Reply to Reviewer #1

This is an interesting work, based on the observation of different patterns in aerosols associated with different wind trajectories. However, some clarifications are needed. Section 3.4 about optical absorption should be revised. Although English is good in general, grammar should be revised. Notation is not always defined or is not self-consistent. Structure (main manuscript/SM) should also be revised.

We are grateful for this reviewer's comments. These comments are all valuable and helpful for improving our paper.

As the reviewer's comments, we moved some descriptions into the Supplementary Materials (Text S1). Moreover, we answered the comments carefully and made corrections in the submitted manuscript and supplementary information. 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. Segregation of information into the Supplementary Material should be done when this information is not essential for understanding the study. In this case, the information in the SM is essential to follow the main manuscript. Authors are suggested to reorganize the information.

Reply: We appreciated the reviewer's comments and moved some descriptions into the Supplementary Materials (Text S1).

P11 L264-267: "Based on the association between $PM_{2.5}$ concentrations and prevailing winds described in Text S1, we inferred that there was a typical transboundary transport process of pollutants from the NCP to the YRD on December 30-31, 2017 and December 7-8, 2020, respectively."

2. Introduction: This reviewer does not agree with some conventions often used in environmental articles, such as the equivalence between black carbon and elemental carbon (BC is a carbonaceous combustion-derived aerosol, while elemental carbon is the major chemical component of BC, but also of any organic

material), or the list of soot sources (why fossil fuels and biomass??; any liquid biofuel, electrofuel, or non-biologic waste material will also emit soot when burned).

Reply: Thanks for the reviewer's comments and we considered them carefully. We revised this sentence as follows.

P4 L96-98: "Soot particles dominated by black carbon, mainly emitted from incomplete burning of fossil, biomass, and other non-biological fuels, are important light absorbing aerosols in fine particles (Bond et al., 2013)."

3. Introduction: This reviewer does not agree that "simulating soot climate effect is readily achievable in models", as stated. The variety of sizes, shapes, compositions, and nanostructures affect the optical properties of soot and makes the simulation very difficult.

Reply: Thanks. To eliminate the misunderstanding, we revised this sentence as follows.

P5 L115-118: "When simulating soot climate effect in models, the complicated microphysical properties of soot particles could be underestimated due to limited studies, thereby introducing considerable uncertainties into the results (Chen et al., 2025; IPCC, 2021)."

4. Section 2.2: If OM/OC ratios in Chinese cities is assumed as 1.91, what organic matter is the remaining 0.91/1.91? Why

Reply: In this study, the OM/OC mass ratio is assumed as 1.91. OC (organic carbon) refers to the total carbon mass in organic compounds. OM (organic matter) refers to the organic compound mass. Thus, the remaining 0.91 refers to the total mass of other elements (e.g., O, H, N, and S) in organic compounds.

We revised this sentence as follows.

P7 L168-170: "Considering the contribution of other elements (e.g., O, H, N, and S) to the mass of organic matter (OM, i.e., organic compound), OM

concentrations were obtained by multiplying organic carbon (OC) concentrations by 1.91 reported by Xing et al. (2013).”

5. Sections 2.2 and 3.2: TEM operates under high vacuum. Therefore, evaporation or sublimation of coatings could occur even in “conventional TEM observations”. What is the change in the beam power to distinguish between “enhanced electron beam observations” and “conventional observations”, and thus between enhanced and conventional absorption? Could authors include TEM images of the same particle before and after enhancing the beam power? Visible bubbles observed in Figures 3 and 4, indicating evaporation, are declared to correspond after enhancing power, but how do these particles look like before? Are diameters “Dp” those obtained with TEM under conventional mode and “Dc” those obtained under enhanced electron beam? Please clarify.

Reply: As the reviewer commented, some coatings could be sublimated under conventional TEM observations. However, the sublimation rate is relatively slow because these coatings usually consist of sulfate and nitrate mixtures, which maintain their original morphology longer than pure ammonium nitrate. Moreover, we used a weak electron beam during conventional TEM observations, which minimized damage to particles and mitigated their sublimation. We also obtained TEM images within five seconds.

To present the morphology of the indiscernible soot more clearly, we enhanced the electron beam via adjusting the beam spot size to accelerate the sublimation of non-refractory coatings. Figure 3c shows TEM images of the same particle before and after exposure to the enhanced electron beam. The TEM images in Figures 3 and 4 were all obtained before the electron beam was increased, except for the sublimated one. The bubbles in some coatings are obvious, which may be related to their chemical compositions and the imaging duration. In this study, only particles containing indiscernible soot were re-imaged under high electron beam conditions. The

D_p and discernible soot D_c were measured under conventional TEM observations, while the indiscernible soot D_c values were measured under TEM observations with enhanced electron beam.

P7 L181-182: "These parameters for indiscernible soot particles were measured under TEM observations with the enhanced electron beam."

P13-14 L348-350: "To observe some indiscernible embedded soot particles more clearly, their non-refractory coatings (e.g., S-rich particles) were sublimed under an enhanced electron beam (Figure 3c)."

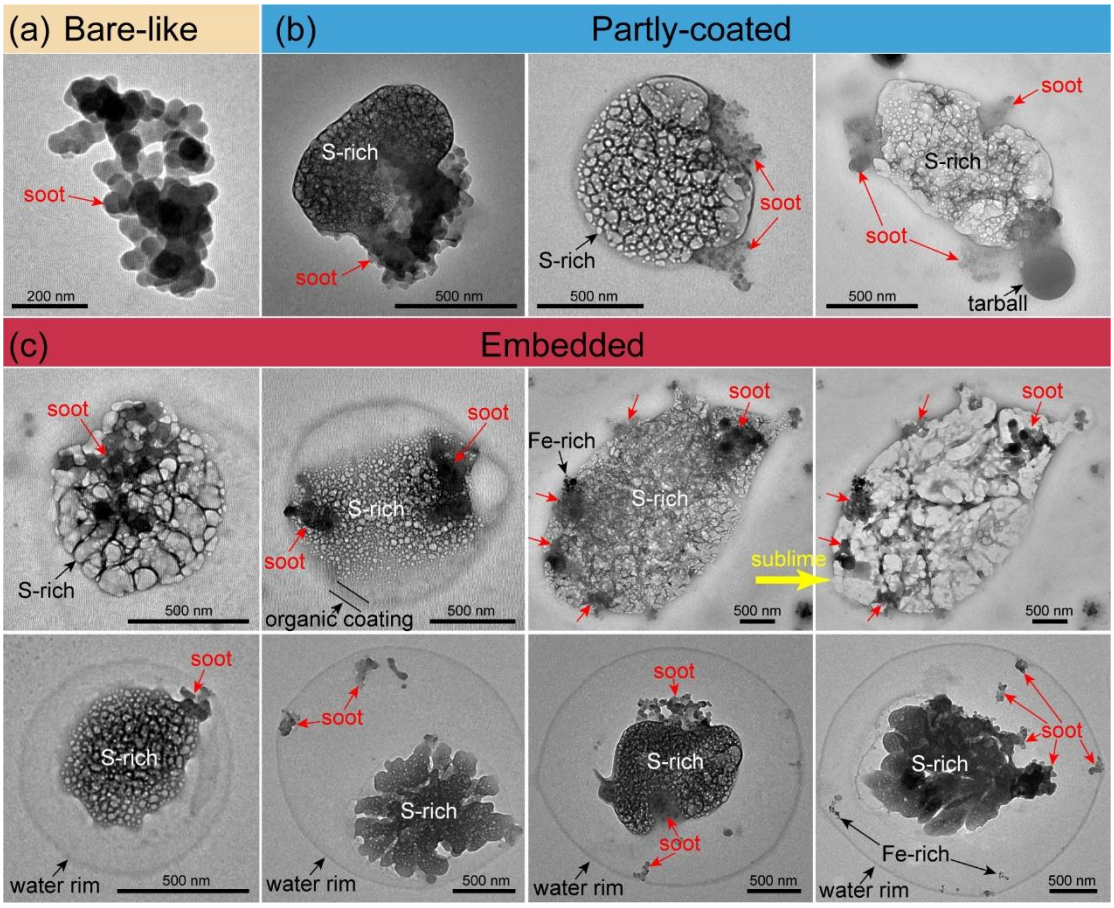

Figure 3. Typical transmission electron microscopy (TEM) images of soot particles in different mixing structures. (a) Bare-like soot particle. (b) Partly-coated soot particles. (c) Embedded soot particles. Some indiscernible embedded soot particles in panel (c) can be clearly observed after their coatings are sublimed under an enhanced electron beam.

6. Sections 2.2 and 3.2: What do authors exactly mean by "mixing states" of soot

particles? Is it an appropriate name? Based on Text S1, it seems that they refer to chemical composition. However, based on Section 3.2 and Figures 3 and 4, it seems that they refer to bare-like, partly-coated or embedded. Please correct or clarify.

Reply: The "mixing state" refers to one type of aerosol particles (e.g., soot) mixing with other aerosols (e.g., S-rich particles). Mixing structure represents morphological mixing between two different types of aerosol particles, including bare-like, partly-coated, and embedded. To emphasize the mixing characteristics of soot with other aerosols, we modified "mixing states of soot particles" to "mixing structures of soot particles".

P7 L175-177: "To better observe soot mixing structures and measure soot geometrical parameters, we enhanced the electron beam to sublime non-refractory coatings of indiscernible soot cores after conventional TEM observations."

P13 L338-339: "The morphology and mixing structures of soot particles can be changed during transport due to atmospheric aging (Li et al., 2024)."

7. Section 2.2: Equations 4 to 7 are written without a brief explanation of their meaning. Authors should at least explain that ignoring the overlap (or sintering or interpenetration) between monomers would lead to underestimation of the fractal prefactor of the power-law relationship (eq. 4). Moreover, publications after 1997 have demonstrated that also this prefactor (not only that in eq. 5) is highly affected by the overlap parameter (see, e.g., Powder Technology 271, 141–154 (2015)).

Reply: Thanks. We added some explanations for equations 4 to 7.

P8 L207-208: "The monomer number in soot particles and the gyration radius of soot particles can be calculated using the following equations."

P9 L212-213: "Because the fractal prefactor is highly affected by the overlap between soot monomers (Lapuerta et al., 2015), an overlap parameter needs to be considered."

8. Section 2.3: Parameters "n" and "W" in equation 9 are not defined. Please check uniformity in the notation.

Reply: We added the definition of parameters "n" and "W" in equation 9.

P10 L238-239: "where W is weighting factor; n is the number of all trajectory endpoints in a grid cell; log(n+1) represents the density of trajectories."

9. Section 3.1: In Figure S4 the content in EC (supposedly associated with soot) is very minor (purple). On the contrary, in Figure S5, the percent of soot-containing particles is very high (light blue). How do these results match?

Reply: As shown in Figure 7, the average D_p/D_c value of soot-containing particles is ~3. Based the D_p/D_c value, we can calculate the volume ratio of soot-containing particles to soot cores at ~27. Moreover, soot sizes are mainly lower than 200 nm (Figures 5 and S8). These suggest that the total volume of soot is very low. As we known, the soot density is also low. Thus, the total mass of soot is very minor although the soot number is large. These results derived from individual particle analysis and bulk analysis are matched.

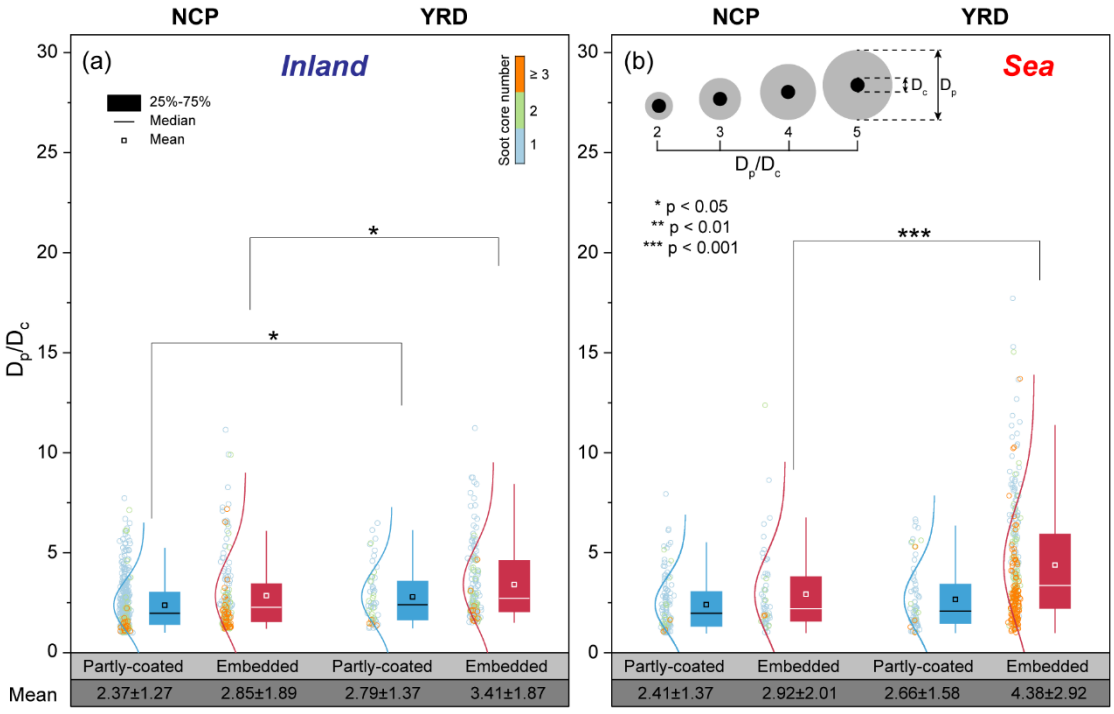

Figure 7. The size ratio of soot-containing particles to their soot cores (D_p/D_c) in two types of transboundary transport models from the NCP to the YRD. (a) D_p/D_c ratios of soot-containing particles transported through the inland pathway. (b) D_p/D_c ratios of soot-containing particles transported 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.

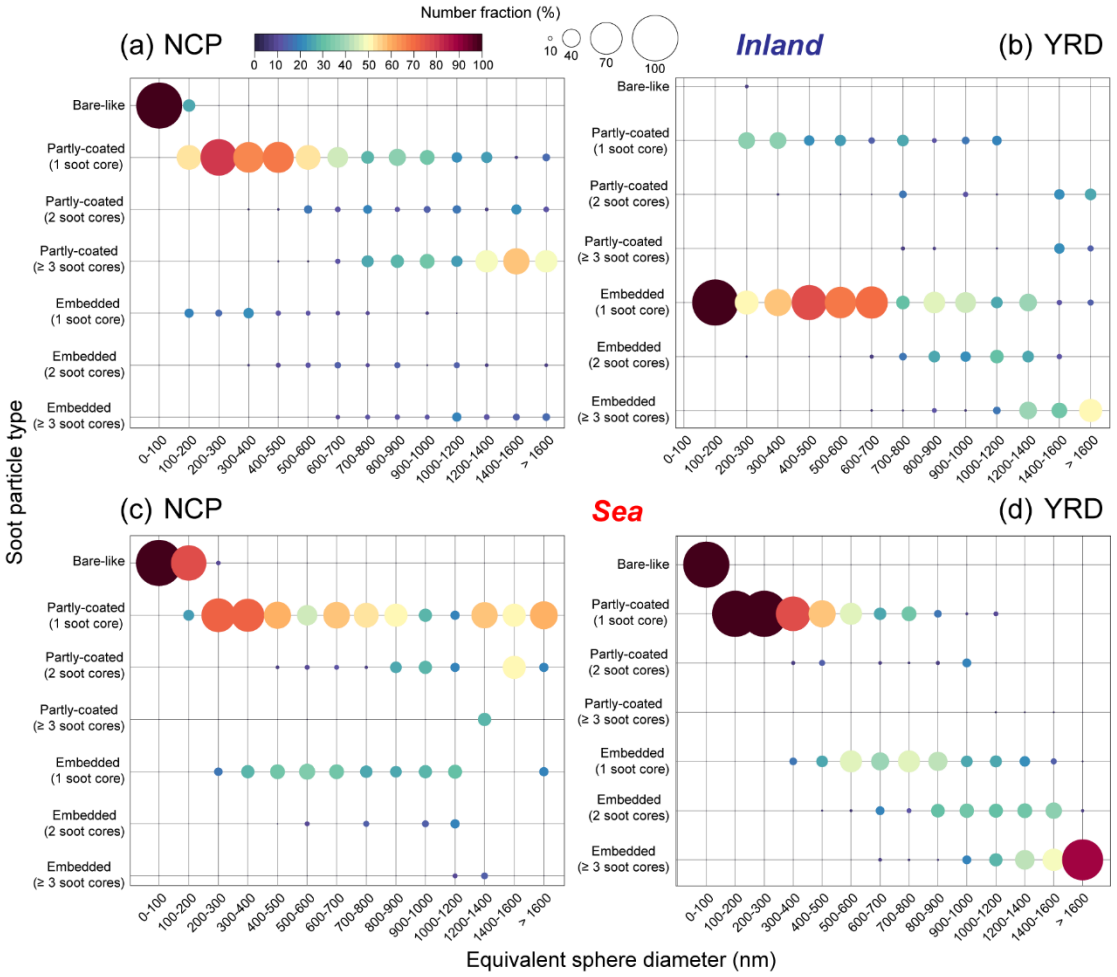

Figure 5. Number fractions of soot-containing particles with different mixing structures and numbers of soot cores in different size bins in two types of transboundary transport models from the NCP to the YRD. (a-b) Soot-containing particles transported through the inland pathway. (c-d) Soot-containing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

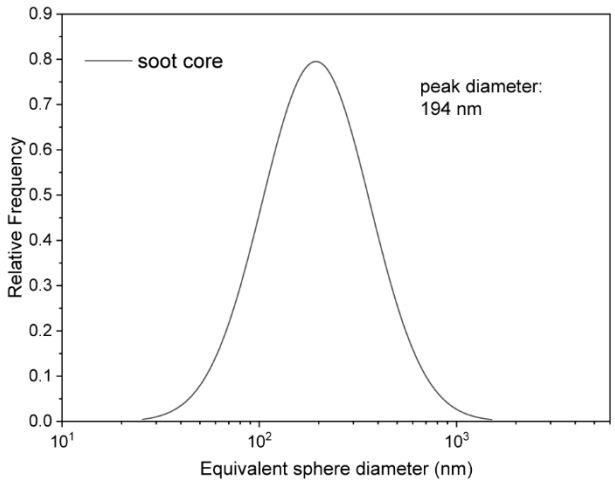

Figure S8. Size distribution of soot cores in individual soot-containing particles.

10. Section 3.3: What is, in the opinion of authors, the dominant reason for the increase in the size, the number of soot cores, the Dp/Dc ratio, and the fractal dimension of soot structures: coalescence between agglomerates (entrainment) or breakage of agglomerates inside the aerosol (collapse)?

Reply: Based on the results and discussion in section 3.3, heterogeneous aging processes and cloud aging processes of soot-containing particles mainly lead to the increases in their sizes, D_p/D_c ratios, and D_f during the transport through the inland pathway and the sea pathway, respectively. Only cloud aging processes can cause the increase in soot core numbers. Also, one recent study observed the similar phenomenon (Chen et al., 2025). No matter what kind of aging process, we believe that the D_f increase is caused by the structural collapse of soot cores.

11. Section 3.4: This reviewer can understand that the energy adsorbed is reduced for the sea pathway with respect to the inland pathway, and even that multiple cores may also contribute to reduce absorption. But does not understand why the energy absorbed from embedded particles is higher than that absorbed from soot cores (Figure 10a). The refractive index of soot is much higher than that of coatings (and specially its imaginary part, related to attenuation of light). Consequently, the ageing process should lead to a decrease in the energy

absorbed. Please, revise, or explain better.

Reply: Thanks for the reviewer's comments. The higher light absorption of embedded particles compared to soot cores can be attributed to the lensing effect. The lensing effect refers to that when soot particles age in the atmosphere and form a core-shell structure, their coatings (e.g., sulfates and nitrates) refract and focus light like a lens, enhancing the absorption capacity of soot particles to solar radiation (Cappa et al., 2012; Fierce et al., 2020). At present, many studies have found the lensing effect of soot particles (Liu et al., 2015; Riemer et al., 2019; Wang et al., 2025). We added some explanations for the lensing effect.

P20 L533-536: "Figure 10a shows the change in the E_{abs} of soot-containing particles following their aging from bare-like to partly-coated, and then to embedded states. Compared to soot cores, partly-coated and embedded soot-containing particles present higher E_{abs} (Figure 10a), due to the lensing effect (Fierce et al., 2020; Wang et al., 2025)."

12. Technical corrections: please correct "If the high-pressure system located" to "If the high-pressure system is located"; "Obviously, there was a bench of data" to "Obviously, there is a bench of data"; "However, transboundary haze pollutants crossed the East China Sea remain unexplored" to "However, transboundary haze pollutants crossing the East China Sea remain unexplored".

Reply: Thanks. We revised them.

P4 L82-85: "If the high-pressure system is located in the interior of the NCP, heavy haze covering the Jing-Jin-Ji region (i.e., Beijing, Tianjin, and Hebei) could move out from inland China to the East China Sea and return into the inland region under prevailing winds, influencing air quality of the YRD (see section 3.1)."

P4 L90-91: "Obviously, there is a bench of data available from national ground monitoring net station of air quality to support the measurements and modelling studies."

P4 L92-93: “However, transboundary haze pollutants crossing the East China Sea remain unexplored.”

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