

1 Sulfur-containing particles emitted by concealed sulfide ore deposits: An
2 unknown source of sulfur-containing particles in the atmosphere

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10 Abstract

11 Sources of sulfur dioxide, sulfates, and organic sulfur compounds, such as fossil fuels,
12 volcanic eruptions, and animal feeding operations, have attracted considerable
13 attention. In this study, we collected particles carried by geogas flows ascending
14 through soil, geogas flows above the soil that had passed through the soil, and geogas
15 flows ascending through deep faults of concealed sulfide ore deposits and analyzed
16 them using transmission electron microscopy. Numerous crystalline and amorphous
17 sulfur-containing particles or particle aggregations were found in the ascending
18 geogas flows. In addition to S, the particles contained O, Ca, K, Mg, Fe, Na, Pb, Hg,
19 Cu, Zn, As, Ti, Sr, Ba, Si, etc. Such particles are usually a few to several hundred
20 nanometers in diameter with either regular or irregular morphology. The
21 sulfur-containing particles originated from deep-seated weathering or faulting
22 products of concealed sulfide ore deposits. The particles suspended in the ascending

23 geogas flow migrated through faults from deep-seated sources to the atmosphere. This
24 is a previously unknown source of the atmospheric particles. This paper reports, for
25 the first time, the emission of sulfur-containing particles into the atmosphere from
26 concealed sulfide ore deposits. The climatic and ecological influences of these
27 sulfur-containing particles and particle aggregations should be assessed.

28 Keywords: sulfur-containing particles, ascending gas flow, unknown source, sulfide
29 ore deposits.

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33 1. Introduction

34 Sources of sulfur oxides, sulfates, and organic sulfur compounds are diverse and
35 associated with natural and anthropogenic activities. Known sources of sulfur are
36 volatile sulfur compounds derived from animal feeding operations (Trabue et al.,
37 2008), and aerobic decomposition of food waste (Wu et al., 2010), biogenic sulfur
38 from rice paddies (Yang et al., 1996; Yang et al., 1998) and the Subantarctic and
39 Antarctic Oceans (Berresheim, 1987), sulfur gas (H_2S and SO_2) from geothermal
40 fields (Kristmannsdottir et al., 2000), organic sulfur compounds from sediments and
41 immature crude oil (Sinninghe Damsté et al., 1988), sulfur oxides from the oxidation
42 of fossil fuels (Soleimani et al., 2007), and sulfur dioxide from acid factories and
43 volcanic eruptions (Wong 1978; Sweeney et al., 2008). Sulfate particles, which are
44 important anthropogenic aerosols and influencing climate (Pósfai et al., 1997;
45 Williams et al., 2001). Furthermore, volcanic activity is a major contributor of sulfur
46 to the atmosphere (Zreda-Gostynska et al., 1993; Graf et al., 1998; Streets et al., 2000;
47 Seino et al., 2004; Bhugwant et al., 2009; Bao et al., 2010; Gieré and Querol, 2010),
48 particularly in countries such as Japan, Indonesia, Réunion Island, the Philippines,
49 Iceland, Guatemala, and New Zealand (Rose et al., 1986; Andres et al., 1993; Streets
50 et al., 2000; Seino et al., 2004; Chenet et al., 2005; Bhugwant et al., 2009).

51 Stratospheric sulfur adds very little to the environmental consequences of the
52 anthropogenic sulfur that is released in the troposphere and deposits within days to
53 weeks (Wong, 1978; Chenet et al., 2005). Existing research shows that SO_2 is
54 oxidized to SO_4^{2-} in both the gas and liquid phases. Moreover, sulfate aerosols can

55 directly affect the climate (Graf et al., 1998). In our previous work, particles carried
56 by an ascending geogas flow in the soil (Holub et al., 1999, 2001; Cao et al., 2009,
57 2010b; Cao et al., 2011; Liu et al., 2011; Wei et al., 2013; [Hu et al., 2015](#)) were
58 studied and found to contain sulfur. Further research showed that sulfur-containing
59 particles carried by ascending geogas flows can be transported through the soil layers
60 and into the atmosphere. Sulfur-containing particles suspended in the ascending
61 geogas flow migrate through faults from deep-seated concealed sulfide ore deposits to
62 the Earth's surface. These particles are a previously unknown source of
63 sulfur-containing particles in the atmosphere. This paper reports, for the first time, the
64 emission of sulfur-containing particles into the atmosphere from concealed sulfide ore
65 deposits. Because concealed sulfide ore deposits are widely distributed, the influence
66 of sulfur-containing particles derived from them is important. The climatic and
67 ecological effects of these particles should be studied.

68 2. Methods

69 Particles carried by an ascending geogas flow above the soil (that had flown through
70 the soil), in the soil, and in deep-seated faults were collected at the Dongshengmiao
71 polymetallic sulfide deposit in the Inner Mongolia Autonomous Region, China.
72 Particles carried by the ascending gas flow in the soil were also collected at other
73 concealed ore deposits containing sulfide minerals, such as the Kafang copper deposit
74 of the southern Yunnan Province, the Yongshengde copper deposit in northeastern
75 Yunnan, and the Qingmingshan copper–nickel sulfide deposit in Guangxi Province,
76 China.

77 Particles transported by the ascending geogas flow above the soil (that had flown
78 through the soil) were sampled using stainless steel tubes and carbon-coated nickel
79 transmission electron microscopy (TEM) grids. The length of the stainless steel tubes
80 was 40 cm and their diameter was 2.8 cm. These tubes were inserted vertically into
81 the soil to a depth of about 30 cm. A carbon-coated nickel TEM grid was fixed at the
82 end of the stainless steel tubes. The ascending geogas flow in the soil moved into the
83 stainless steel tubes and naturally passed through the 30 cm soil layer. Then, the gas
84 flow passed through the 10 cm of the empty stainless steel tubes above the soil.
85 Finally, the geogas flow arrived at the top of the tubes. Particles carried by the geogas
86 flow were adsorbed onto the carbon-coated nickel TEM grid. A protective device was
87 installed on the outside of the steel tubes to ensure that particles sampled were those
88 carried by the ascending geogas flow. The protective device is a cylindrical
89 polyethyleneterephthalate bottle. A small hole at the side of the bottle allowed the
90 outflow of ascending geogas flow; however, adsorption material placed in the hole did
91 not allow the external particles to enter. Sampling devices were installed between July
92 25, and August 23, 2013, and the carbon-coated nickel TEM grids were retrieved on
93 September 8, 2013. Sampling sites were distributed across a fault above the concealed
94 sulfide ore bodies of the Dongshengmiao polymetallic sulfide deposit.

95 Particles transported by the ascending geogas flow in the soil were collected using
96 ordinary plastic funnels. An inverted funnel was inserted in a hole that was 60–80 cm
97 deep and backfilled with soil, and a TEM grid was fixed at the end of the funnel spout
98 with nylon net. The setup was protected from contamination using plastic pipes and

99 cups. The TEM grids were retrieved after 60 days.

100 Particles carried by ascending geogas flows in deep-seated faults were sampled using
101 two methods. The first method used an active sampling device with a vacuum pump,
102 polyvinyl chloride (PVC) pipe and carbon-coated nickel TEM grid as the main
103 components. One end of the PVC pipe was connected with a tubing to the pump. A
104 drilling steel was inserted slantwise into the fault. The inserted depth was 30–50 cm.
105 As the drilling steel was pulled out, the PVC pipe was inserted into the hole. The PVC
106 pipe was compacted using fault gouge. The impurity gases in the PVC pipe were
107 pumped out using the vacuum pump, then, the PVC pipe was quickly sealed. A day
108 later, we connected a tube equipped with a carbon-coated nickel TEM grid to the PVC
109 pipe. The gas was pumped using a vacuum pump and flowed through the TEM grid
110 for 1 to 2 hours. Particles carried by the gas were collected by the TEM grid. Finally,
111 the carbon-coated nickel TEM grid was removed and sealed in a sample cell. The
112 second method did not use a vacuum pump. A carbon-coated nickel TEM grid was
113 fixed to the end of the PVC pipe. The ascending geogas flow in the fault flowed into
114 the PVC pipe and arrived at the top of the PVC pipe naturally. The particles carried by
115 the geogas flows in the faults were adsorbed onto the carbon-coated nickel grid. The
116 sampling devices were installed on August 3–10, 2013, and the TEM grids were
117 retrieved on September 7, 2013.

118 High-resolution TEM analyses were performed using a Tecnai G2 F30 S-TWIN
119 instrument at Yangzhou University, China, using an accelerating voltage of 300 kV.
120 The grids were checked using TEM before sampling to ensure they were devoid of

121 particles.

122 3. Results

123 3.1 Sulfur-containing particles carried by an ascending geogas flow above the soil 124 (that had flown through the soil)

125 According to the TEM analysis, particles containing high levels of S, O, Pb, Zn, Fe,
126 Hg, As, etc, were found in the ascending gas flows above the soil above the
127 Dongshengmiao polymetallic sulfide deposit. Table 1 provides the number of
128 sulfur-containing particles or particle aggregations that were found on the 100 μm \times
129 100 μm TEM grid. In general, one aggregation included more than five particles.
130 Figure 1 shows an elliptical particle (ID: 1) having a diameter of 500 nm. The particle
131 contains 78.17% S and 18.47% O (Table 2). Its O to S atomic ratio is 0.47. Figure 2
132 shows a particle aggregation (ID: 2) that consists of several small particles having a
133 diameter of 3–8 nm. It contains 31.23% S and 59.29% Hg. The spacing of the lattice
134 fringes was measured to be 0.333 nm. Figure 3 shows particle aggregations (ID: 3)
135 with sizes of less than 100 nm. Their O to S atomic ratio is 0.51. The particle
136 aggregations contain 14.48% Pb. The particle (ID: 4) illustrated in Figure 4 is
137 elliptical with a diameter of 200 nm and contains 18.55% As, 54.2% Pb, and 8.34%
138 Zn. The particle (ID: 5) shown in Figure 5 contains 2.25% Co. It is amorphous and
139 has an O to S atomic ratio of 2.91. The particle aggregation (ID: 6) illustrated in
140 Figure 6 contains 62.39% Cu and consists of small particles each having a diameter of
141 5–10 nm. Figure 7 presents a particle aggregation (ID: 7) that consists of many small
142 particles with diameters of about 5 nm, and contains 69.28% Pb.

143 3.2 Sulfur-containing particles carried by an ascending gas flow in the soil

144 Numerous sulfur-containing particles transported by an ascending gas flow were
145 found in the soil over sulfide ore deposits. Figure 8 shows an aggregation of such
146 particles from the Dongshengmiao polymetallic sulfide deposit. The aggregation (ID:
147 8) may be composed of CaSO_4 with trace amounts of K, Mg, Fe, and Si. It is regularly
148 shaped and 300 nm in size. The selected area electron diffraction pattern shows that
149 the aggregation is polycrystalline, possibly gypsum. Figure 9 shows a TEM image of
150 a sulfur-containing particle (ID: 9) from the Kafang copper deposit, South China.
151 Sulfur accounts for 63.99% of the particle (Table 3), and its O to S atomic ratio is 0.83.
152 Its K content is 8.93%, and its size is 330 nm. Figure 10 shows a regularly polygonal
153 particle (ID: 10) from the Yongshengde copper deposit, China. Its O to S atomic ratio
154 is 3.60, and its Fe and F contents are 9.94% and 1.71%, respectively. Figure 11 shows
155 a sulfur-containing particle (ID: 11) from the Qingmingshan Cu–Ni sulfide deposit,
156 Guangxi Province, China. Its O to S atomic ratio is 2.51. The particle contains 2.03%
157 Co and is 300 nm \times 400 nm in size. The selected area electron diffraction pattern
158 shows that the particle is amorphous.

159 3.3 Sulfur-containing particles carried by ascending geogas flows in deep-seated 160 faults

161 Sulfur-containing particles were found in samples obtained using two methods from
162 the deep fault gas of the Dongshengmiao polymetallic sulfide deposit. Figure 12
163 shows a sulfur-containing particle aggregation (ID: 12) that was obtained using the
164 vacuum pump from the deep-seated fault gas near a concealed ore body. The

165 aggregation contains O, Na, Si, S, K, Fe, Zn, and Pb. The S content is 23.8%. Figure
166 13 shows a particle aggregation (ID: 13) that was obtained using a PVC pipe from a
167 fault near a concealed ore body. The ascending gas flow arrived at the top of the PVC
168 pipe naturally, and the particles were adsorbed by a TEM nickel grid. The particle
169 aggregation consists of many small particles that are 4–15 nm in diameter. The small
170 particles are elliptical and crystalline, with 0.302 nm spacing of the lattice fringes, and
171 and their main components are O and S. Figure 14 shows a sulfur-containing particle
172 (ID: 14) that was sampled using a PVC pipe in a fault above a concealed ore body.
173 The vertical distance from the sample to the concealed ore body was 85 m. The
174 vertical distance from the sample to the Earth's surface was 230 m.

175 3.4 Sulfur-containing particles in deep-seated fault gouges and oxidized ores

176 Sulfur-containing particles were also found in deep-seated fault gouges and oxidized
177 zones of the Dongshengmiao polymetallic sulfide deposit. For example, Figure 15
178 shows a sulfur-containing particle (ID: 15) from the oxidized zone. According to its
179 atomic percentage, it contains SO_4^{2-} and may be Sr, Ba sulfate, and Ti oxide. Its size
180 is 200 nm × 400 nm. Figure 16 shows a rhombus-shaped particle (ID: 16) from a
181 deep-seated fault gouge. Its main components are O, S, and Ca, with minor amounts
182 of Fe, Co, and Si.

183 Overall, the sulfur-containing particles or particle aggregations transported by
184 ascending geogas flows can be both regular and irregular in shape and either
185 crystalline or amorphous. The particles or particle aggregations contain Ca, K, Mg, Fe,
186 Na, Pb, Hg, Cu, Zn, As, Ti, Sr, Ba, and Si, as well as O and S.

187 The number of sulfur-containing particles in the ascending geogas flows in
188 non-sulfur-rich areas is much lower than that from the sulfide ore deposits.
189 Furthermore, the overwhelming majority of particles in non-sulfur-rich areas have a
190 low sulphur content. These areas are different from those with the sulfide ore deposits,
191 in which sulfur-containing particles are densely distributed and are present at high
192 levels in the ascending geogas flows.

193 4. Discussion and conclusions

194 Gold particles are formed by post-mineralization fault activity, oxidation, and
195 bacterial weathering of primary minerals (Cao et al., 2010a). Deep-seated gold
196 particles can be transported to the surface by an ascending gas flow, as Brownian
197 motion enables the gold particles in the ascending gas flow to overcome the effect of
198 gravity (Cao et al., 2010a; Cao, 2011). We assume that the same mechanism applies to
199 sulfur-containing particles or particle aggregations. Primary sulfur-containing
200 minerals are transformed into particles by epigenetic reworking, such as
201 post-mineralization fault activity, in which S^{2-} in the sulfide minerals is oxidized to
202 S^{6+} . In this study, the sulfur-containing particles from fault gouges and oxidized ores
203 were found, indicating that these particles were formed by the faulting and oxidation
204 of ores. Faulting and oxidation are well-developed in the Dongshengmiao
205 polymetallic sulfide deposit and other sulfide deposits. This finding indicates that
206 faulting and oxidation play an important role in particle formation.

207 Sulfur-containing particles may be transported to the surface by an ascending geogas
208 flow through faults (Etiope and Martinelli, 2002; Cao et al., 2010a). Material carried

209 by an ascending geogas flow in the soil in the Xuanhan gas field, Sichuan Province,
210 China was sampled and measured using an instrumental neutron activation analysis.
211 Analysis of trace element anomalies has shown the gas-bearing ring fracture structure
212 to be 4000 m deep, suggesting that particles carried by an ascending geogas flow can
213 be transported over long distances (Yang et al., 2000). The gas flow migrates upward
214 because of the temperature difference and the pressure differences between the Earth's
215 interior and its surface is the reason that the gas flow migrate upward (Tong and Li,
216 1999; Etiope and Martinelli, 2002; Cao et al., 2010a). In this study, Sulfide-containing
217 particles suspended in gas above the soil were found, showing that these particles can
218 move through the soil and get into the atmosphere.

219 The probability that these particles are transported by an ascending geogas flow
220 originating in the soil is low. In the study area, the soil consists of kaolinite, halloysite,
221 montmorillonite, illite, chlorite, hematite, quartz, goethite, and similar minerals.
222 Kaolinite is the main mineral, and the sulfur content in the soil is low. Therefore, this
223 soil is clearly not a probable source of sulfur-containing particles transported by an
224 ascending geogas flow. Furthermore, there is no correlation between the numbers of
225 these particles and those of sulfur-containing particles in the soil solid phase.
226 Sulfur-containing particles are clearly enriched in soils above deep sulfur-rich sources
227 because sulfur-containing particles transported by an ascending geogas flow were
228 found in 16 deep sulfide ore bodies that were studied. This result indicates a close
229 relationship between sulfur-containing particles in the gas flow and deep-seated
230 sulfide ore bodies. Other rock types, such as limestone, siltstone, sandstone, and

231 mudstone, do not contain sufficient sulfur to become sources of sulfur-containing
232 particles in an ascending gas flow; for example, the mean sulfur concentrations of the
233 Devonian limestone, mudstone, siltstone, and sandstone in the northern Guangdong
234 Province, China are 610×10^{-6} (68 samples), 80×10^{-6} (25 samples), 160×10^{-6} (33
235 samples), and 110×10^{-6} sulfur (4 samples), respectively.

236 The estimated rate of degassing for the Dongshengmiao deposit calculated to be 2.325
237 $\text{m}^3 \text{s}^{-1}$. The mean sulfur content of the particles carried by the ascending geogas flow
238 for the Dongshengmiao deposit was calculated according to 45 mg/m^3 (Supplement).

239 The estimated annual sulfur emission from particles in the deposit was 3.254 tons. Qi
240 et al. (2007) reported a flue gas amount of $527300 \text{ m}^3 \text{ h}^{-1}$ from the Huhehaote power
241 plant in China and an exit particle concentration of 43.3 mg m^{-3} carried by the flue
242 gas. The SO_3 distribution range in fly ash in 14 power plants (e.g., Tangshan power
243 plant, Gaojing power plant, and Zhengzhou power plant) was reported to range
244 between 0 and 1.05 %. The mean SO_3 and sulfur contents in fly ash were 0.27 % and
245 0.108 %, respectively. On the basis of these mean values, 21.305 tons of annual
246 particulate sulfur emission occurred from the flue gas in the Huhehaote power plant.

247 The annual sulfur emission from the particles carried by ascending geogas flow in the
248 Dongshengmiao deposit was less than carried by the flue gas in the Huhehaote power
249 plant. However, the amount of concealed deposits is much more than that of
250 coal-burning power plants. Moreover, size of the particles carried by the ascending
251 geogas flow from concealed deposits is usually $<500 \text{ nm}$. The mean diameter of the
252 particles carried by the flue gas in 9 samples obtained from four coal-fired power

253 plants in China were 19.71, 3.18, 5.43, 5.67, 130.94, 77.29, 12.99, 11.59, and 236.63
254 μm respectively (Zhang et al. 2007). The sizes of particles carried by the ascending
255 geogas flow from concealed deposits were lesser than those of the particles carried by
256 the flue gas from coal-fired power plants. Within a certain volume, the particles were
257 smaller and the number of particles was more. These small particles are more capable
258 of migration and have a significant health and environmental impact. Therefore,
259 attention must be paid to the particles carried by the ascending geogas flow from
260 concealed deposits.

261 Such sulfur-containing particles enter the atmosphere. Several studies have discussed
262 the direct effects of sulfate particles on the climate (Liu et al., 2009). Some
263 researchers have suggested that sulfur-containing particles can reduce atmospheric
264 temperature or result in climate warming. Streets et al. (2000) suggested that because
265 sulfate aerosols play a vital role in cooling the atmosphere, a reduction in sulfur
266 dioxide emissions in the future would result in increased global warming.
267 Furthermore, aerosol sulfate has been identified as an important contributor to
268 sunlight scattering (Lelieveld and Heintzenberg, 1992; Kim et al., 2001). At the top of
269 the atmosphere above East Asia, SO_4^{2-} radiative forcing is -2 to -10 W m^{-2} over land
270 and -5 to -15 W m^{-2} over ocean (Gao et al., 2014). Niemeier et al. (2011) revealed
271 that an increase in the SO_2 emission rate does not lead to a similar increase in
272 radiative forcing because, as the size of the aerosols increases, their lifetime decreases.
273 It is thus possible that the sulfur-containing particles transported by an ascending
274 geogas flow have an effect on the climate and should, therefore, be evaluated.

275 Sulfate particles can be transported into the lungs leading to respiratory illnesses
276 (World Bank Group, 1999; Soleimani et al., 2007). In particular, the sulfur-containing
277 particles contain high levels of toxic Pb, Hg, Cu, and As. In nature, sulfur usually
278 combines with Pb, Hg, Cu, As, Ni, Cd, and Sb, which are toxic to organisms, to form
279 sulfide deposits. The sulfur-containing particles originating from sulfide deposits
280 commonly contain toxic elements. This phenomenon has been confirmed by EDX
281 analysis of particles. The particle sizes carried by the ascending gas flow are usually
282 less than 500 nm. The size is only one-fifth of the upper size limit of PM_{2.5}. Geogas
283 particles undergo long-distance migration. They can remain in the atmosphere for
284 long periods and in can get into bronchioles and alveoli, affecting the ventilative
285 function of lung. They can also enter the blood. The possible relationship between the
286 occurrence of sulfur-containing particles transported by an ascending geogas flow and
287 endemic diseases in the vicinity of sulfur-containing deposits should be investigated.

288 It is probable that sulfur-containing particles transported by the ascending geogas
289 flows in the soil affect the soil system; for example, sulfur-containing particles can
290 affect both soil biota and enzymatic activities, resulting in changes in the soil structure,
291 nutrient cycling, and organic matter decomposition and retention. Sulfur-containing
292 particles may directly catalyze organic matter decomposition. Furthermore, the
293 potential use of such particles as fertilizers for rice plants needs to be investigated.

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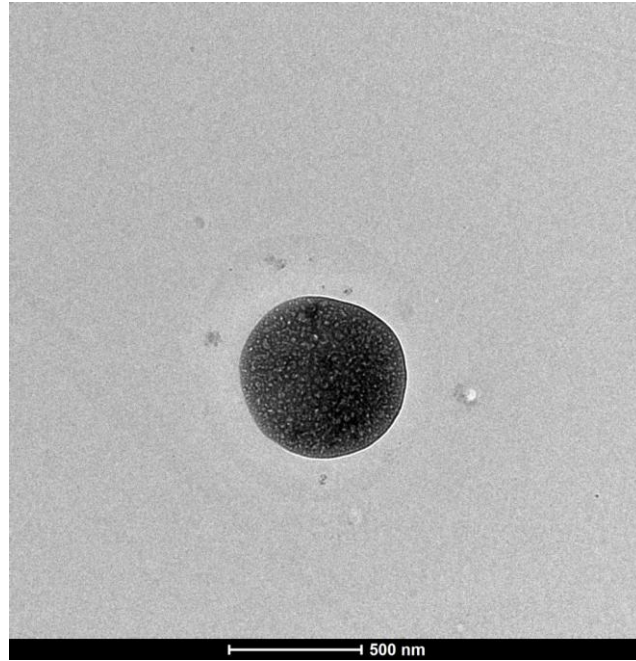
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430 Fig. 1 TEM image of an S-, O-, and Si-containing particle obtained from an ascending
431 gas flow above the soil over the Dongshengmiao deposit.

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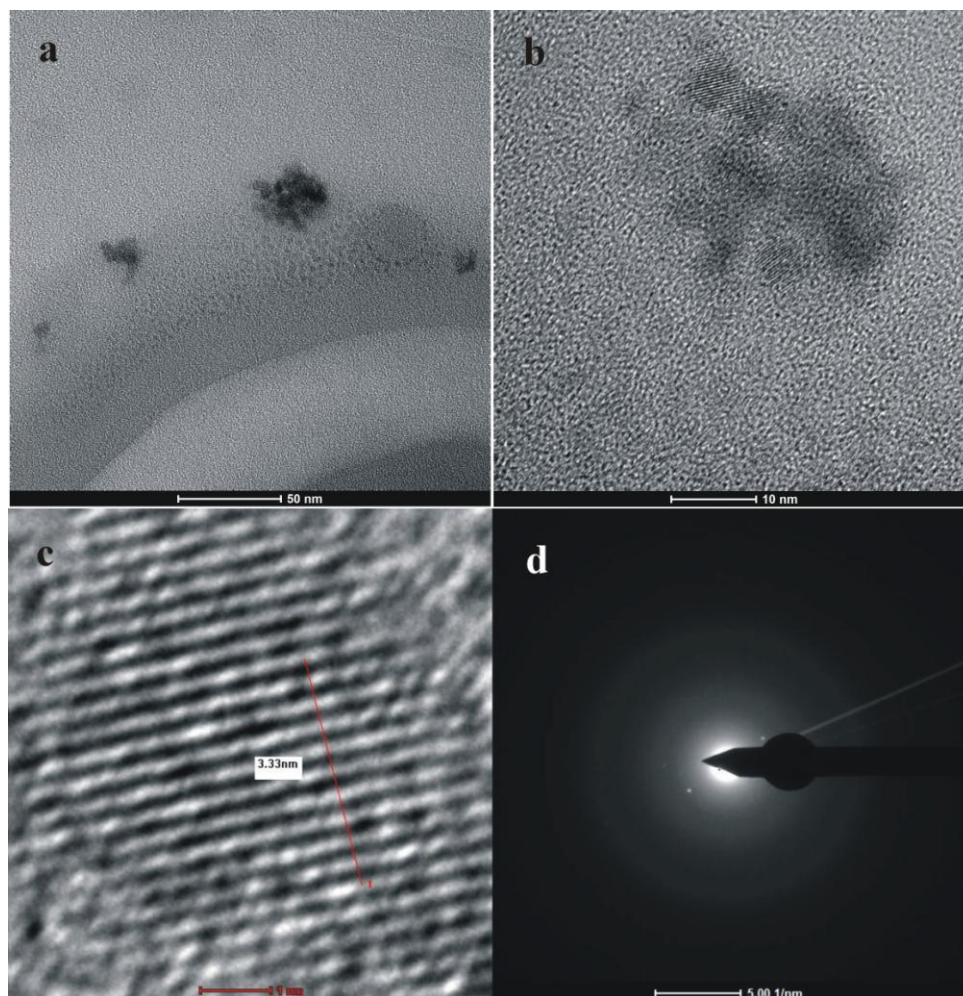
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440 Fig. 2 (a) TEM image, (b, c) high-resolution (HRTEM) images, and (d) selected area
441 electron diffraction (SAED) pattern of an S-, O-, Hg-containing particle aggregation
442 obtained from an ascending gas flow above the soil over the Dongshengmiao deposit.

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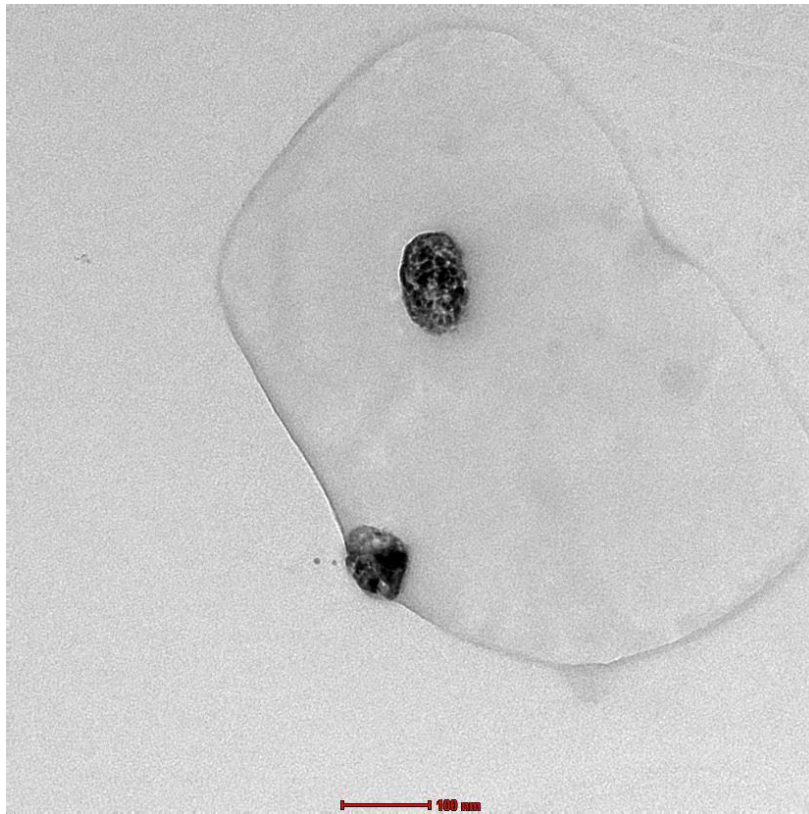
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450 Fig. 3 TEM image of S-, O-, K-, and Pb-containing particle aggregations obtained
451 from an ascending gas flow above the soil over the Dongshengmiao deposit.

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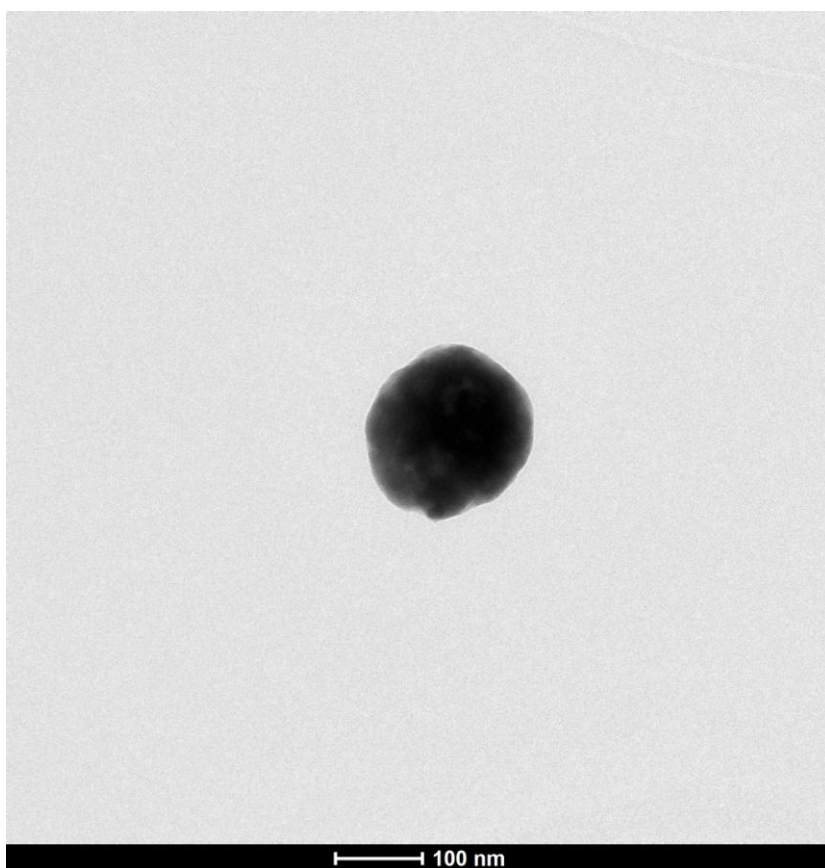
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462 Fig. 4 TEM image of an S-, O-, Na-, Pb-, Zn-, and As-containing particle obtained

463 from an ascending gas flow above the soil over the Dongshengmiao deposit.

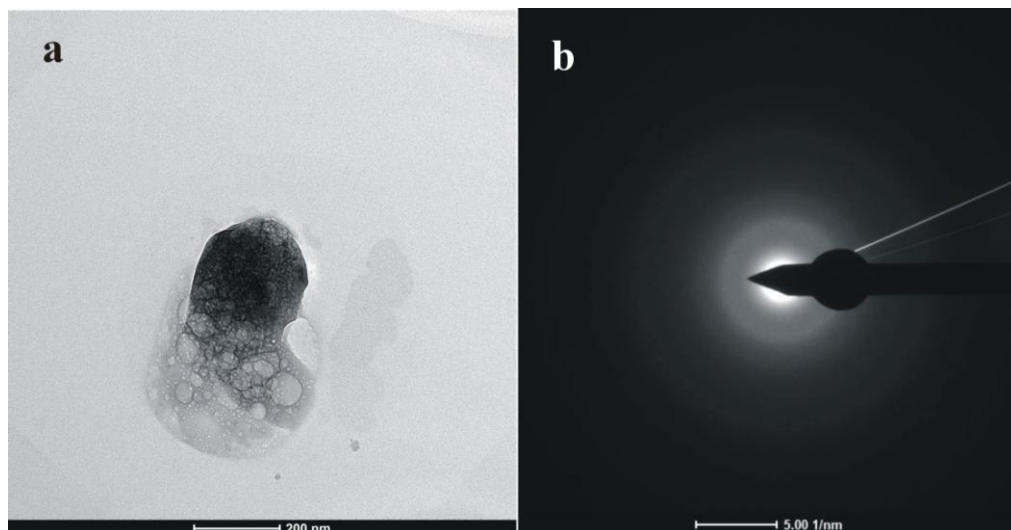
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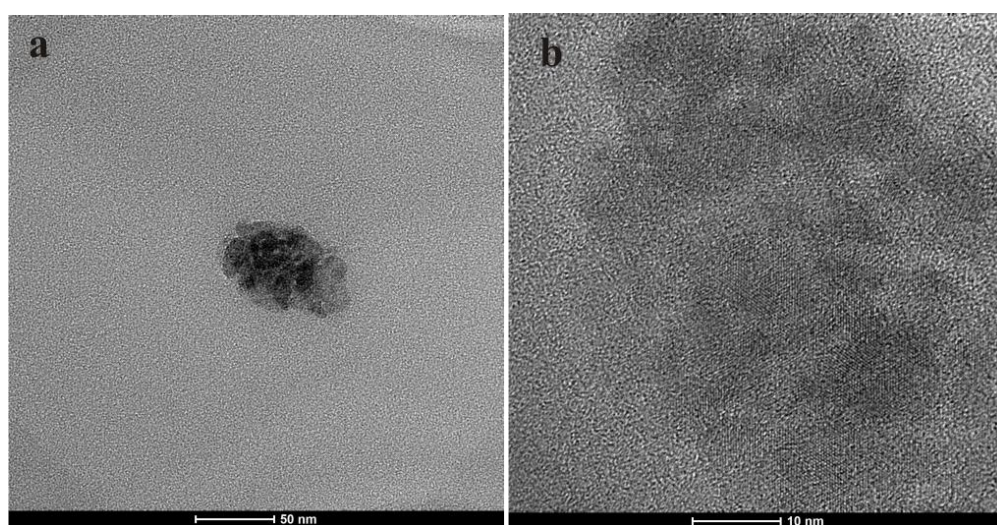
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469 Fig. 5 (a) TEM image and (b) SAED pattern of an S-, O-, K-, Na-, and Pb-containing
 470 particle obtained from an ascending gas flow above the soil over the Dongshengmiao
 471 deposit.

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473 Fig. 6 (a) TEM image and (b) HRTEM image of an O-, Si-, S-, and Cu-containing
 474 particle aggregation obtained from an ascending gas flow above the soil over the
 475 Dongshengmiao deposit.

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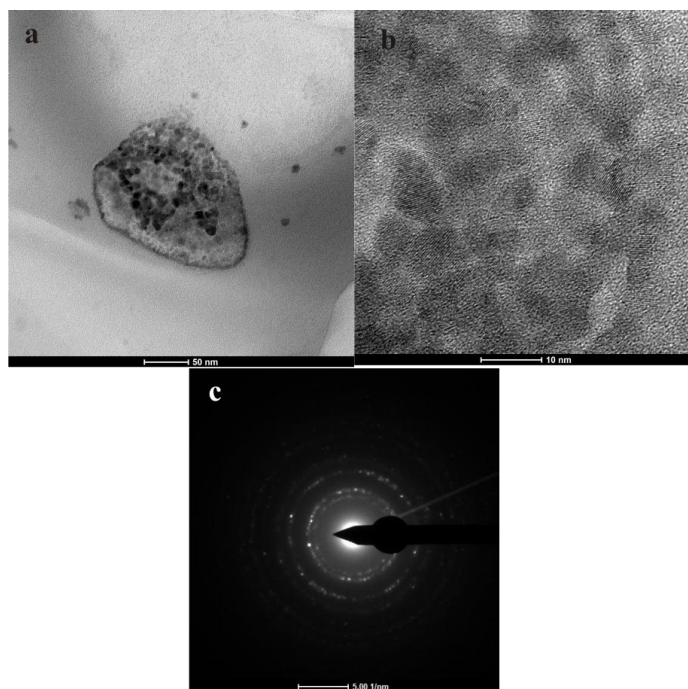
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482 Fig. 7 (a) TEM image, (b)HRTEM image, and (c) SAED pattern of an O-, S-, K-, and
483 Pb-containing particle aggregation obtained from an ascending gas flow above the
484 soil over the Dongshengmiao deposit.

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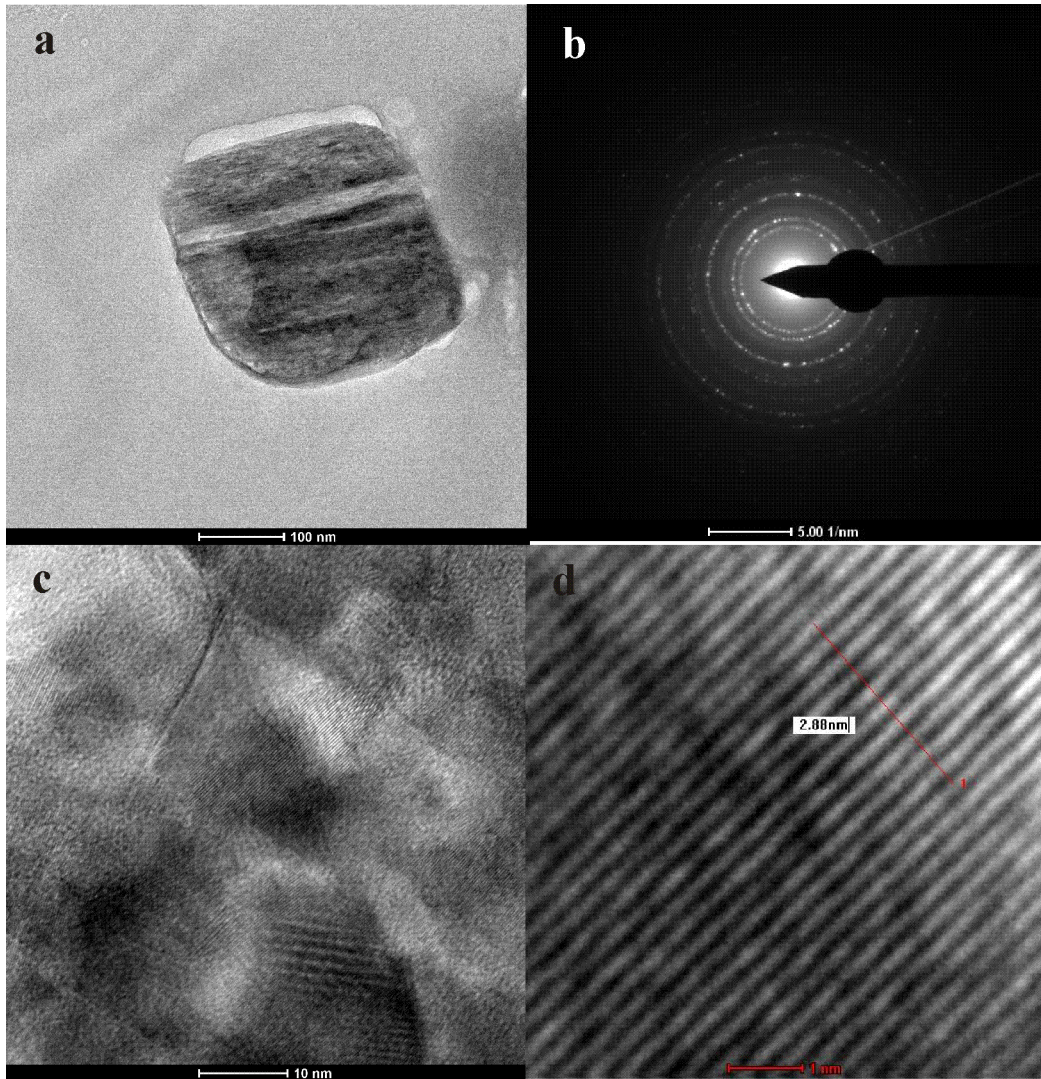
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504 Fig. 8 (a) TEM image, (b) SAED pattern, and (c, d) HRTEM image of an O-, S-, Ca-,
505 and Mg-containing particle obtained from an ascending gas flow in the soil over
506 Dongshengmiao deposit.

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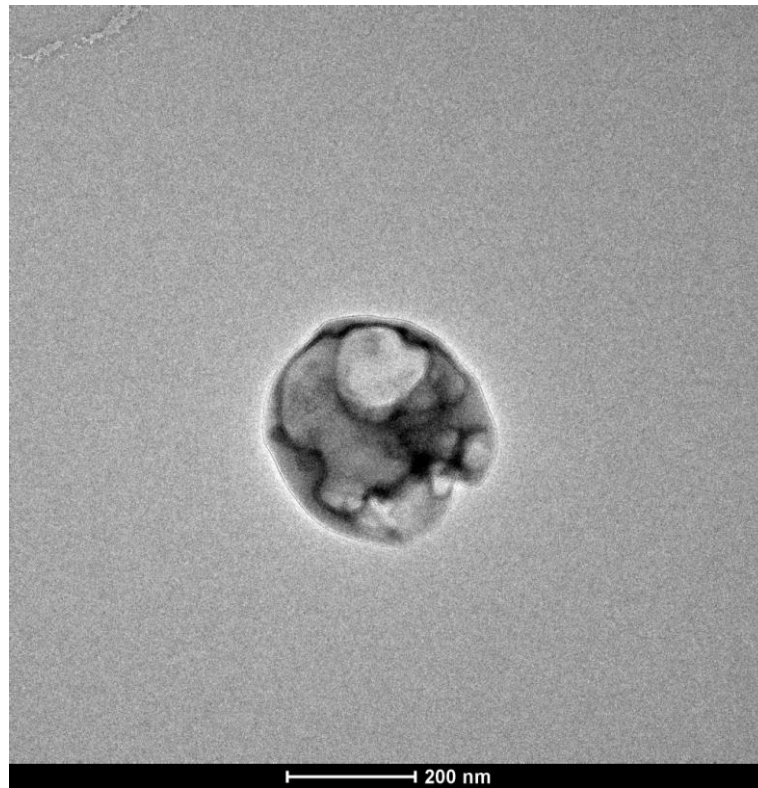
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516 Fig. 9 TEM image of an O-, S-, and K-containing particle obtained from an ascending
517 gas flow in the soil from the Kafang copper deposit, Yunnan Province.

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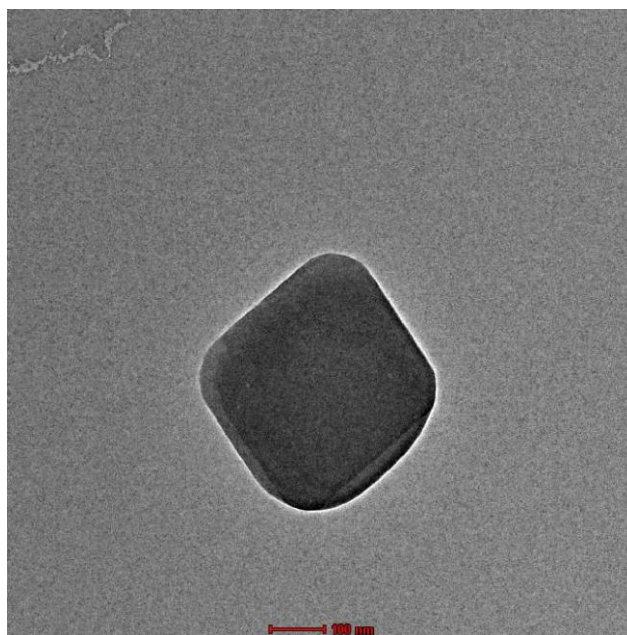
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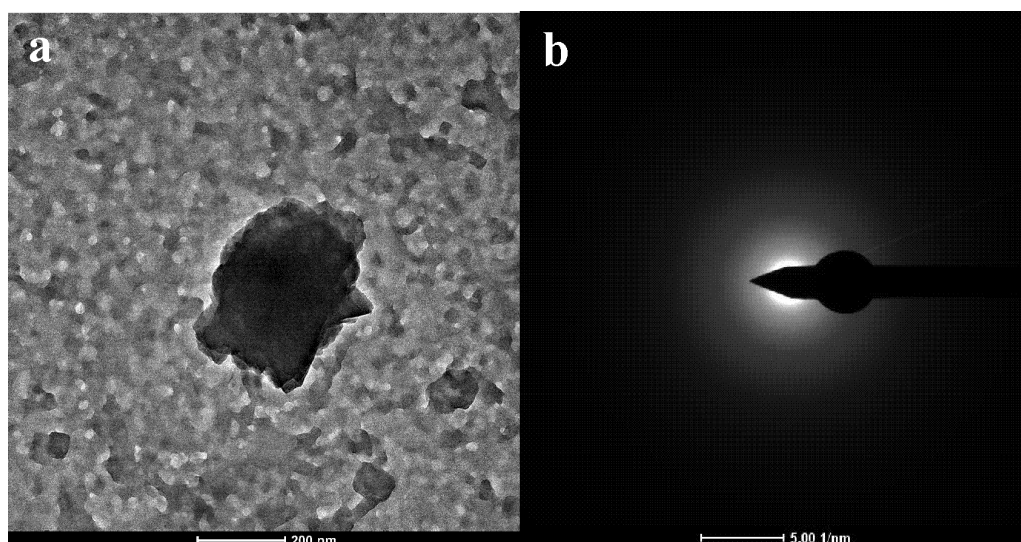
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527 Fig. 10 TEM image of an O-, S-, and Fe-containing particle obtained from an
 528 ascending gas flow in the soil from the Yongshengde copper deposit in northeastern
 529 Yunnan.

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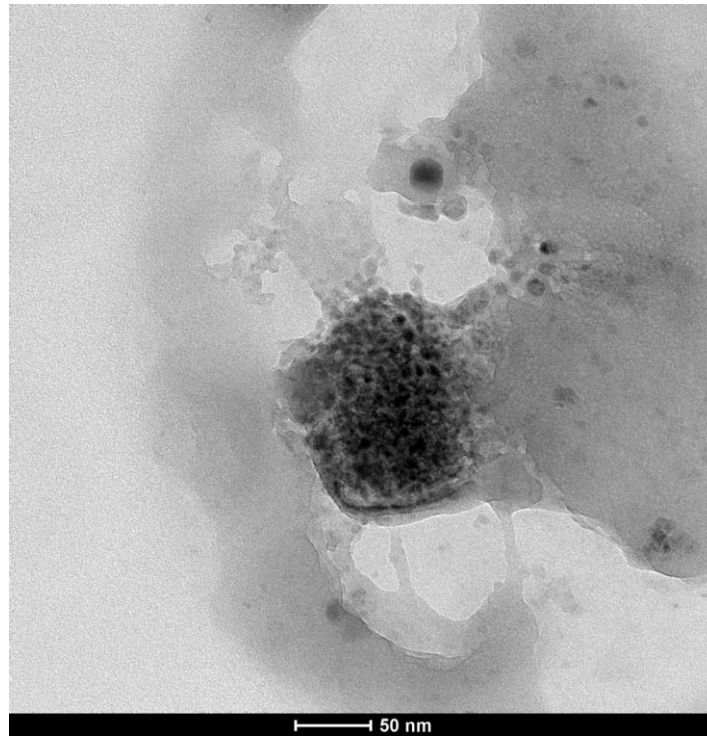
531 Fig. 11 (a) TEM image and (b) SAED pattern of an O-, S-, and Co-containing particle
 532 obtained from an ascending gas flow in the soil from the Qingmingshan Cu–Ni
 533 sulfide deposit, Guangxi Province.

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539 Fig. 12 TEM image of an O-, S-, K-, Pb-, and Na-containing particle sampled using a
540 vacuum pump from the fault gas near a concealed ore body of the Dongshengmiao
541 deposit.

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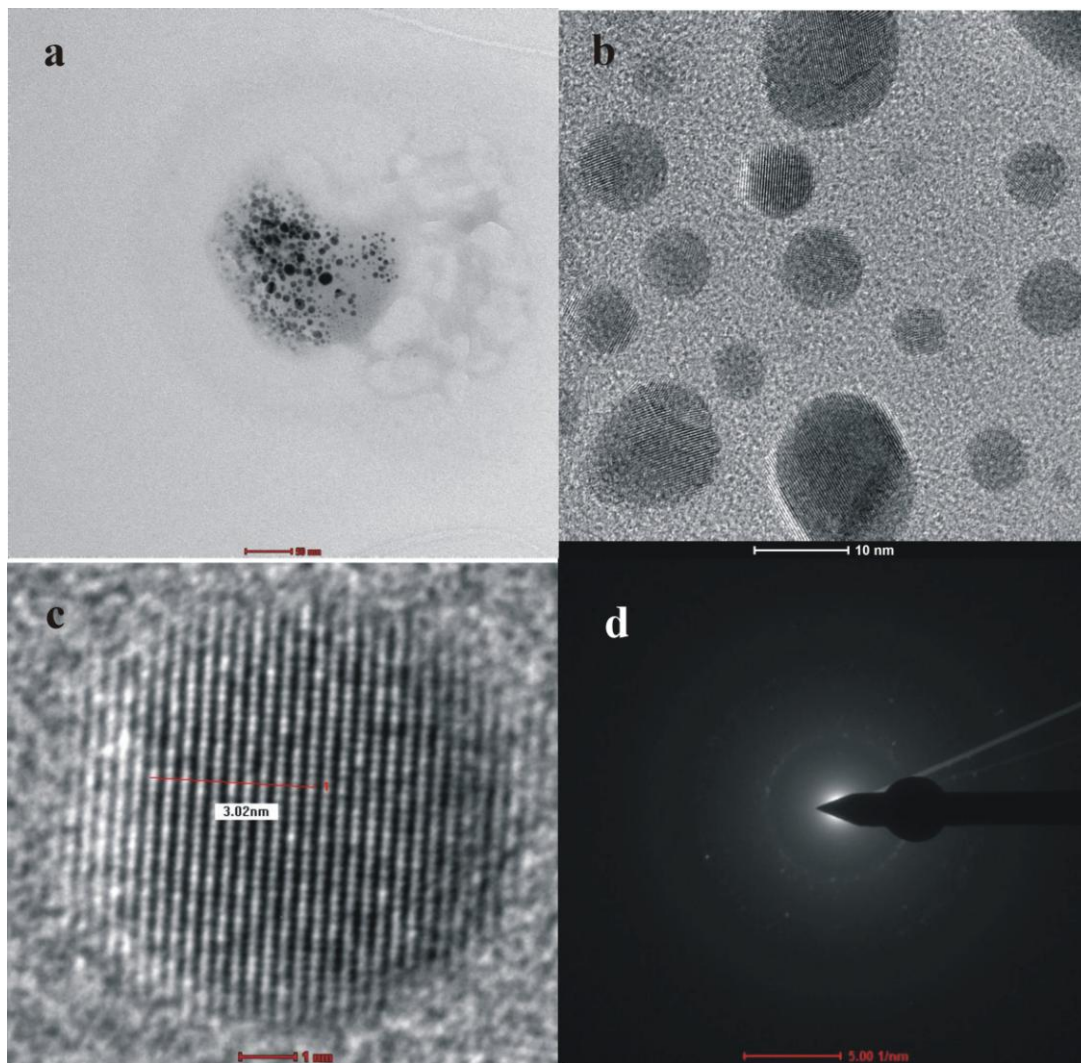
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551 Fig. 13 (a) TEM image, (b, c) HRTEM images, and (d) SAED pattern of an O-, S-,
552 and K-containing particle aggregation sampled using a PVC pipe in a fault near a
553 concealed ore body of the Dongshengmiao deposit.

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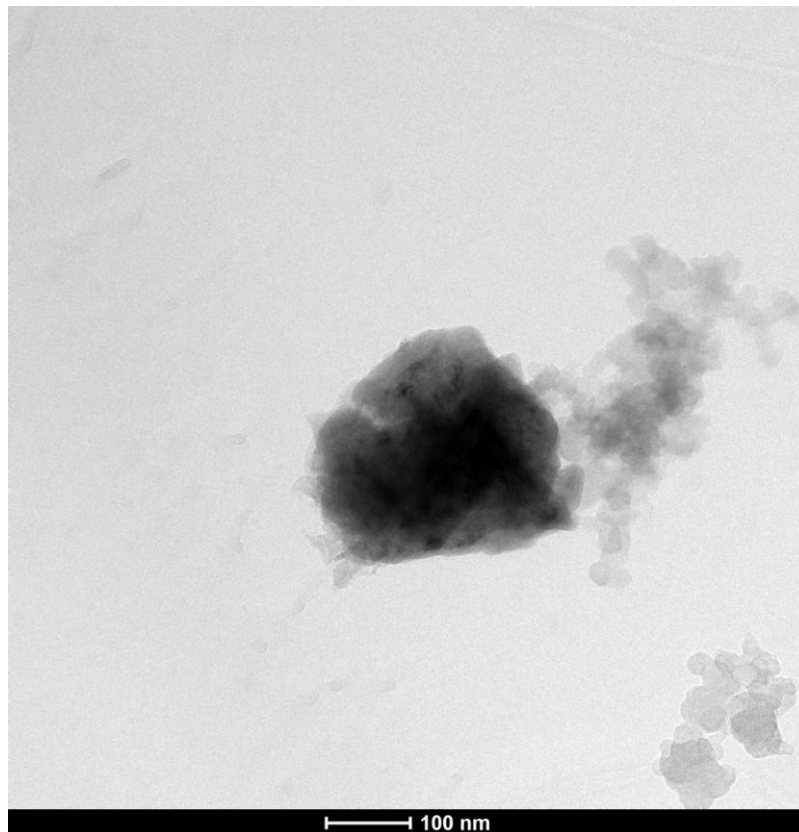
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563 Fig. 14 TEM image of an O-, S-, Fe-, and Mg-containing particle aggregation

564 sampled using a PVC pipe in a fault above a concealed ore body of the

565 Dongshengmiao deposit.

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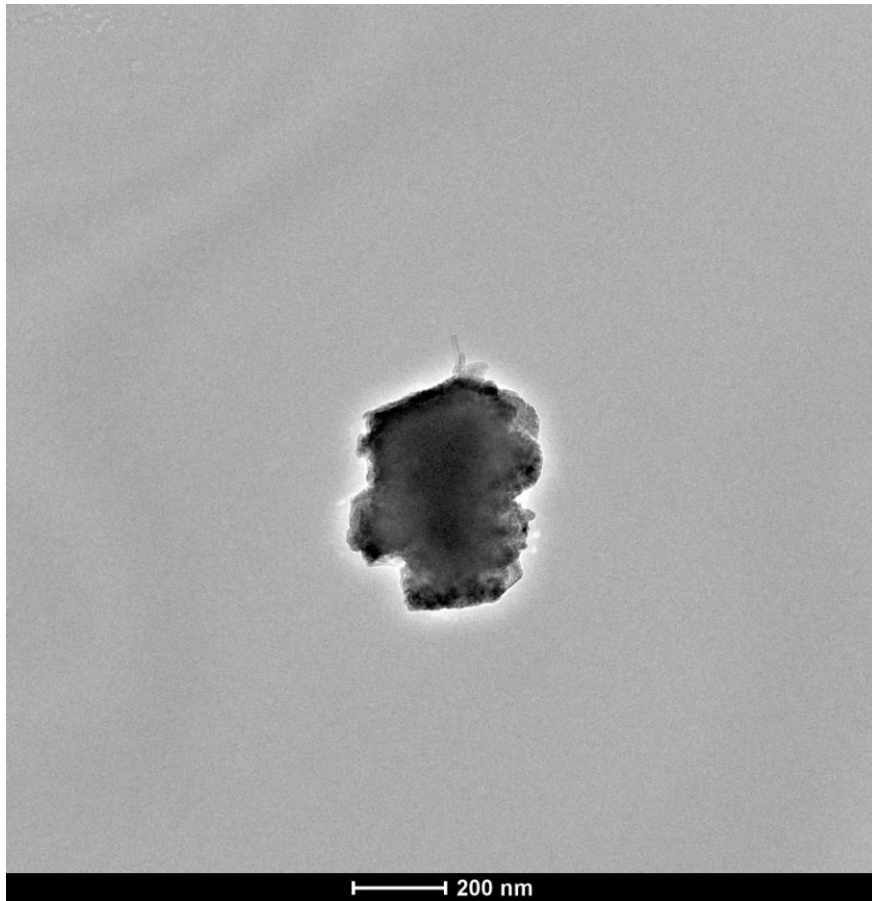
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573 Fig. 15 TEM image of an O-, S-, Ti-, Sr-, and Ba-containing particle from a

574 deep-seated oxidized zone in the Dongshengmiao deposit.

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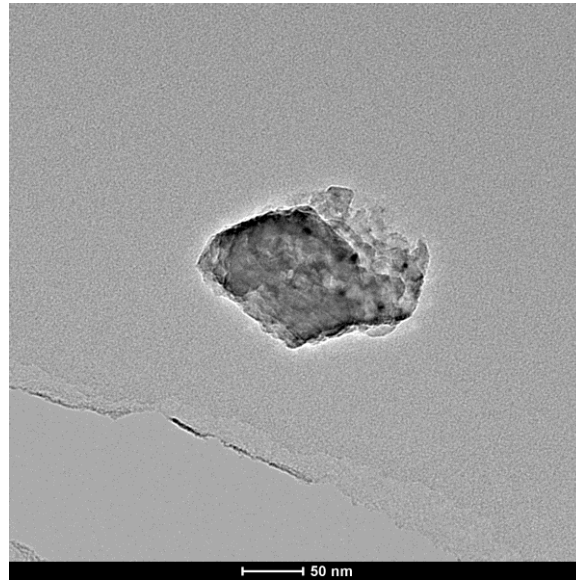
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585 Fig. 16 TEM image of an O-, S-, Fe-, Co-, and Ca-containing particle from a

586 deep-seated fault gouge in the Dongshengmiao deposit.

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Table 1 Number of sulfur-containing particles or particle aggregations number from the Dongshengmiao deposit on 100 μm × 100 μm TEM grids

Sulfur-containing particles or particle aggregations carried by ascending gas flow above the soil (that had flown through the soil)				Sulfur-containing particles or particle aggregations carried by ascending gas flow in deep faults			
Sample	Sample box	Grid	Number	Sample	Sample box	Grid	Number
ND13-1	A1	A1-1	3	NDDW03	A2	A2-2	3
ND13-2	A2	A2-1	2	NDDW05	A4	A4-1	1
ND13-3	A3	A3-2	1			A4-2	29
		A3-3	6	NDDW06	A5	A5-2	1
ND13-4	A4	A4-1	1	NDDW07	B1	B1-1	4
		A4-2	2			B1-2	1
ND13-6	A5	A5-1	1	NDDW19	D3	D3-2	1
		A5-2	3			D3-3	2
		A5-3	1	NDDW26	E4	E4-1	1
ND13-8	B2	B2-1	1			E4-3	1
		B2-2	6	NDDW27	E5	E5-1	2
		B2-3	1			E5-3	2
ND13-9	B3	B3-1	1			E5-4	1
		B3-2	1	NDDW36	G4	G4-1	12
		B3-3	1			G4-3	10
ND13-10	B4	B4-1	1			G4-4	1
		B4-3	6	NDDW37	G5	G5-1	1
ND13-11	B5	B5-1	1				

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Table 2 EDX results for particles 1–8.

Element	Particle number							
	1	2	3	4	5	6	7	8
Weight O%	18.47	9.46	16.02	9.73	15.75	12.9	5.13	51.88
Atomic O%	31.1	31.78	31.12	39.3	34.16	31.35	22.74	69.78
Weight Si%	3.35		1.49	0.5	1.09	3.08		2.19
Atomic Si%	3.21		1.65	1.15	1.34	4.27		1.67
Weight S%	78.17	31.23	63.1	3.82	10.83	21.61	18.25	19.02
Atomic S%	65.68	52.33	61.16	7.7	11.72	26.2	40.32	12.76
Weight Hg%		59.29						
Atomic Hg%		15.87						
Weight K%			4.88		35.75		7.31	0.99
Atomic K%			3.88		31.73		13.25	0.54
Weight Pb%			14.48	54.2	22.5		69.28	
Atomic Pb%			2.17	16.9	3.76		23.67	
Weight Na%				3.1	9.66			
Atomic Na%				8.73	14.58			
Weight Fe%				0.75	2.14			0.21
Atomic Fe%				0.87	1.33			0.08
Weight Co%				0.98	2.25			
Atomic Co%				1.08	1.32			
Weight Zn%				8.34				
Atomic Zn%				8.24				
Weight As%				18.55				
Atomic As%				16				
Weight Cu%						62.39		
Atomic Cu%						38.16		
Weight Mg%								3.86
AtomicMg%								3.42
Weight Ca%								21.82
Atomic Ca%								11.71

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638 Table 3 EDX results for particles 9–16.

Element	Particle number							
	9	10	11	12	13	14	15	16
Weight O%	26.54	56.25	53.66	25.39	67.03	17.21	29.21	40.8
Atomic O%	42.51	73.54	70.2	37.32	80.72	35.83	64.85	62.97
Weight Si%	0.52			0.66	1	0.7		1.5
Atomic Si%	0.47			0.55	0.68	0.83		1.32
Weight S%	63.99	31.3	42.81	23.8	28.01	24.59	10.88	15.03
Atomic S%	51.15	20.42	27.95	17.45	16.83	25.53	12.05	11.58
Weight K%	8.93	0.78		2.01	2.59			
Atomic K%	5.85	0.42		1.21	1.27			
Weight Pb%				4.25				
Atomic Pb%				0.48				
Weight Na%			1.04	40.92		1.35		
Atomic Na%			0.95	41.84		1.96		
Weight Fe%		9.94	0.44	1.11	1.35	51.16	1.27	5.2
Atomic Fe%		3.72	0.16	0.46	0.46	30.5	0.81	2.3
Weight Co%			2.03					6.36
Atomic Co%			0.72					2.66
Weight Zn%				1.82				
Atomic Zn%				0.65				
Weight Mg%						2.74		
Atomic Mg%						3.75		
Weight Ca%						0.28	0.5	31.08
Atomic Ca%						0.23	0.44	19.15
Weight F%		1.71						
Atomic F%		1.88						
Weight Al%						0.25		
Atomic Al%						0.3		
Weight Mn%						1.68		
Atomic Mn%						1.02		
Weight Ti%							10.94	
Atomic Ti%							8.11	
Weight Sr%							10.32	
Atomic Sr%							4.18	
Weight Ba%							36.86	
Atomic Ba%							9.53	

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643 **The comments of the editor's comments**

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645 Please add a reference (e.g. on line 59) to Hu et al., Resource Geology, 2015.

646

647 **Responses to editor's comments**

648

649 "Study of carbon-bearing particles in ascending geogas flows in the Dongshengmiao
650 polymetallic pyrite deposit, Inner Mongolia, China" by Hu, G., Cao, J. J., Hopke, P. K.,
651 and Holub, R. F., Resource Geology, Vol. 65, pp. 13–26, 2015 has been cited.

652

653 **Author's changes in manuscript**

654

655 P. 4, line 57: "; Hu et al., 2015" has been added.

656 P. 17, line 344: "Hu, G., Cao, J. J., Hopke, P. K., Holub, R. F.: Study of
657 carbon-bearing particles in ascending geogas flows in the Dongshengmiao
658 polymetallic pyrite deposit, Inner Mongolia, China, Resour. Geol., 65, 13–26, 2015."
659 has been added.

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