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► **To cite this version:**

Nicolas J. Bonnet, Anicet Beauvais, Nicolas Arnaud, Dominique Chardon, Mudlappa Jayananda. First $^{40}\text{Ar}/^{39}\text{Ar}$ dating of intense Late Palaeogene lateritic weathering in Peninsular India. *Earth and Planetary Science Letters*, 2014, 386, pp.126-137. 10.1016/j.epsl.2013.11.002 . hal-01097320

HAL Id: hal-01097320

<https://hal.science/hal-01097320v1>

Submitted on 2 Jan 2017

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1 **First $^{40}\text{Ar}/^{39}\text{Ar}$ dating of intense Late Palaeogene lateritic weathering**
2 **in Peninsular India**

3

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26 **Abstract**

27 Lateritic surface processes have shaped large platform and cratons of the tropical
28 belt. Constraining the timing of such processes is crucial to decipher their role in
29 cratonic morphogenesis and their response to long-term climatic change and
30 lithospheric deformation. Weathering histories have been documented for South
31 America, Africa and Australia, but precise time constraints of the lateritic weathering
32 processes in South India are still lacking. We present $^{40}\text{Ar}/^{39}\text{Ar}$ ages of supergene
33 cryptomelane (K-Mn oxide) formed in the Sandur Mn ore deposits exposed on the
34 highest lateritic paleolandsurface that once covered the Mysore plateau and the adjacent
35 Deccan Traps. Significant $^{40}\text{Ar}/^{39}\text{Ar}$ ages are estimated between ~ 36 and ~ 26 Ma from
36 well-defined plateaus in step heating ^{39}Ar release spectra and from best-fitted inverse
37 isochrones. These ages constitute firm time constraints that document intense late
38 Eocene-Oligocene lateritic weathering over Peninsular India under the influence of
39 warm and wet climate comparable to that prevailing in tropical humid forests. These
40 results imply that Southern India was weathered between ~ 36 and 26 Ma and may have
41 been dissected mostly in the Neogene.

42

43 **Keywords:** $^{40}\text{Ar}/^{39}\text{Ar}$ radiometric dating; Cryptomelane; Supergene Mn-ore deposits;

44 Lateritic paleolandsurface; Sandur Greenstone Belt; South India

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51 **1. Introduction**

52 Exposed laterites and lateritic duricrusts on relics of paleolandsurfaces in
53 cratonic domains result from successive long-duration chemical weathering and erosion
54 processes, and as such represent morphoclimatic fingerprints of tropical regions (Ollier,
55 1988; Bárdossy and Aleva, 1990; Thomas, 1994; Tardy and Roquin, 1998; Beauvais
56 and Chardon, 2013). Since the first definition and description of laterites in South India
57 (Buchanan, 1807), many studies were conducted on Indian lateritic formations
58 (Subramanian and Mani, 1978; Babu, 1981; Sahasrabudhe and Deshmukh, 1981;
59 Krishna Rao et al., 1989b; Widdowson and Cox, 1996; Widdowson and Gunnell, 1999),
60 including manganiferous supergene ore deposits (Sawkar, 1981; Krishna Rao et al.,
61 1982, 1989a; Dessai, 1985; Roy, 1992; Mohapatra et al., 1996; Sethumadhav et al.,
62 2010). Except few geological stratigraphic constraints, radiometric dating of surface
63 processes involved in the formation and evolution of Indian lateritic paleolandsurfaces
64 are still lacking. Laterites are interlayered with marine sequences of late Eocene and
65 early Miocene age (McGowran et al., 1977; Prasad, 1983). Lateritisation processes have
66 also been indirectly dated with Neogene palynofossils found in colluvial/alluvial
67 sediments overlying in-situ formed Mn ore deposits ~ 100 km south of Belgaum (Fig.
68 1). These fossils constrain intense weathering that led to Mn ore concentration to be
69 Neogene at the latest (Krishna Rao et al., 1989a). The age of lateritic weathering has
70 also been roughly estimated on the basis of the paleomagnetic properties of iron oxides
71 formed in laterites around 16°N (Schmidt et al., 1983), i.e., “Early Tertiary”
72 (Paleogene?) for the high-level Belgaum laterites and “Late Tertiary” (Neogene?) for
73 the low-level laterites found west of the Western Ghats escarpment (Fig.1).

74 Chemical rock weathering commonly resulted in the accumulation of metallic
75 elements (Al, Fe, Mn) in residual laterites and depletion of more soluble elements (Na,
76 K, Si, Ca). Potassium, Na and/or Ba may, however, be retained by supergene Mn oxides
77 in large crystallographic lacuna existing between the MnO_6 octahedra chains that define
78 the typical tunnel-structure of the hollandite manganese oxide group. Cryptomelane
79 belongs to this group with the general stoichiometric formulae $\text{K}_x \text{Mn}^{\text{IV}}_{8-x} \text{Mn}^{\text{III}}_x \text{O}_{16}$, x
80 ranging from 0.2 to 1 and K_x counterbalancing Mn^{III} substitution for Mn^{IV} (Burns and
81 Burns, 1979; Turner and Buseck, 1979; Post and Burnham, 1986; Pasero, 2005).
82 Cryptomelane is a major Mn oxide of many lateritic Mn-ore deposits in South India
83 (Sivaprakash and Phil, 1980; Krishna Rao et al., 1982; Mohapatra et al., 1996;
84 Sethumadhav et al., 2010). This mineral hosts radioactive ^{40}K atoms, which can decay
85 into ^{40}Ar . The high radiogenic ^{40}Ar retentive power of cryptomelane makes this mineral
86 a suitable chronometer for $^{40}\text{Ar}/^{39}\text{Ar}$ step heating dating of weathering processes in
87 lateritic soils including duricrusts and manganiferous ore deposits (Vasconcelos et al.,
88 1992; Vasconcelos et al., 1994a; Ruffet et al., 1996; Vasconcelos, 1999).

89 This study quantifies the radiometric ages of lateritic weathering processes that
90 have affected the rocks of the Dharwar craton on the Mysore Plateau by $^{40}\text{Ar}/^{39}\text{Ar}$
91 dating of cryptomelane from supergene Mn-ore deposits. We focus on the Fe-Mn
92 sedimentary formations belonging to the Sandur Greenstone Belt (SGB) (Figs. 1 and
93 2a), which hosts the biggest Indian manganese reserve (Kameshwar et al., 2000). Our
94 results provide tangible time constraints (Late Eocene – Oligocene) on the
95 paleoweathering processes and morphoclimatic evolution of the potentially oldest
96 lateritic paleolandscape preserved on the Mysore plateau and Traps in southern India.

97

98 2. Geological and geomorphological setting

99 2.1. Sandur Greenstone Belt geology

100 The SGB is a synform-shape greenstone belt that consists of mafic/ultramafic
101 volcanics and sediments (Fig. 2a) having undergone low-grade regional metamorphism
102 during latest Archean orogeny that shaped the Dharwar craton (Roy and Biswas, 1979;
103 Mukhopadhyay and Matin, 1993; Chadwick et al., 1996; Chardon et al., 2008). The
104 sedimentary sequences contain elements of metavolcanic rock, which are relatively rich
105 in Ni, Cr, Co and Sc. The upper part of the greenstone stratigraphic pile includes
106 platform-like sediments bearing stromatolites, Mn formations and carbonaceous Mn
107 phyllites (Fig. 2a).

108 In the eastern part of the belt, the upper part of the sequence includes banded
109 iron formations (BIF), felsic volcanics and terrigenous sediments (Fig. 2a). SHRIMP U-
110 Pb dating of zircon in two interlayered felsic volcanics indicate ages ranging from 2651
111 ± 18 Ma to 2659 ± 40 (Nutman et al., 1996). The BIF and Fe-Mn formations are related
112 to marine transgression and regression cycles, respectively (Roy, 2006). These cycles
113 implied alternating chemical and clastic sedimentation, which is also evidenced by
114 specific iron formation facies and argillites within the BIF (Manikyamba et al., 1993).
115 The lower part of the sequence is confined to the Deogiri region and is essentially
116 composed of manganiferous stromatolitic limestones, dolomites and argillites (Fig. 2a)
117 while the upper part is richer in Fe (Roy, 2000). Iron and Mn fractionation was
118 controlled during eustatic sea-level variations by differentiated redox conditions of two
119 stratified water masses on the shallow and deep shelf (Roy, 1992; Manikyamba and
120 Naqvi, 1997). The Fe-Mn sediments have later been affected by low-grade
121 metamorphism by 2475 ± 65 Ma (whole rock Pb/Pb isochron dating), which is the

122 maximal recrystallisation age of the limestones (Russell et al., 1996; Chadwick et al.,
123 2000). Such a metamorphism led to Mn enrichment of the sediments (Roy, 2000),
124 which are composed of Mn-carbonates, argillites, arenites and phyllites (Fig. 2a), and
125 are the main protorees of the SGB Mn-ore deposits. These deposits grade upward into
126 supergene weathering materials (Roy, 2006).

127

128 2.2. Geomorphology of southern India and Sandur Greenstone Belt

129 The geomorphology of southern India consists of high-elevation surfaces
130 occurring as relics of a differentiated landscape developed on the Dharwar craton and
131 Deccan traps (Fig. 1), and low-elevation surfaces along the western coast
132 (Radhakrishna, 1993), both being overlain by laterites (Widdowson and Cox, 1996;
133 Widdowson, 1997). The regional eastern dip of the high-elevation surfaces is
134 interpreted to result from eastward tilting of Peninsular India due to the Seychelles
135 rifting and Deccan Traps events (~ 65-63 Ma) (Radhakrishna, 1993). On the Deccan
136 Traps, bauxitic lateritic weathering of the highest preserved landsurface must be
137 younger than 62.3 ± 0.6 Ma, which is the age of the youngest basalts (Courtillet et al.,
138 1986; Pande, 2002). On the Traps and the Mysore Plateau, relics of two younger
139 surfaces step below the plateau remnants of the high bauxitic surface (900-1000 m). The
140 three erosion surfaces remain as sparse geomorphic relics at the scale of the Mysore
141 plateau, and have been numbered S2, S3 and S3_d, the latter being a denudation
142 degradation landform of S3 (Gunnell, 1998). Spatially reduced relics of the highest
143 massifs also overhang the Mysore plateau (Fig. 1).

144 Landsurfaces S2, S3 and S3_d are also identified on the SGB relief (Figs. 2b-c).

145 The S2 surface levels the summits of the belt over an altitude range of 900 to 1100 m

146 and exposes deep in situ formed weathering mantles capped by Fe-Mn and Mn
147 duricrusts at altitudes of ~ 1000-1016 m. Our study is mainly focused on the Mn ore
148 open-pit mines carved in this surface (Figs. 2b-c). Relicts of paleo-land surface S3
149 extends below S2 over an altitude range of 750 to 850 m (Fig. 2c), and are covered by
150 in-situ formed lateritic weathering profiles capped by iron duricrust instead of Mn ore
151 concentrations. Relicts of land surface S3_d occur as benches below the S3 surface over
152 an altitude range of 700-750 m above the modern river network (Figs. 2c). The S3_d
153 surface is covered by reworked erosional materials including ferricrete and bedrock
154 clasts originated from S3 lateritic profiles, sharing most characteristics with West
155 African glacis/pediments (Beauvais and Chardon, 2013), but this surface is not affected
156 by a late ferruginous or manganiferous lateritic duricrusting.

157

158 **3. Material and methods**

159 **3.1. Ore bodies description and sampling**

160 We have sampled across the main Mn ore bodies of the SGB deposits, which
161 formed within weathered meta-sedimentary phyllites at different depths below the top
162 of landsurface S2 at 1012-1015 m altitudes (Fig. 3). Description of samples precisely
163 located on open-pit mine benches allowed the establishment of log sections (Fig. 3b and
164 3d). In Kappataswamy pit (N 14°59'59" E 76°32'42", Fig. 3a), the first ore body was
165 sampled between 856 and 893 m of elevation along the E-W log section 1 on the
166 western benches of the pit, and the second one between 904 to 935 m along the N-S log
167 section 2 on the eastern benches (Fig. 3b). The third ore body is located in Channanghi
168 KMK-E pit (N 14°59'50" E 76°34'37", Fig. 3c) at an elevation range of 926 to 994 m
169 (log section 3, Figure 3d). The Mn-ore bodies are composed of different petrographic

170 facies. The float and platy Mn-ores constitute the stratiform ore type, which is
171 conformably bedded within weathered mangan-phyllites preserving this meta-
172 sedimentary protore structure (Mishra, 1978). The massive Mn ore is found between ~
173 850 m and 892 m elevation in the first section, between 904 m and 912 m elevation in
174 the second ore body of the Kappataswamy pit (Fig. 3b), and between 926 and 982 m
175 elevation in Channanghi KMK-E pit (Fig. 3d). The massive ore is a podiform type with
176 cavity fillings (geodes) and botryoidal structures, which formed by secondary Mn
177 supergene enrichments (Mishra, 1978). Iron-Mn duricrust of ~ 10 m thickness caps the
178 two Mn-ore deposits immediately above a segregated/tabular ore body (Figs. 3b-d).

179 The samples collected in the Mn-ore deposits (Fig. 3b-d) were cut with a circular
180 saw to get the best angle for mineralogical observations on metallographic quality
181 polished thin sections (200-300 μm -thick) made from the sawed fragments (Fig. 4a).
182 Symmetrically to the thin sections, 500 μm -thick slabs are prepared for sub-sampling of
183 the target cryptomelane grains suitable for $^{40}\text{Ar}/^{39}\text{Ar}$ dating (Hénocque et al., 1998).

184

185 3.2. Analytical techniques and cryptomelane characterisation

186 Reflected light microscopy and X-ray micro fluorescence ($\mu\text{-XRF}$) analyses
187 allow precise characterisation of K-enriched Mn-oxide (cryptomelane) on the polished
188 thin sections (Fig. 4b-c). Microscopic observations of thin sections were accomplished
189 using a Leica DM-RXP microscope. The $\mu\text{-XRF}$ measurements were completed on
190 slabs using a X-ray microscope (XGT7000, Horiba Jobin Yvon) equipped with a X-ray
191 guide tube producing a finely focused and high-intensity beam with a 100 μm spot size,
192 Rh X-ray tube, accelerating voltage of 30 kV and current of 1 mA. X-ray emission from
193 the irradiated sample was detected via an energy dispersive X-ray (EDX) spectrometer

194 equipped with a liquid-nitrogen-cooled high purity Si detector. Micro mapping of the
195 coupled Mn-K occurrences on slabs contributes to precise identification of
196 cryptomelane, while Fe maps document the occurrence of Fe oxihydroxides (Fig. 4c). A
197 Scanning electron microscope (SEM) has been also used for fine-scale observations of
198 the cryptomelane crystals (Fig. 4d). Electron probe micro chemical analysis (EPMA) of
199 minerals on polished thin sections was completed using a CAMECA SX-100 electron
200 microprobe equipped with five wavelength-dispersive X-ray spectrometers (WDS).

201 Cryptomelane grains were separated from the slabs by hand picking.
202 Cryptomelane separates were crushed to produce powders ($< 64 \mu\text{m}$), which were
203 analyzed by XRD using a Panalytical X'Pert Pro MPD with a Co $K\alpha$ X-ray source ($\lambda =$
204 1.79 \AA) operating at 40 kV and 40 mA. The system is equipped with a linear
205 X'Celerator detector and a flat monochromator. The analysis range was $10\text{-}85^\circ 2\theta$ with
206 a step size of 0.033° and a counting time of 10 s per step.

207 After ultrasonically cleaning in absolute ethanol and conditioning in aluminium
208 foil packets, other separated grains were irradiated for 50 hours in the TRIGA Mark-II
209 reactor of Pavia University (Italia). The standard Taylor Creek Rhyolite sanidine 2
210 (TCRs-2) monitor, with an age of $28.05 \text{ Ma} \pm 0.08$ (Baksi et al., 1996), was analysed
211 after every ten unknown samples to allow determination of the factor J. After a two-
212 month "cooling" period, the irradiated cryptomelane grains were loaded in a double
213 vacuum Staudacher-type furnace for step heating Ar isotopes measurements. The
214 furnace temperature was calibrated by means of a classical thermocouple, and the gas
215 purification was accomplished using a cold trap with liquid air and Al-Zr AP10 getters
216 (one hot, one cold) for 8 minutes before the introduction into the VG3600 mass
217 spectrometer. One minute was allowed for equilibration before analysis. ^{40}Ar and ^{39}Ar

218 were measured on a Faraday cup with a resistor of 10^{11} ohm, while ^{39}Ar , ^{38}Ar , ^{37}Ar ,
219 and ^{36}Ar were analysed using a scintillator and photomultiplier after interaction on a
220 Daly plate. The analytical data are reported in data repositories (Tables DR1 and DR2),
221 and the errors are quoted at the 1σ level. Ages were analysed in two ways: plateaus and
222 inverse isochrones. Plateaus ages were calculated from at least three consecutive steps
223 comprising up to 50% of total $^{39}\text{Ar}_K$ released and overlapping at the 2σ confidence level
224 (Fleck et al., 1977; Maluski, 1985). Alternatively, plateau ages encompassing two steps
225 releasing $\sim 30\text{-}40\%$ ^{39}Ar were also defined as “plateau-like” or “pseudo-plateau” (e.g.,
226 Feng and Vasconcelos, 2007; Vasconcelos et al., 2013). Ages derived from best-fit
227 inverse isochrones in $^{36}\text{Ar}/^{40}\text{Ar}$ vs. $^{39}\text{Ar}/^{40}\text{Ar}$ correlation diagrams (Roddick et al., 1980)
228 were accepted when mean square weighted deviation (MSWD) was less than 2.5 and
229 the $^{40}\text{Ar}/^{36}\text{Ar}$ intercept within 2σ from the $(^{40}\text{Ar}/^{36}\text{Ar})_{\text{atm}}$ value of 298.56 ± 0.31 (Lee et
230 al., 2006; Renne et al., 2009), since supergene cryptomelane crystallised at shallow
231 depth, and thus incorporated excess argon, if any, in the atmospheric ratio. Age spectra
232 also show the variations of $^{40}\text{Ar}^*$ and K/Ca (derived from $^{39}\text{Ar}/^{37}\text{Ar}$) to track the
233 compositional effects on age.

234

235 **4. Results and Discussion**

236 Six samples (KPA-2.1, KPA-2.5, KPA-9, KPA-10, KPA-12a and KPA-12b)
237 from the Kappataswamy pit (Fig. 3b) and one (KMK-3) from the Channanghi KMK-E
238 pit (Fig. 3d) were analysed and dated. KPA-9 and KPA-12 show the platy preserved
239 meta-sedimentary structure with Mn pods, while KPA-2, KPA-10 and KMK-3 are
240 rather massive with cavity crystallisations (geode) and/or botryoidal pod concretions.

241

242 4.1. Mineralogical and geochemical characterisation of cryptomelane

243 Cryptomelane was identified on polished thin sections including μ -XRF and
244 SEM imaging (Fig. 4), and characterized by microchemical (EPMA) and XRD analyses
245 of separated grains (Figs. 5 and 6, Table 1). Samples KPA-9, KPA-12a and KPA-12b
246 are the richest in cryptomelane, which occurs as cryptocrystalline plasmic domains
247 formed within preserved inherited protore microstructures (Figs. 5a-c). Appropriately,
248 sample KPA-9 exhibits a relict of Si-Mn sedimentary protore with residual smoothed
249 small size quartz grains preserved in acicular cryptocrystalline cryptomelane (Fig. 4d).
250 The two different generations of cryptomelane (Fig. 5a) have the same stoichiometric
251 composition (Table 1). Separated grain of KPA-9 mainly contains cryptomelane, but
252 also pyrolusite (βMnO_2) and quartz (Fig. 6). Sample KPA-12a is composed of two
253 cryptomelane generations, the latest one filling the micro porosity developed across the
254 earliest generation and/or the other mineral components such as pyrolusite and Fe
255 oxihydroxides (Fig. 5b). The stoichiometric composition is very close to that of KPA-9
256 (Table. 1). Sample KPA-12b is very rich in cryptomelane (Fig. 5c) with higher K
257 content than KPA-12a (Table. 1). The two separated grains are dominantly composed of
258 cryptomelane but pyrolusite and goethite (γFeOOH) are also present (Fig. 6).
259 Samples KPA-2, KPA-10 and KMK-3 are composed of successive cryptomelane
260 crystallisations organized in banded overgrowth microstructures associated with other
261 Mn oxides (Figs. 5d-f and 6). In the sample KPA-2, the first three bands are pure
262 cryptomelane, the fourth is a cryptocrystalline mixture of nsutite and cryptomelane, and
263 the fifth is dominantly cryptomelane, while pyrolusite crystallized in a cavity (Fig. 5).
264 The cryptomelane crystals analysed in the different bands have different stoichiometric
265 compositions with K varying from 0.45 to 0.57 atoms (Table 1). Sample KPA-10

266 consists of a ~ 5 mm size cryptomelane concretion with a transition zone of less than 1
267 mm separating it from pyrolusite (Fig. 5e). Separated grain of KPA-10 is mainly
268 composed of cryptomelane with 0.5 K atoms (Table 1). Small quantities of pyrolusite,
269 goethite and hematite (Fe_2O_3) are also present (Fig. 6). Sample KMK-3 (Fig. 5f) is a
270 concretion of cryptomelane with less than 1.5 wt.% K (Table 1). The outer rims are
271 composed of lithiophorite $(\text{Al,Li})\text{MnO}_2(\text{OH})_2$ and nsutite (γMnO_2) (Fig. 5f), which also
272 occur as minor fractions in the XRD analysis of the separated grain (Fig. 6).

273

274 4.2. $^{40}\text{Ar}/^{39}\text{Ar}$ ages of cryptomelane

275 Two kinds of $^{40}\text{Ar}/^{39}\text{Ar}$ age spectra are distinguished from the step heating
276 degassing depending on their shape and interpretation. The first group displays regular
277 ^{39}Ar release spectra allowing calculation of plateau ages (Fig. 7a-e), which are
278 commonly defined at the 2σ error and accounting at least for 40% of the total ^{39}Ar
279 released from the sample (Vasconcelos and Conroy, 2003; Feng and Vasconcelos, 2007;
280 Beauvais et al., 2008). The second group corresponds to disturbed ^{39}Ar release spectra
281 (Fig. 8), which are more problematic to interpret. The analytical data are provided in the
282 data repository tables DR1 and DR2.

283

284 4.2.1. Regular $^{40}\text{Ar}/^{39}\text{Ar}$ age spectra

285 The $^{40}\text{Ar}/^{39}\text{Ar}$ age spectra from the first group have been obtained from the first
286 generation of cryptomelane crystallized in the meta-sedimentary protore (Figs. 5a and
287 5c, 7a and 7c), and for cryptomelane crystallisations in banded overgrowth
288 microstructures of botryoids (Figs. 5f and 7d) or geodes (Figs. 5d-e, 7b and 7e). In each

289 case the calculated plateau and pseudo-plateau ages agree with statistically meaningful
290 inverse isochron ages estimated in the correlation diagrams (Fig. 7).

291 However, these spectra present some anomalies at low and high temperatures
292 steps. The age spectrum of KPA-9 (Fig. 7a) shows younger individual ages (steps 10 to
293 13), which may reflect the degassing of a late cryptomelane crystallisation (Figs. 4b and
294 5a). The sample KPA-10 displays an age spectrum with a clear ascending “stair case”
295 pattern at low temperature (steps 1 to 7) with young ages affected by quite a large
296 standard deviation (Fig. 7b). The isochron suggests a moderate excess of ^{40}Ar , which
297 was interpreted as the result of primary mineral weathering (McDougall and Harrison,
298 1988; Vasconcelos et al., 1994a), which is unlikely in this sample (Figs. 5e and 7b,
299 Table 1). The higher amounts of released $^{40}\text{Ar}^*$ and ^{39}Ar in intermediate temperatures
300 (step 8 to 11), however, allow the best estimation of the cryptomelane crystallisation
301 age (Vasconcelos, 1999; Hautmann and Lippolt, 2000; Li and Vasconcelos, 2002;
302 Vasconcelos and Conroy, 2003; Feng and Vasconcelos, 2007). Sample KPA-12b is
303 characterised by well-constrained plateau and isochron ages (Fig.7c).

304 The age spectrum of KMK-3 exhibits an ascending “stair case” pattern at high
305 temperature (steps 8, 9, 10, 13 and 14) with older ages than the plateau age (Fig. 7d). In
306 such a case, it is usually admitted that radiogenic argon is released by two cryptomelane
307 phases, the older one retaining radiogenic ^{40}Ar better than the younger one (Feng and
308 Vasconcelos, 2007) resulting in a maximum plateau age estimate and an older
309 integrated age (Li and Vasconcelos, 2002). The plateau age of KMK-3 has also quite a
310 large error due to relatively low K content (Table 1 and Fig. 7d), and higher
311 atmospheric correction, the isochron being less sensible and thus more precise.

312 The age spectrum of KPA-2.1 (Fig. 7e) is more disturbed and does not allow
313 calculation of a standard plateau age (Fleck et al., 1977). Pseudo-plateaus ages have
314 been, however, previously estimated from similar spectra by considering only two
315 concordant steps including ~ 40% of ^{39}Ar released (e.g., Feng and Vasconcelos, 2007;
316 Vasconcelos et al., 2013). Two pseudo-plateaus occur in intermediate temperature steps
317 4-5 and 7-8, allowing for a concordant age estimate of 35.86 ± 1.10 Ma by combining
318 the four steps at the 2σ level (Fig. 7e). We note that the best-fitted isochron supports a
319 similar age (Fig. 7e). The high temperature steps 13 to 16 suggest a hypogene
320 contaminant (Vasconcelos et al., 1994a; Ruffet et al., 1996; Li and Vasconcelos, 2002),
321 which could have aged the intermediate steps (e.g., the step 6). The low K content
322 extraction in high temperature steps could have also suffered from imprecise blank
323 corrections. Therefore, similarly to the sample KMK-3 (Fig. 7d), the best-fitted isochron
324 might reasonably provide a more precise age (Fig. 7e).

325

326 4.2.2. *Disturbed $^{40}\text{Ar}/^{39}\text{Ar}$ age spectra*

327 The ^{39}Ar release spectra from the second group are characterized by an upward
328 convex shape (Fig. 8) suggestive of a mixing of two or more supergene cryptomelane
329 phases (Vasconcelos et al., 1995; Ruffet et al., 1996; Hautmann and Lippolt, 2000),
330 which may result from the handpicking of single grains. Phase mixing is obvious in
331 sample KPA-12a showing late cryptomelane crystallisation precipitated into the
332 porosity ($\leq 500 \mu\text{m}$) generated by the dissolution of a first plasmic cryptomelane
333 generation (Fig. 5b). Such a process may result in remobilization of low energy sites
334 (Colin et al., 2005; Beauvais et al., 2008). Phase mixing often occurs during separation
335 of grains from multiple overgrowth bands (Vasconcelos and Conroy, 2003). The grain

336 of KPA-2.5 was tentatively separated from a thin band (~ 500 μm) between a late
337 pyrolusite crystallisation in a geodic void and a band composed of a mixing of
338 cryptomelane and nsutite (Fig. 5d). The growth rate of cryptomelane crystals in banded
339 overgrowth structures (botryoids or geodes) is generally very low (Vasconcelos et al.,
340 1992; Dammer et al., 1996; Hénocque et al., 1998) and may result in very small size (≤ 1
341 μm) intimately intergrown cryptomelane phases in a same grain (Vasconcelos et al.,
342 1994b; Vasconcelos, 1999).

343 The pronounced upward convex shape of ^{39}Ar release spectra precludes plateau
344 age calculation, while correlation diagrams only allow estimation of minimum and
345 maximum ages of the oldest and youngest mixing phases, respectively (Fig. 8a-b). Each
346 step of the ^{39}Ar release spectra yields a mixing age of the intergrown cryptomelane
347 generations (Vasconcelos, 1999; Vasconcelos and Conroy, 2003; Feng and
348 Vasconcelos, 2007), implying that the ages in low and high temperature steps are
349 overestimates of the youngest phase, while the ages in intermediate temperature steps
350 are underestimates of the oldest phase (see also Ruffet et al., 1996; Beauvais et al.,
351 2008). This interpretation is mainly based on previous experiments (Vasconcelos et al.,
352 1995), which showed a maximal degassing of $^{40}\text{Ar}^*$ and ^{39}Ar between 700°C and 900°C
353 with a peak at 800°C. In such a range of temperatures, the two isotopic components are
354 release from the mineral structures (Vasconcelos et al., 1994b). Therefore, only
355 maximum and minimum ages might be estimated from upward convex spectra in the
356 temperature range specific of supergene cryptomelane degassing (Fig. 8). For KPA-2.5
357 (Fig. 8a), a minimum age of ~ 46 Ma (step 7) and a maximum age of ~ 30 Ma (step 10)
358 could be at best estimated for the crystallisation of the earliest and latest cryptomelane
359 phases, respectively. For KPA-12a the maximum and minimum ages of ~ 29 Ma (step

360 10) and ~ 34 Ma (step 8) are estimated for the youngest and oldest phases, respectively,
361 in agreement with the estimated isochron age limits, although the MSWD value of the
362 second isochron is quite large (Fig. 8b).

363

364 4.2.3. Cryptomelane $^{40}\text{Ar}/^{39}\text{Ar}$ ages summary

365 Despite mineral phase mixing of some samples, five significant plateau and
366 isochron ages are estimated between ~ 26 and 36 Ma. The older ages (~ 33.5 and 36
367 Ma) are obtained either for the first cryptomelane generation in samples showing the
368 preserved meta-sedimentary protore structure (Figs. 5a-c, 7a-c) or the first
369 crystallisation band of an overgrowth geodic microstructure (Fig. 5d and 7e). The
370 younger ages (~ 26-28 Ma) characterize the late cryptomelane crystallisations of banded
371 overgrowth microstructures (Figs. 5e-f, 7b and 7d). The ages of the different mixing
372 phases of KPA-2.5 and KPA-12a cannot be precisely determined. However, the two
373 cryptomelane phase of KPA-12a (Fig. 5b) may have crystallised between ~ 29 Ma and ~
374 34 Ma (Fig. 8c) that bracket the age of KPA-12b (Fig. 7c) sampled on the same open
375 mine bench. The maximum age (~ 30 Ma) estimated for the youngest cryptomelane of
376 KPA-2.5 is quite realistic compared to the older cryptomelane grain (KPA-2.1)
377 separated from the same sample (Fig. 5d and 7e). Despite their limited number, our
378 results usefully document Late Paleogene weathering history of southern India.

379

380 4.3. Potential implications for lateritic weathering and morphogenesis in southern India

381 The formation of cryptomelane in Mn-ore deposits is enhanced on landsurfaces
382 covered by a humid tropical forest reflecting warm and/or at least wet climatic
383 conditions (Vasconcelos et al., 1994a), like those prevailing for bauxite genesis in

384 tropical cratonic regions (Bárdossy and Aleva, 1990). Such hydroclimatic conditions are
385 required for complete weathering of K-phyllsilicates into kaolinite with release of K^+
386 in Mn^{2+} -rich acidic solutions (pH ~ 4.5-5) that may ultimately lead to cryptomelane
387 crystallisation under appropriate redox conditions (Parc et al., 1989). Therefore, massive
388 formation of cryptomelane in Mn ore deposits and/or lateritic weathering profiles is a
389 good indicator of humid/warm paleoclimatic conditions (Vasconcelos et al., 1994a;
390 Vasconcelos and Conroy, 2003; Beauvais et al., 2008). Under such climatic conditions,
391 wide areas were colonised by humid tropical forests over most of peninsular India (Kent
392 and Muttoni, 2008; Chatterjee et al., 2013) enhancing the pervasive development of
393 lateritic weathering profiles with differential sequestration of Al, Fe and Mn.

394 On the basis of global paleoclimatic reconstructions (e.g., Parrish et al., 1982),
395 Bárdossy and Aleva (1990) argued for intermittent Cenozoic bauxitic weathering in
396 South India. Such a bauxitic weathering intermittency was not observed in West Africa,
397 where bauxites preserved in current landscapes have been dated from Late Palaeocene
398 to Eocene at the latest (Prasad, 1983; Colin et al., 2005; Beauvais et al., 2008; Beauvais
399 and Chardon, 2013). Nevertheless, two stages of lateritic weathering processes have
400 been described in the bauxitic deposits of the Boknur-Navge plateau near Belgaum
401 (Krishna Rao et al., 1989b) (Fig. 1). The first bauxitic stage characterizes the syngenetic
402 lateritic weathering of the Deccan basalts under the action of slightly acidic meteoric
403 waters, whereas the second stage is a late weathering epigenetic process of the early
404 bauxite, under the effect of highly acidic meteoric Si-rich waters (Krishna Rao et al.,
405 1989b). The Boknur-Navge bauxitic plateau is also a remnant of paleolandsurface S2
406 correlated to the S2 remnant of the SGB (Gunnell, 1998). Syngenetic bauxitic
407 weathering on paleolandsurface S2 was likely active in the “Early Tertiary” (Paleogene

408 ?) as suggested by paleomagnetic investigations on Fe-oxihydroxides in bauxites and
409 laterites of the nearby Belgaum area (Schmidt et al., 1983; Kumar, 1986). Although no
410 precise ^{40}Ar - ^{39}Ar age correlative of syngenetic bauxitic weathering of the Traps basalts
411 was obtained in this study, lateritic weathering up to ~ 46 Ma may not be precluded
412 (Fig. 8a). Epigenetic lateritic weathering of paleolandsurface S2 (Krishna Rao et al.,
413 1989b) could however be recorded by our $^{40}\text{Ar}/^{39}\text{Ar}$ ages set.

414 The obtained ages (~26, ~ 28, ~ 33.5 and ~ 36 Ma) attest for intense Late Eocene
415 to Mid-Oligocene lateritic weathering(s) of the highest paleolandsurface (S2) preserved
416 as sparse relics on both the Mysore plateau and the Deccan Traps. These results suggest
417 that the high-elevation landsurface of Peninsular India evolved under a wet and warm
418 climate over a protracted period in the Late Paleogene, essentially during the Oligocene.
419 Repeated lateritic weathering between 26 and 36 Ma apparently correlates with low
420 clastic sediment fluxes and dominant carbonate sedimentation on the Arabian sea
421 margin (Campanile et al., 2008) but the links between inland weathering, erosion and
422 offshore sedimentation are poorly understood. Our $^{40}\text{Ar}/^{39}\text{Ar}$ results might also suggest
423 that the dissection and rejuvenation of the high-elevation lateritic surface of Peninsular
424 India took place in the Neogene as classically inferred (Radhakrishna, 1993). However,
425 the relative contributions of lithospheric deformation and climate change to geomorphic
426 rejuvenation of Peninsular India in the Neogene remain to be investigated.

427

428 **5. Conclusion**

429 We provide time constraints on lateritic weathering having affected Southern
430 India by using $^{40}\text{Ar}/^{39}\text{Ar}$ radiometric dating of supergene cryptomelane in Mn ore
431 deposits underlying a relict paleolandsurface (S2) that once covered the Mysore plateau

432 and Deccan Traps. Well-constrained plateau ages and best-fitted inverse isochrons
433 allow defining $^{40}\text{Ar}/^{39}\text{Ar}$ ages at ~ 26, ~ 28, ~ 33.5 and ~ 36 Ma. These results document
434 intense lateritic weathering in Southern India during the latest Eocene and the Oligocene
435 under the effect of warm and wet climate similar to that prevailing in humid forest
436 tropical environments. More specifically, the highest regional lateritic paleolandscape
437 (S2) of Peninsular India underwent chemical weathering at least between ~ 36 and ~ 26
438 Ma and was possibly dissected and rejuvenated afterwards, which suggests that most of
439 the current relief of South Indian high-elevation terrains was acquired in the Neogene.

440

441 **Acknowledgements**

442 We are thankful to the PhD school (ED251, AMU, OSU Pytheas), which
443 allocated to NB a grant from the French Ministry of Research and Superior Education.
444 We also thank IRD and INSU for financial supports (AB). Bujkesh was our guide
445 during a three weeks field trip. S.R. Sridhar, M.P. Deshpande and K. Raman are
446 gratefully thanked to having allowed us the sampling of the Deogiri Mn ore deposits.
447 Thanks to the assistance of Prakash Babu in the mining open pits. C. Demeurie (Thin
448 section Lab) made the polished thin sections. This work also benefited of technical
449 helps from D. Borschneck (XRD and μXRF), Y. Galy (SEM), J-P. Ambrosi (chemical
450 supplies), D. Merlet (EPMA), P. Münch and F. Lecoœur (Argon radiochronology). Jason
451 Price and an anonymous reviewer are also thanked for useful remarks and suggestions.

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454

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666

667 **Figures and table caption**

668 Figure 1. Simplified map of Peninsular India showing first-order geomorphic and
669 geological features (adapted from Radhakrishna, 1993). B= Bababudan; N= Nilghiris;
670 S= Shevaroy.

671 Figure 2. Simplified geology of the Sandur Greenstone Belt adapted from Chadwick et
672 al. (1996). (b) Corresponding topography (ASTER GDEM30) and delineation of relicts
673 of paleosurfaces S2 and S3. (c) Topographic cross-sections (lines of section are located
674 on Figure 2b).

675 Figure 3. North-looking Google Earth views of (a) Kappataswamy and (b) Channanghi
676 KMK-E pits showing the location of the cross-sections through the Mn ore bodies. (c)
677 Logs of sections 1 and 2. (d) Log of section 3. Black numbered samples are those
678 analysed and dated in the current study.

679 Figure 4. Characterisation of procedure used for separating cryptomelane grains in
680 samples. (a) Polished thin section observed by (b) reflected light microscopy and (c) X-
681 ray micro fluorescence, which yields a composite picture with Mn in blue, K in green
682 and Fe in red. (d) SEM image showing typical cryptomelane acicular crystals in a
683 composite weathering matrix, which preserves altered quartz grains of the protore. P =
684 pyrolusite; C = cryptomelane; Pr = protore; Q = quartz.

685 Figure 5. Reflected-light photomicrographs of mineral assemblages of dated samples
686 showing EPMA numbered targets. (a) Two generations of cryptomelane (C1, C2) and
687 Pyrolusite (P) formed into the Mn protore (Pr). (b) Coexistence of two generations of
688 cryptomelane (C1, C2) with pyrolusite (P) and Fe-oxihydroxide (Fe). (c) Crypomelane

689 (C) and pyrolusite with some Fe-oxihydroxides (Fe). (d) Cavity filling by banded
690 overgrowth of a succession of cryptomelane (C) with pyrolusite (P) and a mixed
691 overgrowth on nsutite+ cryptomelane (NC). (e) Massive cryptomelane and pyrolusite
692 with some oxihydroxides (Fe). (f) Massive banded overgrowth of cryptomelane (C)
693 with pyrolusite (P), lithiophorite (L) and nsutite. Sample names are reported on each
694 photomicrograph.

695 Figure 6. X-ray diagrams of separated cryptomelane sub-samples. C = cryptomelane; L
696 = lithiophorite; n = nsutite; Q = quartz; p = pyrolusite; G = goethite; h = hematite

697 Figure 7. ^{39}Ar - ^{40}Ar age spectra showing a well-defined plateau, with K/Ca (black) and
698 Ar^* (grey) step curves (left), and inverse isochrone diagrams (right) of cryptomelane
699 separates from Kappataswamy open pit samples (a-c and e), and Channanghi KMK-E
700 open pit sample (d). MSWD = mean square weighted deviation.

701 Figure 8. Disturbed ^{39}Ar - ^{40}Ar age spectra with K/Ca (black) and Ar^* (grey) step curves
702 (left) and inverse isochrone diagrams (right) of cryptomelane separates from
703 Kappataswamy open pit samples. (a) Separated phase of KPA-2.5 (target 5 in Figure
704 5d). Samples KPA-2.5 and KPA-12a do not allow calculation of plateau age but only
705 maximum and minimum ages of the mixed youngest and oldest phases, respectively
706 (stippled line depicted from bold numbered steps in the spectra). MSWD = mean square
707 weighted deviation.

708

709 Table 1. Electron Probe Micro Analyses (EPMA) of targeted cryptomelane phases (in
710 bold) and other Mn oxides shown in the Figure 5. Each analyse is the average of three
711 $5\mu\text{m}$ micro spots in a $50\mu\text{m}$ sized micro domain.

Highlights

- * First radiochronological dating of lateritic weathering in southern India
- * Late Palaeogene chemical weathering on the highest paleolandsurface preserved on the Mysore plateau and Deccan Traps
- * Potential rejuvenation and acquisition of the current relief of southern India in the Neogene

Figure. 1

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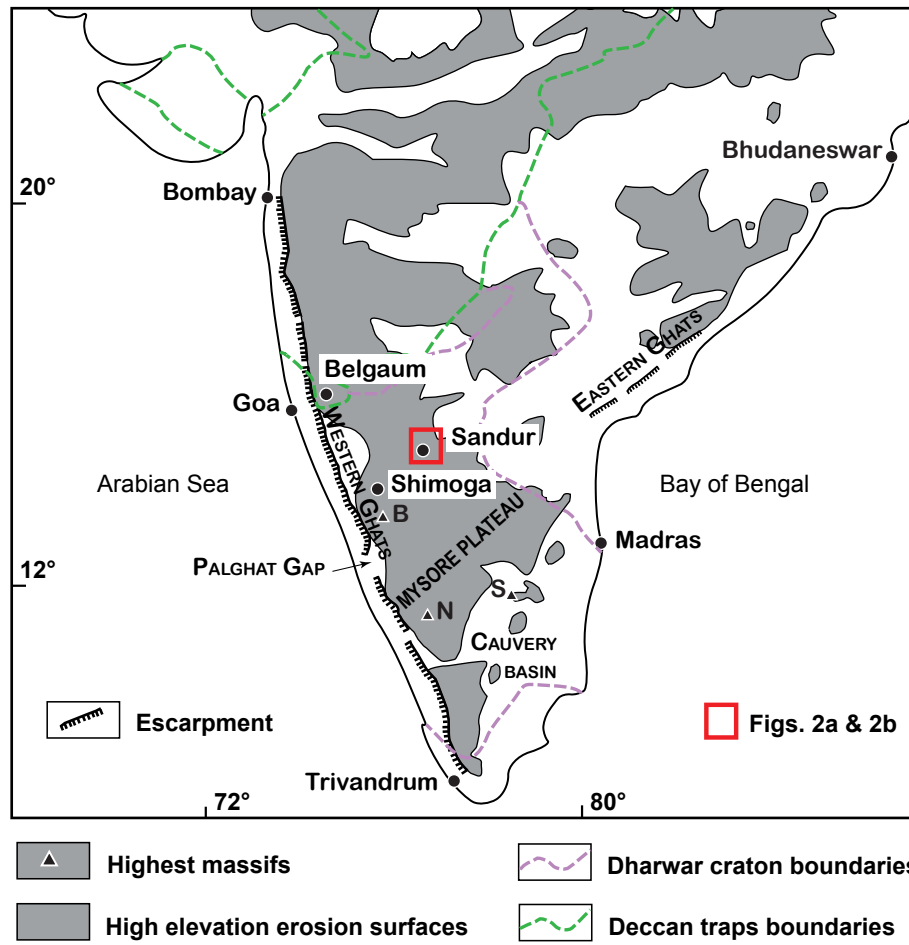


FIG. 1

Figure. 2
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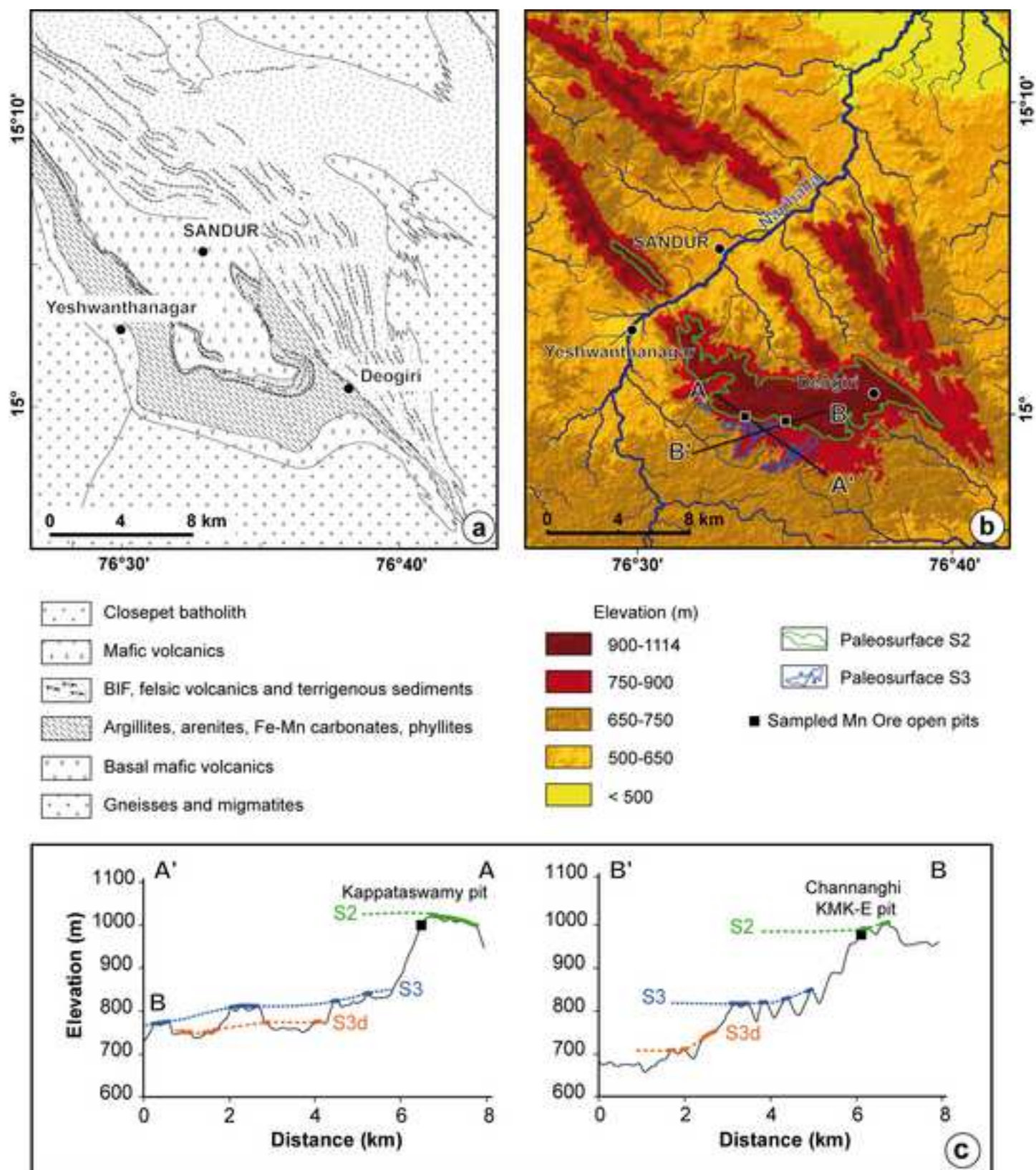
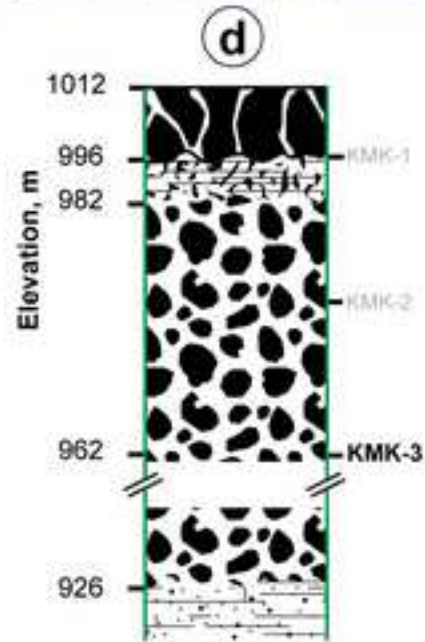
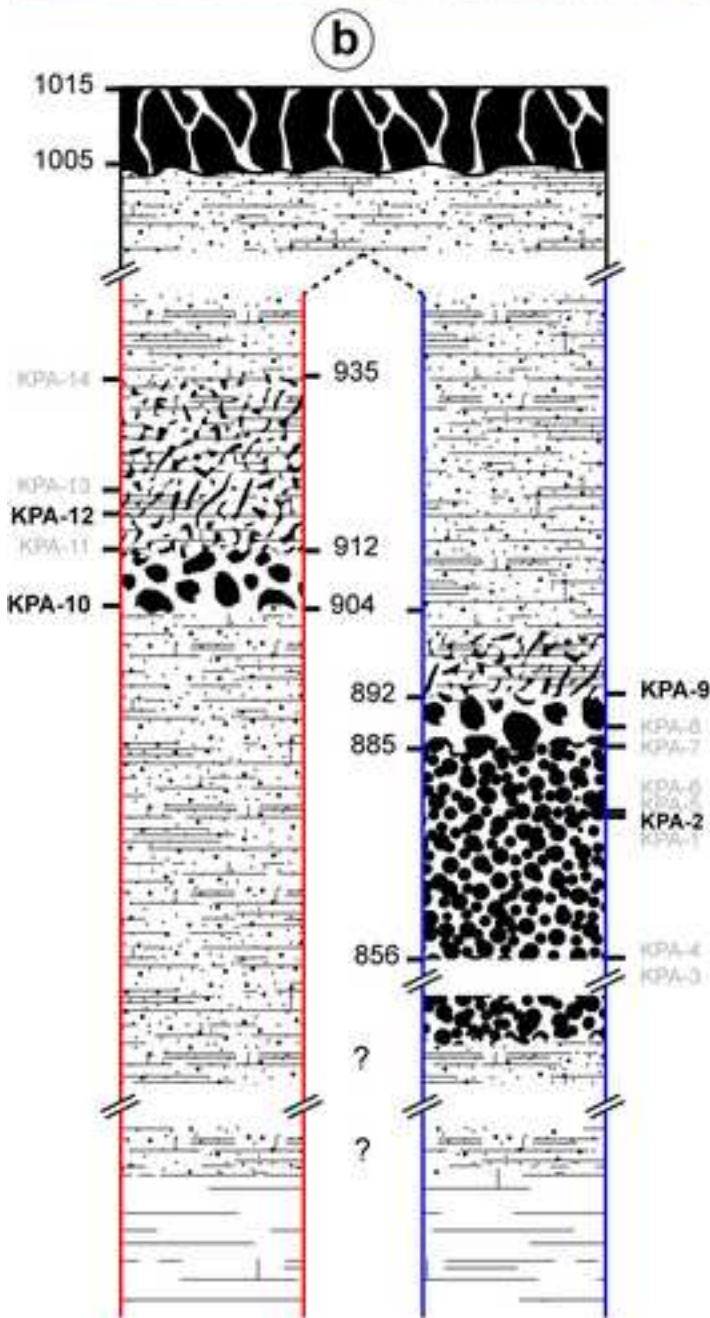
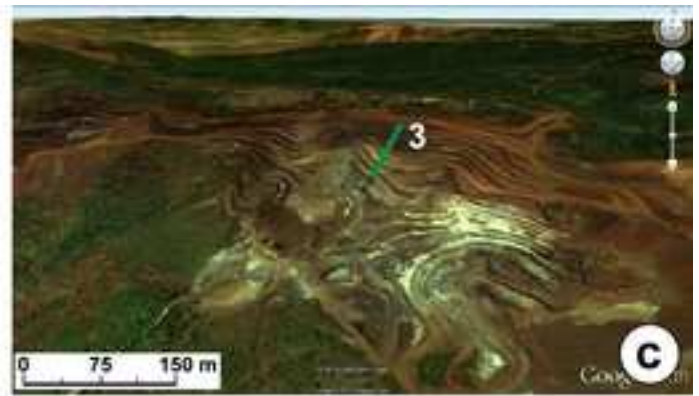
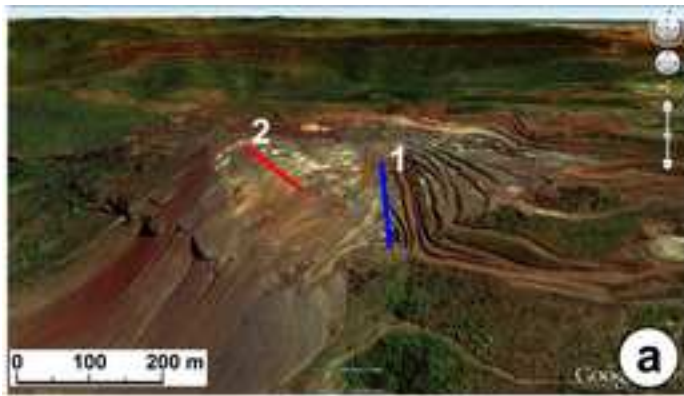


FIG. 2

Figure. 3
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Lateritic Mn ore sequence










-  Fe-Mn massive duricrust
-  Platy Mn-ore with pods
-  Massive Mn-ore with cavities > 1 cm
-  Massive Mn-ore with cavities ≤ 1 cm
-  Float ore
-  Sedimentary protore (Mangan phyllites)
-  Section 1
-  Section 2
-  Section 3

FIG. 3

Figure. 4
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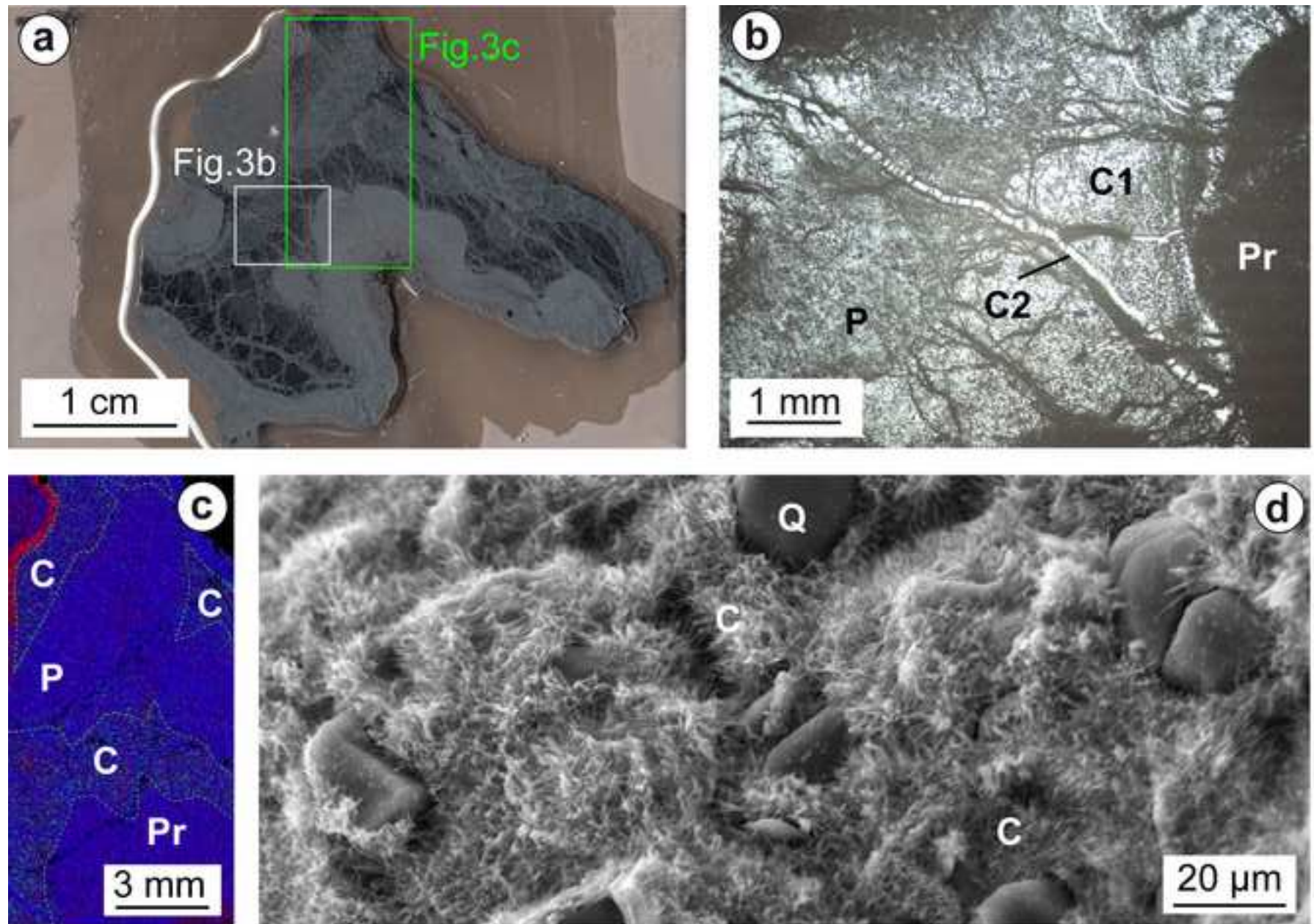


FIG. 4

Figure. 5
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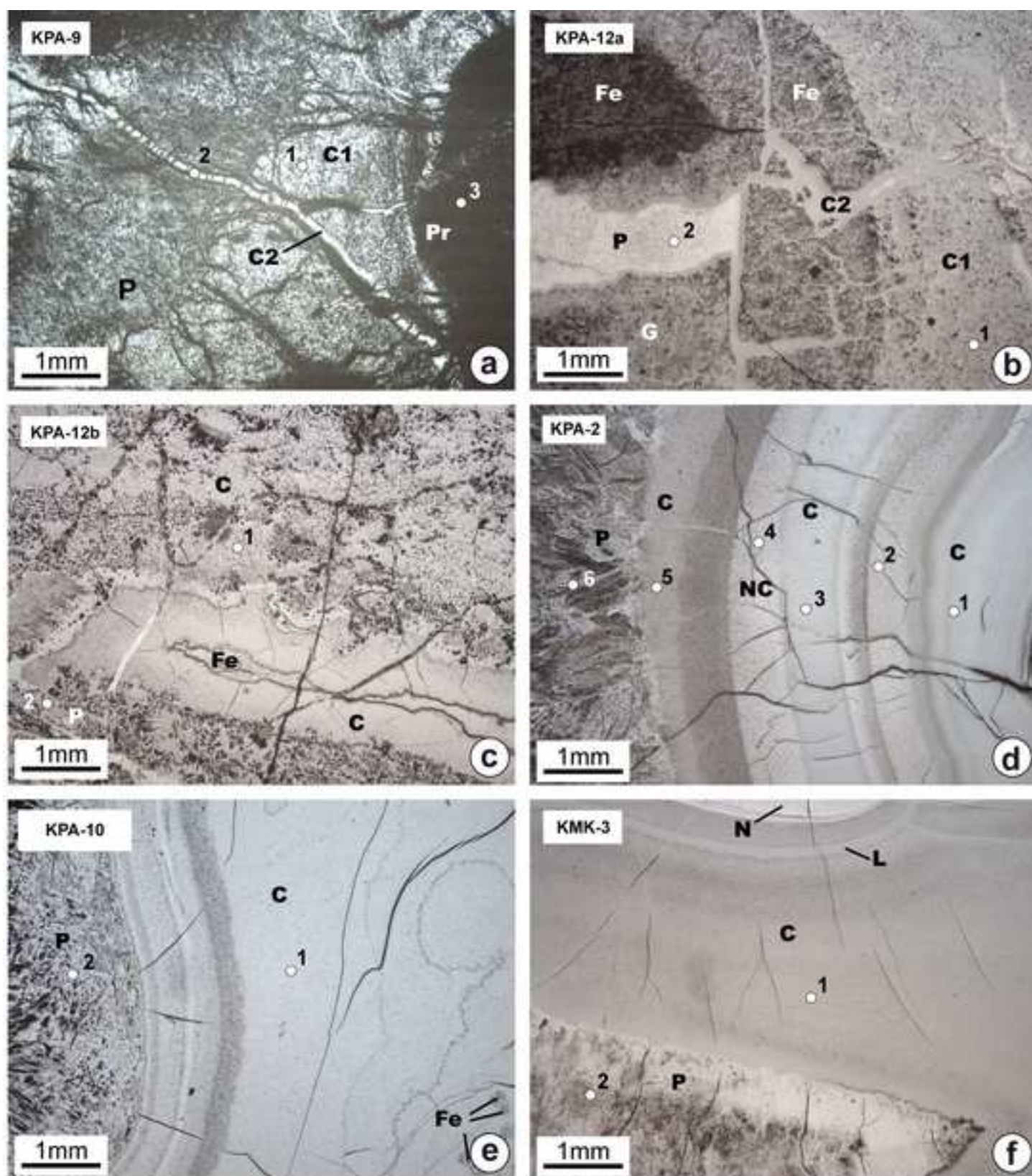


FIG. 5

Figure. 6

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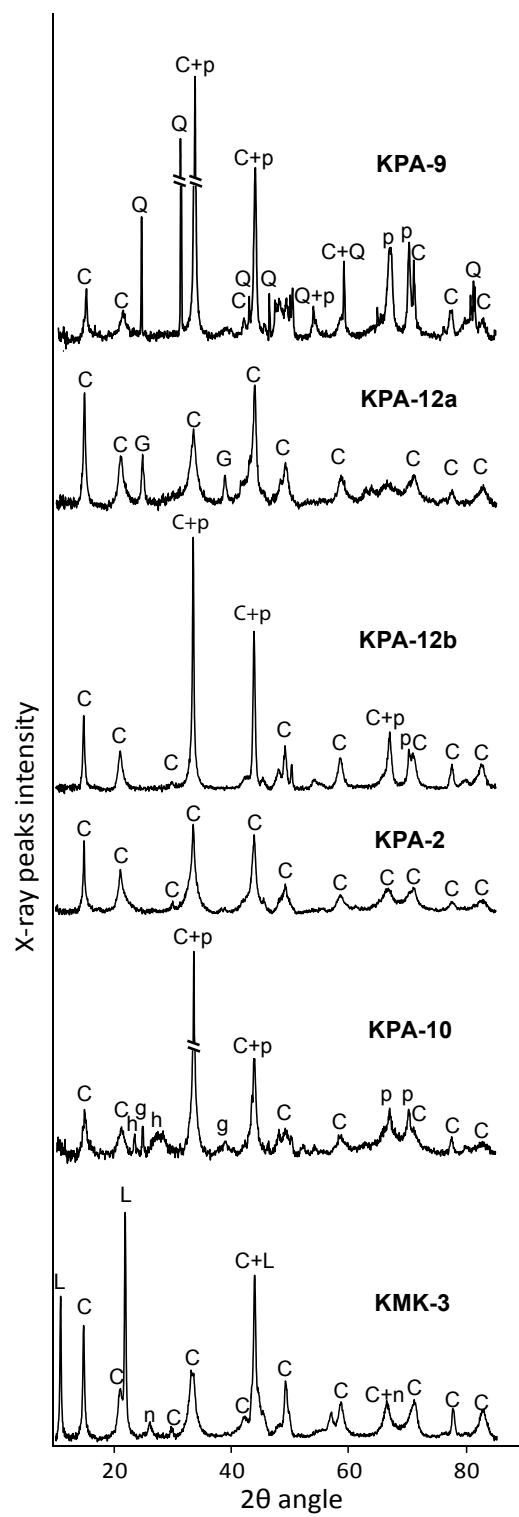


FIG. 6

Figure. 7

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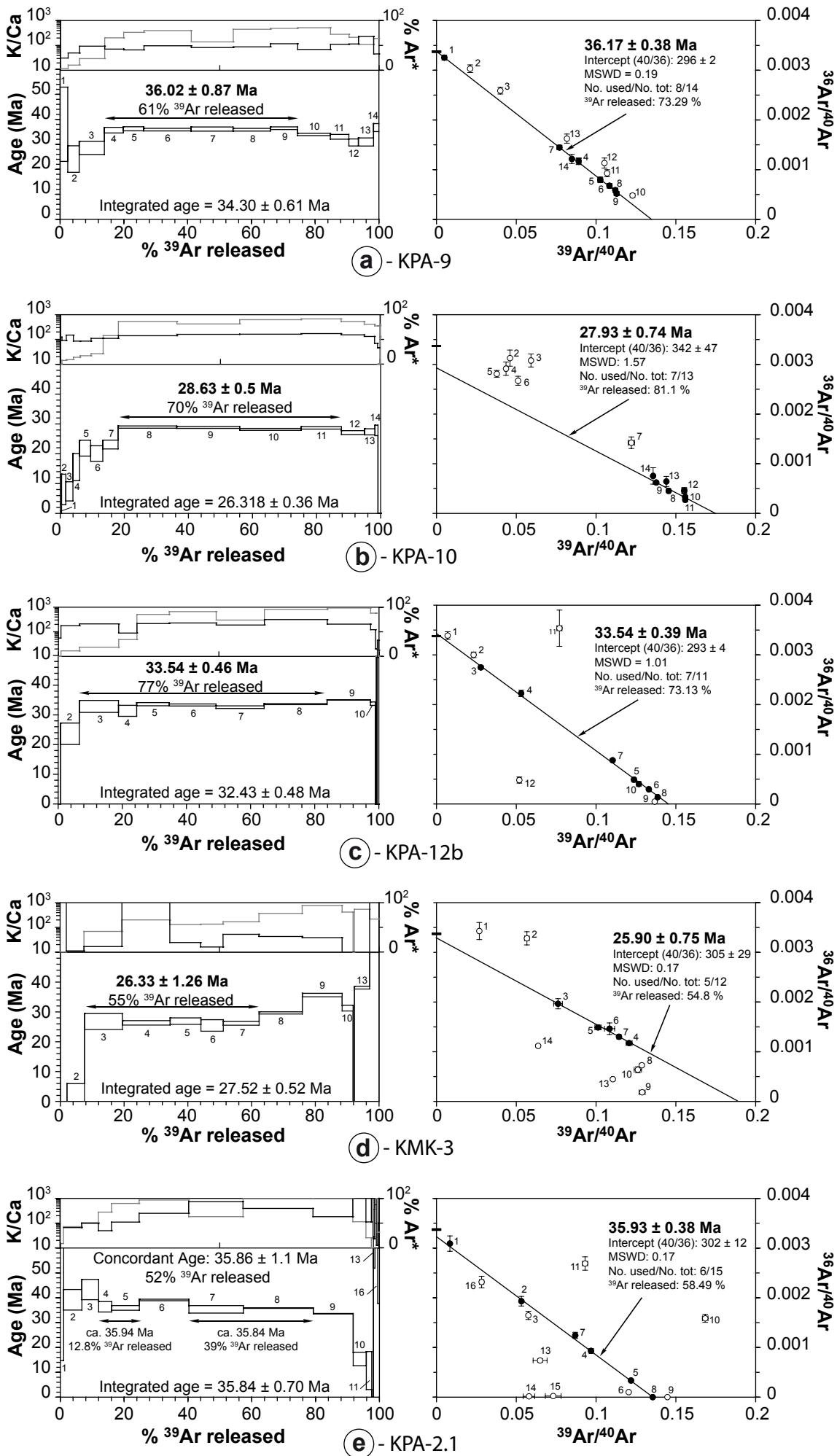


FIG. 7

Figure. 8

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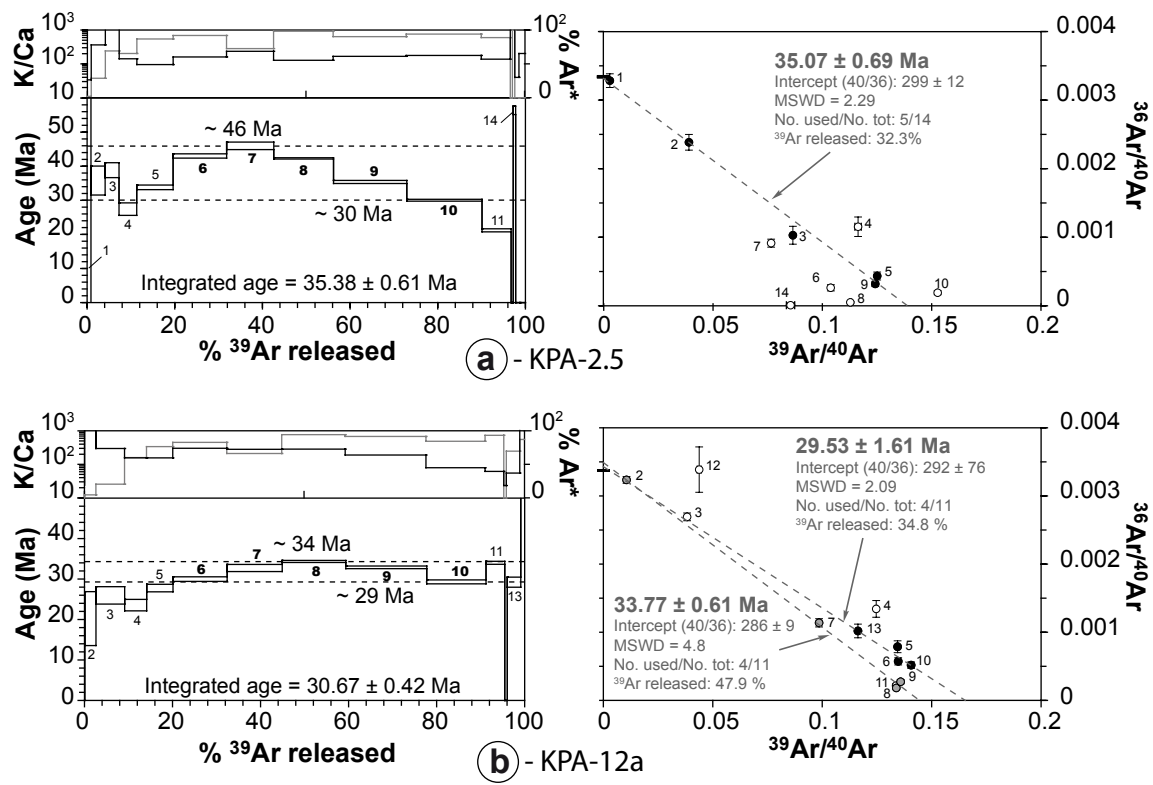


FIG. 8

Table. 1

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Sample ID	KMK-3		KPA-2						KPA-9			KPA-10		KPA-12a		KPA-12b	
	1	2	1	2	3	4	5	6	1	2	3	1	2	1	2	1	2
	C	P	C	C	C	C	C	P	C	C	Pr	C	P	C	P	C	P
wt. %																	
Mn	60.15	60.97	59.72	60.01	59.38	61.07	59.13	62.95	56.62	57.52	12.38	59.11	60.67	59.10	61.86	59.82	60.37
Al	0.15	0.30	0.12	0.30	0.22	0.33	0.20	0.00	1.41	1.24	0.37	0.20	0.29	0.45	0.11	0.18	0.37
Fe	0.13	0.13	0.12	0.13	0.13	0.13	0.13	0.14	0.58	0.51	0.17	0.05	0.13	0.81	0.14	0.17	0.85
K	1.41	0.06	2.49	2.26	2.44	0.94	3.03	-	2.13	2.19	0.50	2.65	0.87	2.19	0.19	3.29	0.29
Na	0.03	0.01	0.09	0.14	0.11	0.03	0.08	-	0.06	0.08	0.01	0.10	0.13	0.08	0.02	0.07	0.01
Zn	-	-	0.01	0.02	0.02	0.01	0.02	-	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Co	-	-	-	-	-	0.01	-	-	0.01	0.01	0.01	-	0.01	0.01	-	0.01	0.01
Ba	0.07	0.02	0.01	-	-	0.01	0.01	0.01	0.01	0.01	-	0.01	-	-	0.01	0.01	0.02
Ni	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Ti	-	-	-	-	-	-	-	-	0.01	0.01	-	-	-	-	-	-	-
Mg	-	0.00	0.01	0.01	0.01	0.01	0.00	-	0.01	0.01	-	0.02	0.01	0.01	-	0.00	0.02
Si	0.06	0.33	0.07	0.08	0.06	0.09	0.07	0.08	0.38	0.20	37.58	0.06	0.10	0.08	0.17	0.07	0.12
Ca	0.02	0.04	0.12	0.10	0.11	0.06	0.11	0.00	0.11	0.12	0.02	0.18	0.10	0.11	0.04	0.13	0.15
O	35.59	36.23	35.60	35.92	35.49	36.24	35.44	36.80	35.35	35.52	50.53	35.36	36.02	35.68	36.44	35.89	36.02
Total	97.62	98.11	98.36	98.98	97.99	98.92	98.24	99.99	96.68	97.41	101.6	97.75	98.35	98.53	98.99	99.64	98.24
Stoichiometric composition of cryptomelane based on 16 oxygens																	
K	0.4		0.45	0.41	0.45	0.17	0.57		0.42	0.4		0.5		0.4		0.61	
Mn ^{IV}	7.6		7.55	7.59	7.55	7.83	7.43		7.58	7.6		7.5		7.6		7.39	
Mn ^{III}	0.4		0.45	0.41	0.45	0.17	0.57		0.42	0.4		0.5		0.4		0.61	

- : not detected ; C = Cryptomelane ; P = Pyrolusite ; Pr = Si-Mn protore

Table. 1

Data repositories Tables

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