

This is an Open Access document downloaded from ORCA, Cardiff University's institutional repository:<https://orca.cardiff.ac.uk/id/eprint/156465/>

This is the author's version of a work that was submitted to / accepted for publication.

Citation for final published version:

Feng, Xiaolei, Shao, Longyi, Jones, Tim , Li, Yaowei, Zhang, Mengyuan, Ge, Shuoyi, Cao, Yaxin, Bérubé, Kelly and Zhang, Daizhou 2023. The changing sulphur content of a northern Chinese dust storm: Initiation, attenuation and culmination. *Atmospheric Environment* 297 , 119606. 10.1016/j.atmosenv.2023.119606

Publishers page: <http://dx.doi.org/10.1016/j.atmosenv.2023.119606>

Please note:

Changes made as a result of publishing processes such as copy-editing, formatting and page numbers may not be reflected in this version. For the definitive version of this publication, please refer to the published source. You are advised to consult the publisher's version if you wish to cite this paper.

This version is being made available in accordance with publisher policies. See <http://orca.cf.ac.uk/policies.html> for usage policies. Copyright and moral rights for publications made available in ORCA are retained by the copyright holders.



1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22

The changing sulphur content of a northern Chinese dust storm: initiation, attenuation and culmination

Xiaolei Feng^a, Longyi Shao^{a*}, Tim Jones^b, Yaowei Li^c, Mengyuan Zhang^a, Shuoyi Ge^a, Yaxin Cao^a, Kelly BeruBe^d, Daizhou Zhang^e

^a State Key Laboratory of Coal Resources and Safe Mining, and College of Geoscience and Surveying Engineering, China University of Mining and Technology (Beijing), Beijing 100083, China

^b School of Earth and Environmental Sciences, Cardiff University, Park Place, Cardiff, CF10, 3YE, UK

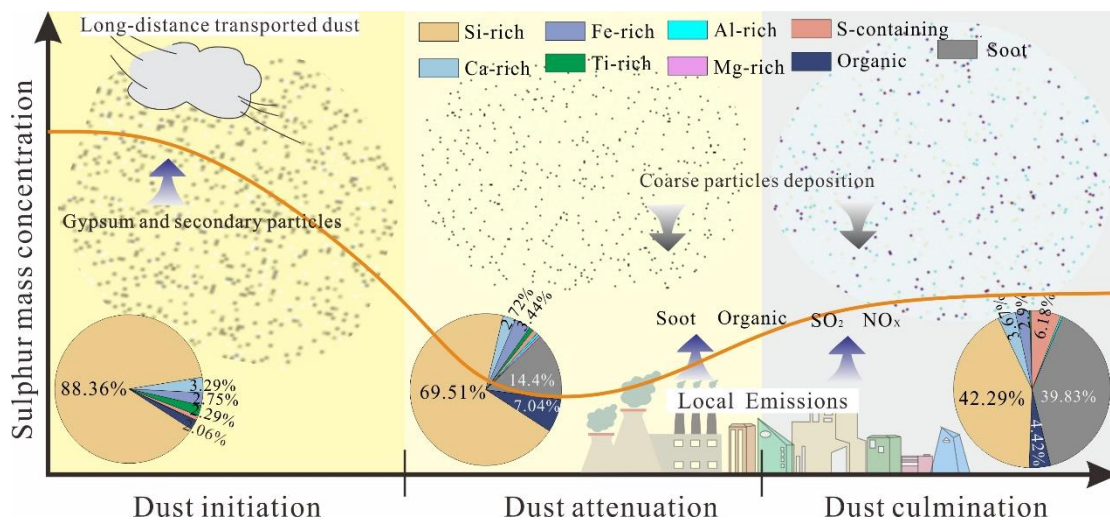
^c Hebei Center for Ecological and Environmental Geology Research, Hebei GEO University, Shijiazhuang, 050031, China

^d School of Biosciences, Cardiff University, Museum Avenue, Cardiff, CF10 3AX, Wales, UK

^e Faculty of Environmental and Symbiotic Sciences, Prefectural University of Kumamoto, Kumamoto, 62-8502, Japan

*Correspondence: ShaoL@cumtb.edu.cn

23 **Graphical abstract**



24

25 **Highlights**

- 26 1. The investigated dust storms have three recognisable stages: initiation,
 27 attenuation and culmination.
 28 2. The high concentration of S in the initiation stage was provided by S containing
 29 minerals from the source area.
 30 3. The secondary chemical reactions in the dust storm are comparable to those in
 31 haze events.

32

33 **Abstract**

34 It is predicted that changing climate trends on land in northern China will be
 35 associated with increasingly frequent and severe dust storms. These regionally
 36 extensive storms could have a serious impact on the environment and human respiratory
 37 health. A severe dust storm event hit Beijing from March 15th to 20th, 2021, and airborne
 38 particles from this dust storm episode were collected. The types, size distributions, and
 39 chemical compositions of the dust storm particles were analysed using individual

40 particle analysis techniques. This dust storm episode was divided into three
41 recognisable stages, which were initiation, attenuation and culmination by the PM10
42 pollution levels and horizontal visibility. Field Emission Scanning Electron Microscopy
43 coupled with an Energy-dispersive X-ray Spectrometer recorded three basic particle
44 groups: minerals, soot, and organic particles, of which minerals were the most abundant.
45 Over the duration of the dust storm, the relative number percentage of minerals
46 continuously decreased, while that of soot and organic particles correspondingly
47 increased. The dust particles were mostly distributed in the 1-2 μm size range. In the
48 attenuation and culmination stages, the number percentage of sub-micrometer particles
49 was higher than in the initiation stage, probably due to coarser minerals being deposited
50 and the finer minerals formed by secondary reactions remaining airborne. In
51 comparison to the initiation stage, the presence of anthropogenic toxic metals increased
52 over the attenuation and culmination stages. Anthropogenic emissions, both particulate
53 and gaseous, were mixed with the crust-derived dust, modifying the dust bulk
54 composition and therefore changing the potential impacts of the dust storm in the later
55 stages. The sulphur concentration was highest in the initiation stage ($27.8 \mu\text{g}/\text{m}^3$), with
56 sulphur concentrations mainly consisting of gypsum from the source area and
57 secondary reactions during early transport. In the attenuation and culmination stages,
58 the overall mass concentration of sulphur decreased significantly, while the various
59 sulphur-containing minerals increased.

60

61 **Keywords**

62 Airborne mineral particles; Sulphur; Secondary chemical reaction; Number-size

63 distributions; Toxic metals

64

65 **1. Introduction**

66 Dust storms are a severe and worsening global environmental issue (Huang et al.,
67 2014; Huang et al., 2015; Soleimani et al., 2020). The Taklimakan Desert, located in
68 Xinjiang northwest China, re-suspended approximately 70 Tg/yr of mineral dust into
69 the atmosphere over the Spring periods of 2007-2011, making this desert the highest
70 airborne emission source in east Asia (Chen et al., 2017). As an important type of
71 atmospheric aerosol, the dust storm particles can affect climate directly through
72 scattering, transmission and absorption of solar radiation, and indirectly by acting as a
73 cloud nucleation nucleus (CNN) when coated with soluble material (Buseck and Posfai,
74 1999; Manktelow et al., 2010; Wen et al., 2021; Shi et al., 2005), and can further
75 influence biogeochemical cycles (Li et al., 2017; Uno et al., 2009). Additionally, the
76 smaller respiratory dust particles can cause serious health issues (Hashizume et al.,
77 2020), whereas the coarser particles are nuisance dusts causing nose, throat and eye
78 irritation. It is reported that the aerosol particles can even act as a carrier to spread the
79 Covid-19 virus (Cao et al., 2021). Dust particles can function as carriers of toxic
80 elements and organics (Cave et al., 2018; Li et al., 2013; Tang et al., 2018; Wen et al.,
81 2021) potentially producing free radicals with the resultant production of oxidative
82 stress in the cardiovascular and respiratory systems (Aghababaeian et al., 2021; Chen
83 et al., 2018; Hasunuma et al., 2021; Sadeghimoghaddam et al., 2021).

84 The compositional characteristics of dust storms depend on two main variables,
85 which are provenance, and the inputs and outputs along the transport route (Li and Shao,
86 2012; Wang et al., 2022; Zhao et al., 2007). The mineral particles in the dust storms are

87 the most abundant; accounting for more than 80% of all particles (Jeong, 2008; Okada
88 et al., 2005; Wang et al., 2022; Shi et al., 2005). The relative abundances of different
89 minerals, such as clays, calcite, quartz, and feldspars will vary by source region and
90 also by the respective mineral grain's sizes and shapes. The larger mineral grains will
91 be less likely to be suspended in the dust storms and will drop out more rapidly as the
92 storm energy levels decrease. Studies have found that there is a significant difference
93 in the mineralogical composition of dust particles in Asia and Africa, and specifically,
94 the illite/kaolinite ratio and chlorite/kaolinite ratio of Asia dust particles were higher
95 than those of Africa (Formenti et al., 2011). The size, morphology and geochemistry of
96 particles will be modified over the long-distance transport of the storm (Li et al., 2018;
97 Li et al., 2014; Xu et al., 2020). Alkaline minerals, typically containing Ca, Mg, K, Mn
98 and Fe, in the dust can react with acidic atmospheric gases by providing an interface
99 for atmospheric chemical reactions (Fussell and Kelly, 2021; Li and Shao, 2009; Shi et
100 al., 2012; Wang et al., 2017). Over long-range transport, the relative percentages of
101 sulphate, nitrate, and chlorine in the dust particles increase when compared to the
102 original composition (Kim et al., 2012; Ma et al., 2001; Li and Shao, 2012). It is
103 recognised that the relative proportions of sulphate minerals and other compounds
104 change in the dust attenuation stage (Wu et al., 2017), and the total sulphur content in
105 the dust can be lower in the culmination of the storm (Li et al., 2018; Wang et al., 2022).
106 However, the variations in the different forms of sulphur-containing minerals (all
107 minerals containing sulphur) over the duration of the dust storm is unclear.

108 The dust storm events sourced from the Gobi Desert have direct harmful impacts

109 on the environment and people of north China (Huang et al., 2014; Shao et al., 2008).
110 Beijing experienced three distinct dust storm events in the spring of 2021 (Fig. S1), and
111 one of them started on 15th of March 2021, and was the most widespread in China in
112 the past decade. The dust influence covered an area of 3.8 million square kilometers in
113 China, affecting more than 19 cities (Filonchyk, 2022; Luo et al., 2022b). The data from
114 China National Environmental Monitoring Centre showed that the PM₁₀ (particulate
115 matter with mean aerodynamic diameter $\leq 10 \mu\text{m}$) concentration in six core districts
116 in Beijing reached $8000 \mu\text{g}/\text{m}^3$ and the PM_{2.5} (particulate matter with mean
117 aerodynamic diameter $\leq 2.5 \mu\text{m}$) concentration exceeded $400 \mu\text{g}/\text{m}^3$ (China Climate
118 Bulletin, 2021). This very large-scale dust storm provided a significant research
119 opportunity for studying the physical and chemical properties of particulate matter in a
120 highly damaging and impactful event.

121 In this study, Scanning Electron Microscopy coupled with Energy Dispersive X-
122 ray Spectrometry (SEM-EDX), a commonly used method of individual particle analysis
123 (Shao et al., 2022), was used to analyse the number-size distributions, types, and
124 elemental compositions of individual particles of PM₁₀ in Beijing's atmosphere during
125 a dust storm episode. The empirical outputs provide data on particle sizes, numbers,
126 morphologies, elemental compositions, and mixing states, which allows interpretation
127 of the particle's provenance and processes that the particulate matter might have
128 undergone while in the storm. During transport, Asian dust particles can react with
129 anthropogenic acidic species through heterogeneous processes to produce large
130 quantities of water-soluble aerosols (Pan et al., 2017; Tang et al., 2017), influencing the

131 direct and indirect effects of dust on climate (Wang et al., 2017). Numerous studies
132 have focused on the level of sulphur and the rate of sulphate reactions during dust and
133 sand processes (Li and Shao, 2008; Wang et al., 2022; Wang et al., 2021; Wu et al.,
134 2017). However, there is still uncertainty about the levels of sulphur in dust storms. The
135 results of many studies have shown that during long-distance transport, the dust plumes
136 undergo heterogeneous reactions in the atmosphere, producing large amounts of
137 sulphur-containing particles (Huang et al., 2010; Nie et al., 2012; Zhao et al., 2011).
138 However, other researchers argue that the sulphur content in dust storms is very
139 consistent (Wang et al., 2022; Wu et al., 2017), where the sulphur in dust particles
140 originates from the source area (Hu et al., 2022), and the sulphur content remains
141 virtually unchanged even over long-distance transport (Wu et al., 2017). This study
142 focuses on the variation of sulphur mass concentrations at different stages during a
143 single dust event, providing a reference for the association between dust and
144 atmospheric sulphur.

145

146 **2. Sampling and Experiments**

147 **2.1 Dust sampling**

148 Tracked by satellite remote sensing, the investigated dust storm was initiated in
149 western Mongolia around 15:00 (UTC+8) on the 14th of March 2021, and then crossed
150 into China by the near-border Chinese town of Erenhot and adjacent areas at around
151 21:00 (UTC+8) on the 14th of March. The dust storm continued to move in a
152 southeasterly direction towards Beijing and arrived in Beijing at around 3:00 (UTC+8)

153 on the 15th of March. During the dust storm, the highest mass concentration of PM₁₀
154 was recorded on the 15th of March, at a daily mass concentration of 1630 µg/m³. On the
155 same day, the daily PM_{2.5} mass concentration and the daily AQI value were 207 µg/m³
156 and 500, respectively. When the dust storm reached the sampling site in Beijing, the
157 atmospheric pressure, temperature, and relative humidity (RH) were measured and
158 recorded every 30 minutes. These meteorological data were recorded using a portable
159 meteorological tracker (Kestrel 5500 Weather LiNK, USA). The temperature and RH
160 varied from 2.9°C to 19.5°C and 9.6% to 90%, respectively. The data for the levels of
161 PM_{2.5}, PM₁₀, and AQI in Fig. 1 were derived from the Wanliu state-owned automatic
162 ambient air monitoring station, which was obtained from the downloaded data of each
163 monitoring station in Beijing (<https://quotsoft.net/air/>). As determined by the levels of
164 PM mass concentration and horizontal visibility (Song et al., 2007), this dust event
165 consisted of three stages, that were initiation, attenuation, culmination. The initiation is
166 on the 15th of March with the PM₁₀ mass concentration larger than 3000 µg/m³ and
167 visibility being less than 500m, the attenuation stage is from the 16th to 17th of March
168 with the daily PM₁₀ mass concentration changing from 200 to 600 µg/m³ and visibility
169 ranging from 1km to 10km. The culmination stage is from the 19th to 20th of March
170 with the daily PM₁₀ mass concentration being lower than 200 µg/m³ and visibility
171 being more than 10km.

172 Airborne particle samples were collected from the 15th of March to the 20th of
173 March 2021. Samples were not collected on the 18th of March due to instrument
174 malfunction. The sampling site was located in the Haidian District, northwest of Beijing,

175 1 km from the fourth Ring Road. The sampler was placed on the flat roof of a building
176 on the campus of the China University of Mining and Technology (Beijing) (CUMTB).
177 The height of the building roof was 18 m above the ground (Fig. S2). Following the
178 National Standard of Determination of Atmospheric PM₁₀ and PM_{2.5} in Ambient Air by
179 Gravimetric Method (HJ 618-2011), the vertical distance between the sampler inlet and
180 the collection surface shall not be less than 1.5 m. The CUMTB campus is surrounded
181 by residential areas and shopping malls. The airborne particles were collected on a
182 polycarbonate (PC; Millipore, UK) filter with apertures of 0.67 μm using a Minivol™
183 sampler (Airmetrics, USA) with a particle PM₁₀ separation device at a flow rate of 5
184 L/min. The sampling efficiency of the particle separation device is 50% for particles
185 with an aerodynamic diameter of 10 μm.

186 Since the study was designed to understand the evolution of PM₁₀ composition and
187 number-size distribution during the dust storm, eight aerosol samples were collected
188 from the 15th of March to the 20th of March 2021. Two samples were collected in the
189 initiation stage on the 15th of March, four samples in the attenuation stage on the 16th
190 to 17th of March, and two samples in the culmination stage on the 19th to 20th of March.
191 The meteorological data recorded during the sample collections are shown in Table 1.

192

193 **2.2 Analysis Methods**

194 **2.2.1 Electron Microscopy**

195 The particle morphology was imaged using Field Emission Scanning Electron
196 Microscopy (FESEM, SUPRA40, Zeiss, German). The elemental composition of

197 individual particles was analysed qualitatively and semi-quantitatively by an Oxford
198 Link Pentafet energy spectrum analysis system, which can detect elements heavier than
199 boron ($Z > 5$). The signal acquisition time varied from 80s to 100s. Each filter was cut
200 into 1 cm² and fixed on a cylindrical copper carrier using conductive adhesive. The
201 carrier and sample were gold coated to approximately 20 nm. An area was randomly
202 selected for each sample and more than 300 particles analysed from each filter. Copper
203 and gold were omitted from the analysis.

204 **2.2.2 Image Analysis**

205 To measure the size of every individual particle analysed, the ‘area measurement’
206 function of Image-J software was used to process the images taken by SEM. The
207 software draws a line around the perimeter of each particle, then automatically converts
208 the measured length of the perimeter to the geometric Equivalent Spherical Diameter
209 (ESD). A total of 3143 particles were analysed and measured.

210

211 **3. Results**

212 **3.1 Morphologies and chemical compositions of particles during the dust storm**

213 The dust particles showed complex elemental compositions under SEM-EDX. In
214 addition to C and O which were detected in all the particles, N, Na, Mg, Al, Si, P, S, Cl,
215 K, Ca, Ti, Mn, Fe, Zn, and Cr were detected in individual particles (Fig. S3). Si and Al
216 had the highest detection frequency, accounting for 96.0% and 88.4% in the 3143
217 analyzed particles, respectively, followed by Fe (33.8%), Mg (29.3%), Cl (26.5%), K
218 (21.6%), Ca (19.8%), Mn (15.7%), Na (10.4%), S (4.2%) and Ti (3.9%). Other elements

219 were present in less than 1%.

220 The morphologies and elemental compositions from the FESEM-EDX analysis,
221 allows the particles to be classified into three main types recorded during the dust storm,
222 which were minerals, soot, and organic particles (Fig. 2). The minerals are mainly
223 composed of Si and Al, with minor amounts of Na, Mg, and Fe. Minerals that have
224 undergone attrition and abrasion in dust storms are likely to have no recognisable
225 crystalline form (Shi et al., 2005; Shao et al., 2022). The morphologies of the minerals
226 were mainly irregular with a small number of more elongate grains. The minerals were
227 extremely stable under the electron beam. Soot and organic particles were mainly
228 composed of C and O, and could be visually recognised by their characteristic shapes.
229 Soot particles are typically chain-shaped aggregates containing microscopic spherical
230 C nanoparticles. Organic particles were typically spherical or nearly spherical, and were
231 stable under the electron beam.

232 In the initiation stage on the 15th of March, the proportion of minerals was the
233 highest, accounting for 97.76%. During the attenuation stage of the 16th and 17th of
234 March, the proportion of minerals decreased when compared with the initiation stage,
235 but was still high, accounting for 78.56%. In the culmination stage of the 19th and 20th
236 of March, the percentage of minerals significantly decreased to 55.75%. The percentage
237 of soot continuously increased from the initiation stage to the culmination stage. In the
238 initiation stage, the percentage of soot is only 0.18%, but in the culmination stage, it
239 has increased to 39.83%. The percentage of organic particles remained low throughout
240 the three stages of the dust storm. The relative percentage of organic particles was the

241 lowest in the initiation stage at only 2.06%. The highest relative percentage of organic
242 particles was during the attenuation stage at 7.04%. Overall, during the whole storm
243 event, the relative number percentage of minerals is the highest, ranging from 47.37%
244 to 99.12%, averaging 77.66%, followed by soot particles, ranging from 0.36% to
245 46.52%, averaging 17.20%, with organic particles being the least abundant type,
246 ranging from 0.88% to 17.96%, averaging 5.14% (Fig. 3a).

247 **3.2 Classification and characteristics of minerals**

248 Airborne minerals not only affect climate, but also play an important role in
249 atmospheric chemical reactions (Shao et al., 2022; Wang et al., 2021). When transported
250 over a long distance, the dust particles can undergo elemental modifications (Li et al.,
251 2012a). As the total mass of minerals in the dust plume will not be constant due to
252 changing inputs and outputs, then this would change the relative percentages of
253 minerals in that plume. Firstly, the original source minerals could undergo chemical
254 change, with heterogeneous reactions resulting in the formation of different mineral
255 types (Aydin et al., 2012; Wang et al., 2017 and 2022; Yu et al., 2020). Secondly, the
256 mineral composition of the dust mass could change as the original minerals settle out
257 of the cloud, and are replaced with freshly sourced mineral grains (Raffaele et al., 2020).

258 Using the results of the elemental analysis, the formula:

$$259 \quad P(X) = X / (Na + Mg + Al + Si + S + Cl + K + Ca + Ti + Fe) \times 100\%$$

260 can be used to divide the mineral particles into different types, where P(X) is the atomic
261 weight ratio of the element (Li and Shao, 2012; Okada et al., 2005; Shao et al., 2017).

262 Through adapting this method and adding Mn and Cr to the formula, a total number of

263 2343 mineral particles are classified by the formula:

$$264 \quad P(X) = X / (\text{Na} + \text{Mg} + \text{Al} + \text{Si} + \text{S} + \text{Cl} + \text{K} + \text{Ca} + \text{Ti} + \text{Fe} + \text{Mn} + \text{Cr}) \times 100\%.$$

265 The minerals were classified as Si-rich, Fe-rich, Ca-rich, Al-rich, Ti-rich, Mg-rich, Mn-
266 rich, Cl-rich and Cr-rich. The minerals containing S are further classified into S-
267 containing minerals (Fig. 3b). In this dust storm event, the most abundant mineral types
268 were Si-rich particles, accounting for 85.51% of the total particles. This was followed
269 by Fe-rich, Ca-rich, and S-containing minerals, accounting for 4.23%, 4.13%, and 4.05%
270 respectively. The Al-rich, Ti-rich, Mg-rich minerals, and other mineral types accounted
271 for 2.37%. The relative number percentage of Si-rich particles decreased over the
272 duration of the dust storm. Fe-rich particles had the highest abundance in the attenuation
273 stage, while Ca-rich particles were the least abundant. The relative number percentage
274 of S-containing minerals increased over the duration of the storm.

275 There are 8 sub-types of Si-rich particles (Table S1), among which Si + Al particles
276 are the most common. According to their atomic weight ratios of Si and Al, minerals
277 can be categorized into several types of alkaline minerals (Li et al., 2018; Lv and Shao,
278 2003; Wang et al., 2022). This presents the opportunity to approximate the percentages
279 of common minerals in the dust cloud when the measured data consists only of
280 elemental data, and no mineralogical data such as X-ray diffraction is available. The
281 mineral with a ratio of Si:Al near 1 may be kaolinite, and when Si:Al is between 1 and
282 2, possible minerals include illite, chlorite, and montmorillonite, in which illite has
283 higher K content, and the value Si:Al of chlorite is closer to 2 (Lv and Shao, 2003;
284 Wang et al., 2022). The Si-rich particles also could consist of potassium feldspar and

285 plagioclase, which have Si:Al approximately equal to 3, and contain K and Na elements,
286 respectively (Lv and Shao, 2003). Si-dominated particles are mainly quartz, which is
287 dominated by Si and O. Si-rich particles are mainly derived from crustal weathering of
288 silicate minerals and quartz, certainly in the initiation stage. (Fig. S4).

289 The majority of the Fe-rich particles were identified as hematite, with peaks of Fe
290 and O elements and an absence of other significant components (Fig. S4). The Ca-rich
291 minerals consist mostly of calcite, dolomite and gypsum. The carbonates, calcite and
292 dolomite, were differentiated according to the relative proportions of Ca and Mg (Fig.
293 S4), with dolomite containing a greater proportion of Mg. The third common Ca-
294 containing mineral was identified as gypsum from the amount of S in the spectra; this
295 is important when considering the chemical evolution of S-containing minerals over the
296 duration of the dust storm from initiation to culmination. There was a second common
297 group of S-containing minerals whose more complex composition of Si, Al, S, and other
298 minor elements was interpreted to be a reaction product between silicate minerals and
299 SO₂ (Jeong 2020; Li et al., 2018; Li and Shao, 2012). These S-containing mixtures were
300 discovered throughout the dust storm. In overall, the relative proportion of total S-
301 containing minerals continually increased over the duration of the dust storm; from 1.06%
302 in the initiation to 6.18% in the culmination.

303 **3.3 Number-size distributions of dust particles during the sampling period**

304 The size distribution of airborne particles, as determined by image analysis of
305 SEM images (Fig. 4), is an important parameter to understand the impacts of dust
306 storms, and is related to the reactive surface area, volume, mass, and size. The particle

307 sizes of all the measured dust samples showed a unimodal distribution with the peak in
308 the 1-2 μm size range. One sample collected on the 19th of March had a peak range of
309 less than 1 μm . It is noted that the collector had a 10 μm cut-off device, however the
310 peak is significantly below this size cut-off. Therefore, the majority of the particles
311 collected were respirable, less than 2.5 μm , and able to be respired deep into the human
312 lung. Clearly given the extremely high total dust load of the storm, much of the mass
313 will have included non-respirable mineral grains acting as a nuisance dust.

314 The number-size distribution (Zhou et al., 2002) of dust particles during the
315 initiation stage on the 15th of March had a peak in the 1-2 μm size range of
316 approximately 150 $\text{dN\%/dlog}(D_p)$, and also the smallest size range of the duration of
317 the storm. During the attenuation stage, the 16th to 17th of March, the size peak dropped
318 to approximately 120 $\text{dN\%/dlog}(D_p)$, and the size range slightly increased. Finally, the
319 culmination stage, the 19th of March to the 20th of March had the lowest peak at
320 approximately 100 $\text{dN\%/dlog}(D_p)$, with those peak particle sizes slightly smaller than
321 the initiation and attenuation stages. In addition, the culmination stage had the largest
322 size range of the three stages. It is conjectured that the secondary reaction particles that
323 formed during the dust transportation and the overall changing nature of the sulphur-
324 containing minerals is evidenced (Fig. 4) by both the chemical SEM-EDX and the
325 relatively smaller secondary particle sizes (Liu et al., 2022; Shao et al., 2021). These
326 results are consistent with previous studies on dust storms and non-dust air pollution
327 periods (Song et al., 2022; Wang et al., 2021; Zhang et al., 2008).

328

329 **4. Discussion**

330 **4.1 Possible sources of the particles during the dust storm period**

331 Previous studies have found that dust storm events in northern China are
332 dominated by minerals, including quartz, clay, feldspar, and carbonates (Formenti et al.,
333 2011; Shao et al., 2008). Studies on the contribution of minerals in dust storms and non-
334 dust periods showed that the number and mass concentrations of airborne mineral
335 particles in Xi'an, Erlianhot, and Zhangbei were higher during the dust storms (Tang et
336 al., 2018; Wu et al., 2017). The NOAA/ARL Hybrid Single-Particle Lagrangian
337 Integrated Trajectory Model (Stein et al., 2015) was employed to calculate the
338 backward trajectories of a dust storm at 500 m elevation that arrived at the Beijing
339 sampling site on the 15th of March and culminated on the 20th of March 2021 (Fig. 5).
340 Fig. 5a shows that the air masses started Mongolia and passed over the deserts of
341 northern China, before reaching Beijing.

342 The ratios of different chemical elements are widely used to identify dust
343 provenance (Zhao et al., 2007). Therefore the Ca:Al ratio can be employed to identify
344 dust source (Huang et al., 2010; Li et al., 2008; Zhao et al., 2007), and the lower Ca:Al
345 ratio ($0.27\pm 0.10\sim 0.73\pm 0.03$) is believed to indicate that the dust originated from a
346 northern source (Song et al., 2022). In this study, the Ca:Al during the dust episode
347 ranged from 0.17 to 0.79, which is a similar ratio range to the northern dust. This result
348 is consistent with the identified source from the backward trajectory.

349 At the dust initiation stage, strong winds suspend crustal mineral particles
350 dominantly Si-rich, from erosion-prone desert surfaces (Fig. 3b). In the following

351 attenuation stage, soot and organic particles mainly derived from airborne emissions
352 from coal-burning, biomass burning, and vehicle emissions (Li et al., 2016; Shao et al.,
353 2022; Xing et al., 2017) mixed into the dust storm. The relative proportions of the
354 crustal minerals decrease, and anthropogenic particles gradually increase during the
355 storm, indicating that anthropogenic emissions are continuously incorporated into the
356 dust storm. March is part of the ‘heating’ season in northern China including Beijing,
357 Tianjin, and Hebei, and these urban areas contribute considerable amounts of
358 anthropogenic air pollution to mix into the storms in the attenuation stage (Fig. 5b). The
359 understanding of the relative contributions of local emissions and the distally-sourced
360 mineral particles is essential to fully elucidate the impacts of these significant
361 environmental events (Huang et al., 2010).

362 Studies on atmospheric chemistry show that reactive gases included in local
363 anthropogenic emissions will modify the composition of dust particles and typically
364 generate sulphates and nitrates (Li and Shao, 2012; Tang et al., 2017; Wu et al., 2017).
365 sulphur-containing mixtures were seen in the initiation stage, which was probably
366 caused by secondary chemical reactions during the dust long-distance transport. The
367 mineral types in the dust storm will be altered constantly as a result of the favorable
368 conditions for heterogeneous chemical reactions and availability of fresh inputs of
369 reactive anthropogenic gasses. Typically, when compared with the common crustal
370 minerals seen in the initiation stage, the altered mineral types in the attenuation stage
371 are more complex in terms of their elemental compositions. In addition, these secondary
372 chemical reactions in the storm will increase the relative number proportions of the

373 particles with a smaller size range (Shao et al., 2021).

374 **4.2 Variations in elemental compositions in different stages of the dust storm**

375 To elucidate the chemical changes in the dust storm particles during the three
376 stages of initiation, attenuation and culmination, the relative atomic weight percentage
377 of elements was analysed. Fig. 6 shows that the elemental atomic weight percentages
378 of Si, Al, K, and Ti decreased, while Ca, Mg, Fe, Zn, Mn, Cr, and S increased over the
379 three stages; with S being of the most significant change. Typically, Si, Al, K, and Ti
380 are considered to be sourced from mineral dust (Huang et al., 2010; Li et al., 2014; Luo
381 et al., 2022; Shao et al., 2017). Ca and Mg are both sourced from crustal inputs (Huang
382 et al., 2010; Song et al., 2022). Zn, Mn, and Cr are considered to largely originate from
383 anthropogenic sources, such as traffic and industrial emissions (Li et al., 2013; Luo et
384 al., 2022). The atomic weight percentages of heavy metals increase during the
385 attenuation stage supporting the other evidence that anthropogenic emissions are mixed
386 into the dust storm. The very significant increase of sulphur atomic weight percentage
387 shows the important role it plays in the alteration of airborne mineral types. The only
388 feasible source of sulphur in airborne particulate matter is the gas to solid phase change
389 from SO₂ to altered mineral chemistries.

390 Heavy metals in airborne particulate matter can be harmful to human health (Luo
391 et al., 2022a; Xiao et al., 2013), especially metals such as Zn, Mn and Cr, which showed
392 a significantly positive correlation with DNA oxidative damage (Feng et al., 2020 and
393 2022). The relative atomic weight of Zn, Mn, and Cr in particles were higher in the
394 attenuation stage than in the initiation stage, and this could be interpreted that the

395 sampling site was strongly influenced by local air masses in Beijing, Tianjin, and Hebei
396 (Fig. 5). Another comparable study on the enrichment factors of heavy metals in dust
397 particles showed that the increase in Zn and Cr was caused by mixing with local
398 aerosols in the polluted area (Luo et al., 2022).

399 The types and relative amounts of heavy metals decreased in the culmination stage
400 when compared to the attenuation stage. A previous study suggested that airborne dust
401 masses have the ability to incorporate and remove pre-existing pollutants (Wang et al.,
402 2021). This could mean that metals were taken-up within the dust mass and then
403 physically transported out of the area as the dust storm continued along its trajectory.
404 Anthropogenic airborne metals can only be incorporated into the dust cloud in a solid
405 phase, however once in the cloud they are able to take part in atmospheric chemical
406 reactions (Huang et al., 2010). These reactions could make the heavy metals more or
407 less bioavailable. To properly understand the potential impact of airborne metal-
408 containing particles in dust storms, it is imperative to understand the mass and size
409 distributions of these particles, and whether or not they are respirable. In addition, it is
410 important to know whether the metals are bioavailable once respired into the human
411 lung. This critical information is currently unknown.

412 **4.3 Sulphur chemistry of the dust storm particles**

413 It is believed that heterogeneous sulphur chemical reactions are limited during dust
414 storm events (Wang et al., 2022; Wang et al., 2021; Wu et al., 2017), which can result
415 in a relatively low SO_4^{2-} content in dust particles. However, it has also been recorded
416 that sulphur content during dust days is significantly higher than that during non-dust

417 days (Huang et al., 2010; Li and Shao, 2012; Xu et al., 2017). Typically, gaseous phase
418 sulphur in the atmosphere will undergo oxidative reactions with dust particles, resulting
419 in the formation of sulphate (Sun et al., 2004; Wu et al., 2021). Therefore,
420 anthropogenic SO₂ reacts with the alkaline minerals in the dust storm, forming the new
421 minerals (Jeong, 2020; Li et al., 2018; Wang et al., 2017).

422 SEM showed that the sulphur-containing minerals consist of rod-shaped gypsum
423 (Fig. 7a) and a mixture of sulphur and clay, of which the mixture of sulphur and clay
424 was the most common in the initiation stage. The crystalline shape of rod-shaped
425 gypsum is probably derived from the erosion of the arid surfaces where well-formed
426 crystallinity results from changing water availability, groundwater levels, and sufficient
427 time being available for the growth of recognisable crystals (Garcia-Guinea et al., 2002;
428 Hu et al., 2022). The most well-known source of the minerals with well-formed
429 crystalline shapes are the macroscopic so-called gypsum ‘desert roses’ or over-lapping
430 crystals commonly called ‘swallowtail’; both morphologies can be seen
431 microscopically under SEM (Jones et al., 2001). The mixture of sulphur and clay could
432 be the product of the reaction between alkaline minerals and SO₂ (Jeong, 2020; Li et
433 al., 2018). During the attenuation stage, the observed gypsum crystals tend to be
434 irregular and possess a granulated surface, and commonly are mixed with clay minerals
435 (Fig. 7b, 7c, 7d and 7e) visually identifiable from their platy morphologies. This trend
436 away from well-formed crystallinity continues into the culmination stage (Fig. 7f, 7g,
437 and 7h).

438 The observation of the overall change in particle morphologies supports the

439 interpretation that there were secondary chemical reactions forming new particle types
440 throughout the dust storm event, with airborne alkaline minerals providing a reaction
441 interface for heterogeneous reactions with SO₂. This speculation was also reinforced by
442 a study of a Beijing dust storm in 2002, which found that the mass percentage of SO₄²⁻
443 during the dust storm in Beijing was higher than the mass percentage in the Gobi Desert
444 (Zhao et al., 2007). Large amounts of sulphur-containing mixtures were also observed
445 under transmission electron microscopy in samples collected during a dust pollution
446 event during the 4th to 5th of May 2017 and spring of 2015 in Beijing (Li et al., 2018;
447 Wang et al., 2022).

448 To estimate the change of sulphur concentration over the duration of the dust storm
449 event, the average atomic weight ratio of sulphur in all individual particles was
450 multiplied by the mass concentration of PM₁₀ to obtain the mass of atmospheric sulphur
451 as µg/m³ (Fig. 8). The mass concentration of sulphur was the highest in the initiation
452 stage, at 27.9 µg/m³, and decreased rapidly over the attenuation stage, at 3.7 µg/m³. A
453 similar trend was recorded for sulphur in March of 2015 in Beijing, in which mass
454 concentration of sulphur in the dust period are higher than that in the following non-
455 dust period (Wang et al., 2022). In the initiation, gypsum from the source area and
456 sulphur added during transport together contribute to the high concentration of sulphur.
457 The rapid decrease of sulphur concentration in the attenuation stage is a possible
458 consequence of the dust plume sweeping away locally emitted air pollutants (Wang et
459 al., 2021). The mass concentration of sulphur increased in the culmination stage, which
460 was caused by secondary chemical reactions in the atmosphere. It also can be seen from

461 Fig. 8 that the mass concentration of SO₂ in the atmosphere is lowest during the
462 attenuation stage, and the initiation and culmination stages are similar. During the three
463 dust stages, meteorological conditions varied by 10.07°C, 12.83°C, 11.61°C for mean
464 temperature, 14.80%, 25.30%, 36.79% for mean relative humidity and 1012.75hPa,
465 1015.42hPa, 1012.35hPa for mean pressure. The data of meteorological conditions are
466 shown in Table 1. There are no special trends for air temperature and pressure, however,
467 humidity increases from the dust initiation stage to the dust culmination stage. Some
468 other studies have shown a negative correlation between PM₁₀ mass concentration and
469 relative humidity (Csavina et al., 2014; Maleki et al. 2022), and a similar trend was
470 found in this dust event. The increase in relative humidity during the dust culmination
471 stage is conducive to the occurrence of sulphate reactions.

472 A comparison with the mass concentration of sulphur in the 2021 dust storm with
473 a dust event in 2015 showed that the mass concentration of sulphur in the 2021 dust
474 storm period was higher than that seen in 2015 (Fig. 8). This could be due to the
475 difference of source areas and transport pathways. Based on the backward trajectory
476 results, the source area of the 2021 dust storm is northern Mongolia, but the 2015 dust
477 originated in southern Mongolia (Wang et al., 2022). The 2021 dust storm was
478 transported over a longer distance than the 2015 dust storm (Wang et al., 2022). The
479 mass concentration of sulphur in the initiation stage in this study was much higher than
480 that on haze and non-haze days. It is well established that there are large amounts of
481 sulphate particles during haze days due to secondary chemical reactions (Cheng et al.,
482 2016; Li et al., 2022). If we exclude the gypsum particles derived from the source area

483 in the initiation stage, the calculated sulphur mass concentration ($8.26 \mu\text{g}/\text{m}^3$) was
484 similar to that recorded during the haze periods such as one that occurred in 2017 (11.83
485 $\mu\text{g}/\text{m}^3$), indicating that secondary chemical reactions were as significant as those during
486 the haze period. The important difference is the presence and impact of the mineral dust
487 particles, and their role as interfaces for sulphates formation and the environmental and
488 adverse human health impacts resulting from dust storms.

489 **5. Conclusions**

490 (1) The dust storms that occurred in Beijing from March 15th to 20th, 2021, consist
491 of three recognisable stages; initiation, attenuation and culmination, with the initiation
492 being associated with the highest PM_{10} mass concentration and lowest horizontal
493 visibility. The dust storm particles can be subdivided into types consisting of minerals,
494 soot, and organic particles identified microscopically. The relative number percentage
495 of minerals continuously decreased, while that of soot and organic particles
496 correspondingly increased. The number-size of particles showed a unimodal
497 distribution with the peak in the 1-2 μm size range during the initiation and attenuation
498 stages, whereas the particles were mainly less than 1 μm in the culmination stage.

499 (2) Dust plumes consisted of mineral particles from the source area, and later
500 added particles during the transportation. Minerals provide an interface for atmospheric
501 chemical reactions, which leads to the modification of particulate composition and an
502 increase in the types of minerals. The predominant mineral type in the initiation stage
503 was Si-rich particles, followed by Ca-rich, Fe-rich, and Ti-rich particles, with minor
504 amounts of S-containing minerals. During the attenuation and culmination stages, Al-

505 rich, Mg-rich, and other types of minerals became evident. The relative number
506 percentage of S-containing particles was increasing and the sum of the several
507 remaining mineral particles was decreasing.

508 (3) The number percentages of metal-containing dust particles were relatively low
509 in the initiation stage, but in the attenuation stage, they increased significantly,
510 demonstrating an increasing trend of anthropogenic emissions mixing into the dust
511 storm. The heavy metals in dust storm particles should be considered because their
512 bioavailability and potential oxidative damage to human health.

513 (4) Over the duration of the dust storm, the mass concentration of sulphur was
514 highest in the initiation stage, with the main type of sulphur being the crustal mineral
515 gypsum and a mixture of sulphur and clay. The rapid decrease of sulphur concentration
516 in the attenuation stage was caused by the dust plume sweeping away air pollutants.
517 The mass concentration of sulphur-containing mixtures in the initiation stage was
518 similar to that during haze pollution, suggesting that the secondary chemical reactions
519 during dust storm periods were as evident as in haze. In the dust culmination stage, the
520 mass concentration of sulphur slightly increased.

521

522 **Acknowledgments**

523 This study is supported by the National Natural Science Foundation of China
524 (Grant No. 42075107), the Fundamental Research Funds for the Central Universities,
525 and the Yueqi Scholar fund of China University of Mining and Technology (Beijing).

526

527 **References**

- 528 Aghababaeian, H., Ostadtaghizadeh, A., Ardalan, A., Asgary, A., Akbary, M.,
529 Yekaninejad, M.S., Stephens, C., 2021. Global health impacts of dust storms: A
530 systematic review. *Environmental health insights* 15(11786302211018390-
531 11786302211018390. 10.1177/11786302211018390.
- 532 Aydin, F., Aydin, I., Erdogan, S., Akba, O., Isik, B., Hamamci, C., 2012. Chemical
533 characteristics of settled particles during a dust-storm. *Polish Journal of*
534 *Environmental Studies* 21(3), 533-537.
- 535 Buseck, P.R., Posfai, M., 1999. Airborne minerals and related aerosol particles: Effects
536 on climate and the environment. *Proceedings of the National Academy of Sciences*
537 *of the USA* 96, 3372-3379.
- 538 Cao, Y.X., Shao, L.Y., Jones, T.J., Oliveira, M.L.S., Ge, S.Y., Feng, X.L., Silva, L.F.O.,
539 BéruBé, K.A., 2021, Multiple relationships between aerosol and COVID-19: A
540 framework for global studies. *Gondwana Research* 93, 243-251.
541 <https://doi.org/10.1016/j.gr.2021.02.002>
- 542 Cave, M.R., Wragg, J., Beriro, D.J., Vane, C., Thomas, R., Riding, M., Taylor, C., 2018.
543 An overview of research and development themes in the measurement and
544 occurrences of polyaromatic hydrocarbons in dusts and particulates. *Journal of*
545 *Hazardous Materials* 360, 373-390.
- 546 Chen, Q.C., Wang, M.M., Sun, H.Y., Wang, X., Wang, Y.Q., Li, Y.G., Zhang, L.X., Mu,
547 Z., 2018. Enhanced health risks from exposure to environmentally persistent free
548 radicals and the oxidative stress of PM_{2.5} from Asian dust storms in Erenhot,

549 Zhangbei and Jinan, China. *Environment International* 121, 260-268.
550 10.1016/j.envint.2018.09.012.

551 Chen, S., Huang, J., Li, J., Jia, R., Jiang, N., Kang, L., Ma, X., Xie, T., 2017.
552 Comparison of dust emissions, transport, and deposition between the Taklimakan
553 Desert and Gobi Desert from 2007 to 2011. *Science China Earth Sciences* 60(7),
554 1338-1355. 10.1007/s11430-016-9051-0.

555 Cheng, Y.F., Zheng, G.J., Wei, C., Mu, Q., Zheng, B., Wang, Z.B., Gao, M., Zhang, Q.,
556 He, K.B., Carmichael, G., Poschl, U., Su, H., 2016. Reactive nitrogen chemistry
557 in aerosol water as a source of sulfate during haze events in China. *Science*
558 *Advances* 2(12), ARTN e1601530. 10.1126/sciadv.1601530.

559 China Climate Bulletin, 2021. China Climate Bulletin
560 <http://www.cma.gov.cn/zfxxgk/gknr/qxbg/>. (Accessed 8 March 2022) (in Chinese).

561 China Meteorological Administration, 2021. China Meteorological Administration.
562 [http://www.nmc.cn/publish/environment/National-Bulletin-atmospheric-](http://www.nmc.cn/publish/environment/National-Bulletin-atmospheric-environment.htm)
563 [environment.htm](http://www.nmc.cn/publish/environment/National-Bulletin-atmospheric-environment.htm). (in Chinese).

564 Csavina J, Field J, Félix O, Corral-Avitia AY, Eduardo Sáez A, Betterton EA. Effect of
565 wind speed and relative humidity on atmospheric dust concentrations in semi-arid
566 climates. *Sci Total Environ*. 2014;487:82–90.

567 Feng, X.L., Shao, L.Y., Xi, C.X., Jones, T., Zhang, D.Z., BeruBe, K., 2020. Particle-
568 induced oxidative damage by indoor size-segregated particulate matter from coal-
569 burning homes in the Xuanwei lung cancer epidemic area, Yunnan Province, China.
570 *Chemosphere* 256. 127058-127058. 10.1016/j.chemosphere.2020.127058.

571 Feng, X.L., Shao, L.Y., Jones, T., Li, Y.W., Cao, Y.X., Zhang, M.Y., Ge, S.Y., Yang, C
572 - X., Lu, J., BeruBe, K., 2022. Oxidative potential and water-soluble heavy metals
573 of size-segregated airborne particles in haze and non-haze episodes: Impact of the
574 "Comprehensive Action Plan" in China. *Science of the Total Environment*
575 814(152774). 10.1016/j.scitotenv.2021.152774.

576 Filonchyk, M., 2022. Characteristics of the severe March 2021 Gobi Desert dust storm
577 and its impact on air pollution in China. *Chemosphere* 287.
578 10.1016/j.chemosphere.2021.132219.

579 Formenti, P., Schütz, L., Balkanski, Y., Desboeufs, K., Ebert, M., Kandler, K., Petzold,
580 A., Scheuven, D., Weinbruch, S., Zhang, D., 2011. Recent progress in
581 understanding physical and chemical properties of African and Asian mineral dust.
582 *Atmospheric Chemistry and Physics* 11(16), 8231-8256. 10.5194/acp-11-8231-
583 2011.

584 Fussell, J.C., Kelly, F.J., 2021. Mechanisms underlying the health effects of desert sand
585 dust. *Environment International* 157. 106790. 10.1016/j.envint.2021.106790.

586 Garcia-Guinea, J., Morales, S., Delgado, A., Recio, C., Calaforra, J.M., 2002.
587 Formation of gigantic gypsum crystals. *Journal of the Geological Society* 159,
588 347-350.

589 Hashizume, M., Kim, Y., Ng, C.F.S., Chung, Y., Madaniyazi, L., Bell, M.L., Guo, Y.L.,
590 Kan, H., Honda, Y., Yi, S.M., Kim, H., Nishiwaki, Y., 2020. Health Effects of
591 Asian Dust: A Systematic Review and Meta-Analysis. *Environmental Health*
592 *Perspectives* 128(6). 66001. 10.1289/EHP5312.

593 Hasunuma, H., Takeuchi, A., Ono, R., Amimoto, Y., Ha Hwang, Y., Uno, I., Shimizu,
594 A., Nishiwaki, Y., Hashizume, M., Askew, D.J., Odajima, H., 2021. Effect of Asian
595 dust on respiratory symptoms among children with and without asthma, and their
596 sensitivity. *Science of the Total Environment* 753 (ARTN 141585).
597 10.1016/j.scitotenv.2020.141585.

598 Hu, T.F., Wu, F., Song, Y.P., Liu, S.X., Duan, J., Zhu, Y.Q., Cao, J.J., Zhang, D.Z., 2022.
599 Morphology and mineralogical composition of sandblasting dust particles from
600 the Taklimakan Desert. *Science of the Total Environment* 834, 155315.
601 10.1016/j.scitotenv.2022.155315

602 Huang, J.P., Wang, T.H., Wang, W.C., Li, Z.Q., Yan, H.R., 2014. Climate effects of dust
603 aerosols over East Asian arid and semiarid regions. *Journal of Geophysical*
604 *Research-Atmospheres* 119(19), 11398-11416. 10.1002/2014jd021796.

605 Huang, K., Zhuang, G., Li, J., Wang, Q., Sun, Y., Lin, Y., Fu, J.S., 2010. Mixing of
606 Asian dust with pollution aerosol and the transformation of aerosol components
607 during the dust storm over China in spring 2007. *Journal of Geophysical Research*
608 115(10.1029/2009jd013145).

609 Huang, R.J., Cheng, R., Jing, M., Yang, L., Li, Y., Chen, Q., Chen, Y., Yan, J., Lin, C.,
610 Wu, Y., Zhang, R., El Haddad, I., Prevot, A.S.H., O'Dowd, C.D., Cao, J., 2018.
611 Source-specific health risk analysis on particulate trace elements: coal combustion
612 and traffic emission as major contributors in wintertime Beijing. *Environmental*
613 *Science & Technology* 52(19), 10967-10974. 10.1021/acs.est.8b02091.

614 Huang, Z.W., Huang, J.P., Hayasaka, T., Wang, S.S., Zhou, T., Jin, H.C., 2015. Short-

615 cut transport path for Asian dust directly to the Arctic: a case study. *Environmental*
616 *Research Letters* 10(11), Artn 114018. 10.1088/1748-9326/10/11/114018.

617 Jeong, G.Y., 2008. Bulk and single-particle mineralogy of Asian dust and a comparison
618 with its source soils. *Journal of Geophysical Research-Atmospheres* 113(D2), Artn
619 D02208. 10.1029/2007jd008606.

620 Jeong, G.Y., 2020. Mineralogy and geochemistry of Asian dust: dependence on
621 migration path, fractionation, and reactions with polluted air. *Atmospheric*
622 *Chemistry and Physics* 20(12), 7411-7428.

623 Jones, T.P., Williamson, B.J., BeruBe, K.A., Richards, R.J., 2001. Microscopy and
624 chemistry of particles collected on TEOM filters: Swansea, south Wales, 1998-
625 1999. *Atmospheric Environment* 35 (21), 3573-3583.

626 Kim, W.H., Song, J.M., Ko, H.J., Kim, J.S., Lee, J.H., Kang, C.H., 2012. Comparison
627 of chemical compositions of size-segregated atmospheric aerosols between Asian
628 dust and non-Asian dust periods at background area of Korea. *Bulletin of the*
629 *Korean Chemical Society* 33(11), 3651-3656. 10.5012/bkcs.2012.33.11.3651.

630 Li, J., Shao, L.Y., Chang, L.L., Xing, J.P., Wang, W.H., Li, W.J., Zhang, D.Z., 2018.
631 Physicochemical characteristics and possible sources of individual mineral
632 particles in a dust storm episode in Beijing, China. *Atmosphere* 9(7), ARTN 269.
633 10.3390/atmos9070269.

634 Li, W.J., Shao, L.Y., 2012a. Chemical modification of dust particles during different
635 dust storm episodes. *Aerosol and Air Quality Research* 12(6), 1095-1104.
636 10.4209/aaqr.2011.11.0188.

637 Li, W.J., Shao, L.Y., Li, Z.P., Li, H., Gao, J., Li, J.J., Zhang, H., Zhang, Z.Z., Silva,
638 L.F.O., Zhang, M.Y., Chen, Y.Z., Oliveira, M.L.S., 2022. Surface chemistry of
639 atmospheric nanoparticles during a haze episode in Beijing by TOF-SIMS.
640 *Gondwana Research* 110, 305-318. 10.1016/j.gr.2022.02.013.

641 Li, W.J., Wang, T., Zhou, S.Z., Lee, S., Huang, Y., Gao, Y., Wang, W.X., 2013.
642 Microscopic observation of metal-containing particles from chinese continental
643 outflow observed from a non-industrial site. *Environmental Science & Technology*
644 47(16), 9124-9131. 10.1021/es400109q.

645 Li, W.J., Shao, L.Y., Shi, Z.B., Chen, J.M., Yang, L.X., Yuan, Q., Yan, C., Zhang, X.Y.,
646 Wang, Y.Q., Sun, J.Y., Zhang, Y.M., Shen, X.J., Wang, Z.F., Wang, W.X., 2014.
647 Mixing state and hygroscopicity of dust and haze particles before leaving Asian
648 continent. *Journal of Geophysical Research-Atmospheres* 119(2), 1044-1059.
649 10.1002/2013jd021003.

650 Li, W.J., Xu, L., Liu, X.H., Zhang, J.C., Lin, Y.T., Yao, X.H., Gao, H.W., Zhang, D.Z.,
651 Chen, J.M., Wang, W.X., Harrison, R.M., Zhang, X.Y., Shao, L.Y., Fu, P.Q., Nenes,
652 A., Shi, Z.B., 2017. Air pollution-aerosol interactions produce more bioavailable
653 iron for ocean ecosystems. *Science Advance* 3(3), e1601749.
654 10.1126/sciadv.1601749.

655 Li, W.J., Shao, L.Y., 2009. Observation of nitrate coatings on atmospheric mineral dust
656 particles. *Atmospheric Chemistry and Physics* 9, 1863-1871.

657 Li, W.J., Shao, L.Y., Zhang, D.Z., Ro, C.U., Hu, M., Bi, X.H., Geng, H., Matsuki, A.,
658 Niu, H.Y., Chen, J.M., 2016. A review of single aerosol particle studies in the

659 atmosphere of East Asia: morphology, mixing state, source, and heterogeneous
660 reactions. *Journal of Cleaner Production* 112(1330-1349).
661 10.1016/j.jclepro.2015.04.050.

662 Liu, P.J., Shao, L.Y., Li, Y.W., Wang, W.H., Zhang, M.Y., Yang, C.X., Niu, H.Y., Feng,
663 X.L., Zhang, D.Z., 2022. Compositions, sources, and aging processes of aerosol
664 particles during winter hazes in an inland megacity of NW China. *Atmosphere* 13
665 (4), ARTN 521. 10.3390/atmos13040521.

666 Luo, H., Wang, Q., Guan, Q., Ma, Y., Ni, F., Yang, E., Zhang, J., 2022a. Heavy metal
667 pollution levels, source apportionment and risk assessment in dust storms in key
668 cities in Northwest China. *Journal of Hazardous Materials* 422(126878).
669 10.1016/j.jhazmat.2021.126878.

670 Luo, J., Huang, F., Gao, S., Liu, S., Liu, R., Devasthale, A., 2022b. Satellite monitoring
671 of the dust storm over Northern China on 15 March 2021. *Atmosphere* 13(2),
672 10.3390/atmos13020157.

673 Lv, S.L., Shao, L.Y., 2003. Mineral compositions of individual particulates in PM10
674 over Beijing. *ACTA Petrologica et Mineralogica* 22(4), 421-424. DOI:
675 10.3969/j.issn.1000-6524.2003.04.023. (in Chinese with English abstract)

676 Ma, C.J., Kasahara, M., Holler, R., Kamiya, T., 2001. Characteristics of single particles
677 sampled in Japan during the Asian dust-storm period. *Atmospheric Environment*
678 35(15), 2707-2714. Doi 10.1016/S1352-2310(00)00410-6.

679 Maleki, H., Sorooshian, A., Alam, K., Fathi, A., Weckwerth, T., Moazed, H., Jamshidi,
680 A., Babaei, A.A., Hamid, V., Soltani, F., Goudarzi, G., 2022. The impact of

681 meteorological parameters on PM10 and visibility during the Middle Eastern dust
682 storms. *Journal of Environmental Health Science and Engineering* 10.1007/s40201-
683 022-00795-110.1007/s40201-022-00795-1.

684 Manktelow, P.T., Carslaw, K.S., Mann, G.W., Spracklen, D.V., 2010. The impact of dust
685 on sulfate aerosol, CN and CCN during an East Asian dust storm. *Atmospheric*
686 *Chemistry and Physics* 10(2), 365-382. 10.5194/acp-10-365-2010.

687 Nie, W., Wang, T., Xue, L. K., Ding, A. J., Wang, X. F., Gao, X. M., Xu, Z., Yu, Y. C.,
688 Yuan, C., Zhou, Z. S., Gao, R., Liu, X. H., Wang, Y., Fan, S. J., Poon, S., Zhang,
689 Q. Z., Wang, W. X., Asian dust storm observed at a rural mountain site in southern
690 China: chemical evolution and heterogeneous photochemistry, *Atmos. Chem.*
691 *Phys.*, 12, 11985–11995, <https://doi.org/10.5194/acp-12-11985-2012>, 2012.

692 Okada, K., Qin, Y., Kai, K., 2005. Elemental composition and mixing properties of
693 atmospheric mineral particles collected in Hohhot, China. *Atmospheric Research*
694 73(1-2), 45-67. 10.1016/j.atmosres.2004.08.001.

695 Pan, X.L., Uno, I., Wang, Z., Nishizawa, T., Sugimoto, N., Yamamoto, S., Kobayashi,
696 H., Sun, Y.L., Fu, P.Q., Tang, X., Wang, Z.F., 2017. Real-time observational
697 evidence of changing Asian dust morphology with the mixing of heavy
698 anthropogenic pollution. *Scientific Reports* 7 (ARTN 335).10.1038/s41598-017-
699 00444-w.

700 Raffaele, L., Bruno, L., Sherman, D.J., 2020. Statistical characterization of
701 sedimentation velocity of natural particles. *Aeolian Research* 44.
702 10.1016/j.aeolia.2020.100593.

703 Shao, L.Y., Hu, Y., Fan, J., Wang, J., Wang, J., Ma, J., 2017. Physicochemical
704 characteristics of aerosol particles in the Tibetan Plateau: Insights from TEM-EDX
705 analysis. *Journal of Nanoscience and Nanotechnology* 17(9), 6899-6908.
706 10.1166/jnn.2017.14472.

707 Shao, L.Y., Li, J., Zhang, M.Y., Wang, X., Li, Y.W., Jones, T., Feng, X.L., Silva, L.F.O.,
708 Li, W.J., 2021. Morphology, composition and mixing state of individual airborne
709 particles: Effects of the 2017 Action Plan in Beijing, China. *Journal of Cleaner*
710 *Production* 329. 10.1016/j.jclepro.2021.129748.

711 Shao, L.Y., Liu, P.J., Jones, T., Yang, S.S., Wang, W.H., Zhang, D.Z., Li, Y.W., Yang,
712 C.-X., Xing, J.P., Hou, C., Zhang, M.Y., Feng, X.L., Li, W.J., Bérubé, K., 2022. A
713 review of atmospheric individual particle analyses: Methodologies and
714 applications in environmental research. *Gondwana*
715 *Research* .10.1016/j.gr.2022.01.007.

716 Shao, L.Y., Li, W.J., Xiao, Z.H., Sun, Z.Q., 2008. The mineralogy and possible sources
717 of spring dust particles over Beijing. *Advances in Atmospheric Sciences* 25, 395-
718 403.

719 Shi, Z.B., Shao, L.Y., Jones, T.P., Lu, S.L., 2005. Microscopy and mineralogy of
720 airborne particles collected during severe dust storm episodes in Beijing, China.
721 *Journal of Geophysical Research* 110 (d1), pp. 1-10.

722 Soleimani, Z., Teymouri, P., Bolorani, A.D., Mesdaghinia, A., Middleton, N., Griffin,
723 D.W., 2020. An overview of bioaerosol load and health impacts associated with
724 dust storms: A focus on the Middle East. *Atmospheric Environment* 223(ARTN

725 117187). 10.1016/j.atmosenv.2019.117187.

726 Song, L.L., Bi, X.H., Zhang, Z.Y., Li, L.X., Dai, Q.L., Zhang, W.H., Li, H., Wang, X.H.,
727 Liang, D.N., Feng, Y.C., 2022. Impact of sand and dust storms on the atmospheric
728 environment and its source in Tianjin-China. *Science of the Total Environment*
729 825(10.1016/j.scitotenv.2022.153980).

730 Song, Z.X., Wang, J.Y., Wang, S.G., 2007. Quantitative classification of northeast Asian
731 dust events. *Journal of Geophysical Research* 112. 10.1029 /2006JD007048.

732 Stein, A.F., Draxler, R.R., Rolph, G.D., Stunder, B.J.B., Cohen, M.D., Ngan, F., 2015.
733 NOAA's HYSPLIT atmospheric transport and dispersion modeling system. *Bulletin of*
734 *the American Meteorological Society* 96(12), 2059-2077. 10.1175/BAMS-D-14-
735 00110.1.

736 Sun, Y.L., Zhuang, G.S., Yuan, H., Zhang, X.Y., Guo, J.H., 2004. Characteristics and
737 sources of 2002 super dust storm in Beijing. *Chinese Science Bulletin* 49(7), 698-
738 705. 10.1360/03wb0157.

739 Tang, K., Huang, Z.W., Huang, J.P., Maki, T., Zhang, S., Shimizu, A., Ma, X.J., Shi,
740 J.S., Bi, J.R., Zhou, T., Wang, G.Y., Zhang, L., 2018. Characterization of
741 atmospheric bioaerosols along the transport pathway of Asian dust during the
742 Dust-Bioaerosol 2016 Campaign. *Atmospheric Chemistry and Physics* 18(10),
743 7131-7148. 10.5194/acp-18-7131-2018.

744 Tang, M.J., Huang, X., Lu, K.D., Ge, M.F., Li, Y.J., Cheng, P., Zhu, T., Ding, A.J.,
745 Zhang, Y.H., Gligorovski, S., Song, W., Ding, X., Bi, X.H., Wang, X.M., 2017.
746 Heterogeneous reactions of mineral dust aerosol: implications for tropospheric

747 oxidation capacity. *Atmospheric Chemistry and Physics* 17(19), 11727-11777.
748 10.5194/acp-17-11727-2017.

749 Uno, I., Eguchi, K., Yumimoto, K., Takemura, T., Shimizu, A., Uematsu, M., Liu, Z.Y.,
750 Wang, Z.F., Hara, Y., Sugimoto, N., 2009. Asian dust transported one full circuit
751 around the globe. *Nature Geoscience* 2(8), 557-560. 10.1038/Ngeo583.

752 Wang, W.H., Shao, L.Y, Zhang, D.Z., Li, Y.W., Li, W.J., Liu, P.J., Xing, J.P., 2022.
753 Mineralogical similarities and differences of dust storm particles at Beijing from
754 deserts in the north and northwest. *Science of the Total Environment* 803(149980).
755 10.1016/j.scitotenv.2021.149980.

756 Wang, W.H., Shao, L.Y., Guo, M.L., Hou, C., Xing, J.P., Wu, F., 2017. Physicochemical
757 properties of individual airborne particles in Beijing during pollution periods.
758 *Aerosol and Air Quality Research* 17(12), 3209-3219.

759 Wang, Z., Hu, W., Niu, H., Hu, W., Wu, Y., Wu, L., Ren, L., Deng, J., Guo, S., Wu, Z.,
760 Zhang, D., Fu, P., Hu, M., 2021. Variations in physicochemical properties of
761 airborne particles during a heavy haze-to-dust episode in Beijing. *Science of the*
762 *Total Environment* 762, 10.1016/j.scitotenv.2020.143081.

763 Wang, Z., Pan, X.L., Uno, I., Chen, X.S., Yamamoto, S., Zheng, H.T., Li, J., Wang,
764 Z.F., 2018. Importance of mineral dust and anthropogenic pollutants mixing
765 during a long-lasting high PM event over East Asia. *Environmental Pollution* 234
766 368-378. 10.1016/j.envpol.2017.11.068.

767 Wen, H., Zhou, Y., Xu, X.Y., Wang, T.S., Chen, Q.L., Chen, Q.C., Li, W.J., Wang, Z.,
768 Huang, Z.W., Zhou, T., Shi, J.S., Bi, J.R., Ji, M.X., Wang, X., 2021. Water-soluble

769 brown carbon in atmospheric aerosols along the transport pathway of Asian dust:
770 Optical properties, chemical compositions, and potential sources. *Science of the*
771 *Total Environment* 789, 10.1016/j.scitotenv.2021.147971.

772 Wu, F., Zhang, D.Z., Cao, J.J., Guo, X., Xia, Y., Zhang, T., Lu, H., Cheng, Y., 2017.
773 Limited production of sulfate and nitrate on front-associated dust storm particles
774 moving from desert to distant populated areas in northwestern China. *Atmospheric*
775 *Chemistry and Physics* 17(23), 14473-14484. 10.5194/acp-17-14473-2017.

776 Wu, Z.X., Hu, T.F., Hu, W., Shao, L.Y., Sun, Y.Z., Xue, F.L., Niu, H.Y., 2022. Evolution
777 in physicochemical properties of fine particles emitted from residential coal
778 combustion based on chamber experiment. *Gondwana Research* 110, 252-263.
779 10.1016/j.gr.2021.10.017.

780 Xiao, Z.H., Shao, L.Y., Zhang, N., Wang, J., Wang, J.Y., 2013. Heavy metal
781 compositions and bioreactivity of airborne PM10 in a valley-shaped city in
782 Northwestern China. *Aerosol and Air Quality Research* 13(3), 1116-1125.
783 10.4209/aaqr.2012.10.0287.

784 Xing, J.P., Shao, L.Y., Zheng, R., Peng, J.F., Wang, W.H., Guo, Q., Wang, Y.H., Qin,
785 Y.H., Shuai, S.J., Hu, M., 2017. Individual particles emitted from gasoline engines:
786 Impact of engine types, engine loads and fuel components. *Journal of Cleaner*
787 *Production* 149, 461-471. <https://doi.org/10.1016/j.jclepro.2017.02.056>.

788 Xu, L., Fukushima, S., Sobanska, S., Murata, K., Naganuma, A., Liu, L., Wang, Y.Y.,
789 Niu, H.Y., Shi, Z.B., Kojima, T., Zhang, D.Z., Li, W.J., 2020. Tracing the evolution
790 of morphology and mixing state of soot particles along with the movement of an

791 Asian dust storm. *Atmospheric Chemistry and Physics* 20(22), 14321-14332.
792 10.5194/acp-20-14321-2020.

793 Xu, P., Zhang, J.K., Ji, D.S., Liu, Z.R., Tang, G.Q., Hu, B., Jiang, C.S., Wang, Y.S.,
794 2017. Evaluating the effects of springtime dust storms over Beijing and the
795 associated characteristics of sub-micron aerosol. *Aerosol and Air Quality Research*
796 17(3), 680-692. 10.4209/aaqr.2016.05.0195.

797 Yu, Z., Jang, M., Kim, S., Bae, C., Koo, B., Beardsley, R., Park, J., Chang, L.S., Lee,
798 H.C., Lim, Y.-K., Cho, J.H., 2020. Simulating the impact of long-range-
799 transported Asian mineral dust on the formation of sulfate and nitrate during the
800 KORUS-AQ campaign. *ACS Earth and Space Chemistry* 4 (7), 1039–1049.

801 Zhang, R., Han, Z.W., Shen, Z.X., Cao, J., 2008. Continuous measurement of number
802 concentrations and elemental composition of aerosol particles for a dust storm
803 event in Beijing. *Advances in Atmospheric Sciences* 25(1), 89-95.
804 10.1007/s00376-008-0089-2.

805 Zhao, J., Zhang, F., Xu, Y., Chen, J., Yin, L., Shang, X., Xu, L., Chemical characteristics
806 of particulate matter during a heavy dust episode in a coastal city, Xiamen, 2010,
807 *Aerosol Air Qual. Res.*, 11, 299–308, <https://doi.org/10.4209/aaqr.2010.09.0073>,
808 2011.

809 Zhao, X.J., Zhuang, G.S., Wang, Z.F., Sun, Y.L., Wang, Y., Yuan, H., 2007. Variation of
810 sources and mixing mechanism of mineral dust with pollution aerosol - revealed
811 by the two peaks of a super dust storm in Beijing. *Atmospheric Research* 84(3),
812 265-279. 10.1016/j.atmosres.2006.08.005.

813 Zhou, J.C., Swietlicki, E., Hansson, H.C., Artaxo, P., 2002. Submicrometer aerosol
814 particle size distribution and hygroscopic growth measured in the Amazon rain
815 forest during the wet season. *Journal of Geophysical Research-Atmospheres* 107
816 (D20),
817

Figures and Tables:

Figures:

Fig. 1 The mass concentrations of PM₁₀ and PM_{2.5} and the value of air quality index (AQI) in dust episode

Fig. 2 The relative number percentage of individual particles in PM₁₀ during the dust sampling period

Fig. 3 The different types of mineral particles in dust pollution

Fig. 4 Number-size distribution of dust particle

Fig. 5 Air mass backward trajectories in March 2021 at the sampling site in Beijing

Fig. 6 Number-size distribution of mineral particles in different dust pollution stages

Fig. 7 Atomic weight ratio of elements in dust particles at different pollution stages

Fig. 8 Mass concentration of S in different dust storm stages and haze pollution in Beijing. Data for the dust storm of 2015 were from Wang et al. (2022) and data for the haze and non-haze days of 2017 were from Shao et al., (2021).

Tables:

Table 1. Sampling periods and meteorological parameters during the dust episodes

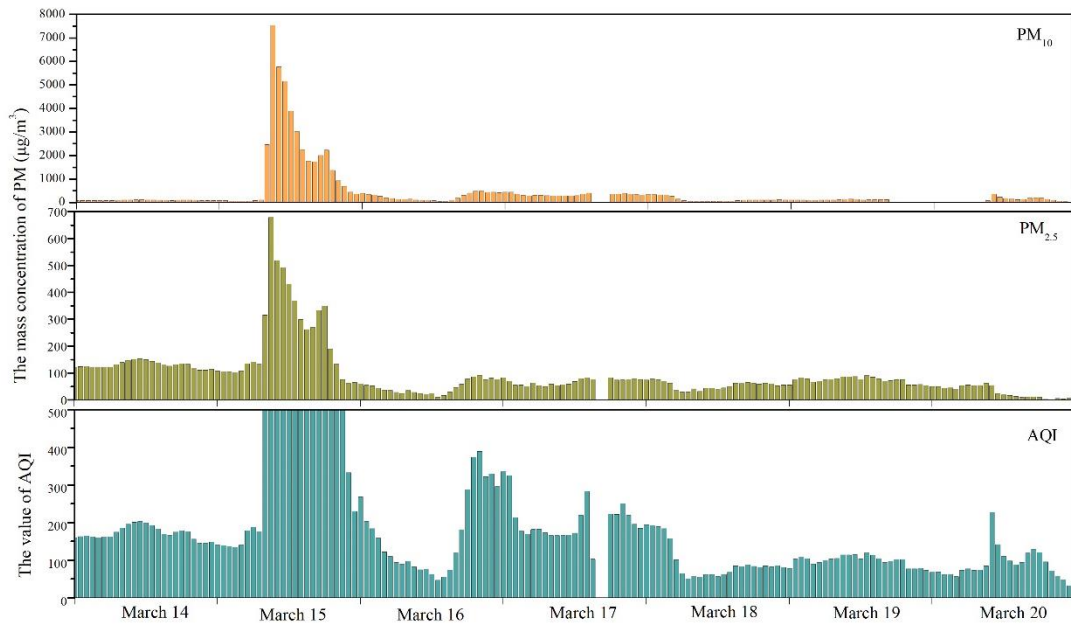


Fig. 1. The hourly mass concentrations of PM₁₀ and PM_{2.5} and the value of air quality index (AQI) in dust episode

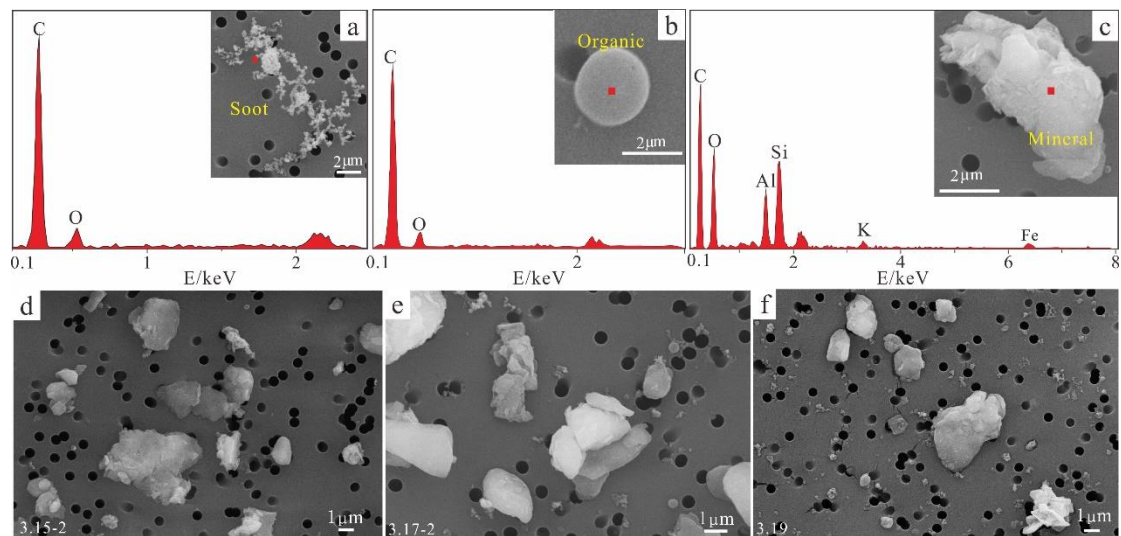


Fig. 2. Morphology and elemental composition of different particle types during dust pollution. (a) soot is mainly composed of C and O, whose morphology is mainly a chain aggregate being composed of carbon spheres from Sample 3.16-2; (b) organic particles are also composed of C and O and the SEM image showed spherical or nearly spherical particles from Sample 3.17-1; (c) mineral particles are mainly composed of Si and Al,

with a small amount of K, Fe. and their morphologies are mainly irregular shape from Sample 3.17-1; (d) the sample collected in the dust initiation stage, when the mineral particles were the dominant. Sample 3.15-2 (number percentage of mineral particles is 97.76%); (e) the sample collected in the dust attenuation stage. The relative number percentage of mineral particles was lower than that in dust initiation stage. Samples 3.17-2 (number percentage of mineral particles is 78.56%, with minor organic particles); (f) the samples collected in the dust culmination stage, with obviously lowered number percentage of minerals. Samples 3.19 (number percentage of mineral particles is 55.75%).

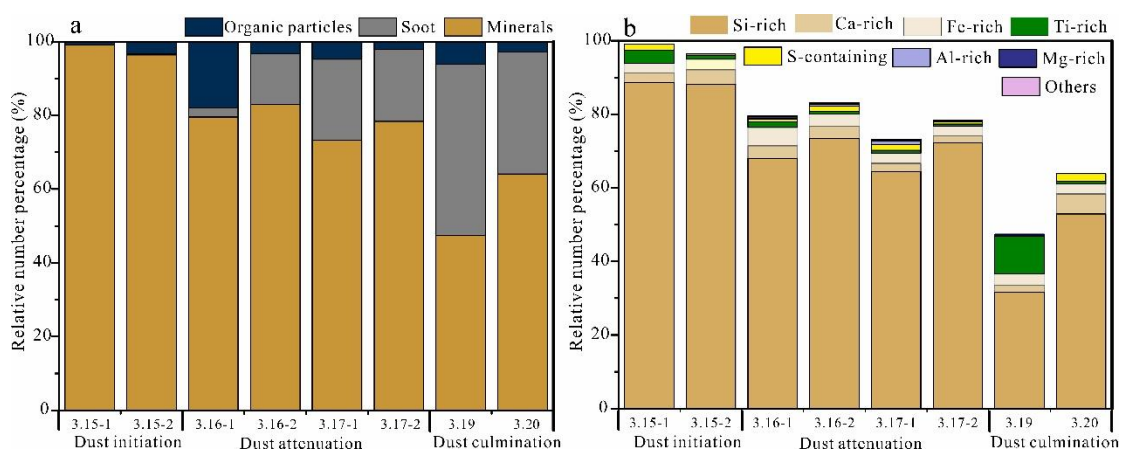


Fig. 3. The relative number percentage of individual particles in PM₁₀ during the dust sampling period. (a) refers to the relative proportions of mineral, soot and organic particles in the total analysed particles. (b) refers to the relative proportions of different types of mineral particles in the various pollution stages.

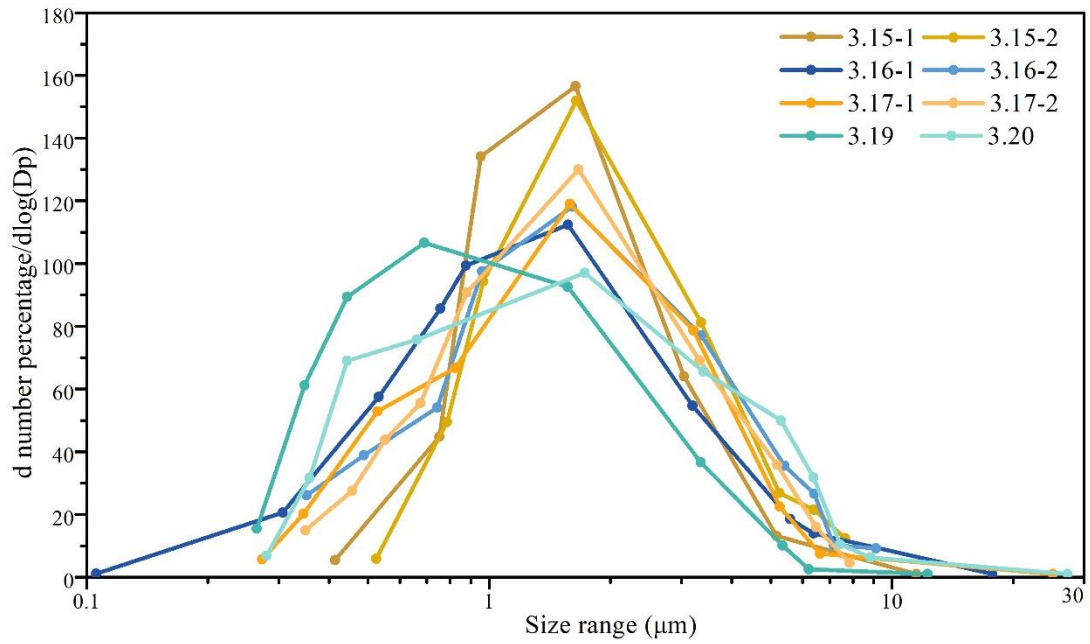


Fig. 4. Number percentage-size distribution of dust particle in PM₁₀. In the 1-2 μm size range, the number of individual particles in the dust storm period (3.15) was highest and the number in 3.17 was higher than that in 3.16, indicating that dust mass carried the coarse particles. In less than 1 μm size range, the number of particles was highest in non-dust period, followed by that in 3.17, which suggested that secondary reactions product the smaller particles

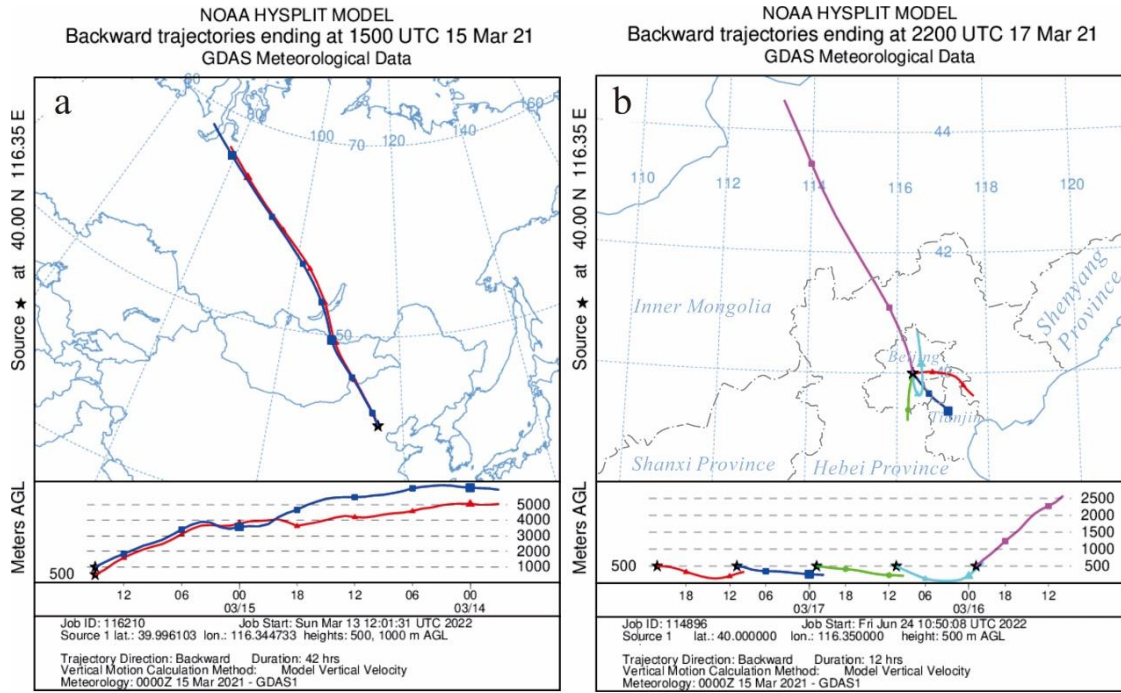


Fig. 5. Air mass backward trajectories from March 15 to March 18, 2021, at the sampling site in Beijing

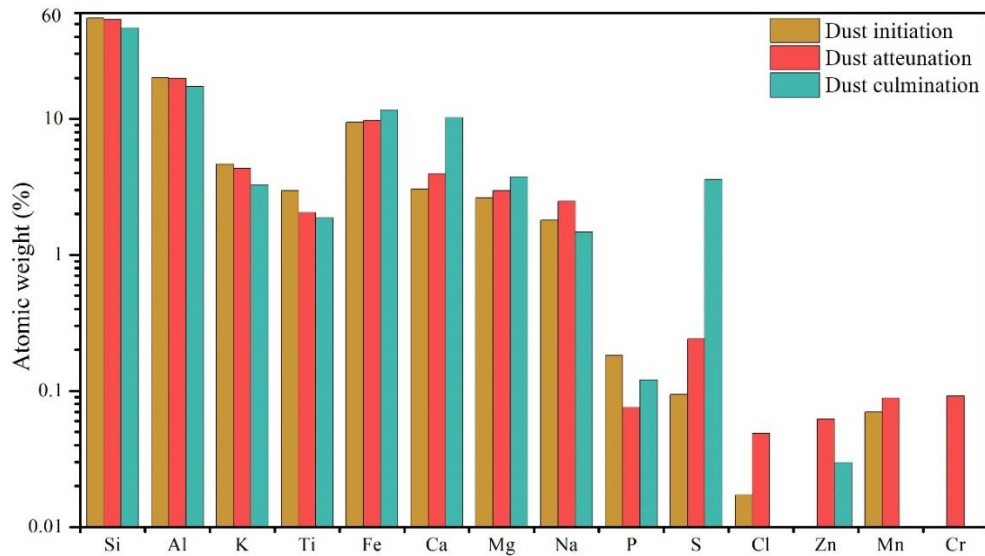


Fig. 6. Atomic weight ratio of elements in PM_{10} during different dust pollution stages. The elemental atomic weight ratios of Si, Al, K, and Ti were decreasing during the dust storm. Fe, Ca, and Mg were increasing during the dust storm. Zn, Mn, and Cr were least in the dust initiation stage. S was increasing in the dust storm.

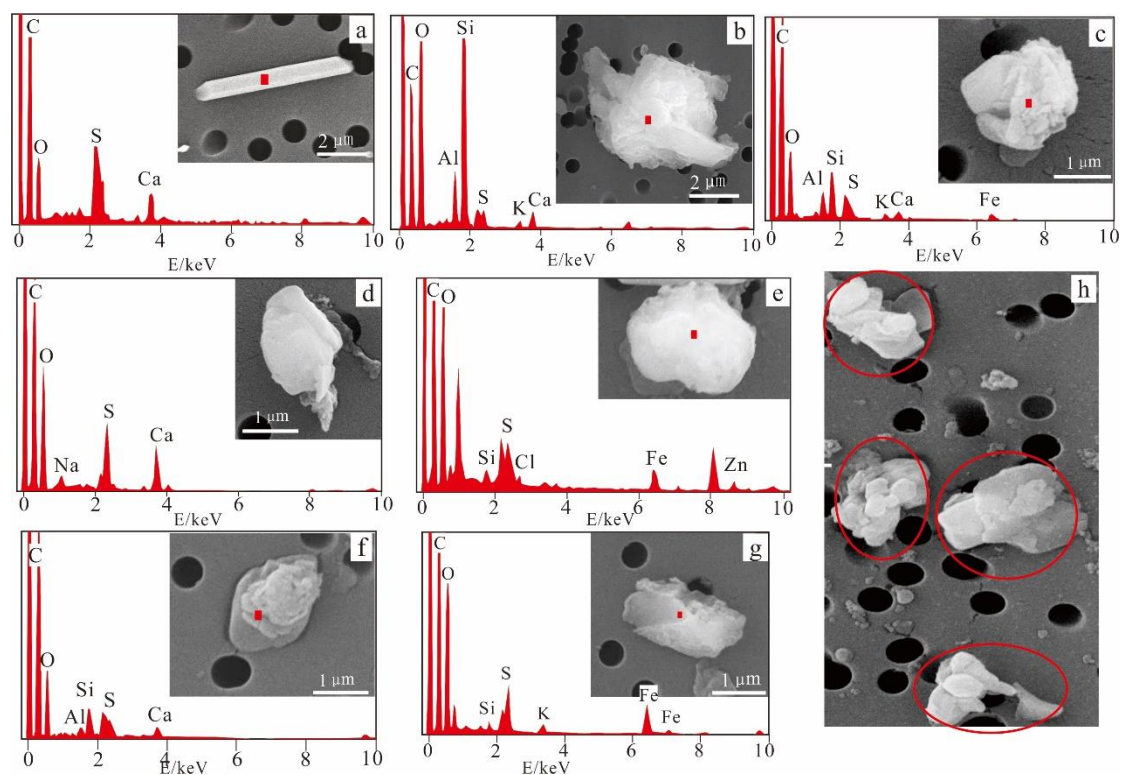


Fig. 7. SEM images and elemental compositions of S-containing minerals. (a) Rod-shaped gypsum collected in March 15; (b) clay mineral with a small amount of S collected in March 16; (c) the mixture of S and clay mineral collected in March 17; (d) irregular gypsum and (e) the mixture of S and clay minerals, both collected in March 16; (f) gypsum attached in clay mineral and (g) S attached in clay mineral collected in March 20; (h) the mixture of S and clay mineral collected on March 19

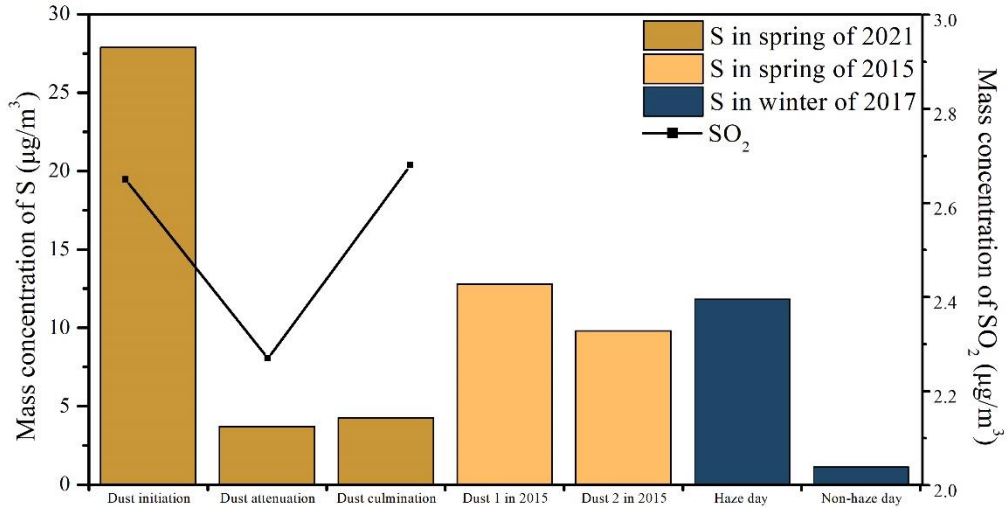


Fig. 8. Mass concentration of S in different dust storm stages and haze pollution in Beijing. Data for the dust storm of 2015 were from Wang et al. (2022) and data for the haze and non-haze days of 2017 were from Shao et al., (2021).

Table 1 Sampling periods and meteorological parameters during the dust episodes

	Sampling ID	Sampling Period, Beijing time	Meteorological Parameters		
			T (°C)	RH (%)	P (hPa)
Dust initiation	3.15-1	15:30-17:30	11.16	13.94	1010.80
	3.15-2	21:00-2:00	8.98	15.65	1014.70
Dust attenuation	3.16-1	8:40-12:10	13.16	13.03	1017.94
	3.16-2	20:30-2:30	12.26	22.86	1013.85
	3.17-1	8:30-12:30	13.14	36.16	1015.44
	3.17-2	15:13-19:00	12.75	29.13	1014.44
Dust	3.19	10:40-16:40	11.42	47.28	1012.51
culmination	3.20	9:30-21:30	11.80	26.29	1012.18

Beijing time: UTC plus 8 hours, T: temperature, RH: relative humidity, P: pressure.

Supplement material

Figures and Tables:

Fig. S1 The daily mass concentrations of PM₁₀ and PM_{2.5} and the value of air quality index (AQI) in Beijing in spring 2021. The value of AQI and PM came from China National Environmental Monitoring Centre (<http://www.cnemc.cn/>). The dust pollution data came from China Meteorological Administration (China Meteorological Administration, 2021).

Fig. S2 The distribution map of Wanliu state-owned automatic air quality monitoring station and sampling site

Fig. S3 Overall detection frequencies of elements in dust particles at different pollution stages

Fig. S4 The different types of mineral particles in dust pollution

Table S1 The number of mineral particles in different dust pollution stages

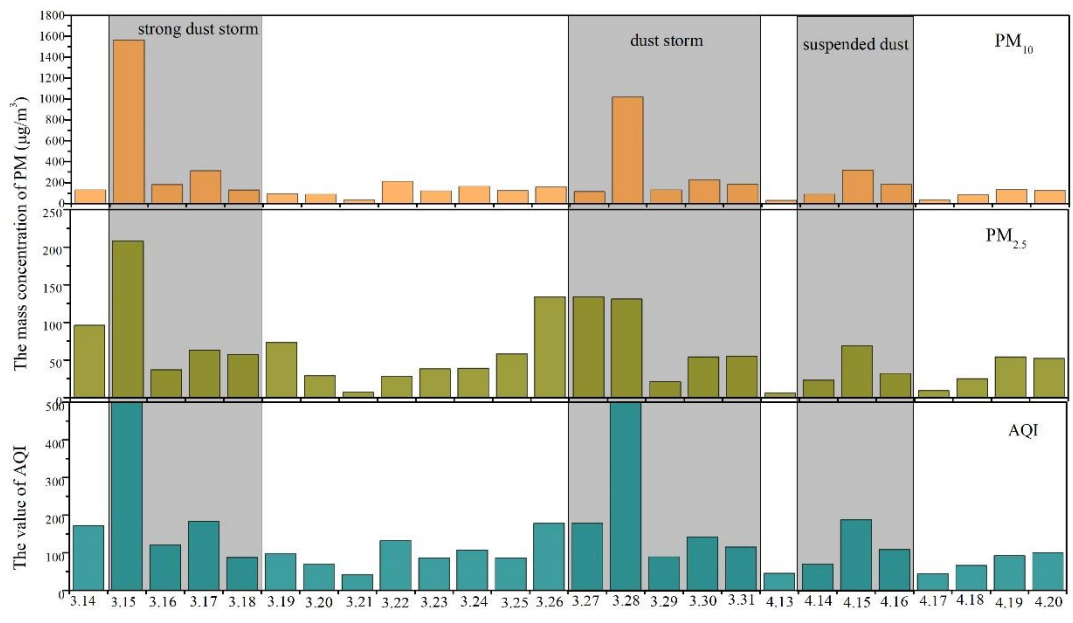


Fig. S1 The daily mass concentrations of PM₁₀ and PM_{2.5} and the value of air quality index (AQI) in Beijing in spring 2021. The value of AQI and PM came from China National Environmental Monitoring Centre (<http://www.cnemc.cn/>). The dust pollution data came from China Meteorological Administration (China Meteorological Administration, 2021).



Fig. S2 The distribution map of Wanliu state-owned automatic air quality monitoring station and sampling site

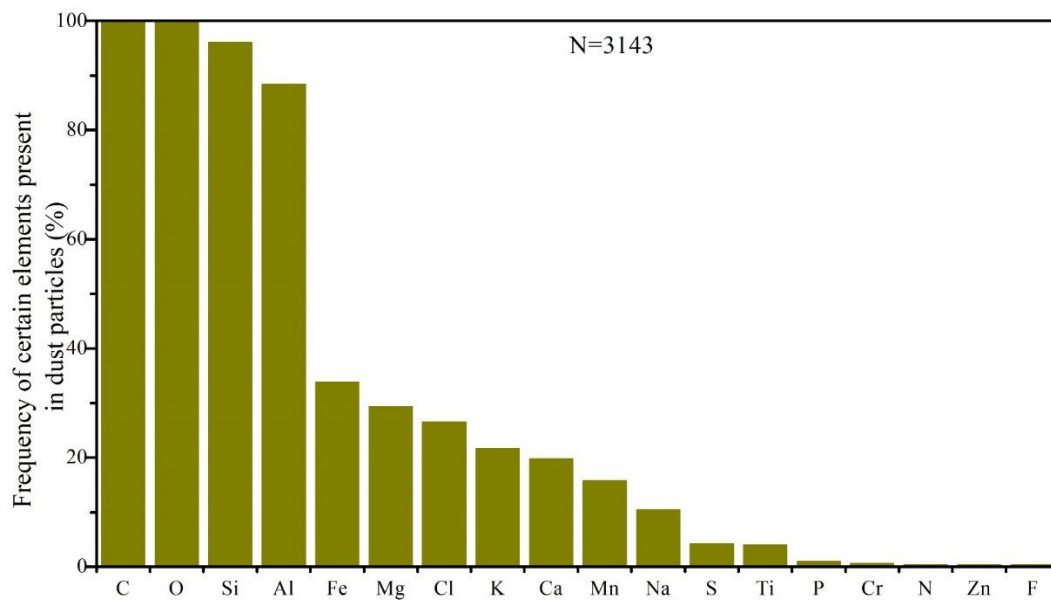


Fig. S3 Overall detection frequencies of elements in dust particles

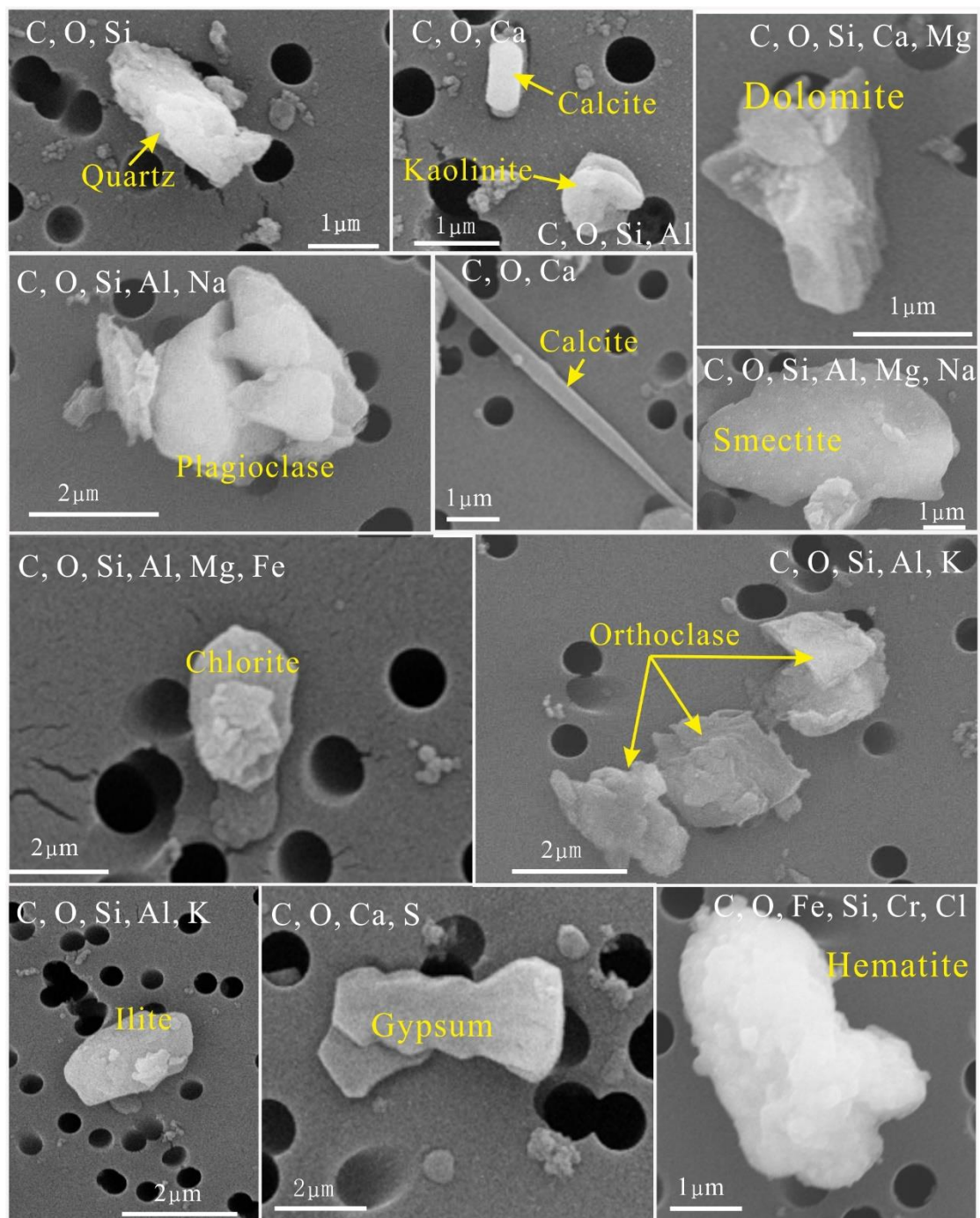


Fig. S4 The different types of mineral particles in dust pollution

Table S1 The number of mineral particles in different dust pollution stages

	Ca-rich				Fe-rich			Ti-rich		Al-rich		Mg-rich	
	Ca-	Ca+	Ca+	Oth	Fe-	Fe+	Oth	Ti-	Ti+	Al-	Al+	Mg-	Mg+
	dominated	Mg	Si	er	dominated	Si	er	dominated	Si	dominated	Si	dominated	Si
Dust initiation	4	2	7	1	7	4		5	2				
Dust attenuation	21	8	10	1	21	24	5	11	2	4	4	3	1
Dust culmination	22	5	1	1	12	11	4	2		1	3		

	Si-rich									S-containing	others
	Si-dominated	Si+Al	Si+Fe	Si+Ca	Si+Na	Si+Mg	Si+K	Other			
Dust initiation	89	219	20	6	3	1	6	2		3	
Dust attenuation	253	645	62	8	34	2	25	3		16	2

Dust culmination

96

204

30

11

6

8

5

67
