



1 **Tracing the evolution of morphology and mixing state of soot**
2 **particles along with the movement of an Asian dust storm**

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23 **Abstract**

24 Tracing the aging progress of soot particles during transport is highly challenging. An
25 Asian dust event could provide an ideal opportunity to trace the continuous aging
26 progress of long-range transported soot particles. Here, we collected individual aerosol
27 particles at an inland urban site (T1) and a coastal urban site (T2) in China and a coastal
28 site (T3) in southwestern Japan during an Asian dust event. Microscopic analysis
29 showed that the number fraction of soot-bearing particles increased from 19% to 22%
30 from T1 to T2 in China but surprisingly increased to 56% at T3 in Japan. The dominant
31 fresh soot (71%) at T1 became partially embedded (70%) at T2 and fully embedded
32 (84%) at T3. These results indicated that the soot particles had lower deposition than
33 other aerosol types and became more aged from T1 to T3. The fractal dimension of the
34 soot particles slightly changed from 1.74 at T1 and 1.78 at T2 but significantly became
35 1.91 at T3. We found that the soot morphology compressed depending on secondary
36 coating thickness and relative humidity. Moreover, we observed a unique mixing
37 structure at T3 that tiny soot particles were seemingly broken from large ones cross the
38 East China Sea and distributed in organic coatings instead of sulfate core in particles.
39 Our study provide important constraints of the morphological effects to better
40 understand changes of microscopic structures of soot. These new findings will be
41 helpful to improve optical calculation and modeling of soot particles and their regional
42 climate effects in the atmosphere.



43 **1. Introduction**

44 Soot (i.e., black carbon (BC)) is a type of carbonaceous material with graphitic
45 structures emitted from the incomplete combustion of fossil fuels and biomass. Soot
46 particles exhibit a chain-like aggregation morphology with a diameter of 10 nm to 100
47 nm (Buseck et al., 2014). Because of its strong capacity to absorb solar radiation, soot
48 is considered the second greatest contributor to global warming after carbon dioxide
49 (IPCC., 2013; Bond et al., 2013). Soot is an important particulate pollutant in fine
50 particles (i.e., PM_{2.5}) in urban polluted air, which adversely affects the respiratory health
51 of citizens and induces generally unwanted heating in the planetary boundary layer
52 (West et al., 2016; Ding et al., 2016).

53 Fresh soot particles are hydrophobic but are converted into a hydrophilic state
54 following their aging through physical and chemical processes (Li et al., 2016b; Riemer
55 et al., 2010; Perring et al., 2017). Aged soot particles containing secondary coating
56 aerosols (e.g., ammonium sulfate, ammonium nitrate, and organic matter) can be
57 activated as cloud condensation nuclei (CCN) (Zhang et al., 2008; Wang et al.,
58 2010; Ding et al., 2019; Shiraiwa et al., 2007; Lee et al., 2019). These coatings can
59 significantly change the optical scattering and absorption capacity of soot particles (Liu
60 et al., 2017; Moffet and Prather, 2009; Matsui et al., 2018; He et al., 2015; Zhang et al.,
61 2018a). Numerical model simulations have estimated that light absorption by internally
62 mixed soot is enhanced by a factor of 2 over externally mixed soot (Jacobson, 2001).
63 In contrast, Cappa et al. (2012) reported in situ observations of soot absorption
64 enhancement of only 6% in ambient air. This discrepancy between simulation and



65 observation could be attributed to the complex mixing structure and various
66 morphologies of soot particles in the air (Adachi et al., 2016; Li et al., 2016a; Wu et al.,
67 2018).

68 In aged air masses, soot particles tend to be internally mixed with secondary
69 aerosols such as sulfates, nitrates, and secondary organic matter (Li et al., 2016b).
70 Especially in the East Asian region, one of the most polluted areas in the world, soot is
71 internally mixed with secondary aerosols in polluted urban, rural, and remote air
72 (Adachi et al., 2016; Zhang et al., 2013; Yuan et al., 2019; Zhang et al., 2018b). However,
73 most of these studies have focused on the aging and mixing state of soot particles at one
74 or multiple isolated sites. These results have not traced the detailed aging processes
75 (e.g., morphology and mixing structure) from fresh to aged soot particles during their
76 transport.

77 Although great progress has been made in the field of soot aging, it is highly
78 challenging to trace the aging processes of soot particles during transport. Asian dust
79 storms carry both dust and anthropogenic aerosols across East Asia into the North
80 Pacific Ocean (Li et al., 2014; Geng et al., 2014; Zhang et al., 2005). This presents an
81 ideal environment to study the aging processes of soot particles during long-range
82 transport. Compared to previous publications, the present study quantified the variation
83 in mixing structures and fractal dimension of soot particles and further explored how
84 shape of soot particles changed following the dust storm movement from East China to
85 Japan.

86 Using transmission electron microscopy (TEM), we investigated the morphology,



87 mixing structure, relative abundance, and size distribution of individual soot particles.
88 Furthermore, we evaluated the morphological differences of individual soot particles at
89 three sampling sites. Finally, a conceptual model was proposed to better understand the
90 aging processes of long-rang transported soot particles.

91 **2. Experimental methods**

92 **2.1 Aerosol sampling**

93 Three sampling sites were chosen for aerosol collections: an inland urban site in
94 Jinan city (T1, 36.67°N 117.06°E), China, a coastal urban site in Qingdao city (T2,
95 36.10°N 120.46°E), China, and a coastal rural site at Amakusa (T3, 32.30°N 130.00°E)
96 in southwestern Japan (Figure 1). A dust storm outbreak was observed in East Asia.
97 Detailed information about this dust storm will be discussed in Section 3.1. We
98 collected aerosol particles during dust transport from 18 to 19 March 2014 at the three
99 sampling sites (Figure S1-S5). In total, seven dust samples were collected within 30
100 hours after the dust storm arrival. The details about the sampling dates, times,
101 meteorological conditions, and PM (particulate matter) concentrations for the samples
102 are listed in Table S1.

103 A DKL-2 sampler (Genstar Electronic Technology, China) was used to collect
104 individual aerosol particles on copper TEM grids covered by carbon film (carbon type-
105 B, 300-mesh copper; Tianld Co., China) with an air flow of 1.0 L/min. A single-stage
106 impactor with a 0.5 mm diameter jet nozzle was installed on the sampler. This impactor
107 has a collection efficiency of 100% at an aerodynamic diameter of 0.5 μm with an
108 assumed particle density of 2 g/cm^3 . The sampling duration varied from 1 min to 10



109 min according to the visibility, PM concentration, and particle distribution on the
110 substrate. All samples were placed in sealed, dry plastic capsules and stored in a
111 desiccator at 25 °C and $20 \pm 3\%$ relative humidity (RH) for further analysis.

112 **2.2 Electron microscopic analyses**

113 A JEOL JEM-2100 transmission electron microscope (TEM) operated at 200 kV
114 was used to analyze individual particles. Elemental composition was determined
115 semiquantitatively by using an energy-dispersive X-ray spectrometer (EDS) (Oxford
116 Instruments, UK) that can detect elements heavier than carbon ($Z \geq 6$). The distribution
117 of aerosol particles on TEM grids was not uniform, with coarser particles occurring
118 near the center and finer particles occurring on the periphery (Xu et al., 2019). Therefore,
119 to ensure that the analyzed particles were representative of the entire size range, three
120 areas were chosen from the center to the periphery of the sampling spot on each grid.
121 iTEM software (Olympus Soft Imaging Solutions GmbH, Germany) was used to
122 analyze the TEM images and obtain the projected area, perimeter, aspect ratio, and
123 equivalent circle diameter (ECD) of individual aerosol particles. In total, we analyzed
124 412, 486, and 887 aerosol particles for T1, T2, and T3 site, respectively.

125 **2.3 AFM analysis**

126 Atomic force microscopy (AFM) is an analytical method used for studying the
127 surface structure of solid materials. AFM (Dimension Icon, Germany) can determine
128 the three-dimensional morphology of particles in tapping mode. The AFM settings
129 consisted of imaging forces between 1 and 1.5 nN, scanning rates between 0.5 and 0.8
130 Hz, and a scanning range of 10 μm with a resolution of 512 pixels per length. The



131 bearing areas (A) and bearing volumes (V) of the particles were directly obtained from
132 NanoScope Analysis software. Their equivalent circle diameters (ECDs) and equivalent
133 volume diameters (EVDs) were calculated according to the formulas described by Chi
134 et al. (2015).

135 The correlations of ECDs and EVDs are shown in Figure S6 in the Supporting
136 Information. Therefore, the ECD of individual aerosol particles measured from the
137 iTEM software can be further converted into an EVD based on this correlation.

138 **2.4 Air mass backward trajectories**

139 Forty-eight hour backward trajectories were calculated for the three sites using the
140 NOAA HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) trajectory
141 model (Stein et al., 2015). We selected an altitude of 1500 m as the end point in each
142 backward trajectory.

143 We measured the actual duration from the Beijing-Tianjin-Hebei (BTH) area to T1
144 and T2 according to the backward trajectories in Figure 1. It was approximately 12
145 hours between BTH and T1 and 15 hours between BTH and T2. The interval between
146 T1 and T2 was three hours. The duration between the air mass leaving T2 and reaching
147 T3 was approximately 30 hours.

148 **2.5 Morphological analysis of soot particles**

149 The fractal dimension (D_f) calculated by the scaling law is used to characterize the
150 morphology of soot particles (Koeylue et al., 1995).

$$151 \quad N = k_g \left(\frac{2R_g}{d_p} \right)^{D_f} \quad (1)$$

152 where N is the total number of soot monomers, R_g is the radius of gyration of the soot



153 particle, d_p is the diameter of soot monomer, k_g is the fractal prefactor, and D_f is the
154 mass fractal dimension of an individual soot particle.

155 D_f and k_g in Equation 1 are estimated from a power law fit of a scatter plot of N
156 versus the values of $2R_g/d_p$. N can also be calculated by Equation 2.

$$157 \quad N = k_a \left(\frac{A_a}{A_p} \right)^\alpha \quad (2)$$

158 where A_a is the projected area of the soot particle, A_p is the mean projected area of the
159 soot monomer, k_a is a constant, and α is an empirical projected area exponent.

160 The values of α and k_a in Equation 2 depend on the overlap parameter (δ) calculated
161 using Equation 3 (Oh and Sorensen, 1997).

$$162 \quad \delta = \frac{2a}{l} \quad (3)$$

163 where a is the soot monomer radius and l is the monomer spacing.

164 The radius of gyration of the soot particle R_g is obtained by the simple
165 correlation in Equation 4 developed by Brasil et al. (1999)

$$166 \quad L_{\max}/(2R_g) = 1.50 \pm 0.05 \quad (4)$$

167 where L_{\max} is the maximum length of the soot particle.

168 The values of d_p , A_a , A_p , a , l , and L_{\max} can be directly obtained from TEM images.

169 In addition to D_f , we also used the aspect ratio (AR) to further quantify the
170 roundness of soot particles. The aspect ratio is the maximum ratio between the length
171 and width of a bounding box (Equation 5). An aspect ratio of 1 (the lowest value)
172 indicates that a particle is not elongated in any direction.

$$173 \quad AR = \frac{L_{\max}}{W_{\max}} \quad (5)$$

174 where L_{\max} is the maximum length of a soot particle and W_{\max} is the maximum width



175 of a soot particle.

176 **3. Results and discussion**

177 **3.1 The Asian dust storm event**

178 Figure 2 displays variations in PM₁₀ and PM_{2.5} concentrations before, during, and
179 after the dust storm event at the Jinan, Qingdao, and Amakusa sampling sites. The dust
180 storm air mass started to influence T1 at approximately 14:00 on 03/17 (BJT, Beijing
181 Time, UTC+8). The concentration of PM₁₀ at T1 increased rapidly to a maximum value
182 of 834 µg/m³. The air mass reached T2 at 17:00 on 03/17, and the highest PM₁₀
183 concentration was recorded at 721 µg/m³. After the arrival of a cold front at T2, the air
184 mass continued moving approximately 1000 km to T3 at 17:00 on 03/18. The
185 concentration of PM₁₀ reached 87 µg/m³ at T3 (Figure 2). During this study, the
186 meteorological data (e.g., temperature and air pressure) measured at the three sampling
187 sites also confirm the arrival time of the dust storm (Figures S3-S5). All seven dust
188 samples were collected after the arrival of the dust storm, thus confirming the sampling
189 of the same dust storm event (Figures 1b and S2).

190 Figure 1 indicates that all the air masses during the dust storm event originated
191 from Mongolia, moving southeastward via the BTH area, reaching T1 and T2,
192 respectively, within a 3-hour interval. The BTH, as the largest city cluster in China,
193 contains one of the largest anthropogenic emission sources (e.g., heavy industries, coal-
194 fired power plants, and vehicles) in the world (Li et al., 2016b). The ground PM and
195 meteorological measurements at the three sampling sites (Figure 2 and S3-S5) coupled
196 with air mass back trajectories (Figure 1) and a dust storm simulation in East Asia
197 (Figure S1) together verified that the dust storm event, under the force of a strong cold
198 front, transported across the large BTH city cluster to the downwind area. Therefore,



199 this dust storm movement provides a unique opportunity to study particles in the same
200 air mass and thus trace physical and chemical changes in aerosol particles.

201 **3.2 Classification and mixing state of soot-bearing particles**

202 Soot particles with a typical chain-like structure can be easily distinguished from
203 other aerosol components (e.g., sulfate, organic, metal, and mineral particles) by their
204 morphology. TEM observation is a convenient way to determine whether soot is
205 associated with other aerosol components (Li et al., 2016b; Laskin et al., 2019). During
206 the dust storm period, 56% of the analyzed particles within a size range of 50 nm to 2.4
207 μm included soot particles at T3, approximately three times higher than those at T1
208 (19%) and T2 (22%). This high percentage of internally mixed soot particles was also
209 shown by Ueda et al. (2016) in an Asian outflow at Noto Peninsula, Japan, based on
210 single-particle soot photometer (SP2) analyses. Our results show that the dust storm
211 event not only carried large amounts of dust particles from the Gobi Desert in
212 northwestern China but that this dust-laden air mass also incorporated many soot
213 particles from polluted East Asia (Figure 2 and Figure 3a-d). This is consistent with Pan
214 et al. (2015), who showed that dust storms in East Asia contain and transport
215 anthropogenic pollutants from urban areas.

216 Based on the mixing structures between soot and sulfate on the substrates, three
217 groups of soot particles were defined in this study: fresh, partially embedded and fully
218 embedded (Figure 3).

219 *Fresh soot.* The soot particles were not obviously mixed with secondary aerosol
220 components (Figure 3a). Although surfaces of the fresh soot particles could contain
221 minor organic matter, the organic film was insufficient to change soot morphology and
222 optical properties (Buseck et al., 2014).



223 *Partially embedded soot.* Part of the soot particle was coated by secondary aerosols
224 (Figure 3b).

225 *Fully embedded soot.* The entire soot particle was encapsulated by secondary
226 aerosols (Figure 3c). We also noticed that some soot particles were only embedded in
227 the organic coating instead of the sulfur-rich core (Figure 3d).

228 TEM images show that the fully embedded soot particles with a clear rim on the
229 substrate displayed a droplet-like shape (Figure 3c-d), suggesting that these secondary
230 particles were in an aqueous phase in ambient air (Li et al., 2016b).

231 Based on the three mixing structures of soot particles, we further obtained their
232 relative abundance at the three sampling sites (Figure 4). Seventy-one percent of soot-
233 bearing particles were fresh at T1, decreasing to 16% at T2. In contrast, partially
234 embedded soot increased from 14% at T1 to 70% at T2 when the cold front moved from
235 T1 to T2. It should be noted that fresh soot disappeared at T3 after crossing the East
236 China Sea, and the fully embedded soot dominated soot-bearing particles (84%).

237 Following the dust storm movement, we found that the number fraction of total
238 soot-bearing particles increased to 56% among all the analyzed particles from T1 to T3,
239 suggesting that soot particles had lower deposition than other aerosol types in the cold
240 front. Indeed, soot particles normally have smaller sizes and densities than mineral dust,
241 metal, sulfate, and nitrate particles (Peng et al., 2017), suggesting that soot particles can
242 be transported over longer distances during Asian dust storms.

243 **3.3 Quantifying the morphology of soot particles**

244 The fractal dimension (D_f) of soot particles is a key parameter used to reflect soot



245 morphological structure; e.g., compact soot particles usually have larger D_f than lacy
246 aggregates (China et al., 2015; Wang et al., 2017; China et al., 2013). Therefore, D_f can
247 be used to understand soot aging processes in the atmosphere. Figure 5a shows that the
248 D_f sequence of soot particles is T1 (1.74 ± 0.10) < T2 (1.78 ± 0.16) < T3 (1.91 ± 0.04).
249 The D_f of soot particles at T1 and T2 (1.74 ± 0.10 and 1.78 ± 0.16) is much closer to
250 the values of soot emitted from sources, such as the D_f from biomass burning in the
251 range of 1.68–1.74 (Chakrabarty et al., 2006) and the D_f from diesel burning in the
252 range of 1.56–1.68 (Wentzel et al., 2003). The D_f of soot particles at T3 (1.91 ± 0.04)
253 is similar to that of aged soot (1.81–1.90) in remote marine air (China et al., 2015) and
254 polluted air in North China (Wang et al., 2017).

255 At the three sampling sites, the highest D_f value at T3 suggests a more compacted
256 structure of the soot particles. Moreover, we obtained the aspect ratios of soot particles
257 at the three sampling sites, which can indicate the roundness of the particle shape (Yuan
258 et al., 2019). The average aspect ratio of soot particles at T3 was 1.56, much lower than
259 1.72 at T1 and 1.66 at T2 (Figure 5b). These two parameters show that the soot
260 morphology became more compact and had a rounder shape following the dust storm
261 movement.

262 **3.4 Soot-bearing particle size growth following soot aging**

263 The average ratio (D_p/D_{core}) of the diameter of the internally mixed particle (D_p) to
264 its corresponding soot core (D_{core}) during the dust storm period was 1.42 at T1, 1.78 at
265 T2, and 2.49 at T3 (Figure 5b). The D_p/D_{core} values in this study are much higher than
266 the reported values in fresh emissions (e.g., average value 1.24 for fossil fuel (Sahu et
267 al., 2012)) but close to ~2.0 in aged aerosols in background and polluted air (Dahlkötter



268 et al., 2014; Raatikainen et al., 2015; Metcalf et al., 2012). Recently, Peng et al. (2017)
269 reported a high growth rate in urban Beijing and a derived average D_p/D_{core} value of
270 1.97 (1.34-2.61). The D_p/D_{core} value in urban Beijing air is much higher than our
271 reported values of 1.42-1.78 at T1 and T2 during the dust storm period. This is
272 understandable considering the weak secondary aerosol formation in the dust storm in
273 the continental air as a result of acidic gases being scavenged by the large amounts of
274 mineral dust particles (Li et al., 2016b).

275 Based on the air mass backward trajectories, we can infer that it took approximately
276 three hours for the cold front to move between T1 and T2 and 30 hours from T2 to T3
277 (Figure 1). Here, we calculated the coating volume of aged soot particles based on the
278 values of D_p and D_{core} of individual particles and found a 152% increase in the coating
279 volume from T1 to T2 and a 609% increase from T2 to T3.

280 **3.5 Aging mechanism of soot particles**

281 We noticed that the partially embedded soot particles significantly increased from
282 14% at T1 to 70% at T2 (Figure 4), indicating that the fresh soot particles aged during
283 the dust storm movement from the inland to the coastal area. However, we found that
284 the D_f value at T1 only slightly changed from 1.74 at T1 to 1.78 at T2. These results
285 indicate that the morphological structures of soot particles underwent slight changes,
286 although large amounts of fresh soot converted into partially embedded soot particles
287 from T1 to T2.

288 Figure 4 shows that the fresh soot particles disappeared at T3, and the number
289 fraction of fully embedded soot particles increased to 84%. Moreover, the D_f of soot
290 particles had a large change from 1.78 at T2 to 1.91 at T3, which suggests that the
291 morphology structure of soot particles changed from chain-like to compact when the
292 air masses crossed the East China Sea (Figure 5a). This large change in soot



293 morphology from T2 to T3 is different from the slight change in soot particles from T1
294 to T2. The contrasting results suggest that soot particles underwent more complicated
295 aging processes in marine air than in continental air.

296 Secondary aerosol formation on soot particles can significantly change their fractal
297 morphology into a compact shape (China et al., 2015; Wang et al., 2017; Ma et al.,
298 2013; Pei et al., 2018). The thick coating of soot particles occurred when air masses
299 crossed the East China Sea (Figure 5b), suggesting that secondary aerosol coating
300 formation can significantly compress the fractal morphology of soot particles. Recently,
301 Yuan et al. (2019) further found that the phase change of secondary aerosols (due to RH
302 variation) in aged soot particles could further compress the fractal shape of soot
303 aggregates. The high humidity in marine air (T2 to T3) should lead to phase changes of
304 secondary aerosols and further cause the morphological compactness of soot
305 aggregations. These two reasons can explain why the large change in soot fractal
306 dimension occurred from T2 to T3 instead of T1 to T2 (Figure 5a).

307 TEM observations present a particular mixing structure of the fully embedded soot
308 at T3: organic coating instead of sulfate contains several typical soot particles, and the
309 organic coating spreads on the substrate (named droplet-like particles (O'Brien et al.,
310 2015; Li et al., 2011)) (Figure 3d). The droplet-like coating morphology of soot can
311 reflect that these secondary particles were in an aqueous phase at T3 in the air. A
312 previous study has shown that secondary aerosol particles begin to acquire aqueous
313 shells at RH 60% (Sun et al., 2018). Once secondary aerosols change from a solid to
314 liquid phase following an RH increase in marine air, soot particles tend to adhere to the
315 liquid phase through coagulation (Li et al., 2016b). Figure 3d shows the phase
316 separation of the organic coating and sulfate core on the substrate under the
317 phenomenon of liquid-liquid phase separations (You et al., 2012). Recently,



318 Brunamonti et al. (2015) found that soot particles tend to redistribute into the organic
319 coating during liquid-liquid phase separation. Therefore, the soot distribution in the
320 organic coating indicates that aerosol particles in the air mass at T3 underwent an
321 aqueous aging process over the East China Sea, which is different from the continental
322 aerosol particles at T1 and T2. It must be noted that several tiny soot particles were
323 distributed in the organic coating at T3 (Figure 3d), which did not occur at T1 and T2.
324 Our findings suggest that the complex cloud-aqueous process of individual particles in
325 marine air could result in scattered soot particles.

326 Tracing the soot particles during the dust storm, we can clarify that the morphology
327 change of soot particles depends on the secondary coating thickness and relative
328 humidity in the air. Moreover, the cloud-aqueous process and the phase separation of
329 organic and sulfate components in the soot-bearing particles likely break the chain-like
330 soot and change soot distribution within individual secondary aerosol particles. The
331 microscopic changes between soot and coating could change their optical absorption,
332 which is different from the core-shell absorption (He et al., 2015). Our study proposes
333 that BC-related optical models should not only consider the mixing state of soot
334 particles but also incorporate the morphological structure of soot particles in different
335 environmental air.

336 Based on the results and discussion above, we propose a conceptual model to
337 summarize the evolution of morphology and mixing state of soot particles along with
338 the movement of an Asian dust storm (Figure 6). Dust storms in East Asia could carry
339 soot and other anthropogenic pollutants from urban areas to downwind areas. During
340 the transport, the dominated mixing structure of individual soot particles changed from
341 fresh to partially embedded and finally to fully embedded. Meanwhile, the chain-like
342 soot compressed and had a rounder shape depending on secondary coating thickness



343 and relative humidity.

344

345 **4. Conclusions**

346 Individual aerosol particles were collected from 18 to 19 March 2014 during an
347 Asian dust storm event. Three sampling sites along with the pathway of the dust storm
348 were chosen to study soot aging, including an inland urban site in Jinan city, China (T1),
349 a coastal urban site in Qingdao city, China (T2), and a coastal rural site at Amakusa in
350 southwestern Japan (T3). Soot-bearing particles were classified into three types: fresh,
351 partially embedded, and fully embedded. There was a noticeable difference in the
352 mixing structure of soot particles during long-range transport, with 71% fresh soot in
353 the analyzed soot particles (by number) at T1, 70% partially embedded soot at T2, and
354 84% fully embedded soot at T3. The fractal dimension (D_f) of soot particles at T3 (1.91)
355 was higher than that at the other two sites (1.74 and 1.78), suggesting that soot particles
356 converted from chain-like to compact shapes during long-range transport. This study
357 showed that an increasing number of soot particles were internally mixed with
358 secondary aerosol particles and significantly aged during transport. The average ratio
359 of D_p/D_{core} during the dust storm period was 1.42 at T1, 1.78 at T2, and 2.49 at T3,
360 indicating increasing coating thickness. By comparing the soot fractal dimension in
361 continental air and marine air, we found that secondary coating thickness and relative
362 humidity both can significantly change the fractal morphology of soot particles in the
363 air. Individual particle analysis showed that several tiny soot particles only observed in
364 organic coatings instead of sulfate in individual soot-bearing particles at T3, suggesting



365 that the complicated aging processes of individual particles can break the chain-like
366 soot formation.

367

368 **Data availability**

369 All data presented in this paper are available upon request from the corresponding
370 author (liweijun@zju.edu.cn).

371 **Supporting information**

372 Table S1 and Figures S1-S6

373 **Author contributions**

374 LX and WL conceived the study and wrote the manuscript. The field campaign was
375 organized and supervised by WL and DZ. SF, KM, AN, and TK collected aerosol
376 particles. LX, SS, LL, YW, HN, and ZS contributed sample and data analyses. All
377 authors reviewed and commented on the paper.

378 **Competing interests**

379 The authors declare that they have no conflict of interest.

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385

386 **References**

387 Adachi, K., Moteki, N., Kondo, Y., and Igarashi, Y.: Mixing states of light-absorbing particles measured
388 using a transmission electron microscope and a single-particle soot photometer in Tokyo, Japan, J.



- 389 Geophys. Res.: Atmos., 121, 9153-9164, <https://doi.org/10.1002/2016JD025153>, 2016.
- 390 Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Bernsten, T., DeAngelo, B. J., Flanner, M. G.,
391 Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G.,
392 Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K.,
393 Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T.,
394 Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific
395 assessment, *J. Geophys. Res.: Atmos.*, 118, 5380-5552, <https://doi.org/10.1002/jgrd.50171>, 2013.
- 396 Brasil, A. M., Farias, T. L., and Carvalho, M. G.: A recipe for image characterization of fractal-Like
397 aggregates, *J. Aerosol Sci.*, 30, 1379-1389, [https://doi.org/10.1016/S0021-8502\(99\)00026-9](https://doi.org/10.1016/S0021-8502(99)00026-9), 1999.
- 398 Brunamonti, S., Krieger, U. K., Marcolli, C., and Peter, T.: Redistribution of black carbon in aerosol
399 particles undergoing liquid - liquid phase separation, *Geophys. Res. Lett.*, 42, 2532-2539,
400 <https://doi.org/10.1002/2014GL062908>, 2015.
- 401 Buseck, P. R., Adachi, K., Gelencsér, A., Tompa, É., and Pósfai, M.: Ns-Soot: A Material-Based Term for
402 Strongly Light-Absorbing Carbonaceous Particles, *Aerosol Sci. Technol.*, 48, 777-788,
403 <https://doi.org/10.1080/02786826.2014.919374>, 2014.
- 404 Cappa, C. D., Onasch, T. B., Massoli, P., Worsnop, D. R., Bates, T. S., Cross, E. S., Davidovits, P., Hakala,
405 J., Hayden, K. L., Jobson, B. T., Kolesar, K. R., Lack, D. A., Lerner, B. M., Li, S.-M., Mellon, D.,
406 Nuaaman, I., Olfert, J. S., Petäjä, T., Quinn, P. K., Song, C., Subramanian, R., Williams, E. J., and
407 Zaveri, R. A.: Radiative Absorption Enhancements Due to the Mixing State of Atmospheric Black
408 Carbon, *Science*, 337, 1078-1081, <https://doi.org/10.1126/science.1223447>, 2012.
- 409 Chakrabarty, R. K., Moosmüller, H., Garro, M. A., Arnott, W. P., Walker, J., Susott, R. A., Babbitt, R. E.,
410 Wold, C. E., Lincoln, E. N., and Hao, W. M.: Emissions from the laboratory combustion of wildland
411 fuels: Particle morphology and size, *J. Geophys. Res.: Atmos.*, 111,
412 <https://doi.org/10.1029/2005JD006659>, 2006.
- 413 Chi, J. W., Li, W. J., Zhang, D. Z., Zhang, J. C., Lin, Y. T., Shen, X. J., Sun, J. Y., Chen, J. M., Zhang, X.
414 Y., Zhang, Y. M., and Wang, W. X.: Sea salt aerosols as a reactive surface for inorganic and organic
415 acidic gases in the Arctic troposphere, *Atmos. Chem. Phys.*, 15, 11341-11353,
416 <https://doi.org/10.5194/acp-15-11341-2015>, 2015.
- 417 China, S., Mazzoleni, C., Gorkowski, K., Aiken, A. C., and Dubey, M. K.: Morphology and mixing state
418 of individual freshly emitted wildfire carbonaceous particles, *Nat. Commun.*, 4, 2122,
419 <https://doi.org/10.1038/ncomms3122>, 2013.
- 420 China, S., Scarnato, B., Owen, R. C., Zhang, B., Ampadu, M. T., Kumar, S., Dzepina, K., Dziobak, M.
421 P., Fialho, P., and Perlinger, J. A.: Morphology and mixing state of aged soot particles at a remote
422 marine free troposphere site: Implications for optical properties, *Geophys. Res. Lett.*, 42, 1243-1250,
423 <https://doi.org/10.1002/2014GL062404>, 2015.
- 424 Dahlkötter, F., Gysel, M., Sauer, D., Minikin, A., Baumann, R., Seifert, P., Ansmann, A., Fromm, M.,
425 Voigt, C., and Weinzierl, B.: The Pagami Creek smoke plume after long-range transport to the upper
426 troposphere over Europe – aerosol properties and black carbon mixing state, *Atmos. Chem.*
427 *Phys.*, 14, 6111-6137, <https://doi.org/10.5194/acp-14-6111-2014>, 2014.
- 428 Ding, A. J., Huang, X., Nie, W., Sun, J. N., Kerminen, V. M., Petäjä, T., Su, H., Cheng, Y. F., Yang, X.
429 Q., Wang, M. H., Chi, X. G., Wang, J. P., Virkkula, A., Guo, W. D., Yuan, J., Wang, S. Y., Zhang, R.
430 J., Wu, Y. F., Song, Y., Zhu, T., Zilitinkevich, S., Kulmala, M., and Fu, C. B.: Enhanced haze pollution
431 by black carbon in megacities in China, *Geophys. Res. Lett.*, 43, 2873-2879,
432 <https://doi.org/10.1002/2016GL067745>, 2016.



- 433 Ding, S., Liu, D., Zhao, D., Hu, K., Tian, P., Zhou, W., Huang, M., Yang, Y., Wang, F., Sheng, J., Liu, Q.,
434 Kong, S., Cui, P., Huang, Y., He, H., Coe, H., and Ding, D.: Size-Related Physical Properties of Black
435 Carbon in the Lower Atmosphere over Beijing and Europe, *Environ. Sci. Technol.*, 53, 11112-11121,
436 <https://doi.org/10.1021/acs.est.9b03722>, 2019.
- 437 Geng, H., Hwang, H., Liu, X., Dong, S., and Ro, C. U.: Investigation of aged aerosols in size-resolved
438 Asian dust storm particles transported from Beijing, China, to Incheon, Korea, using low-Z particle
439 EPMA, *Atmos. Chem. Phys.*, 14, 3307-3323, <https://doi.org/10.5194/acp-14-3307-2014>, 2014.
- 440 He, C., Liou, K. N., Takano, Y., Zhang, R., Levy Zamora, M., Yang, P., Li, Q., and Leung, L. R.: Variation
441 of the radiative properties during black carbon aging: theoretical and experimental intercomparison,
442 *Atmos. Chem. Phys.*, 15, 11967-11980, <https://doi.org/10.5194/acp-15-11967-2015>, 2015.
- 443 IPCC.: Clouds and Aerosols. In: *Climate Change 2013: The Physical Science Basis, Contribution of*
444 *Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change.*
445 *Intergovernmental Panel on Climate Change (IPCC)*, 571- 657, 2013.
- 446 Jacobson, M. Z.: Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols,
447 *Nature*, 409, 695-697, <https://doi.org/10.1038/35055518>, 2001.
- 448 Koelyue, U., Xing, Y., and Rosner, D. E.: Fractal Morphology Analysis of Combustion-Generated
449 Aggregates Using Angular Light Scattering and Electron Microscope Images, *Langmuir*, 11, 4848-
450 4854, <https://doi.org/10.1021/la00012a043>, 1995.
- 451 Laskin, A., Moffet, R. C., and Gilles, M. K.: Chemical Imaging of Atmospheric Particles, *Acc. Chem.*
452 *Res.*, 52, 3419-3431, <https://doi.org/10.1021/acs.accounts.9b00396>, 2019.
- 453 Lee, A. K. Y., Rivellini, L.-H., Chen, C.-L., Liu, J., Price, D. J., Betha, R., Russell, L. M., Zhang, X., and
454 Cappa, C. D.: Influences of Primary Emission and Secondary Coating Formation on the Particle
455 Diversity and Mixing State of Black Carbon Particles, *Environ. Sci. Technol.*, 53, 9429-9438,
456 <https://doi.org/10.1021/acs.est.9b03064>, 2019.
- 457 Li, J., Liu, C., Yin, Y., and Kumar, K. R.: Numerical investigation on the Ångström exponent of black
458 carbon aerosol, *J. Geophys. Res.: Atmos.*, 121, 3506-3518, <https://doi.org/10.1002/2015JD024718>,
459 2016a.
- 460 Li, W., Zhou, S., Wang, X., Xu, Z., Yuan, C., Yu, Y., Zhang, Q., and Wang, W.: Integrated evaluation of
461 aerosols from regional brown hazes over northern China in winter: Concentrations, sources,
462 transformation, and mixing states, *J. Geophys. Res.: Atmos.*, 116, D09301,
463 <https://doi.org/10.1029/2010JD015099>, 2011.
- 464 Li, W., Shao, L., Shi, Z., Chen, J., Yang, L., Yuan, Q., Yan, C., Zhang, X., Wang, Y., Sun, J., Zhang, Y.,
465 Shen, X., Wang, Z., and Wang, W.: Mixing state and hygroscopicity of dust and haze particles before
466 leaving Asian continent, *J. Geophys. Res.: Atmos.*, 119, 1044-1059,
467 <https://doi.org/10.1002/2013JD021003>, 2014.
- 468 Li, W., Sun, J., Xu, L., Shi, Z., Riemer, N., Sun, Y., Fu, P., Zhang, J., Lin, Y., Wang, X., Shao, L., Chen,
469 J., Zhang, X., Wang, Z., and Wang, W.: A conceptual framework for mixing structures in individual
470 aerosol particles, *J. Geophys. Res.: Atmos.*, 121, 13784-13798,
471 <https://doi.org/10.1002/2016JD025252>, 2016b.
- 472 Liu, D., Whitehead, J., Alfarra, M. R., Reyes-Villegas, E., Spracklen, D. V., Reddington, C. L., Kong, S.,
473 Williams, P. I., Ting, Y.-C., Haslett, S., Taylor, J. W., Flynn, M. J., Morgan, W. T., McFiggans, G.,
474 Coe, H., and Allan, J. D.: Black-carbon absorption enhancement in the atmosphere determined by
475 particle mixing state, *Nat. Geosci.*, 10, 184-188, <https://doi.org/10.1038/ngeo2901>, 2017.
- 476 Ma, X., Zangmeister, C. D., Gigault, J., Mulholland, G. W., and Zachariah, M. R.: Soot aggregate



- 477 restructuring during water processing, *J. Aerosol Sci.*, 66, 209-219,
478 <https://doi.org/10.1016/j.jaerosci.2013.08.001>, 2013.
- 479 Matsui, H., Hamilton, D. S., and Mahowald, N. M.: Black carbon radiative effects highly sensitive to
480 emitted particle size when resolving mixing-state diversity, *Nat. Commun.*, 9, 3446,
481 <https://doi.org/10.1038/s41467-018-05635-1>, 2018.
- 482 Metcalf, A. R., Craven, J. S., Ensberg, J. J., Brioude, J., Angevine, W., Sorooshian, A., Duong, H. T.,
483 Jonsson, H. H., Flagan, R. C., and Seinfeld, J. H.: Black carbon aerosol over the Los Angeles Basin
484 during CalNex, *J. Geophys. Res.: Atmos.*, 117, D00V13, <https://doi.org/10.1029/2011JD017255>,
485 2012.
- 486 Moffet, R. C., and Prather, K. A.: In-situ measurements of the mixing state and optical properties of soot
487 with implications for radiative forcing estimates, *Proc. Natl. Acad. Sci. U.S.A.*, 106, 11872-11877,
488 <https://doi.org/10.1073/pnas.0900040106>, 2009.
- 489 O'Brien, R. E., Wang, B., Kelly, S. T., Lundt, N., You, Y., Bertram, A. K., Leone, S. R., Laskin, A., and
490 Gilles, M. K.: Liquid-Liquid Phase Separation in Aerosol Particles: Imaging at the Nanometer Scale,
491 *Environ. Sci. Technol.*, 49, 4995-5002, <https://doi.org/10.1021/acs.est.5b00062>, 2015.
- 492 Oh, C., and Sorensen, C. M.: The Effect of Overlap between Monomers on the Determination of Fractal
493 Cluster Morphology, *J. Colloid Interface Sci.*, 193, 17-25, <https://doi.org/10.1006/jcis.1997.5046>,
494 1997.
- 495 Pan, X., Uno, I., Hara, Y., Kuribayashi, M., Kobayashi, H., Sugimoto, N., Yamamoto, S., Shimohara, T.,
496 and Wang, Z.: Observation of the simultaneous transport of Asian mineral dust aerosols with
497 anthropogenic pollutants using a POPC during a long-lasting dust event in late spring 2014, *Geophys.*
498 *Res. Lett.*, 42, 1593-1598, <https://doi.org/10.1002/2014GL062491>, 2015.
- 499 Pei, X., Hallquist, M., Eriksson, A. C., Pagels, J., Donahue, N. M., Mentel, T., Svenningsson, B., Brune,
500 W., and Pathak, R. K.: Morphological transformation of soot: investigation of microphysical
501 processes during the condensation of sulfuric acid and limonene ozonolysis product vapors, *Atmos.*
502 *Chem. Phys.*, 18, 9845-9860, <https://doi.org/10.5194/acp-18-9845-2018>, 2018.
- 503 Peng, J., Hu, M., Guo, S., Du, Z., Shang, D., Zheng, J., Zheng, J., Zeng, L., Shao, M., Wu, Y., Collins,
504 D., and Zhang, R.: Ageing and hygroscopicity variation of black carbon particles in Beijing measured
505 by a quasi-atmospheric aerosol evolution study (QUALITY) chamber, *Atmos. Chem. Phys.*, 17,
506 10333-10348, <https://doi.org/10.5194/acp-17-10333-2017>, 2017.
- 507 Perring, A. E., Schwarz, J. P., Markovic, M. Z., Fahey, D. W., Jimenez, J. L., Campuzano-Jost, P., Palm,
508 B. D., Wisthaler, A., Mikoviny, T., Diskin, G., Sachse, G., Ziemba, L., Anderson, B., Shingler, T.,
509 Crosbie, E., Sorooshian, A., Yokelson, R., and Gao, R.-S.: In situ measurements of water uptake by
510 black carbon-containing aerosol in wildfire plumes, *J. Geophys. Res.: Atmos.*, 122, 1086-1097,
511 <https://doi.org/10.1002/2016JD025688>, 2017.
- 512 Raatikainen, T., Brus, D., Hyvärinen, A. P., Svensson, J., Asmi, E., and Lihavainen, H.: Black carbon
513 concentrations and mixing state in the Finnish Arctic, *Atmos. Chem. Phys.*, 15, 10057-10070,
514 <https://doi.org/10.5194/acp-15-10057-2015>, 2015.
- 515 Riemer, N., West, M., Zaveri, R., and Easter, R.: Estimating black carbon aging time-scales with a
516 particle-resolved aerosol model, *J. Aerosol Sci.*, 41, 143-158,
517 <https://doi.org/10.1016/j.jaerosci.2009.08.009>, 2010.
- 518 Sahu, L. K., Kondo, Y., Moteki, N., Takegawa, N., Zhao, Y., Cubison, M. J., Jimenez, J. L., Vay, S.,
519 Diskin, G. S., Wisthaler, A., Mikoviny, T., Huey, L. G., Weinheimer, A. J., and Knapp, D. J.: Emission
520 characteristics of black carbon in anthropogenic and biomass burning plumes over California during



- 521 ARCTAS-CARB 2008, *J. Geophys. Res.: Atmos.*, 117, D16302,
522 <https://doi.org/10.1029/2011JD017401>, 2012.
- 523 Shiraiwa, M., Kondo, Y., Moteki, N., Takegawa, N., Miyazaki, Y., and Blake, D. R.: Evolution of mixing
524 state of black carbon in polluted air from Tokyo, *Geophys. Res. Lett.*, 34, L16803,
525 <https://doi.org/10.1029/2007GL029819>, 2007.
- 526 Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., and Ngan, F.: NOAA's
527 HYSPLIT Atmospheric Transport and Dispersion Modeling System, *Bull. Amer. Meteor. Soc.*, 96,
528 2059-2077, <https://doi.org/10.1175/bams-d-14-00110.1>, 2015.
- 529 Sun, J., Liu, L., Xu, L., Wang, Y., Wu, Z., Hu, M., Shi, Z., Li, Y., Zhang, X., Chen, J., and Li, W.: Key
530 Role of Nitrate in Phase Transitions of Urban Particles: Implications of Important Reactive Surfaces
531 for Secondary Aerosol Formation, *J. Geophys. Res.: Atmos.*, 123, 1234-1243,
532 <https://doi.org/10.1002/2017JD027264>, 2018.
- 533 Ueda, S., Nakayama, T., Taketani, F., Adachi, K., Matsuki, A., Iwamoto, Y., Sadanaga, Y., and Matsumi,
534 Y.: Light absorption and morphological properties of soot-containing aerosols observed at an East
535 Asian outflow site, Noto Peninsula, Japan, *Atmos. Chem. Phys.*, 16, 2525-2541,
536 <https://doi.org/10.5194/acp-16-2525-2016>, 2016.
- 537 Wang, J., Cubison, M. J., Aiken, A. C., Jimenez, J. L., and Collins, D. R.: The importance of aerosol
538 mixing state and size-resolved composition on CCN concentration and the variation of the importance
539 with atmospheric aging of aerosols, *Atmos. Chem. Phys.*, 10, 7267-7283, <https://doi.org/10.5194/acp-10-7267-2010>, 2010.
- 541 Wang, Y., Liu, F., He, C., Bi, L., Cheng, T., Wang, Z., Zhang, H., Zhang, X., Shi, Z., and Li, W.: Fractal
542 Dimensions and Mixing Structures of Soot Particles during Atmospheric Processing, *Environ. Sci.
543 Technol. Lett.*, 4, 487-493, <https://doi.org/10.1021/acs.estlett.7b00418>, 2017.
- 544 Wentzel, M., Gorzawski, H., Naumann, K. H., Saathoff, H., and Weinbruch, S.: Transmission electron
545 microscopical and aerosol dynamical characterization of soot aerosols, *J. Aerosol Sci.*, 34, 1347-1370,
546 [https://doi.org/10.1016/S0021-8502\(03\)00360-4](https://doi.org/10.1016/S0021-8502(03)00360-4), 2003.
- 547 West, J. J., Cohen, A., Dentener, F., Brunekreef, B., Zhu, T., Armstrong, B., Bell, M. L., Brauer, M.,
548 Carmichael, G., Costa, D. L., Dockery, D. W., Kleeman, M., Krzyzanowski, M., Künzli, N., Liousse,
549 C., Lung, S.-C. C., Martin, R. V., Pöschl, U., Pope, C. A., Roberts, J. M., Russell, A. G., and
550 Wiedinmyer, C.: "What We Breathe Impacts Our Health: Improving Understanding of the Link
551 between Air Pollution and Health", *Environ. Sci. Technol.*, 50, 4895-4904,
552 <https://doi.org/10.1021/acs.est.5b03827>, 2016.
- 553 Wu, Y., Cheng, T., Liu, D., Allan, J. D., Zheng, L., and Chen, H.: Light Absorption Enhancement of Black
554 Carbon Aerosol Constrained by Particle Morphology, *Environ. Sci. Technol.*, 52, 6912-6919,
555 <https://doi.org/10.1021/acs.est.8b00636>, 2018.
- 556 Xu, L., Zhang, D., and Li, W.: Microscopic comparison of aerosol particles collected at an urban site in
557 North China and a coastal site in Japan, *Sci. Total Environ.*, 669, 948-954,
558 <https://doi.org/10.1016/j.scitotenv.2019.03.163>, 2019.
- 559 You, Y., Renbaum-Wolff, L., Carreras-Sospedra, M., Hanna, S. J., Hiranuma, N., Kamal, S., Smith, M.
560 L., Zhang, X., Weber, R. J., Shilling, J. E., Dabdub, D., Martin, S. T., and Bertram, A. K.: Images
561 reveal that atmospheric particles can undergo liquid-liquid phase separations, *Proc. Natl. Acad. Sci.
562 U.S.A.*, 109, 13188-13193, <https://doi.org/10.1073/pnas.1206414109>, 2012.
- 563 Yuan, Q., Xu, J., Wang, Y., Zhang, X., Pang, Y., Liu, L., Bi, L., Kang, S., and Li, W.: Mixing State and
564 Fractal Dimension of Soot Particles at a Remote Site in the Southeastern Tibetan Plateau, *Environ.*



565 Sci. Technol., 53, 8227-8234, <https://doi.org/10.1021/acs.est.9b01917>, 2019.

566 Zhang, D., Iwasaka, Y., Shi, G., Zang, J., Hu, M., and Li, C.: Separated status of the natural dust plume
567 and polluted air masses in an Asian dust storm event at coastal areas of China, *J. Geophys. Res.:
568 Atmos.*, 110, D06302, <https://doi.org/10.1029/2004JD005305>, 2005.

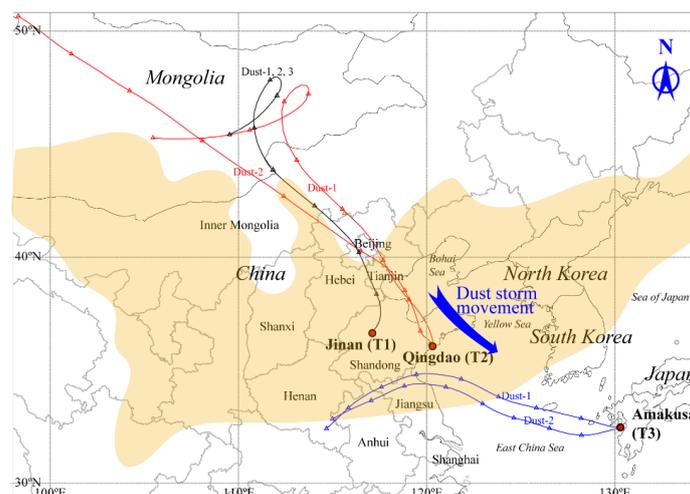
569 Zhang, G., Bi, X., Li, L., Chan, L. Y., Li, M., Wang, X., Sheng, G., Fu, J., and Zhou, Z.: Mixing state of
570 individual submicron carbon-containing particles during spring and fall seasons in urban Guangzhou,
571 China: a case study, *Atmos. Chem. Phys.*, 13, 4723-4735, <https://doi.org/10.5194/acp-13-4723-2013>,
572 2013.

573 Zhang, R., Khalizov, A. F., Pagels, J., Zhang, D., Xue, H., and McMurry, P. H.: Variability in morphology,
574 hygroscopicity, and optical properties of soot aerosols during atmospheric processing, *Proc. Natl.
575 Acad. Sci. U.S.A.*, 105, 10291-10296, <https://doi.org/10.1073/pnas.0804860105>, 2008.

576 Zhang, X., Mao, M., Yin, Y., and Wang, B.: Numerical Investigation on Absorption Enhancement of
577 Black Carbon Aerosols Partially Coated With Nonabsorbing Organics, *J. Geophys. Res.: Atmos.*, 123,
578 1297-1308, <https://doi.org/10.1002/2017JD027833>, 2018a.

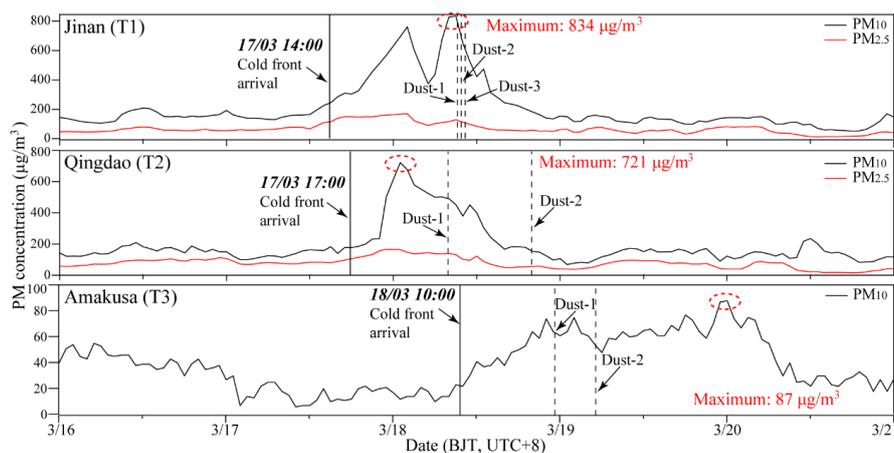
579 Zhang, Y., Su, H., Ma, N., Li, G., Kecorius, S., Wang, Z., Hu, M., Zhu, T., He, K., Wiedensohler, A.,
580 Zhang, Q., and Cheng, Y.: Sizing of Ambient Particles From a Single-Particle Soot Photometer
581 Measurement to Retrieve Mixing State of Black Carbon at a Regional Site of the North China Plain,
582 *J. Geophys. Res.: Atmos.*, 123, 7778-7795, <https://doi.org/10.1029/2018JD028810>, 2018b.

583



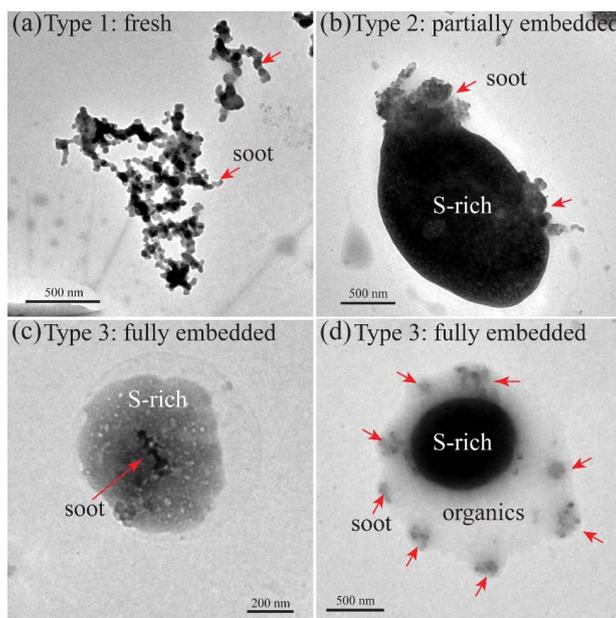
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585 Figure 1. The locations of the three sampling sites and HYSPLIT forty-eight hour air
586 mass backward trajectories arriving at 1500 m above ground level at T1, T2, and T3
587 sites. The interval between two triangle symbols is six hours. The yellow shadow is
588 derived from Figure S1, which represents the area influenced by the dust storm at 08:00
589 on 2014/03/18 (BJT, UTC+8).



590

591 Figure 2. Time series of PM (particulate matter) concentrations at T1, T2, and T3 during
592 sampling. Data sources: T1 and T2: The Ministry of Ecology and Environment of the
593 People's Republic of China, <https://www.aqistudy.cn/>; T3: National Institute for
594 Environmental Studies of Japan, <https://www.nies.go.jp/igreen/>.

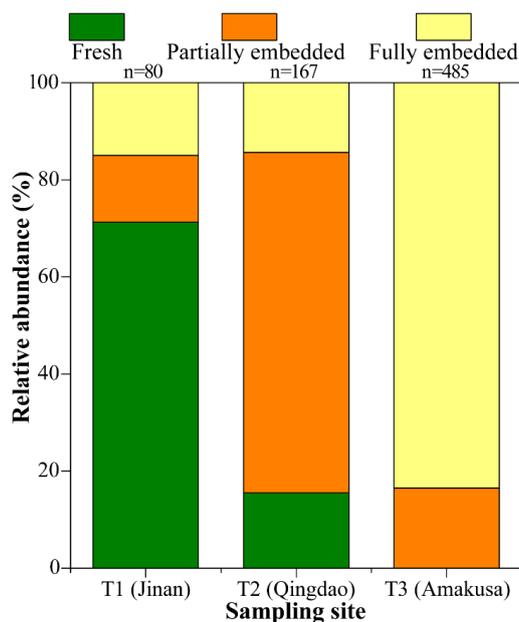


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596 Figure 3. Morphology and relative abundance of soot-bearing aerosol particles: (a)
597 fresh chain-like soot aggregates with no visible coating; (b) partially embedded soot:
598 part of the soot particle was coated by secondary aerosols; (c) fully embedded soot: the
599 whole soot particle was encapsulated by secondary aerosols; (d) a subtype of fully
600 embedded soot: individual soot particles were only embedded in the organic coating on
601 a sulfur-rich particle.

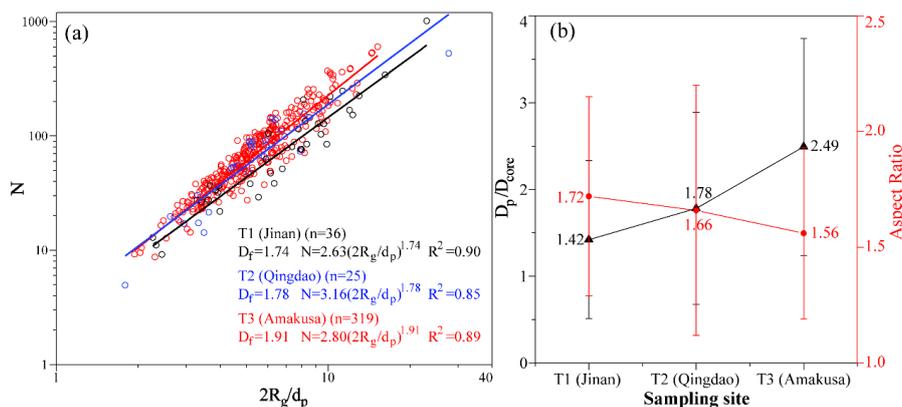
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605 Figure 4. Relative abundance of three types of soot-bearing aerosol particles at the three
 606 sampling sites. The number of analyzed soot-bearing particles is shown above the
 607 column.

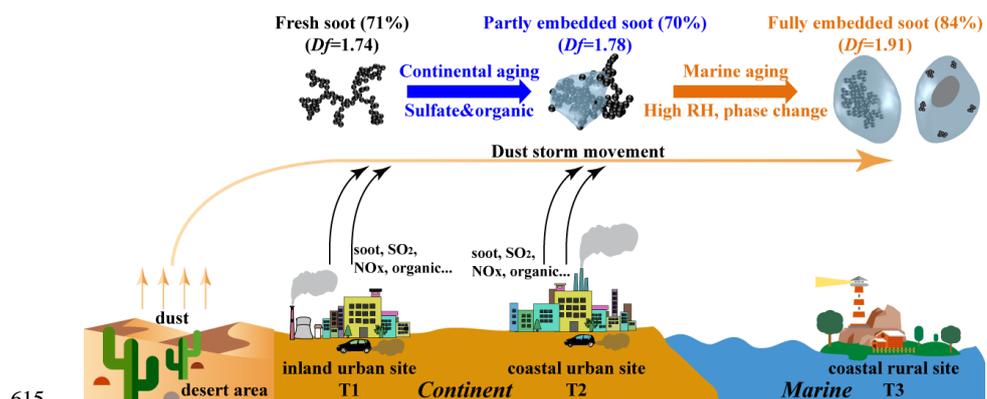


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609 Figure 5. (a) Fractal dimension of soot-bearing particles at the three sampling sites. The
 610 parameter n in parentheses represents the total number of soot particles analyzed for
 611 each site to calculate D_f and k_g . (b) The particle-to-soot core diameter ratio (D_p/D_{core})
 612 and aspect ratio of soot-bearing particles at the three sampling sites.

613

614



615

616 Figure 6. Schematic diagram showing the evolution of morphology and mixing state of

617 soot particles along with the movement of an Asian dust storm