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2 (final accepted version)

3 # Magnetite–apatite deposit from Sri Lanka:

4 # implications on Kiruna-type mineralization

5 # associated with ultramafic intrusion and mantle

6 # metasomatism

7 **Xiao-Fang He[1,2], M. Santosh[1,2,3]*, T. Tsunogae[4,5], Sanjeewa P.K. Malaviarachchi[6]**

8 [1] *School of Earth Sciences and Resources, China University of Geosciences Beijing, No. 29 Xueyuan Road,*
9 *Haidian District, Beijing, 100083, China*

10 [2] *Department of Earth Sciences, School of Physical Sciences, University of Adelaide, Adelaide, SA 5005,*
11 *Australia*

12 [3] *State Key Laboratory of Continental Dynamics, Department of Geology, Northwest University, Xi'an 710069,*
13 *China*

14 [4] *Faculty of Life and Environmental Sciences, University of Tsukuba, Ibaraki 305-8572, Japan*

15 [5] *Department of Geology, University of Johannesburg, Auckland Park 2006, South Africa*

16 [6] *Department of Geology, Faculty of Science, University of Peradeniya, Peradeniya 20400, Sri Lanka*

17 *Corresponding author e-mail: msantosh.gr@gmail.com*
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19 # ABSTRACT

20 Kiruna-type iron oxide–apatite associations occur in a variety of rock types and their

21 origin has remained controversial. Most of the Kiruna-type deposits are associated with

22 intermediate to felsic rocks, and in rare cases with ultramafic rocks. Here we investigate the

23 Seruwila iron oxide–apatite deposit at the contact between the Highland and Vijayan

24 complexes which has been defined as the 'eastern suture' in Sri Lanka which formed during

25 the late Neoproterozoic assembly of the Gondwana supercontinent. The ore deposit is hosted

26 in an ultramafic intrusion and comprises massive and disseminated mineralization. The ore-

27 bearing rocks are mainly composed of low-Ti magnetite and chlor-fluorapatite. Our

28   petrological and geochemical studies suggest a magmatic–hydrothermal model for the

29   mineralization wherein: 1) the Cl-rich magmatic–hydrothermal fluid scavenged iron and P

30   from the ultramafic magma, transported iron to shallower levels in the crust and deposited

31   along the suture zone to form the massive type magnetite and apatite; and 2) the cooling of

32   the hydrothermal fluids resulted in the growth of disseminated magnetite and the precipitation

33   of sulfide minerals, followed by a calcic metasomatism (scapolitization and actinolitization).

34   This model is in conformity with the genetic relation between Kiruna-type deposits and iron

35   oxide–copper–gold (IOCG) deposits. We also report LA–ICP–MS zircon U–Pb ages from the

36   host ultramafic intrusion suggesting its emplacement at ca. 530 Ma, which is younger than

37   the regional high-grade metamorphism associated with the collisional assembly of the crustal

38   blocks in Sri Lanka at ca. 540 Ma. By analogy with the common occurrence of Kiruna-type

39   deposits in extensional tectonic settings, and the geochemical features of the studied rocks

40   including low silica, high Mg, Fe, Ca with high field strength elements (HFSEs) (Nb, Ta, Zr,

41   Hf, Ti) depletion and strong LREE, F enrichment, we envisage that the ultramafic

42   magmatism occurred in a post-collisional extensional setting derived from a volatile- and

43   LREE-rich metasomatized lithospheric mantle.

44   **Key words:** Kiruna-type; IOA deposits; magnetite; apatite; ultramafic intrusion; Sri Lanka

INTRODUCTION

46   Kiruna-type iron oxide–apatite (IOA) deposits are important sources of Fe and P, and

47   occur in a variety of geological settings such as those in Sweden, Iran, American Cordillera,

48   and China (Herz and Valentine 1970; Hou et al. 2012; Jonsson et al. 2013; Sabet-Mobarhan-

49   Talab et al. 2015). They share common mineralogical features dominated by low-Ti

50   magnetite with variable amounts of apatite but poor in quartz (Tornos et al. 2016). They also

51   generally display a close relationship with alkali-calcic intermediate to felsic rocks. The

52   origin of IOA deposit is still debated. Some workers favor a magmatic model due to the close

53   genetic relationship of the Kiruna-type magnetite–apatite mineralization with igneous suites

54   (Park 1961; Henriquez and Martin 1978; Naslund et al. 1998), whereas others favor a

55   hydrothermal origin based on evidence from fluid inclusions, replacement texture and

56   metasomatic zones (Barton and Johnson 1996; Murray and Oreskes 1997; Simon et al. 2004;

57   Dare and Beaudoin 2015). Recent studies have proposed an integrated magmatic–

58   hydrothermal model (e.g. Taghipour et al. 2015; Knipping et al. 2015; Tornos et al. 2016),

59   which also confirms the genetic connection between Kiruna-type IOA and iron oxide–

60   copper–gold (IOCG) deposits, in which IOA deposits represent the deeper roots of the IOCG

61   systems (e.g. Sillitoe, 2003).

62   Occurrence of Kiruna-type iron oxide–apatite (IOA) rocks in ultramafic rocks are rare.

63   Only a few examples have been reported including fluoro-hydroxyl-apatite associated with

64   chlorite-talc schists located in the margin of small ultramafic bodies in Georgia and Maryland,

65   USA (Herz and Valentine 1970), and small concentrations of magnetite–apatite rocks in

66   ophiolite complexes (Orthys Complex, Greece; Mitsis and Economou-Eliopoulos 2001;

67   Lizard Complex, UK; Hopkinson and Roberts 1999). However, most of these are small-scale

68   deposits.

69   The Seruwila magnetite–apatite deposit occurs along the tectonic contact between the

70   Vijayan and Highland Complexes which is interpreted as a mineralized domain along a

71   Neoproterozoic suture zone, also referred to as the "eastern suture" in Sri Lanka, (e.g. He et

72   al. 2016). The ore-bearing rocks are mainly composed of magnetite and apatite in various

73   proportions, hosted in an ultramafic intrusion with cumulate features. Although this deposit

74   has no world-class economic value given its relatively small size as compared to other iron

75   deposits, it stands as the unique example of a Kiruna-type IOA deposit associated with

76   ultramafics within a Gondwana-aged suture. In addition, the Seruwila IOA deposit is

77 important in addressing the long-standing controversy over the magmatic vs. hydrothermal

78 origin of such deposits in general.

79   Although the Seruwila IOA deposit was first discovered by the Sri Lankan

80 Geological Survey more than thirty years ago, there is only very limited information about

81 the classification or genesis of this deposit. The age of the ultramafic host rocks and the

82 geochemical characteristics of this association also remain unknown. In this study, we

83 provide the first systematic investigation on this rare IOA–ultramafic association. We present

84 the petrology, mineral chemistry, zircon U–Pb geochronology and Lu–Hf isotopic studies

85 from the ultramafic host and ore-bearing rocks in order to gain insights on the genesis of this

86 possible Kiruna-type mineralization. The new age data that we report from the ultramafic

87 rocks also provide better constraints on the tectonic setting of Sri Lanka within the Gondwana

88 supercontinent during the Late Neoproterozoic–Cambrian transition.

GEOLOGICAL BACKGROUND

89

The Vijayan and Highland Complexes

90

91   The metamorphic basement of Sri Lanka is subdivided into three major approximately

92 north–south trending complexes based on stratigraphic correlation and metamorphic grade.

93 These are the Vijayan Complex (VC) in the east, the Highland Complex (HC) in the centre,

94 and the Wanni Complex (WC) in the west (Fig. 1). The Vijayan Complex (VC) is mainly

95 composed of Neoproterozoic (1060–620 Ma) granitoid gneisses, migmatites and minor

96 metasedimentary enclaves of quartzite and calc-silicate rocks (Kröner et al. 1991). Most of

97 the metagranitoids range in composition between diorite and granite with the dominant

98 compositions being granodiorite and granite (Kröner et al. 1991; Milisenda et al. 1994; Fig.

99 2a). The compositional characteristics of the VC granitoid gneisses were ascribed to

100 subduction-related arc magmatism (Kröner et al. 2013; He et al. 2016; Ng et al. 2017). The

101 Highland Complex (HC) is dominantly composed of Archean–Paleoproterozoic

102 metasedimentary rocks, with interbedded quartzites and pelites in the metasedimentary belt,

103 which were intruded by both felsic and mafic sills and dykes (Kröner and Jaeckel, 1994;

104 Braun and Kriegsman, 2003; Santosh et al. 2014; Takamura et al. 2015). Rocks of both VC–

105 HC series underwent high-grade metamorphism during Late Neoproterozoic to early

106 Cambrian (Dharmapriya et al. 2015; He et al. 2016 and references therein).

107 The Vijayan–Highland Complex (VC–HC) preserves evidence for typical rock

108 association developed in subduction-related settings including metachert, serpentinised

109 ultramafic units, and metabasalts (He et al. 2016). Cr–Ni serpentinites (Fig.1) near Ussangoda

110 (Rajapaksha et al. 2012; He et al. 2016), Cu-sulphide deposits (Dissanayake and

111 Weerasooriya 1986), massive magnetite–hematite deposit at Wellawaya, also occur along the

112 boundary between the VC and HC in addition to the IOA-type deposit at Seruwila

113 (Jayawardena, 1982). The abundance of mineralization support the notion that the suture is a

114 persistently mineralized domain.

The Seruwila iron oxide–apatite deposit ore geology

116 The study area of Seruwila is located in the Trincomalee District about 275 kilometers

117 from Colombo (Fig. 1). The dominant basement rocks in this area are charnockites

118 (orthopyroxene-bearing anhydrous granulites) and quartzite (HC) and metagranites and

119 amphibole–biotite gneisses to the southeast (VC). The mineralization at Seruwila occurs at

120 the eastern boundary between the HC and the VC (Fig. 2a).

121 The massive or scattered ores in Seruwila occur as well-defined lenticular pods within

122 ultramafic rocks. They are cut across by a group of normal faults (Fig. 2b). The magnetite-

123 bearing ultramafic rocks occur as discontinuous and disrupted layers with length ranging

124 from 1 to 5 meters, and the weathered surfaces contain secondary copper minerals such as

125 malachite and azurite (Fig. 2b). Coarse scapolite crystals range up to 2 cm in length in the

126 ultramafic intrusion–ore deposit transition zone. Clinopyroxene-bearing intermediate rocks

127 (enderbite) constitute the major basement rock in the deposit area showing a sharp contact

128 with the ultramafic rocks and contains no ore mineral (Jayawardena, 1982). The

129 clinopyroxene-bearing rocks show alkaline features and has an emplacement age of 924 ± 10

130 Ma with metamorphism at 541 ± 14 Ma (He et al. 2016). A layer of coarse-grained calc-

131 silicate rock and coarse-grained anorthosite occur at the contact of the mineralized zone

132 (Jayawardena, 1982). The calc-silicate is mainly composed of calcite, apatite and olivine

133 (fayalite) and the anorthosite is mainly composed of plagioclase with labradorite composition

134 (Pathirana, 1980). Secondary calc-silicate veins containing sulphides occur along fractures of

135 the section. Secondary veins containing serpentinite, anhydrite or gypsum are also present

136 (Jayawardena, 1982).

137   The massive magnetite ore bodies range in thickness from 1–10 meters and consists of

138 highly coarse-grained magnetite and apatite in which the magnetite contains diopside

139 inclusions. The disseminated magnetite ores formed within the calc-silicate veins together

140 with minor sulphides, and are mainly composed of magnetite, chalcopyrite, pyrrhotite, and

141 pyrite together with apatite and scapolite, tremolite, diopside and minor actinolite and calcite.

142 The chalcopyrite is better-developed in the disseminated ore compared with the massive ores.

143 ## PETROLOGY

144   We summarize below brief field relations and salient petrographic features of the

145 different rock types from which twelve representative samples were analyzed in this study

146 (Table 1). The representative field photographs are shown in Fig. 3.

147   The magnetite–apatite ore and host rock samples were collected from a disused mine

148 in Block-C of the Seruwila deposit. According to the mineral assemblages, the rocks can be

149 divided into: 1) ultramafic host rock; 2) massive magnetite–apatite ore rock; 3) disseminated

150 magnetite–apatite ore and 4) transitional zone ore-bearing ultramafic rock and 5)

151 clinopyroxene-bearing enderbitic basement rock (He et al. 2016). Neither metamorphic fabric

152 nor deformational features occur in the studied rocks.

153 **Group 1: ultramafic host rocks.** Sample VC16-1 and VC18-4 represent the ultramafic host

154 rocks of the magnetite–apatite ore. In hand specimen, the rocks display medium to coarse-

155 grained texture with clinopyroxene, amphibole, as the dominant minerals with minor fine-

156 grained magnetite and apatite. The pale green clinopyroxene occurs as medium to coarse-

157 grained (1–2 mm) euhedral crystals and some carry amphibole inclusions (Fig. 4a). They

158 show cumulus textures including grain triple junctions and large dihedral angles (~120°) (Fig.

159 4a, b). Texturally two types of amphiboles are identified; the first is represented by coarse-

160 grained (0.5–1 mm), pale green euhedral amphibole which is free of inclusions. The second

161 type is fine-grained (<0.1 mm) and brownish, occurring as anhedral inclusions in

162 clinopyroxene. The main opaque mineral is magnetite, the distribution of which is limited in

163 these samples (Fig. 4a), with smaller grain size and irregular morphology compared to the

164 magnetite crystals in sample VC18-3 and VC18-6. Zircon grains usually occur as inclusions

165 in the silicate minerals (Fig. 4b).

166 **Group 2: Massive magnetite–apatite ore rocks.** Samples VC18-3, VC16-4 are mainly

167 composed of coarse-grained magnetite with abundant apatite (Fig. 3a). The mineral

168 assemblage consists of magnetite (30–40 vol%), apatite (20–30 vol%), amphibole (15–20

169 vol%) and clinopyroxene (5–10 vol%) with minor spinel. The rock shows a magmatic texture

170 in the absence of foliation and carries euhedral magnetite with size up to 3–5 cm and coarse-

171 grained euhedral apatite more than 3 cm in diameter (Fig. 4c, d). Magnetite contains

172 inclusions of apatite and green euhedral spinel. Spinel usually occurs along magnetite

173 boundaries and shows hexagonal shape. Euhedral apatite grains are also common as

174 inclusions in magnetite.

175 **Group 3: Disseminated magnetite–apatite ore rocks.** Samples VC16-2, 18-1 and 18-6 are

176 apatite-bearing disseminated magnetite ores (Fig. 3b, c). The mineral assemblage is magnetite,

177 apatite, and scapolite with minor amphibole, clinopyroxene, and calcite. The dominant

178 amphibole is brownish, coarse-grained (1–2 mm) and subhedral and is interpreted to be the

179 primary mineral in the protolith (Fig. 4e, f). Clinopyroxene occurs in two textural

180 associations in both groups of samples: one as coarse greenish grains (size 1–2 mm in size)

181 and the other as subhedral brownish grains with amphibole rims (Fig. 4f). Brown colored

182 anhedral actinolite surrounding clinopyroxene (0.2–0.5 mm) may represent a late stage

183 mineral (Fig. 4e), and the dark brown to red thin band occurring along the boundary of

184 magnetite and clinopyroxene probably formed during later alteration. The other textural

185 occurrence of clinopyroxene is as smaller grains mainly surrounded by actinolite, possibly

186 formed through hydrous alteration. Apatite is white to light gray in color and occurs as

187 euhedral to subhedral crystals. Apatite also occurs as inclusions within magnetite, and most

188 apatite grains contain abundant fluid inclusions. Magnetite in disseminated ore samples

189 shows is characterized by smaller and anhedral grain shapes, and is mostly found surrounding

190 silicates as aggregates and less in spinel exsolution (Fig. 4d) compared to the massive

191 magnetite ores. The apatite to magnetite proportion is highly variable. Chalcopyrite occurs as

192 anhedral grains along with magnetite and scapolite. Most calcite and actinolite occur at the

193 grain boundary, surrounding magnetite or amphiboles, and are generally anhedral. Scapolite

194 is characterized by replacement textures cutting across magnetite and silicates (Fig. 4).

195 **Group 4: Transitional zone ore-bearing ultramafic rocks.** Sample VC16-3, 16-5, 16-6,

196 18-2, 18-5 are from the transitional zone of the ore body and ultramafic rocks (Fig. 3d). They

197 are generally medium to coarse-grained with equigranular massive texture, and are dark

198 colored in hand specimen. The rocks are composed of clinopyroxene, magnetite, amphibole,

199 scapolite, and apatite with accessory hematite. The pale green colored clinopyroxene occurs

200 as subhedral crystals (Fig. 4g, h). Amphibole is fine to medium grained and subhedral, and

201 the clinopyroxene and amphibole are free of inclusions (Fig. 4g, h). Scapolite is light green in

202 color, medium to coarse-grained (0.5–1.5 cm) and contains abundant needle-like apatite

203 inclusions. Apatite not included in scapolite occurs as medium to coarse-grained colorless

204 crystals (0.5–1.5 cm). Magnetite and minor chalcopyrite are the dominant opaque minerals

205 with size ranging in size from medium to coarse-grained (1–3 mm) and occur as anhedral

206 disseminated crystals.

ANALYTICAL METHODS

207

208 Mineral chemical analyses were carried out using an electron microprobe analyzer (JEOL

209 JXA8530F) at the Chemical Analysis Division of the Research Facility Center for Science

210 and Technology, the University of Tsukuba. Whole-rock major-oxides and trace-elements

211 including rare earth elements (REE) of sixteen representative samples were analyzed by XRF

212 and LA–ICP–MS instruments at the National Research Centre of Geoanalyses, Beijing

213 (China). U–Pb dating and trace element analysis of zircon were simultaneously conducted by

214 laser ablation–inductively coupled plasma–mass spectrometry (LA–ICP–MS) at the Key

215 Laboratory of Continental Collision and Plateau Uplift, Institute of Tibetan Plateau Research,

216 Chinese Academy of Science, Beijing, China. In situ zircon Hf isotopic analyses were

217 conducted on the same spots or in the adjacent domains where U–Pb dating was done.

218 The details of analytical techniques relating to petrography, mineral chemistry, whole rock

219 chemistry, and zircon U-Pb and Lu-Hf isotopic analyses are given in Supplementary file:

220 Appendix 1.

RESULTS

221

222 The details of mineral chemistry, whole rock chemistry, zircon U–Pb and Lu–Hf isotopic

223 analyses results are given in Supplementary file: Appendix 1. The related figures are Figs. 5–

224   9. The data table are in Table 2 and Supplementary Table 1–3.

225   **DISCUSSION**

226   **Petrogenesis of Seruwila ultramafics: a CO_2 and LREE enriched**

227   **metasomatized mantle beneath VC–HC boundary?**

228   The ultramafic samples (clinopyroxenite) are distinct from the surrounding

229   Neoproterozoic (ca. 924 Ma, He et al. 2016) granitic basement rocks which are strongly

230   deformed and metamorphosed at upper amphibolite- to granulite-facies conditions. Neither

231   metamorphic fabric nor deformational features are observed in the ultramafic host or their

232   ore-bearing zones (Fig. 3). The close relationship of the ultramafics with ore deposit,

233   particularly the massive ores which occur within the ultramafic rock suggests a genetic link

234   between the ultramafic rocks and the mineralization. The high-Mg bulk chemistry, cumulus

235   texture including grain triple junctions and the large dihedral angle (~120°, Fig. 4a, b)

236   (Holness et al. 2005), the coarse-grained euhedral crystal shape of pyroxene and amphibole,

237   and the association of anorthosite in this deposit (Jayawardena, 1982) suggest fractionation in

238   a volatile-rich magma chamber. Apatite chemistry characterized by moderately high

239   concentration of F and Cl further support the volatile-rich signauture (Fig. 5b).

240   All the studied samples are characterized by low silica, high Mg, Fe, Ca and a narrow

241   range in Sm/Nd ratios consistently less than the bulk Earth value. They show general HFSE

242   depletion including negative Zr, Hf, Ti, Y anomalies (Fig. 6). The chondrite-normalized REE

243   compositions and primitive-mantle-normalized trace element compositions (Fig. 6) are

244   characterized by LREE enrichment with variable total LREE concentrations and HREE

245   depletion. Jayawardena (1982) analyzed the bulk chemistry of the adjacent calc-silicates and

246   reported very low values of metallic elements as compared to those in the massive and

247   disseminated ores. However, the calc-silicate rocks show comparatively high manganese

248   concentrations (437–754 ppm) and high Y, La, Sr and Ba contents. The calc-silicate rock was

249   likely the source for the late-stage calcic metasomatism (scapolitization and actinolitization)

250   and probably was not the major source of Fe in the ore deposit.

251   The combined petrological and geochemical features suggest that the ultramafics in

252   Seruwila represent the cumulate phase of a highly fractionated volatile-rich magma. The

253   depleted HREE contents indicates that the primitive magma was likely generated from deep

254   level, where the residual phases are dominated by garnet rather than plagioclase or spinel.

255   The enrichment of LREE, high contents of volatiles and the low SiO_2, Cr, Ni, and low Sm/Nd

256   ratios indicate that the Seruwila ultramafics were derived from an enriched mantle source

257   region beneath VC–HC boundary. This boundary extends SW from Seruwila towards

258   Dehiattakandiya, east of the Mahaweli and up to Buttala where several serpentinite and

259   magnetite deposits have been identified. Along its southeast extension, Ni and Cr bearing

260   serpentinites have also been detected. He et al. (2016) carried out a detailed study on the

261   serpentinites and reported abundant magnesite veins and pods suggesting CO_2-induced

262   mantle metasomatism in the mantle wedge. We thus interpret that the Seruwila ultramafics

263   were likely derived from a CO_2 and LREE enriched metasomatized mantle. This

264   interpretation is consistent with experimental data that CO_2-rich vapor is a suitable

265   transporting medium for LREE, iron and P (Harrison 1979).

266   **Age data implications**

267   Zircon grains from the transition zone sample VC16-3 and ultramafic host rock sample

268   VC18-4 are mostly homogeneous without clear core-rim texture (Fig. 7) and they yield

269    $^{206}Pb/^{238}U$ age of 527 ± 3 Ma and 530 ± 4 Ma, respectively (Fig. 8). However, the

270   interpretation of these ages is not straight forward because most ultramafic rocks are highly

271   Zr-unsaturated (< 50 ppm Zr) and Si-poor (Erlank et al. 1978). Thus, zircon seldom

272   crystallizes from ultramafic magma because of less silica activity and low Zr content

273   (Heaman et al. 1990; Finch and Hanchar 2003; Wang et al. 2016 and references there in).

274   In spite of the common Zr-depleted compositions for ultramafic rocks, the occurrence of

275   zircon grains have been reported from several localities including metasomatized peridotites

276   from the Kokchetav Massif, Kazakhstan (e.g., Katayama et al. 2003), serpentinites and

277   rodingites within serpentinized mantle peridotites (Tsujimori et al. 2005) and garnet peridotite

278   (Liati et al. 2004). This suggests that metasomatic fluids can contribute additional Zr to the

279   generally Zr-poor lithospheric mantle, thereby enriching mafic/ultramafic melts derived from

280   such sources. Also, the higher Zr content in the rocks of present study (135–372 ppm)

281   compared to typical gabbroic rocks (Erlank et al. 1978) and the presence of zircon grains as

282   inclusions in euhedral clinopyroxene suggest a magmatic origin. Furthermore, the relatively

283   anhedral zircon crystal with patchy zoning is also typical of zircon grains in ultramafic rocks

284   (Fig. 7) (Grieco et al. 2001; Corfu et al. 2003; Zheng et al. 2006; Lei et al. 2016). Their high

285   Th/U ratios further support this interpretation as Th is usually depleted in hydrothermal fluid.

286   Therefore, these two identical ages and are taken to represent the emplacement timing of the

287   ultramafic intrusion during Early Cambrian. This age is younger than the timing of regional

288   high-grade metamorphism reported in this area (580–540 Ma) (Widanagamage 2011; Kröner

289   et al. 2013; He et al. 2016; Ng et al. 2017).

290   The zircon Lu–Hf data from both samples show εHf(t) values around zero and plot on the

291   CHUR line (Fig. 9). The tight (^{176}Hf/^{177}Hf)$_i$ range of 0.282339 to 0.282482 indicates that the

292   zircon Lu–Hf systematics is unlikely to have been modified by later stage fluid. The two-

293   stage Hf model ages (T_{DM}) varies from 1263 to 1064 Ma, suggesting melt derived from late

294   Mesoproterozoic to early Neoproterozoic depleted mantle with no or limited crustal

295   contamination.

296   In summary, the 530 Ma ages of the ultramafics from the Seruwila area on the VC–HC

297 boundary is younger than the age of high-grade metamorphism associated with the

298 amalgamation of the Wanni and Vijayan Complex during Gondwana assembly. Therefore, the

299 530 Ma is likely to represent a post-collisional ultramafic magmatism during early Cambrian

300 triggered by lithospheric extension.

301 **Implications**

302 Kiruna-type associated with ultramafics

303 Iron oxide–apatite (IOA) deposits occur within a variety of host rocks and tectonic

304 settings. Nelsonites (Fe–Ti oxide apatite rock) and Kiruna-type iron oxide–apatite rocks are

305 the two typical varieties. Nelsonites usually form massive Fe–Ti oxide ore deposits within

306 anorthosites (Owens and Dymek 1992). They have been interpreted by some as the product

307 of liquid (rich in Ti, P, ± Zr) immiscibility from magmas of ferrodioritic composition

308 (Philpotts 1967, 1981; Watson and Green 1981; Ashwal 1993; Darling and Florence 1995),

309 or as cumulate zones within oxide ores (locally mobilized as dike like bodies), although their

310 host Fe–Ti oxide ores may represent immiscible oxide liquids (Owens and Dymek 1992;

311 Dymek and Owens 1996, 2001). Geological, textural, mineralogical, and geochemical data

312 collectively indicate that the magnetite–apatite association in the Seruwila area differs from

313 typical nelsonite in terms of the nature of the host rock, the Ti-poor compositions of

314 magnetite, the highly variable proportion of apatite and magnetite with presence of silicates,

315 and the significant REE fractionation in all the samples (Dymek and Owens 2001).

316 The Kiruna-type IOA rocks contain variable quantities of magnetite–apatite mostly

317 related to intermediate alkaline or calc-alkaline rocks (Hilderband, 1986, Nyström and

318 Henriquez, 1994). Typical examples include the Kiruna deposit in Sweden (e.g. Jonsson et al.

319 2013), the El Laco deposit in Chile (e.g. Dare et al. 2015), and Bafq area in Iran (Sabet-

320 Mobarhan-Talab et al. 2015). Rare examples of Kiruna-type mineralization associated with

321   mafic/ultramafic intrusions have been reported in the Upper Zone of the Bushveld Complex

322   where the IOA deposit is hosted in layered mafic intrusions (Von Gruenewaldt 1994), in

323   Georgia and Maryland of the USA associated with chlorite–talc schists located in ultramafic

324   bodies (Herz and Valentine 1970), in Lizard Ophiolite Complex, UK related to Fe–Ti oxide-

325   rich gabbro (Hopkinson and Roberts 1995) and in Orthys Ophiolite Complex, Greece

326   associated with peridotite (plagioclase lherzolite) (Mitsis and Economou-Eliopoulos 2001).

327   The Kiruna-type IOA deposits are generally attributed to extensional sub-domains such as

328   rifts and/or back-arcs in active convergent (Andean-type) continental margins, and to

329   intracontinental (intracratonic) rifts within a subaerial to shallow marine basinal sequences

330   floored by crystalline basement (Hilderband 1986; Nyström and Henriquez, 1994; Jonsson et

331   al. 2013; Taghipour et all. 2015; Knipping et al. 2015). However, the origin of Kiruna-type

332   rocks remains equivocal. Some researchers favor a magmatic origin (Naslund et al. 1998;

333   Jonsson et al. 2013) and others a hydrothermal origin (Barton and Johnson 1996; Dare et al.

334   2015; Sabet-Mobarhan-Talab et al. 2015). Recent studies propose an integrated model of

335   magmatic–hydrothermal system for the Kiruna-type mineralization (Jonsson et al. 2013;

336   Taghipour et al. 2015; Knipping et al. 2015), providing new insights on the genesis of

337   Kiruna-type IOA deposit, and suggesting a genetic relation with iron-oxide–copper–gold

338   (IOCG) deposits.

339   The iron oxide–apatite deposit at Seruwila shows many similarities with the Kiruna-type

340   iron oxide apatite deposits, particularly with respect to the mineral assemblages, structure of

341   the iron ores, and chemical composition of ore minerals and occurrence of the alteration

342   zones. However, it differs from other IOA deposit hosted in mafic/ultramafic host rocks

343   including the examples mentioned above. In all of these cases, only small apatite crystals are

344   associated with Fe–Ti oxides. The lack of Ti-oxides (ilmenite, rutile) is also a major

345   difference. In the present case, the large magnetite with chlor-fluorapatite crystals hosted in

346 an ultramafic body along a major tectonic boundary, with nearly end-member composition of

347 magnetite and distinct REE-enrichment may suggest a new type of IOA association

348 developed within a post collisional tectonic setting. This is probably the first report of a

349 Kiruna-type IOA deposit associated with ultramafics within a suture system. Our study

350 suggests significant potential for the occurrence of similar deposits elsewhere in the

351 Gondwana suture systems.

352 <u>The genesis of iron mineralization: magmatic or hydrothermal?</u>

353 The close spatial relation, similarity of the trace element signatures, REE distribution patterns

354 and zircon Lu–Hf isotopic data of all studied samples indicate that the Seruwila iron oxide–

355 apatite ore and host ultramafic are genetically related. The fluorine-rich nature of apatite

356 indicates contributions from a volatile-enriched magma. However, depletion in elements

357 considered relatively immobile in hydrothermal fluids (such as Ti, Al, Cr, Zr, Hf and Y)

358 suggests the involvement of hydrothermal processes. We interpret the genesis of Seruwila

359 deposit reflecting both magmatic and hydrothermal processes (also termed as "ortho-

360 magmatic"). The presence of Ca-rich scapolite and actinolite replacement textures indicate a

361 late-stage calcic metasomatism, as also does the occurrence of calc-silicate layers/veins

362 within the ore-bearing rocks. The composition of scapolite is Cl-bearing Meionite (Ca-rich),

363 and previous workers have suggested that scapolites associated with the carbonate

364 metasomatism tend towards the meionitic ($CaCO_3$-rich) end-member (e.g. Pan, 1998; Shaw,

365 1960a). Therefore, we interpret that scapolite formed by calcic metasomatism through

366 interaction with the surrounding calc-silicate rocks.

367 The lithologic and petrological features of the Seruwila IOA deposit point to two

368 different stages of mineralization: 1) the massive magnetite–apatite mineralization; and 2) the

369 disseminated magnetite–apatite–copper sulfide mineralization. The massive magnetite ore

370 samples are characterized by coarse-grained euhedral pure magnetite with ubiquitous spinel

371   exsolution and cumulus textures, whereas the apatite occurs as coarse euhedral crystals free

372   of inclusions, consistent with high T environment. In contrast, the disseminated magnetite ore

373   samples are mostly composed of subhedral to anhedral magnetite with little spinel exsolution

374   but commonly containing silicate mineral aggregates, and the associated apatite is finer-

375   grained with abundant fluid-inclusions indicating that the disseminated mineralization formed

376   after the massive-type magnetite at a lower temperature. The high Al and low Cr contents of

377   spinel, and the low Ti, Al, and Mn of magnetite exclude formation from a pure magmatic

378   melt derived from peridotitic mantle (e.g., Kubo, 2002). Instead, the involvement of high

379   temperature fluids exsolved from the ultramafic magma is suggested. Therefore, we interpret

380   a high-T "magmatic–hydrothermal" origin for the massive type magnetite and a low-T

381   hydrothermal origin for the disseminated magnetite.

382   Knipping et al. (2015) proposed that metals preferred wetting of magnetite in a Cl-

383   bearing magmatic-hydrothermal system, causing buoyant segregation of the magnetite

384   suspension as chloride complexes. Experimental data suggest that Ca, water and volatiles (F,

385   Cl, P) strongly fractionate into the iron-rich melt (Simon et al. 2004; Bell and Simon, 2011).

386   This model is consistent with our data as indicated by the highest Cl content in apatite and

387   magnetite in massive type ore samples and lowest in ultramafic host samples, which could

388   also result from seawater recycling of the subducted slab (Philippot et al. 1998). In terms of

389   the minor Cu-bearing sulfide minerals occurring with disseminated magnetite, it has been

390   commonly observed that the precipitation of sulfide-oxide usually happens near-surface or at

391   shallow level due to the physicochemical changes within the hydrothermal system (Foose et

392   al. 1985; Scott et al. 1990; Eliopoulos et al. 1998).

393   Based on the evidence presented in our study, we propose a two-stage model for the

394   mineralization of Seruwila IOA deposit as follows (Fig. 10). 1) The Cl-rich magmatic–

395   hydrothermal fluid scavenged iron and P from the ultramafic magma, transported iron as

396 magnetite suspensions and ascended due to positive buoyancy. With the sudden decrease of

397 pressure and temperature in the post-collision extensional setting after the collision of VC

398 and HC, magnetite suspensions accumulated to form the massive type magnetite with

399 abundant apatite. 2) Progressive cooling and ascending caused the eventual growth of

400 disseminated magnetite and the precipitation of sulfide mineral at shallower level and trapped

401 abundant fluid-inclusions aided by hydrothermal fluid at shallower depth, forming the

402 disseminated ore body, followed by calcic metasomatism (scapolitization and actinolitization)

403 possibly through interaction with the proximal carbonate rocks.

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Figure captions:

Fig. 1 Generalized geological and tectonic framework of Sri Lanka showing the major crustal blocks, their boundaries, serpentinite localities and the sample localities. (After Cooray, 1994; Rajapaksha et al. 2012; He et al. 2016).

Fig. 2 (a) Detailed geological map of the study area showing the northern part of the boundary between Highland and Vijayan Complexes, together with sample localities. (Modified after Corray, 1994); (b) Geological map of Seruwila copper-magnetite deposit (Modeified after Seneviratne et al. 1998 and Brooks et al. 1985).

Fig. 3 Representative field photographs of the ore and host rocks from Seruwila magnetite deposit. (a) and (b) the contact zone of ultramafic host rock with magnetite; (c) apatite-bearing disseminated magnetite ore sample; (d) massive crystalline magnetite ore sample with abundant clinopyroxene;

Fig, 4 Photomicrographs showing textures of representative samples from Seruwila magnetite deposit. (a) euhedral amphibole, clinopyroxene from ultramafic host rock (VC18-4); (b) zircon grains occur as inclusions in the silicate minerals. (c) massive magnetite ore sample with assemblage of coarse-grained apatite, clinopyroxene and magnetite (sample VC18-3); (d) back scattered electron(BSE) image of spinel exsolution occurring within and along the magnetite grain boundary (VC18-3); (e) secondary actinolite growth surrounding clinopyroxene and apatite (VC18-6); (f) back scattered electron(BSE) image of spinel exsolution occurring along anhedral magnetite grain boundary (sample VC18-6); (g) clinopyroxene, magnetite, amphibole, scapolite, and apatite assemblage from Seruwila deposit transitional boundary (sample VC18-5), euhedral scapolite replaces and cuts the magnetite; (h) scapolite replace magnetite and clinopyroxene, green colored spinel exsolution from magnetite (sample VC16-3);

Fig. 5 Compositional diagrams showing chemistry of representative minerals. (a) Si (pfu)

688   versus X_{Mg} diagram showing compositions of calcic amphibole (after Leake et al. 1997);

689   (b) wollastonite-enstatite-ferrosilite diagram showing compositions of clinopyroxene

690   and orthopyroxene (after Morimoto, 1988); (c) plots of the halogen content in apatite (in

691   molar proportions); (d) plots of Ti vs. Ni/Cr diagram for magnetite (after Dare et al.

692   2015).

693 **Fig. 6** Chondrite-normalized REE distribution diagram (a) and primitive-mantle-normalized

694   multi-element variation diagram (b) for studied samples from Seruwila deposit.

695   Normalizing values for (a) and (b) are from McDonough and Sun (1995) and Sun and

696   McDonough (1989), respectively.

697 **Fig. 7** Cathodoluminescence (CL) images for representative zircons from the Seruwila

698   deposite. Spot numbers, $^{207}Pb/^{235}U$ ages (with 2 sigma error), and εHf values where

699   available, are also shown. The small circle represents location of LA–ICP–MS U–Pb

700   analyses and the larger circles represent Hf isotopic analyses. All the scale bars represent

701   100 μm.

702 **Fig. 8** Tera-Wasserburg U–Pb concordia plots and weighted average plots for ore rock sample

703   VC16-3 (a) and (b), for ultramafic host rock sample VC18-4 (c) and (d). Ellipses with

704   light grey color are excluded from weighted mean age calculation. All data point

705   uncertainties are 2σ. The two different uncertainty values shown represent

706   uncertainties with and without the systematic error propagated respectively.

707 **Fig. 9** Zircon Hf isotopic evolution diagram from Seruwila deposit ore and host rocks.

708   CHUR-chondritic uniform reservoir. The corresponding lines of crustal extraction are

709   calculated by using the $^{176}Lu/^{177}Hf$ ratio of 0.015 for the average continental crust

710   (Griffin et al. 2004).

711 **Fig. 10** (a) Tectonic model of Seruwila ultramafic intrusion where the magma is formed by

712   partial melting of metasomatized volatile-rich mantle in a post-collisional extension

713   environment. (b) Schematic model for the genesis of Seruwila magnetite–apatite

714   deposit: 1) at depth, the iron-rich melt likely separated from the parental ultramafic

715   magma perhaps due to contrasts in density, viscosity, or volatile content; 2) tectonic

716   stress changes (post-collisional extension/faults) cause an efficient ascent of the

717   magnetite suspension and accumulation as massive-type magnetite; 3) progressive

718   cooling aided by hydrothermal fluid caused the eventual growth of disseminated

719   magnetite; 4) calcic metasomatism (scapolitization and actinolitization) developed

720   through interaction with the proximal calc-silicates. VC–Vijayan Complex, HC–

721   Highland Complex

722 **Tables:**

723

724

725

Table 1. *Localities, rock types and mineral assemblage of samples in this study from Seruwila magnetite–apatite deposit.*

Sample No.	Co-ordinates	Localities	Mineral assemblage
VC16/1	N08°20'45.4", E81°17'24.5"	host ultramafic rock	amp, cpx, mag, ap, zrn
VC18/4	N08°20'39.5", E81°18'39.4"	host ultramafic rock	amp, cpx, mag, ap, zrn
VC16/4	N08°20'45.4", E81°17'24.5"	massive magnetite ores	cpx, mag, spl, amp, scp, ap, zrn
VC18/3	N08°20'39.5", E81°18'39.4"	massive magnetite ores	cpx, mag, spl, amp, scp , ap, zrn
VC16/2	N08°20'45.4", E81°17'24.5"	disseminated magnetite ores	cpx, mag, amp, scp, ap, zrn
VC18/1	N08°20'39.5", E81°18'39.4"	disseminated magnetite ores	cpx, mag, amp, scp, ap, zrn
VC18/6	N08°20'39.5", E81°18'39.4"	disseminated magnetite ores	cpx, mag, spl amp, scp, ap, zrn
VC16/3	N08°20'45.4", E81°17'24.5"	transitional zone of host rock and ore body	cpx, mag, spl, amp, scp, ap, zrn
VC16/5	N08°20'45.4", E81°17'24.5"	transitional zone of host rock and ore body	cpx, mag, amp, scp, ap, zrn
VC16/6	N08°20'45.5", E81°17'24.6"	transitional zone of host rock and ore body	cpx, mag, amp, scp, ap, zrn
VC18/2	N08°20'39.5", E81°18'39.4"	transitional zone of host rock and ore body	cpx, mag, amp, scp, ap, zrn
VC18/5	N08°20'39.5", E81°18'39.4"	transitional zone of host rock and ore body	cpx, mag, amp, scp, ap, zrn

726   Mineral abbreviations follow Whitney & Evans, 201

727

Table 2 *LA-ICP-MS zircon Lu–Hf isotope data of for samples in this study.*

Sample spots	Age (Ma)	$^{176}Yb/^{177}Hf$	$^{176}Lu/^{177}Hf$	$^{176}Hf/^{177}Hf$	2s	$^{176}Hf/^{177}Hf_i$	$e_{Hf}(0)$ *	$e_{Hf}(t)$ †	T_{DM} (Ma) ‡	2s T_{DM} (Ma) §	$f_{Lu/Hf}$ ¶
VC16-3-01	541.5	0.00386	0.000135	0.282382	0.000018	0.282381	-13.8	-1.9	1203	1623	-1.00
VC16-3-05	521.0	0.003341	0.000124	0.282368	0.000018	0.282367	-14.3	-2.9	1222	1667	-1.00
VC16-3-06	524.7	0.004586	0.000168	0.282356	0.000018	0.282354	-14.7	-3.2	1239	1692	-0.99
VC16-3-07	516.9	0.004907	0.000179	0.282392	0.000022	0.282390	-13.4	-2.1	1190	1617	-0.99
VC16-3-08	532.3	0.004478	0.000166	0.282339	0.000018	0.282337	-15.3	-3.7	1263	1726	-1.00
VC16-3-09	536.0	0.004616	0.000175	0.282398	0.000020	0.282396	-13.2	-1.5	1182	1591	-0.99
VC18-4-03	517.1	0.002037	0.00008	0.282406	0.000022	0.282405225	-12.9	-1.6	1168	1583	-1.00
VC18-4-04	539.9	0.002387	0.000098	0.282482	0.000016	0.282481	-10.3	1.6	1064	1399	-1.00
VC18-4-06	584.1	0.015184	0.000584	0.282439	0.000014	0.282432603	-11.8	0.9	1138	1479	-0.98
VC18-4-07	531.2	0.003274	0.000135	0.282471	0.000022	0.282469656	-10.6	1.0	1081	1430	-1.00
VC18-4-18	522.8	0.007562	0.000288	0.282447	0.000022	0.282444178	-11.5	-0.1	1118	1492	-0.99

*: $\varepsilon Hf(0)=((^{176}Hf/^{177}Hf)_S/(^{176}Hf/^{177}Hf)_{CHUR,0}-1)\times10,000$;

† $\varepsilon Hf(t)=((^{176}Hf/^{177}Hf)_S-(^{176}Lu/^{177}Hf)_S\times(e^{\lambda t}-1))/((^{176}Hf/^{177}Hf)_{CHUR,0}-(^{176}Lu/^{177}Hf)_{CHUR}\times(e^{\lambda t}-1))-1)\times10,000$;

‡ Single-stage depleted mantle model age: T_{DM} (Ma)$=10000/\lambda\times Ln(1+((^{176}Hf/^{177}Hf)_S-(^{176}Hf/^{177}Hf)_{DM})/((^{176}Lu/^{177}Hf)_S-(^{176}Lu/^{177}Hf)_{DM}))$;

§ Two-stage depleted mantle model age: 2s T_{DM} (Ma)$=T_{DM1}-(T_{DM1}-t)\times((^{176}Lu/^{177}Hf)_{cc}/(^{176}Lu/^{177}Hf)_{CHUR}-(^{176}Lu/^{177}Hf)_S/(^{176}Lu/^{177}Hf)_{CHUR})/((^{176}Lu/^{177}Hf)_{cc}/(^{176}Lu/^{177}Hf)_{CHUR}-(^{176}Lu/^{177}Hf)_{DM}/(^{176}Lu/^{177}Hf)_{CHUR})=1/\lambda\times Ln(1+((^{176}Hf/^{177}Hf)_{S,t}-(^{176}Hf/^{177}Hf)_{DM,t})/((^{176}Lu/^{177}Hf)_S-(^{176}Lu/^{177}Hf)_{DM}))+t$;

¶ $f_{Lu/Hf}=(^{176}Lu/^{177}Hf)_S/(^{176}Lu/^{177}Hf)_{CHUR}-1$; (t=crystallization time of zircon (in Ma),$\lambda=1.865\times10^{-11}$ year^{-1} (Scherer et al., 2001; Söderlund et al. 2004); $(^{176}Lu/^{177}Hf)_{cc}=0.015$, $(^{176}Lu/^{177}Hf)_{DM})=0.0384$, $(^{176}Hf/^{177}Hf)_{DM}=0.28325$ (Griffin et al. 2000); $(^{176}Lu/^{177}Hf)_{CHUR}=0.0332$, $((^{176}Hf/^{177}Hf)_{CHUR,0}=0.282772$ (Blichert-Toft and Albare'de 1997); $(^{176}Lu/^{177}Hf)_S$ and $(^{176}Hf/^{177}Hf)_S$ are measured values of samples).
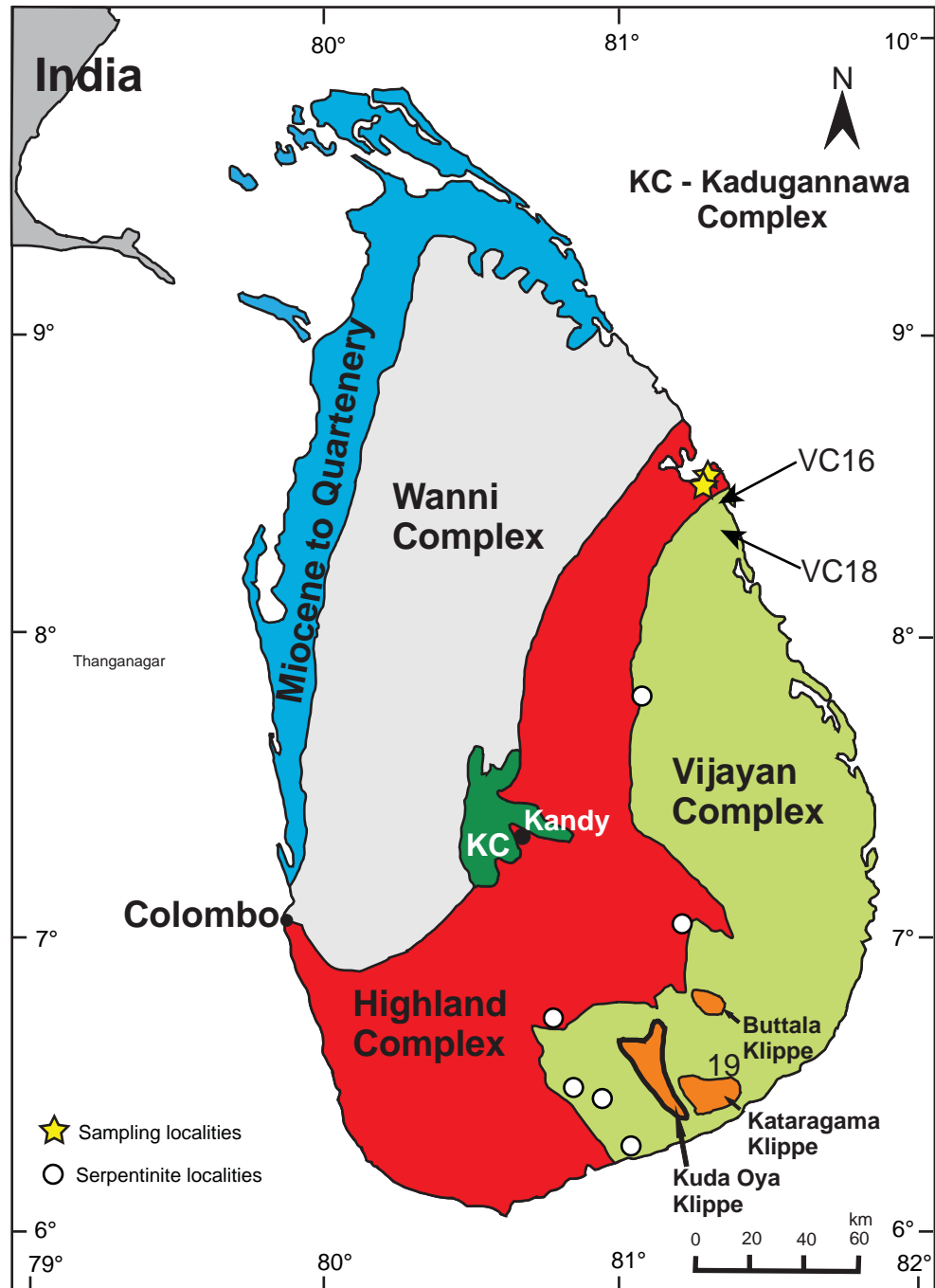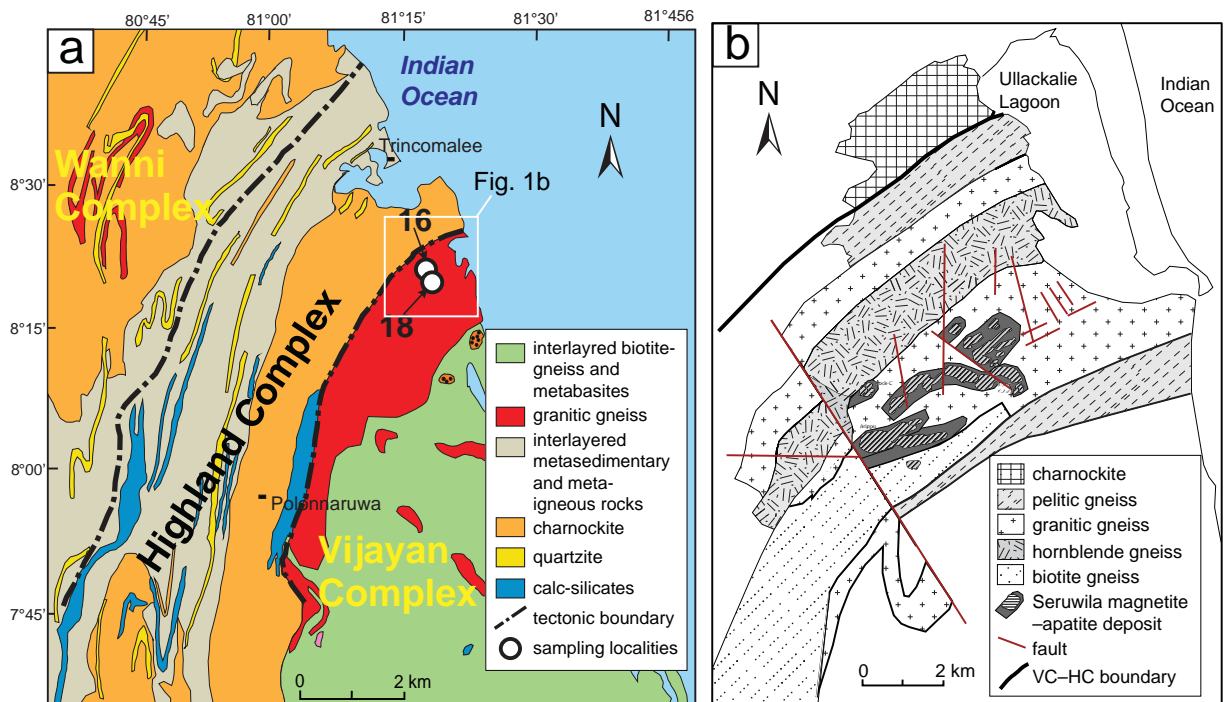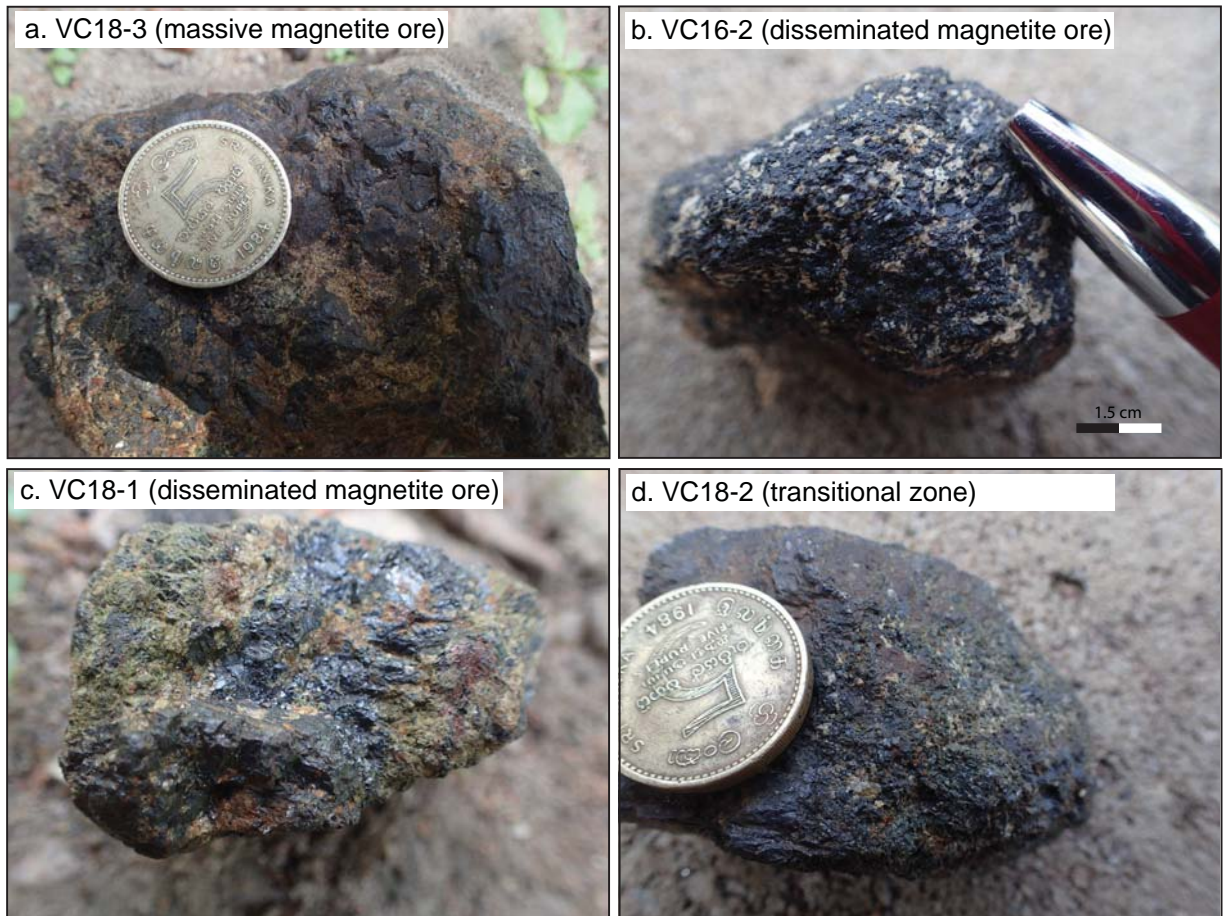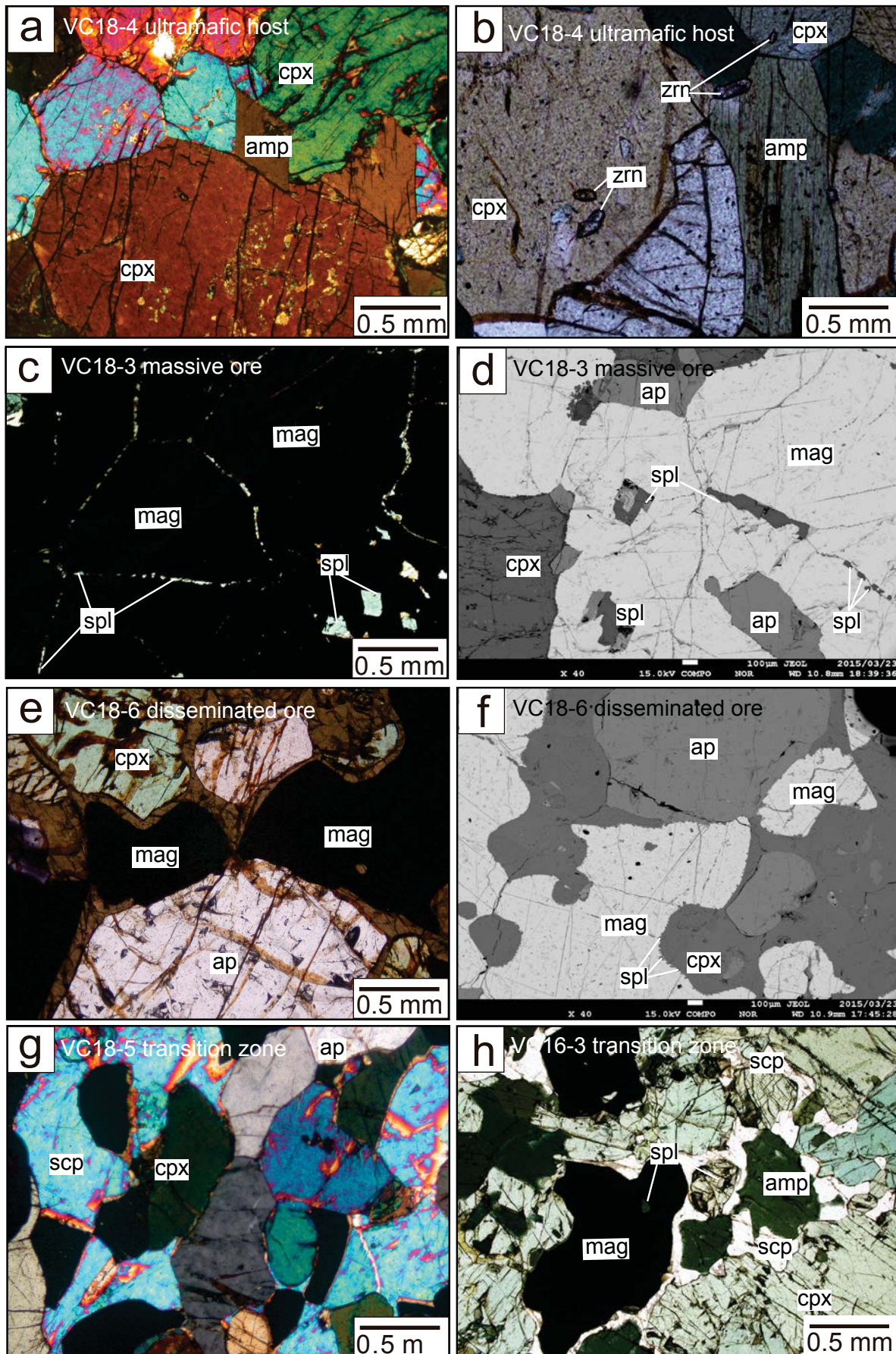
728

Fig. 1

Fig. 2

Fig. 3

a. VC18-3 (massive magnetite ore)

b. VC16-2 (disseminated magnetite ore)
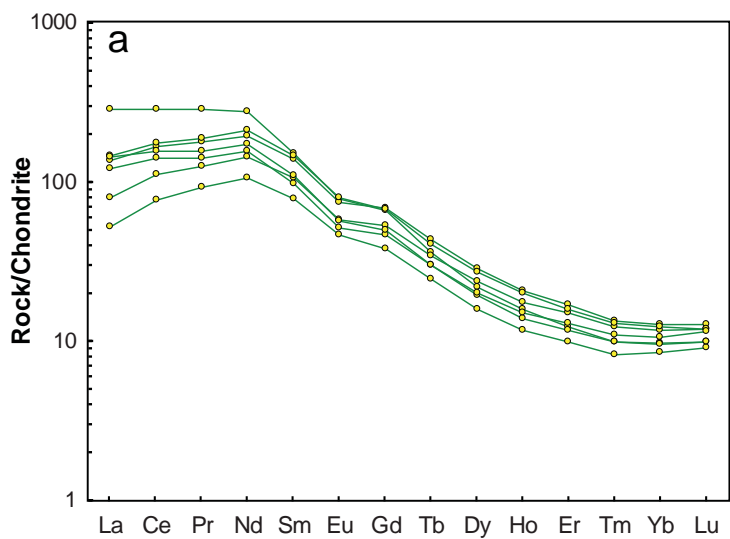
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c. VC18-1 (disseminated magnetite ore)

d. VC18-2 (transitional zone)

Fig. 4
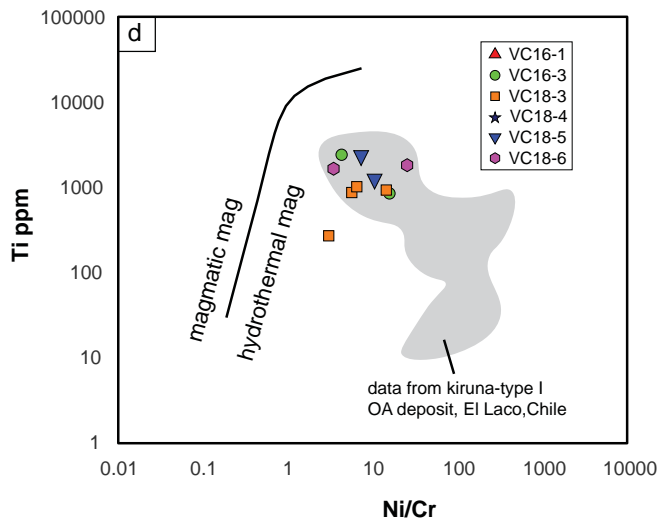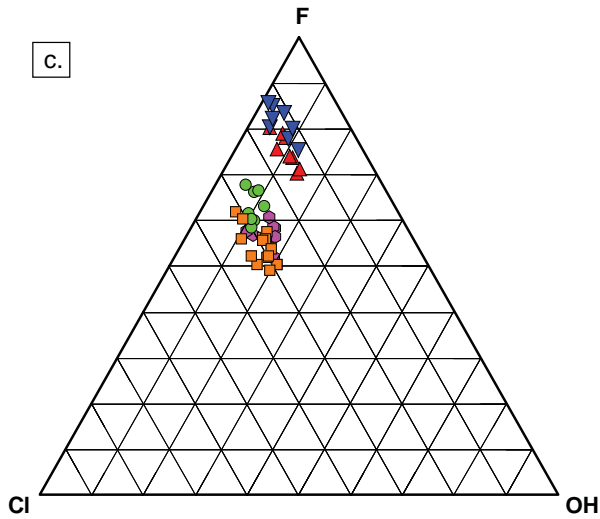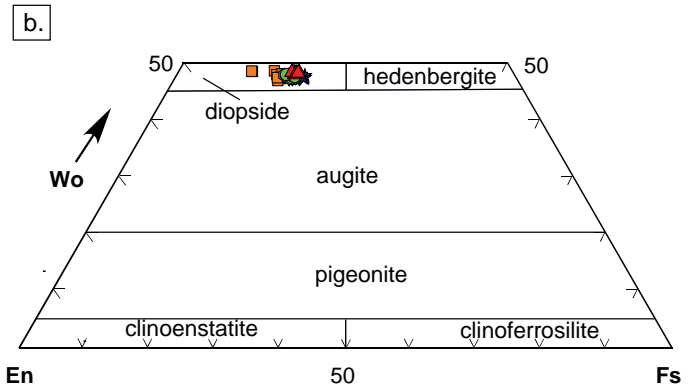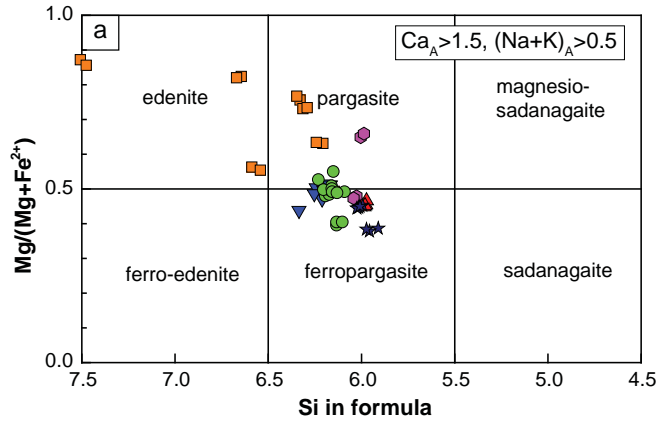
Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10

(a)

HC

oceanic crust

Seruwila Kiruna-type deposit
& ultramafic intrusion

VC

granitic gneiss

continental crust

oceanic lithospheric mantle

metasomatized SCLM partial melting

manle metasomatism

volatile rich fluids (CO_2, Cl-rich)

**sub-continental
lithospheric mantle**

(b)

calcic metsomatism

massive type
magnetite segregation
along the fault

disseminated
type
magnetite

accumulation of
primary magnetite
as well as
scavenging of
Fe into the
high-salinity fluid

ascending of iron-rich
fluid as chloride
complexes (suspension)

seperation of
iron-rich fluid and
silicate melts (P, F, Cl-rich)

ultramafic magma

⊙ magnetite suspension   ⬤ calcic-metasomatism
🜨 massive magnetite ore   ▱ calc-silicates
🜨 disseminated magnetite ore ↑ buoyancy force