differences in pollutant emissions and their implications.

P18 L482-487: "We further noticed that soot-containing particles did not pass areas with high emissions during transboundary transport through the sea pathway compared to those transported through the inland pathway (Figures 1a-b). These findings suggest that the aging process of soot-containing particles was primarily driven by the meteorological change (i.e., cloud), with minimal contribution from additional industrial and urban emissions along the sea pathway."

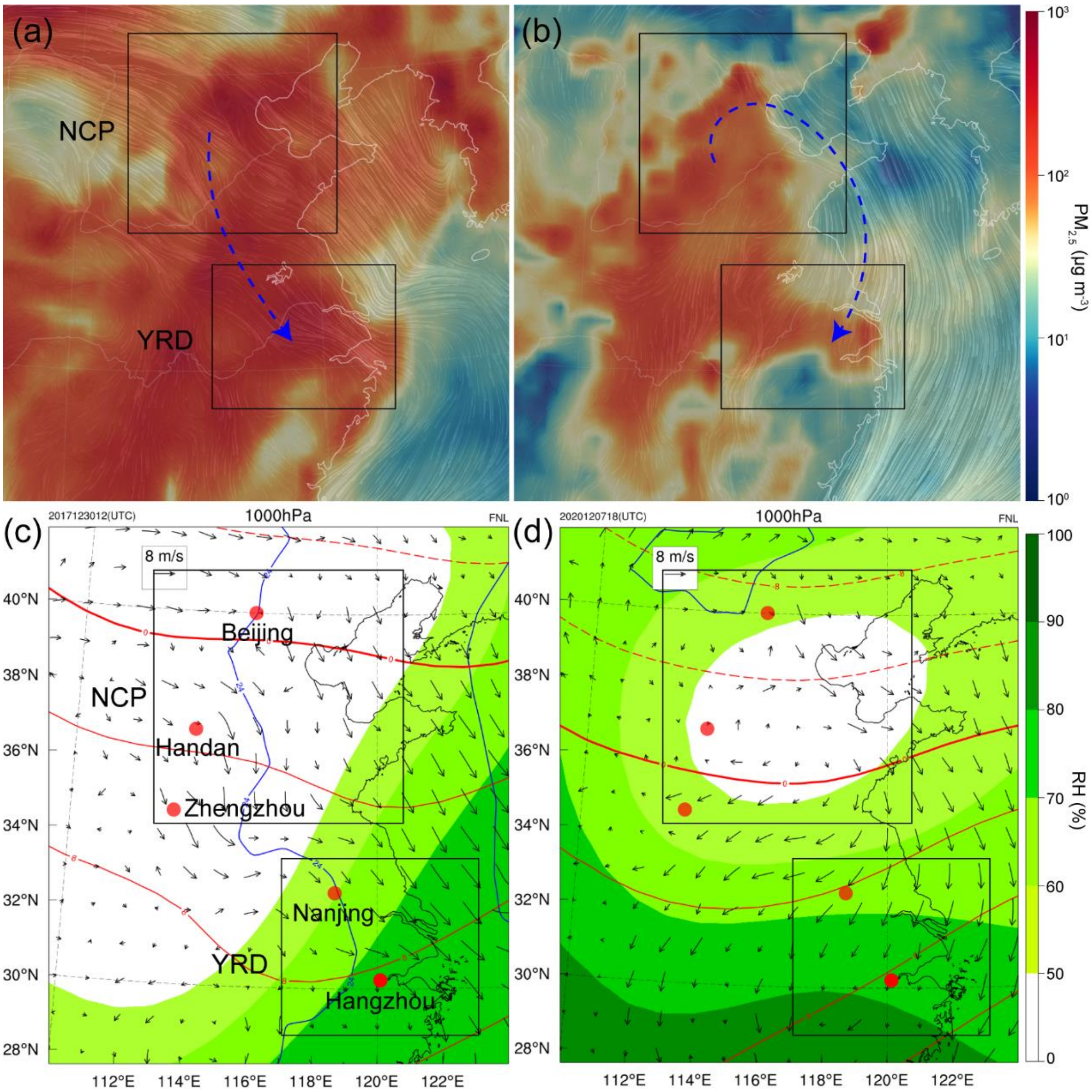

Figure 1. Meteorological fields in eastern China during the observation period. (a-b) Wind fields combined with surface $PM_{2.5}$ concentrations at 20:00 (local time) on December 30, 2017 and at 2:00 on December 8, 2020 derived from European Centre for Medium-Range Weather Forecasts (ECMWF, https://earth.nullschool.net/). The

blue arrow dashed lines indicate prevailing wind direction. (c-d) Meteorological fields covering observation sites in the North China Plain (NCP) and Yangtze River Delta (YRD) at 1000 hpa.

2. Line 327: Figure 3 and 4 show particles with soot cores very close to the particle edge. How exactly are partly-coated and embedded particles differentiated. Is there a specific threshold for how much soot is exposed for it to be considered partly-coated? Similarly, how did the authors categorize multi-core particles where individual cores were embedded and partly-coated in the same particle.

   Reply: In this study, soot cores completely coated by S-rich particles were considered to be embedded types and other internally mixed soot cores were considered to be partly-coated types.

   Based on TEM observations, less than 10% of soot-containing particles had both embedded and partly-coated soot cores. To categorize these particles, we classified those with more than 95% of the total soot volume embedded in host particles as embedded soot-containing particles, and the remainder as partly-coated types. Because these particles were relatively few, they had a limited impact on the statistical results. We added the explanation as follows.

   P14 L360-365: "In this study, less than 10% of soot-containing particles had both embedded and partly-coated soot cores. To categorize these particles, we classified those with more than 95% of the total soot volume embedded in host particles as embedded soot-containing particles, and the remainder as partly-coated types. Because these particles were relatively few, they had a limited impact on the statistical results."

3. Line 392: Are the differences in coating thickness between NCP and YRD statistically significant?

   Reply: Yes, the differences in coating thicknesses of soot-containing particles between the NCP and the YRD are significant. We calculated the *P* value to show it (Figure 7). The weak difference in partly-coated soot-containing

particles transported through the sea pathway may be attributed to the lack of additional pollutants involved in their aging processes and their lack of activation.

P16 L423-426: "Following the transboundary transport of soot-containing particles through the inland pathway, the mean D_p/D_c ratios of partly-coated and embedded soot-containing particles increased from 2.37 ± 1.27 and 2.85 ± 1.89 in the NCP to 2.79 ± 1.37 and 3.41 ± 1.87 in the YRD (*P* < 0.05, Figure 7a)."

P17 L446-449: "When soot-containing particles were transported from the NCP to the YRD through the sea pathway, the partly-coated D_p/D_c ratio slightly increased from 2.41 ± 1.37 to 2.66 ± 1.58, but the embedded D_p/D_c ratio significantly increased from 2.92 ± 2.01 to 4.38 ± 2.92 (*P* < 0.001, Figure 7b)."

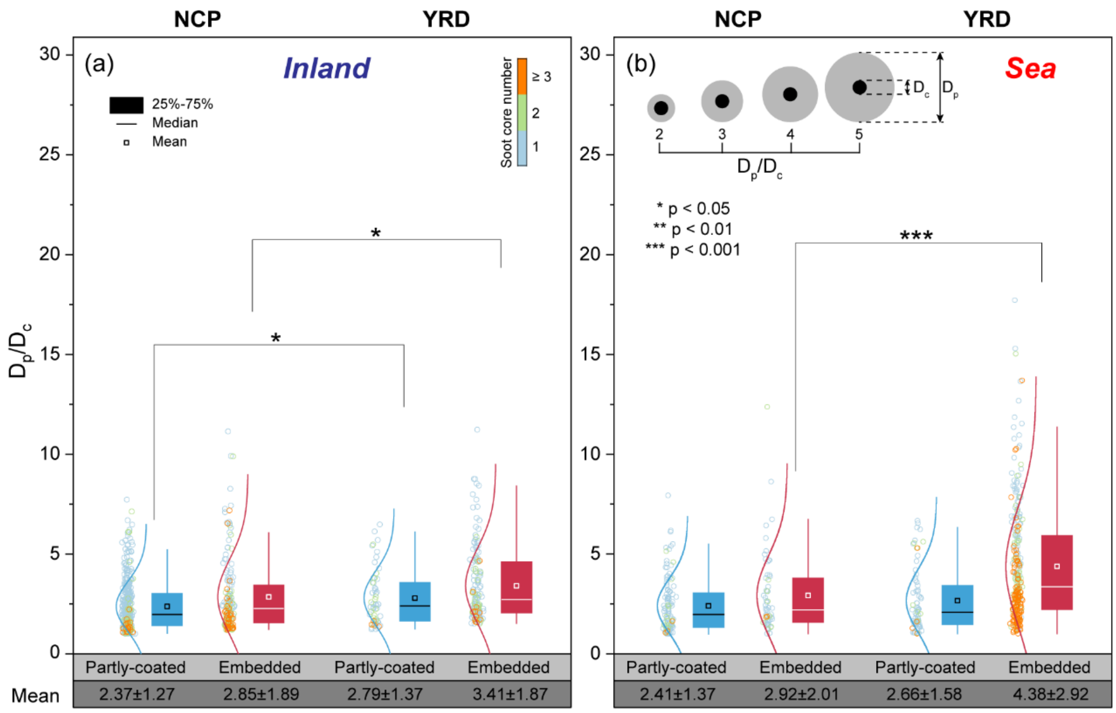

Figure 7. The size ratio of soot-containing particles to their soot cores (D_p/D_c) in two types of transboundary transport models from the NCP to the YRD. (a) D_p/D_c ratios of soot-containing particles transported through the inland pathway. (b) D_p/D_c ratios of soot-containing particles transported through the sea pathway. A schematic model of the D_p/D_c ratio of soot-containing particles with the core-shell structure is exampled.

4. Can the authors expand on their discussion of the water rim observed in some particles, as this seems to be an important piece of evidence for aqueous phase processing (i.e. line 427). It would be helpful to clarify if this is a marker for particles that have undergone aqueous-phase processing (as mentioned at line 431), or for particles that contained an aqueous-phase when analyzed with TEM (line 429). Put another way: If the particles underwent aqueous-phase processing during transport, then were subjected to lower humidity conditions and effloresced, would the rim still be present.

Reply: Yes, the water rim will still exist after aqueous-phase particle efflorescence. The previous studies have observed water rims after aqueous-phase particles are dehydrated using an individual particle hygroscopicity system (Sun et al., 2018). We added a discussion on the water rim.

P17 L462-465: "Laboratory studies have also observed water rims after aqueous-phase particles are dehydrated (Sun et al., 2018). In other words, if particles undergo aqueous-phase processing during transport, and then effloresce under low RH conditions, the water rim will be present as a marker."

5. Line 437: Is there satellite or meteorological evidence of clouds being present along the back trajectory of the airmass. If possible, It would be useful to differentiate between cloud processing and high humidity (but still subsaturated)

Reply: Thanks for the reviewer's comments. We added a satellite image combined with the backward trajectory to reflect the presence of clouds during the transport of haze masses through the sea pathway (Figure S7).

P18 L475-478: "Figure S7 shows the satellite image combined with the backward trajectory of haze masses during December 7-8, 2020. We found the presence of clouds over the East China Sea during the transport of haze masses through the sea pathway (Figure S7)."

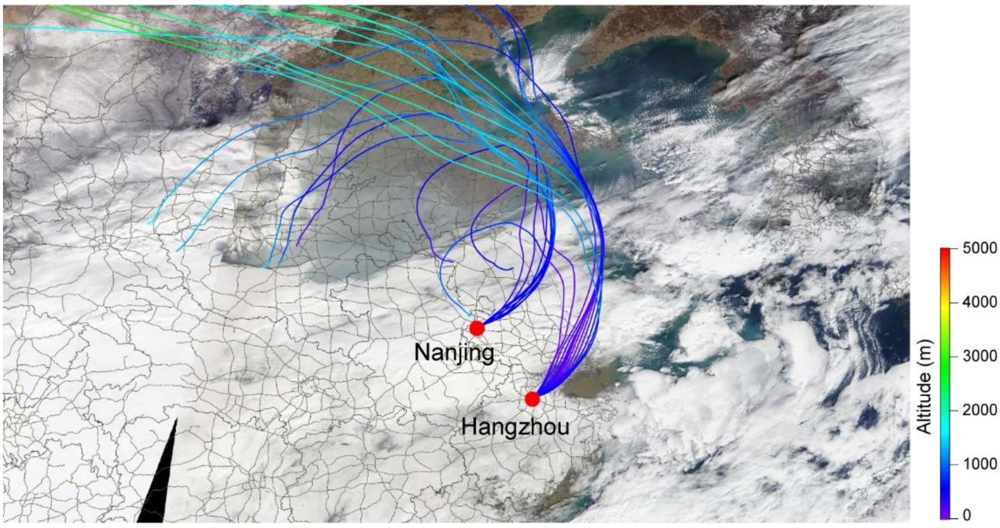

Figure S7. A satellite image combined with the backward trajectory of haze masses before arriving at Nanjing and Hangzhou sites during December 7-8, 2020.

6. Line 512: Can the authors clarify why entrainment of multiple soot cores results in lower ΔEabs/Δ(Dp/Dc).

Reply: Yes, the lower $\Delta E_{abs}/\Delta(D_p/D_c)$ of soot-containing particles transported through the sea pathway may be attributed to the larger change in their D_p/D_c and smaller change in their E_{abs} compared with the inland pathway. We added some discussion on it.

P21 L560-572: "Previous studies have revealed that the E_{abs} of soot-containing particles first increases and then tends to stabilize with their coating thickness (e.g., D_p/D_c) increases (Beeler et al., 2024; Fu et al., 2022). We found that the mean D_p/D_c of embedded soot-containing particles exhibited a large value at 4.38 when haze masses were transported through the sea pathway (Figure 7b). In addition, cloud processes induced multiple soot cores within single particles during the transboundary transport through the sea pathway in contrast to the inland pathway, reducing their optical absorption (Figure 10a). Beeler et al. (2024) also found consistent results that much lower E_{abs} variation for soot-containing particles with the thickening of coatings in pyrocumulonimbus clouds compared with urban air. Therefore, the larger D_p/D_c change and the smaller E_{abs} change of soot-containing particles transported through the sea

pathway should result in the lower $\Delta E_{abs}/\Delta(D_p/D_c)$ compared to those transported through the inland pathway."

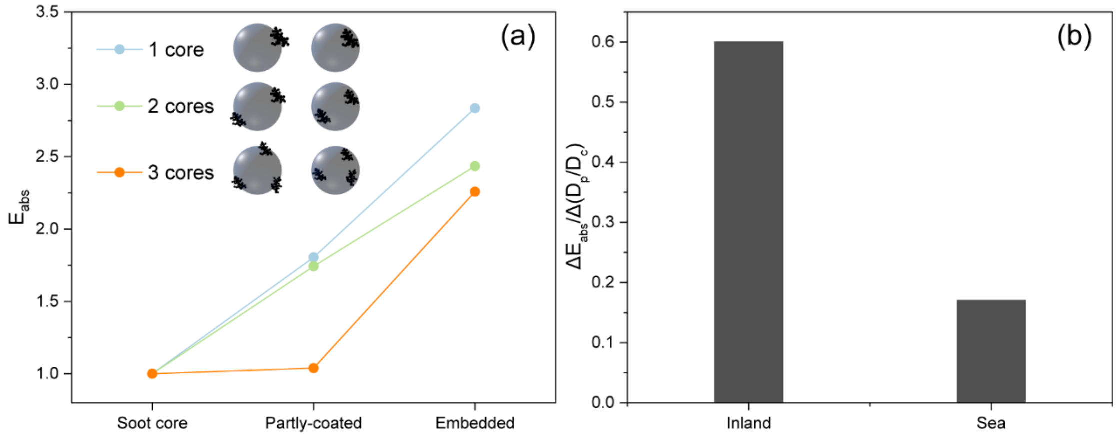

Figure 10. Variation in the optical absorption of soot-containing particles. (a) The light absorption enhancement (E_{abs}) of partly-coated and embedded soot-containing particle models relative to their soot cores. (b) The change in E_{abs} per unit the change in D_p/D_c ($\Delta E_{abs}/\Delta(D_p/D_c)$) of soot-containing particles during two transboundary transport events through the inland and the sea pathways. Partly-coated and embedded soot-containing particle models constructed by the Electron-Microscope-to-BC-Simulation (EMBS) tool were exampled in panel (a).

7. Most figures (Figure 2, 4, 5, 6, 7, 8, 9) have panels for the results of each pathway, however it is not immediately clear which one is which. Although this information is in the caption, I would recommend labeling the row or panels with the different pathways.

Reply: Thanks. We added labels (Inland and Sea) in these Figures.

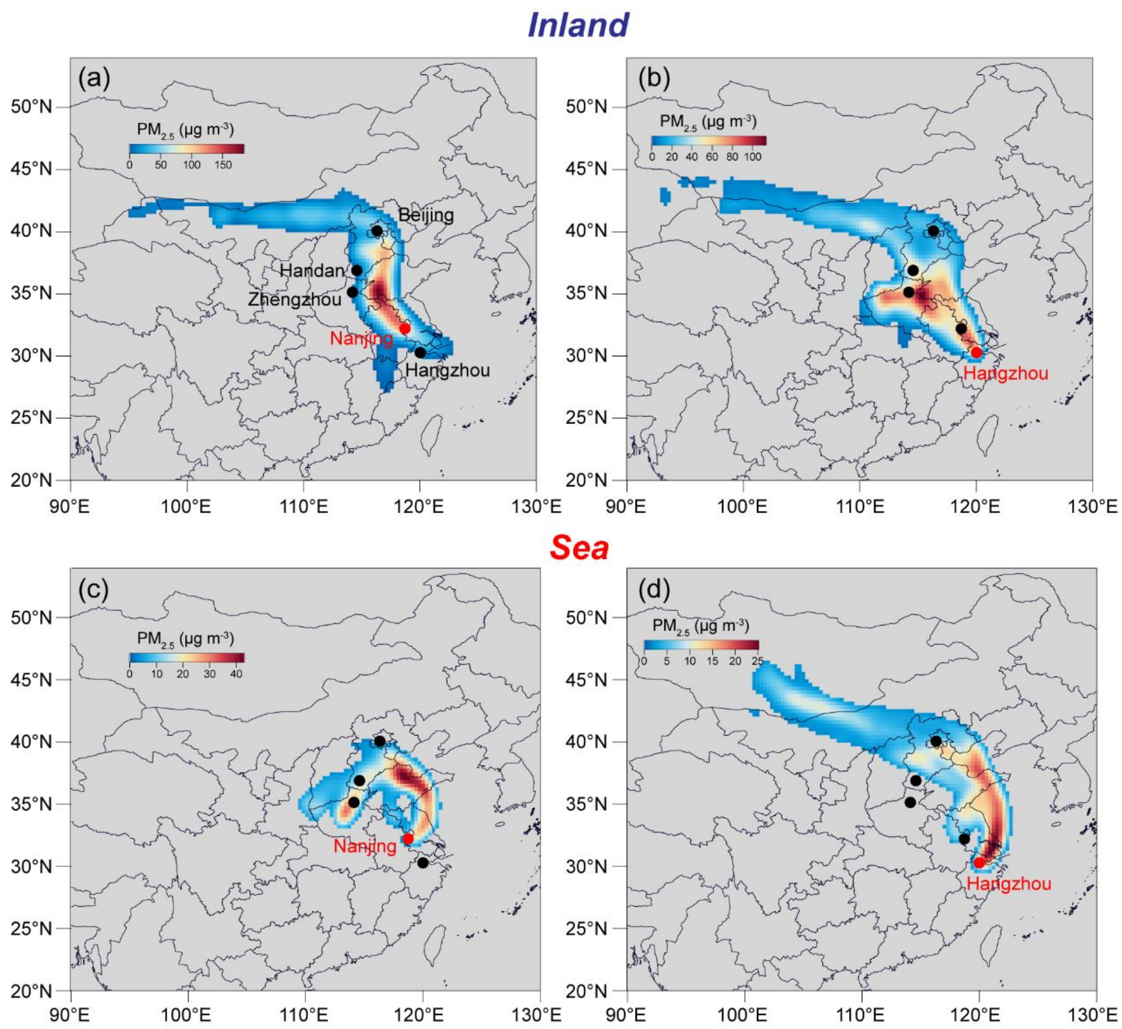

Figure 2. Concentration-weighted trajectory (CWT) plots of $PM_{2.5}$ before arriving at observation sites (Nanjing and Hangzhou) in the YRD. (a-b) Transboundary transport through the inland pathway during December 30-31, 2017. (c-d) Transboundary transport through the sea pathway during December 7-8, 2020.

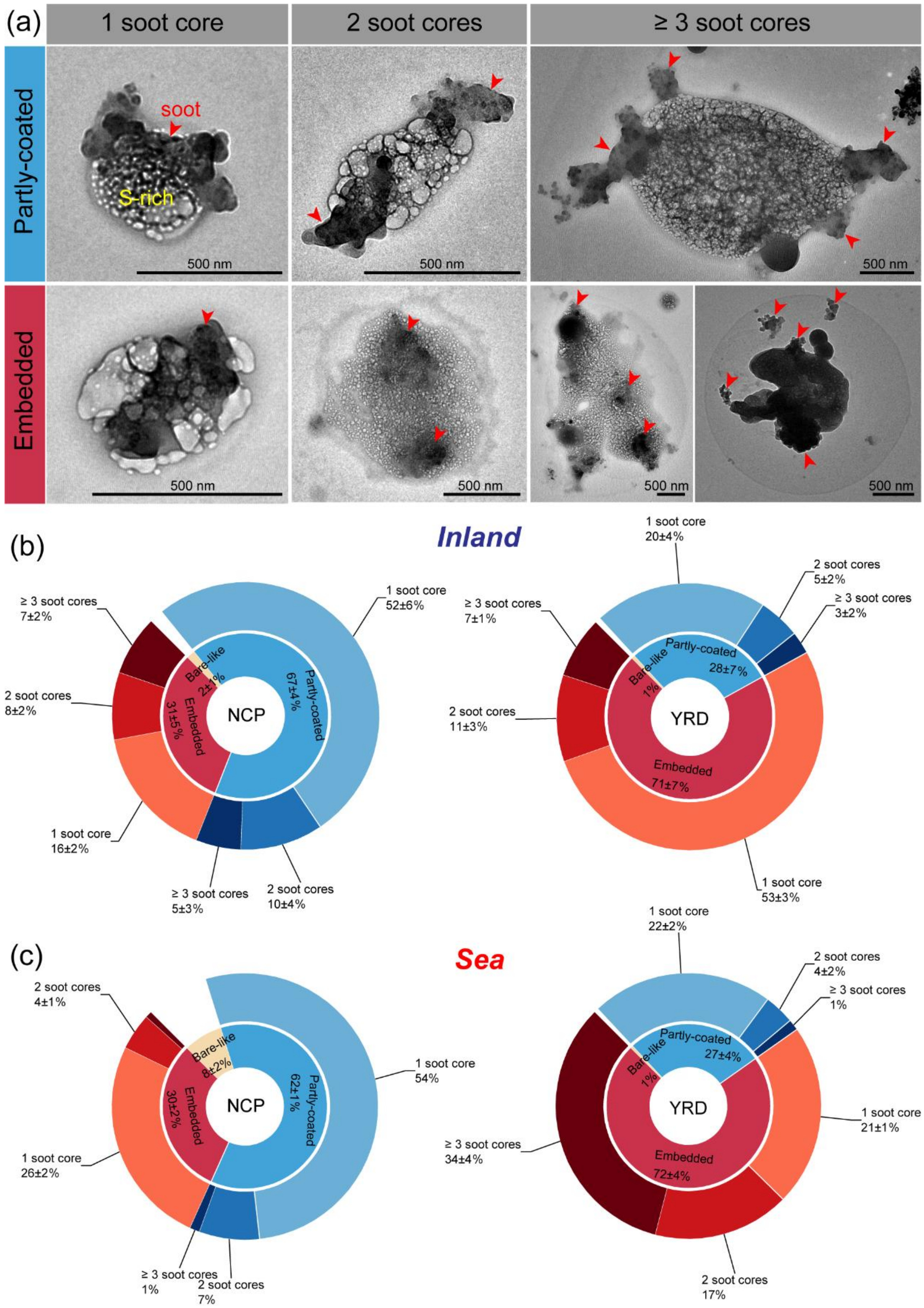

Figure 4. Typical TEM images and number fractions of soot-containing particles with different mixing structures and soot core numbers in two types of transboundary transport models from the NCP to the YRD. (a) Partly-coated and embedded soot-containing particles with different numbers of soot cores. (b)

Variation in the number fraction of soot-containing particles during the transboundary transport through the inland pathway. (c) Variation in the number fraction of soot-containing particles during the transboundary transport through the sea pathway.

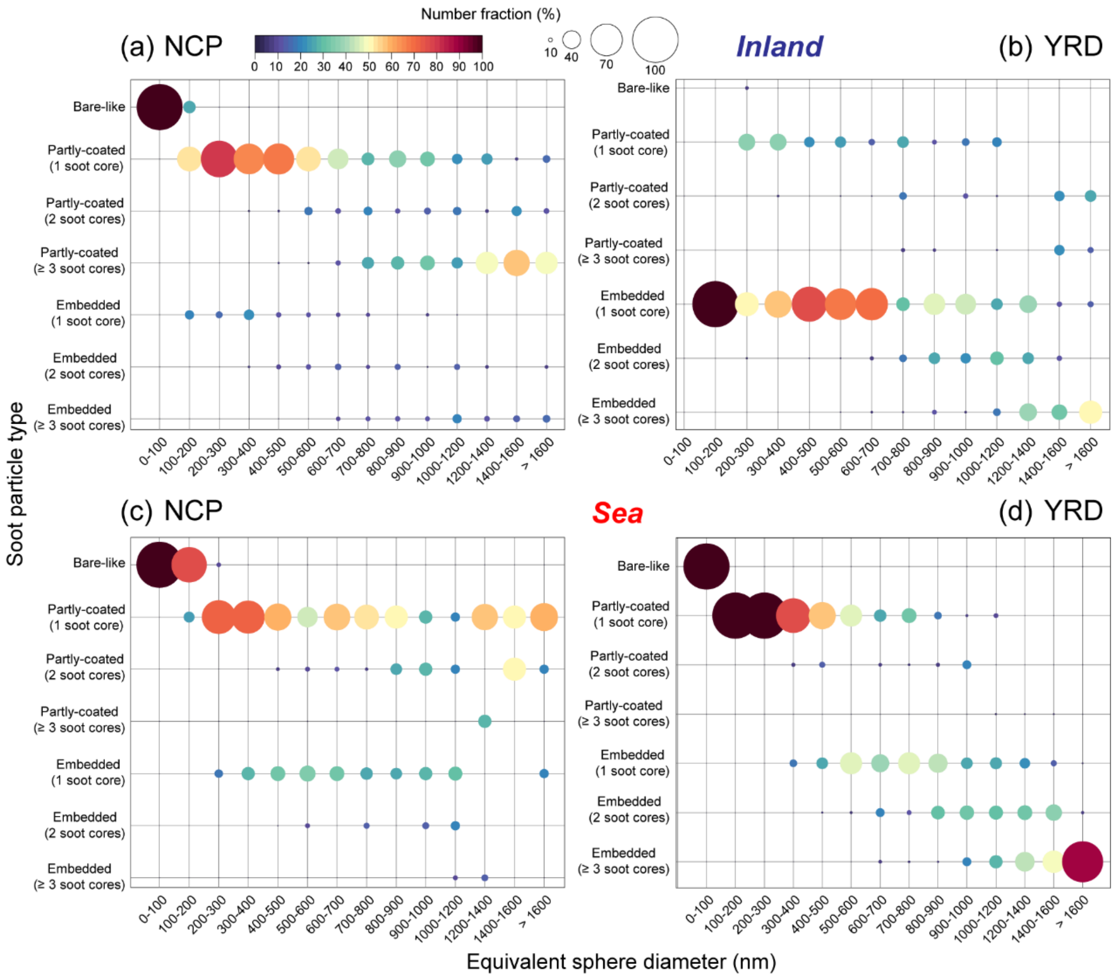

Figure 5. Number fractions of soot-containing particles with different mixing structures and numbers of soot cores in different size bins in two types of transboundary transport models from the NCP to the YRD. (a-b) Soot-containing particles transported through the inland pathway. (c-d) Soot-containing particles transported through the sea pathway.

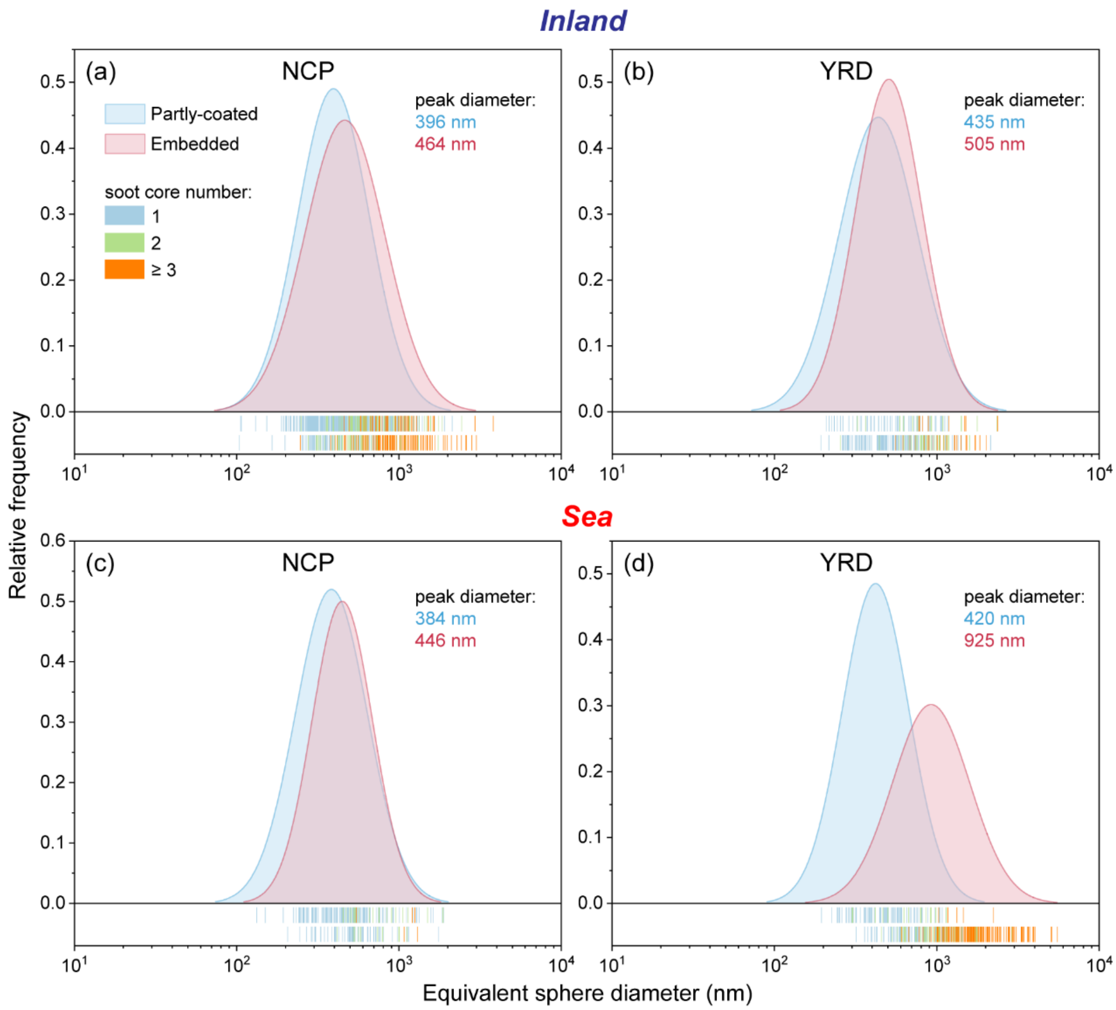

Figure 6. Number size distribution of soot-containing particles in two types of transboundary transport models from the NCP to the YRD. (a-b) Size distribution of soot-containing particles transported through the inland pathway. (c-d) Size distribution of soot-containing particles transported through the sea pathway.

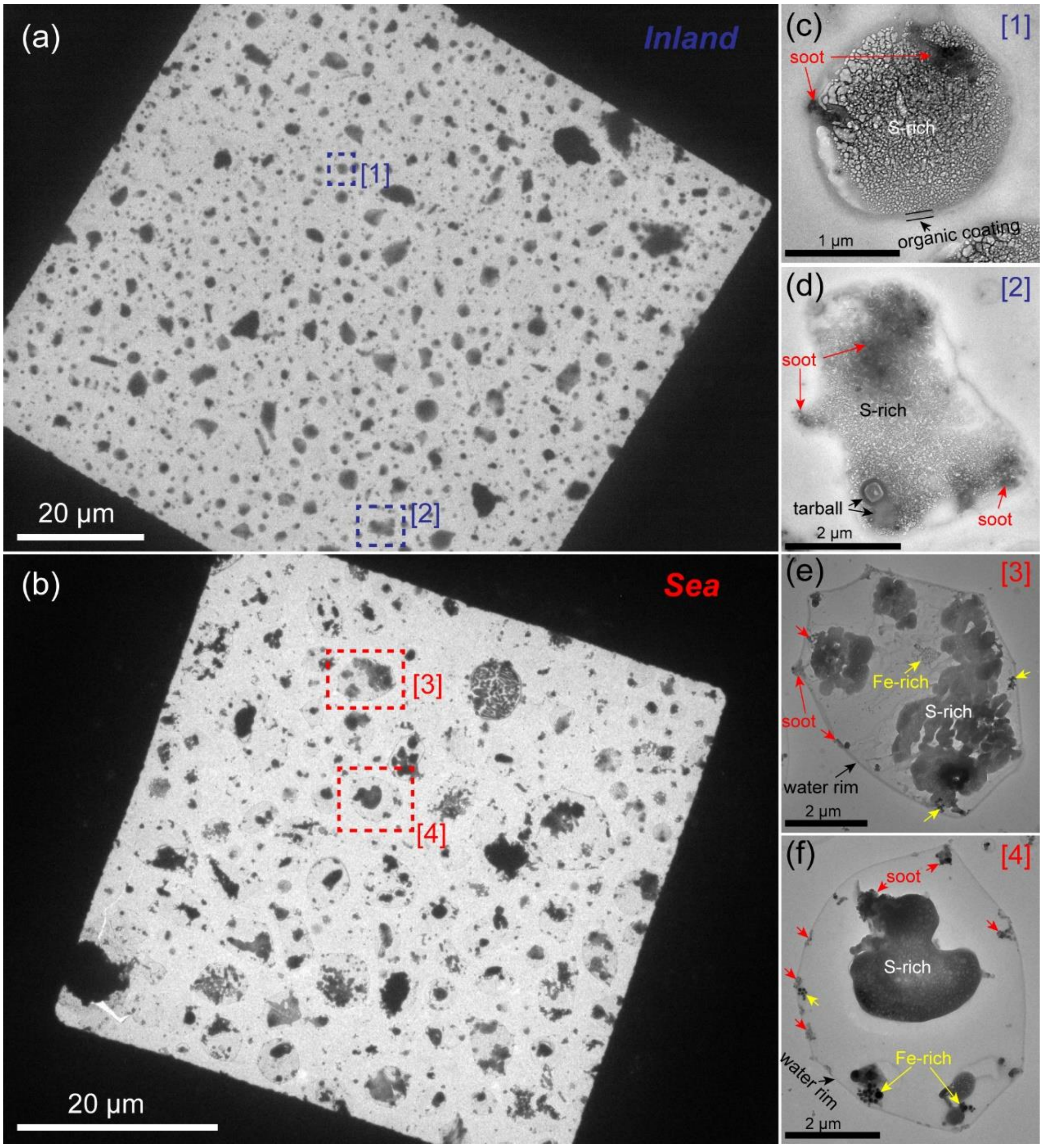

Figure 8. Low magnification TEM images of soot-containing particles in the YRD during two transboundary transport. (a) Soot-containing particles transported through the inland pathway. (b) Soot-containing particles transported through the sea pathway. (c-d) Magnified TEM images for soot-containing particles in panel (a). (e-f) Magnified TEM images for soot-containing particles in panel (b).

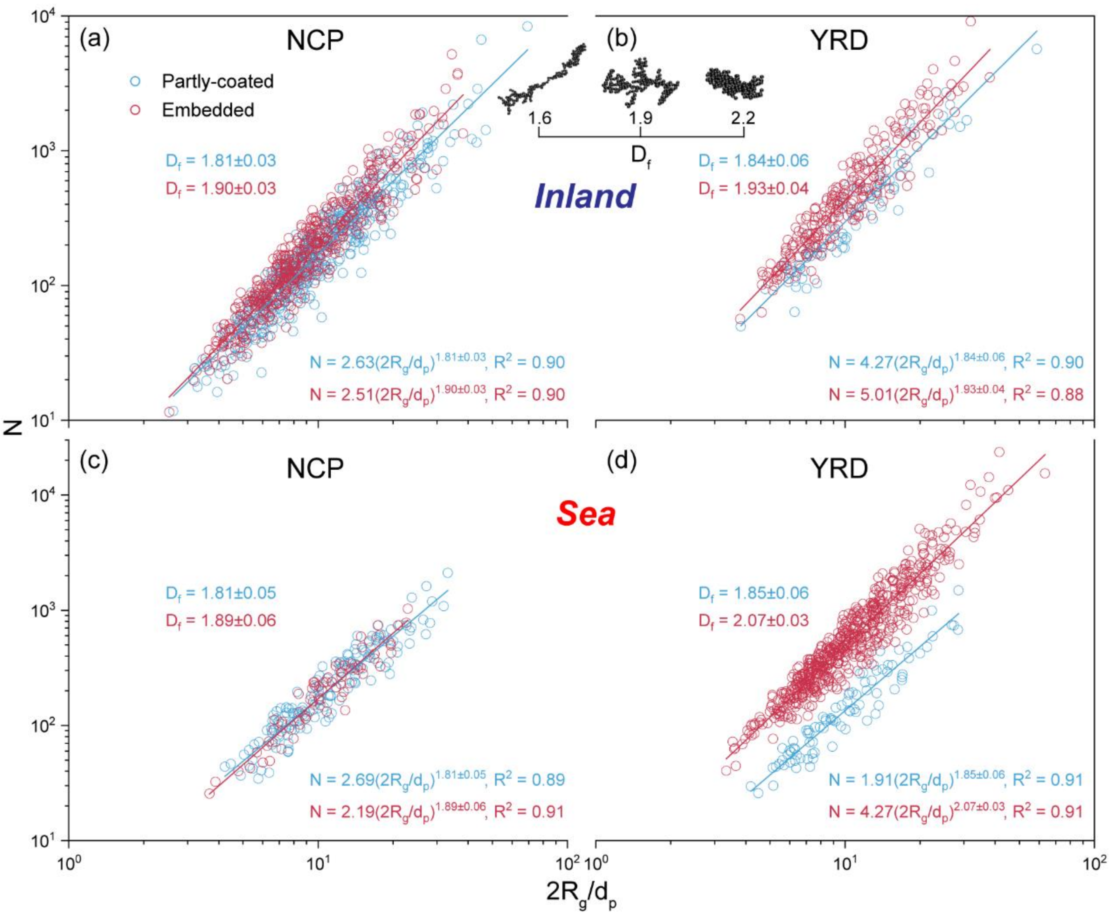

Figure 9. Variation in the fractal dimension (D_f) of partly-coated and embedded soot particles during their transboundary transport from the NCP to the YRD. (a-b) D_f of soot particles transported through the inland pathway. (c-d) D_f of soot particles transported through the sea pathway. A schematic model of the soot D_f is exemplified.

8. Line 193: When calculating Dp/Dc, how are multiple soot cores handled. Is ESDsoot the sum of all soot cores?

Reply: When we calculated the D_p/D_c of soot-containing particles with multiple soot cores, the total volume of soot cores was first computed. Then, based on the total volume, we calculated the diameter of the soot core and the D_p/D_c of soot-containing particles with multiple soot cores.

9. Line 278: It would be helpful for context to include an average (or range of) transport time based on the back trajectories.

Reply: We added the simulated time for backward trajectories.

P11 L285-287: "To determine whether the transport pathway of pollutants was consistent with the wind field, the $PM_{2.5}$ transport pathway was simulated based on the 72 hr CWT analysis (Figure 2)."

10. Line 98: "…exerting favorable effects on global warming in the atmosphere". I would recommend changing from "favorable" to "positive radiative forcing" or similar.

Reply: We revised it as follows.

P4 L98-99: "They exert positive radiative forcing effects on global warming in the atmosphere (Cappa et al., 2012; Jacobson, 2001)."

11. Line 293: "These results suggest that massive primary and secondary aerosols including EC (i.e., soot) were transported from the NCP to the YRD under cold fronts, both through the inland and the sea pathways". The word "massive" here is confusing. I would recommend changing it unless the authors are referring to the size of the particles.

Reply: Thanks, we revised this word.

P12 L303-305: "These results suggest that many primary and secondary aerosols including EC were transported from the NCP to the YRD under cold fronts, both through the inland and the sea pathways."

References

Beeler, P., Kumar, J., Schwarz, J. P., Adachi, K., Fierce, L., Perring, A. E., Katich, J. M., and Chakrabarty, R. K.: Light absorption enhancement of black carbon in a pyrocumulonimbus cloud, Nat. Commun., 15, 6243, https://doi.org/10.1038/s41467-024-50070-0, 2024.

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