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4   **Geochemical characteristics of mineral inclusions in the Luobusa**

5   **chromitite (Southern Tibet): Implications for an intricate geological setting**

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ABSTRACT

The Luobusa chromitite and ophiolite present a captivating geological feature marked by peculiar mineralogical and geochemical characteristics. Abundant platinum-group minerals (PGM), base-metal sulfides (BMS), and PGE-sulfides and alloys in the chromitite unveil a multi-stage genesis, encompassing partial mantle melting, melt-rock interactions, and dynamic shifts in oxygen and sulfur fugacity (fO_2, fS_2). This study explores the geochemical signatures and PGE patterns of these mineral inclusions to elucidate the evolutionary process of the Luobusa ophiolite, tracing its transition from a sub-ridge environment to a sub-arc setting. The variable ΣPGE values (40 - 334 ppb) in chromitite, coupled with notably lower ΣPGE values (10 - 63 ppb) in dunite imply extensive melt fractionation and melt-rock interactions. Coexisting well-crystalline Os-Ir alloys alongside interstitial BMS likely reflect low fS_2 and high temperatures during the early formational stages, whereas abundant anhedral sulfarsenide and pyrite inclusions in chromite point to lower temperatures and higher fS_2 during the late stages. The trace element composition of pyrite inclusions resonates with the characteristics of mid-ocean ridge (MOR) and oceanic island rocks, manifesting interplay of diverse magmatic sources during the evolution of the Luobusa ophiolite.

Keywords: *Luobusa ophiolite, SW Tibet, chromitite, PGM and BMS inclusions, genetic model.*

INTRODUCTION

Podiform chromitites commonly occur as lenticular or ellipsoidal bodies associated with ophiolitic nappes (Arai 1997 and references therein). Despite ongoing scientific research and data accumulation, the origin of these chromitites has been the subject of continued debate (e.g., He et al. 2021; Yang et al. 2021; Yao et al. 2023). Early studies proposed that podiform chromitites result from

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differential gravitational separation of mantle-derived magmas (Dickey 1975; Mondal and Mathez 2007; Maier et al. 2013). Other hypotheses suggest reactions between basaltic melts and mantle peridotites (e.g., Zhou et al. 1994; Arai and Miura 2016) or crystallization of chromite from immiscible melts (Matveev and Ballhaus 2002; Su et al. 2020, 2021a, b). High- Al chromitites are believed to form in mid-ocean ridge environments (MOR) with MORB melt, while high-Cr chromitites are typically associated with supra-subduction zone (SSZ) processes by melt reaction with boninitic melt (e.g., Zhou et al. 2005; Arai and Miura 2016). The coexistence of high-Al and high-Cr ores may reflect the long-term evolution of the associated ophiolitic complexes (Xiong et al. 2017, 2018; Ullah et al. 2020, 2022; Zhang et al. 2023).

Integration of the geochemical properties of ophiolitic chromitites with detailed mineralogical analyses of platinum group element (PGE) alloys and base metal sulfide (BMS) inclusions can provide valuable information for reconstructing the ore-forming processes (Melcher 2000; Uçurum et al. 2006; Rollinson 2008; Uysal et al. 2009). Chromitites commonly exhibit significantly higher PGE contents compared to the peridotites hosting them, with micrometer-sized PGM inclusions, such as sulfides of the laurite-erlichmanite series, alloys in the Os-Ir-Ru system, and sulfarsenides of Ir, Ru, and Os, contributing to the PGE budget (Tsoupas and Economou-Eliopoulos 2021).

Ultrahigh-pressure (UHP) mineral inclusions in mantle peridotites and chromitites from several orogenic belts suggest a multistage genesis of ophiolite host rocks under deep to shallow mantle conditions (Yang et al. 2014; Xiong et al. 2015, 2022a, b). However, the origin of podiform chromitites in subduction-related environments is challenging to reconcile with the presence of UHP and super-reduced (SuR) mineral phases in many chromitites (Robinson et al. 2004, 2015; Yang et al. 2007; Xu et al. 2009). This suggests recycling of chromitites and their host rocks in upper mantle regions (Robinson et al. 2004; Yamamoto et al. 2009).

The Luobusa ophiolite in the Indus—Yarlung Zangbo suture of southern Tibet was interpreted as a forearc fragment of Neo-Tethyan oceanic lithosphere (Zhou et al. 1996, 2005; Bai et al. 2000; Shi et al. 2007; Yang et al., 2007). It contains the largest chromite deposit in China, with reserves over several million metric tons

93   of ore, with Cr_2O_3 as high as 50 wt% and $Cr_2O_3/FeO > 4$ (Li et al. 1995; He et al.

94   2021). According to Zhou et al. (1996), the Luobusa ophiolite is characterized by

95   a distinctive mantle sequence comprising harzburgite and diopsidic harzburgite

96   with abundant dunite lenses and podiform chromitites. These harzburgites

97   represent the residual rocks formed after tholeiitic magma extraction and contain

98   accessory chromite grains, which exhibit variations in Cr and Al contents and low

99   Ti levels, aligning them with mid-ocean ridge basalt (MORB)-type peridotites

100   (Zhou et al. 2005). On the other hand, podiform chromitites in Luobusa likely

101   originated from the crystallization of boninitic melts during a late stage of melting

102   (Zhou et al. 1996). Similarly, several authors (e.g., Yang et al. 2007; Zhou et al. 2005;

103   Wang et al., 2010; Xu et al. 2015; Zhang et al. 2017, 2019, 2020; Xiong et al., 2022)

104   have suggested that the Luobusa ophiolite formed at a mid-ocean ridge and,

105   subsequently was partially melted to generate boninitic magmas in a SSZ setting.

106   In the Luobusa podiform chromite, researchers have discovered an array of

107   UHP minerals, including microdiamonds and coesite, as distinct inclusions (e.g.,

108   Bai et al. 2000, 2004; Yang et al. 2007; Xiong et al. 2015). This suggests that the

109   formation of chromite likely commenced at significant depths, possibly within the

110   transition zone or lower mantle (Xiong et al. 2015). The formation process of

111   Luobusa podiform chromite appears to involve both deep-seated and shallow

112   processes, potentially spanning multiple stages of evolution, including events in

113   the transition zone, mid-ocean ridges, subduction zones, and even late-stage

114   surface modifications, as discussed in Xu et al. (2015) and Xiong et al. (2015). This

115   makes the Luobusa ophiolite a case study for gaining insights into the origin and

116   evolution of significant podiform chromite deposits in forearc ophiolites.

117   In this contribution, we investigate the mineral chemistry of Luobusa

118   chromitite and its host peridotites, offering a comprehensive analysis of

119   petrographic and geochemical attributes concerning PGM and BMS inclusions. We

120   examine the complex formation process involving deep-seated and shallow

121   processes across various stages of geological evolution of significant podiform

122   chromite deposits within forearc ophiolites, positioning the Luobusa ophiolite as

123   a compelling case study in understanding the intricate dynamics shaping such

124   geological phenomena.

4

GEOLOGICAL SETTING

The Yarlung Zangbo suture zone in southern Tibet extends over 2000 km, separating the Indian subcontinent from the Eurasian subcontinent. Among several ophiolitic massifs, the Luobusa nappe occurs along the Yarlung Zangbo suture zone and is located approximately 200 km southeast of Lhasa. It extends for about 40 km along strike and is separated from Triassic flysch by a reverse fault. The Luobusa ophiolite tectonically overlies the Tertiary Luobusa Formation and Gangdese arc granite to the north (Fig. 1) (Yang et al. 2007). This ophiolitic nappe consists of peridotite and subordinate mafic cumulates. At the base of the nappe, a thin zone contains tectonically admixed volcanic rocks and cherts (Yang et al. 2007). The peridotites include harzburgite, refertilized harzburgites, and dunite. The mafic cumulate rocks include gabbro, wehrlite, and pyroxenite (Wang et al. 1987) (Fig. 1). A dunite zone about 150 m thick underlies the ophiolitic nappe and is cut by N-S normal faults.

In the Luobusa ophiolite nappe, chromitite ore bodies are commonly found in association with harzburgite and are typically enclosed by dunitic rims, as observed in previous studies (Zhou et al. 1996). These chromitite deposits occur as lenticular, podiform, or irregular masses, typically measuring less than 100 m in length. They are located in the upper part of the mantle section or are associated with the transitional dunite (Xiong et al. 2015). Various modes of occurrence were observed, including massive, nodular, and disseminated chromitite (Fig. 2). The chromitite ores in Luobusa are typically 0.5 to 3 m long, although some can reach up to 20 m. The ore bodies primarily consist of magnesiochromite, accounting for up to 95% of the modal composition (Xiong et al. 2015). The chromitite ores are surrounded by dunite, which predominantly comprises granular olivine and amoeboid magnesiochromite (Fig. 2).

The geochronological data available for the ophiolitic rocks in the Luobusa area indicate a diachronous evolutionary history. It is suggested that around 177 ± 33 Ma the prevailing environment was a mid-ocean ridge setting (Zhou et al. 2002). A geodynamic shift occurred at ~ 126 Ma, and the ophiolites were imprinted by a supra-subduction zone environment (Malpas et al. 2003). This transition imitates the tectonic processes associated with the subduction of

157 oceanic crust beneath an overriding plate. During the late stages of ophiolite

158 evolution, the MORB-like mantle peridotites in Luobusa were likely fertilized by

159 melts derived from supra-subduction zone settings (e.g., Xu et al. 2011, 2023; Li et

160 al. 2012). These incompatible element-rich melts infiltrated the mantle peridotites

161 and contributed to the formation of chromitite ore bodies (Zhou et. 2014).

162

163 **SAMPLES AND METHODS**

164 *Sampling*

165 Sampling in the Luobusa region presents a unique challenge due to the

166 widespread distribution of chromite ore bodies. To date, ~ 300 chromite ore

167 bodies of varying sizes and morphologies have been discovered in this area, with

168 chromite deposits occurring across different lithological zones, as detailed in

169 Xiong et al. (2015). Our primary focus centers on specific chromitite ore bodies in

170 Luobusa, particularly the Cr-11 and Cr-31 ore bodies. These ore bodies exhibit a

171 rich variety of ore structure types, including disseminated chromitite, banded

172 chromitite, nodular, anti-nodule, and massive chromitite, among others. The

173 massive chromite displays pristine composition and textural features, which

174 qualify it to gain insights into its genesis and evolution (Xiong et al. 2015).

175

176 *Mineral chemistry*

177 Following petrographic investigation of 60 polished thin sections, the

178 mineralogical composition and textural characteristics of the studied chromitites

179 and associated peridotites were analyzed with a JEOL JXA-8100 electron

180 microprobe at the State Key Laboratory Breeding Base of Nuclear Resources and

181 Environment, East China Institute of Technology. The measuring conditions

182 included an accelerating voltage of 15 kV, beam current of 20 nA, and beam

183 diameter of 1 μm. The detection limits for elements in the chromitite grains and

184 PGM were ~0.01 wt %. Synthetic and natural metal alloys and minerals were used

185 as standards. A pure cobalt reference standard was analyzed every five analyses

186 in order to check for any drift in the analytical conditions. The BMS minerals and

6

187   PGM found were analyzed using the $L\alpha$ lines for Ru, Ir, Rh, Pd, Pt and As, the $M\alpha$

188   line for Os and the $K\alpha$ lines for S, Ni, Fe and Cu. A comprehensive set of standards

189   including pure metals for PGE, Ni and Cu, arsenopyrite for As, and pyrite for Fe

190   and S were used to calibrate the EPMA.

191   Because PGM typically occur as a few μm-large inclusions in chromite grains

192   in the peridotite samples (Capobianco and Drake 1990; Gonzalez-Jimenez et al.

193   20012, 2014), we used the backscattered electron images to distinguish the PGE

194   minerals, and then to determine their chemical composition (size of 10-100μm)

195   by EMPA. Fifteen representative samples of massive chromitite were selected to

196   study the PGM inclusions. We note that several PGM grains were either too small

197   (≤ 2 μm) or inhomogeneous for quantitative determination.

198

199   *Trace element composition of pyrite inclusions*

200   The chemical compositions of pyrite inclusions were determined by the laser

201   ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) at the

202   GEOMAR Helmholtz Centre for Ocean Research Kiel, Germany. This instrument is

203   a 193 nm Excimer laser ablation system (Coherent, GeoLasPro) that is coupled to

204   a double-focusing magnetic sector mass spectrometer (Nu Instruments, AttoM)

205   under hot plasma conditions (NAI = 24 - 35; ThO/Th = 0.016 - 0.022 %; Fietzke

206   and Frische 2016). Spot analyses were performed as 30 s ablation under Helium

207   carrier gas at a laser repetition rate of 5 Hz, a spot diameter of 44 μm, and a fluence

208   of 2 and 5 J cm^{-2} for sulfides and silicate glasses, respectively. The USGS glass

209   standard BHVO-2G (Wolf and Wilson 2007), the USGS sulfide standard MASS-1

210   (Wilson et al. 2002), and the synthetic sulfide standard PGE-Ni7b (Wohlgemuth-

211   Ueberwasser et al. 2007) were used as reference materials. The NISTSRM610

212   glass (Wise and Watters 2012) was used for the sake of mass calibration, but due

213   to the lack of suitable reference values for S, Ru, Os, Ir and Hg, the MASS-1 was

214   used to calibrate S, Ir and Hg and PGE_Ni7b to calibrate Ru and Os. 60 - 80 s of gas

215   background data were collected prior to each ablation. The data evaluation was

216   performed by applying the linear regression slope method proposed by Fietzke et

217   al. (2008). Data are corrected for the following interferences: $^{65}CuH^+$ on $^{66}Zn^+$;

218   $^{63}Cu^{40}Ar^+$ on $^{103}Rh^+$; $^{65}Cu^{40}Ar^+$ on $^{105}Pd^+$ and $^{115}Sn^+$ on $^{115}In^+$. The exact argide

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219   and hydride formation rates were determined by ablating elemental Cu (line-scan,

220   60 - 120 s, 32 μm spot size, 5 J cm^{-2}). Fe contents in pyrite grains as determined by

221   the EPMA were used for internal standardization of the LA-ICP-MS data.

222

223

224   *Bulk-rock PGE geochemistry*

225   The PGE concentrations in the peridotite and chromitite samples were

226   determined at the National Research Center of Geoanalysis, Chinese Academy of

227   Sciences (CAS) using the NiS fusion pre-concentration technique followed by ICP-

228   MS measurement. The CAS uses a modified NiS-fire assay method (Sun and Sun

229   2005; Zhou et al. 2005) in which the samples are mixed with sodium carbonate,

230   sodium borate, borax, glass powder, nickel powder, iron powder, and sulfur. The

231   mixtures are transferred into fire-clay crucibles, with an appropriate amount of

232   ^{190}Os spike solution and covered with a thin layer of $Na_2B_4O_7$. After fusing at

233   1150°C for ~1 h and drying under an infrared lamp, the crucibles are cooled and

234   the Ni-Fe buttons are removed. The buttons are then placed in a glass beaker

235   containing 60 mL H_2O for a minimum of 10 h. To better dissolve the button, about

236   30 mL HCl is added and the beaker is heated at ~110 °C for ~30 min, and left to

237   cool 10 h until the solution becomes clear and the residue is seen. The solution is

238   then filtered using a millipore filter membrane (diameter 25 mm, 0.45 mm), and

239   then the precipitate is collected and washed five times with ~15 mL water. The

240   residue, together with the membrane, is transferred to a 7 mL Teflon beaker,

241   sealed together with 1 mL HCl + 1 mL H_2O_2, and heated at ~110 °C for 1.5 h. Upon

242   cooling to room temperature, the beaker was opened and the solution was

243   transferred and diluted with H_2O to a total volume of 10 mL in preparation for ICP-

244   MS analysis (Sun et al. 1998a, b; Sun and Sun 2005). The detection limit was 0.2

245   ng/g for Pt and Pd, 0.001 ng/g for Ir, Rh, and Os, and 0.1 ng/g for Ru. The precision

246   of the analyses was estimated to be better than 5% for Rh, Pd, and Ir, and 10% for

247   the other elements.

248

249   **RESULTS**

250   *Mineralogical characteristics of PGM and BMS inclusions*

251   Numerous BMS sulfide inclusions were observed by backscattered electron

252   images in the examined samples of the Luobusa massive chromitites. Abundant

253   pyrite, arsenopyrite, sphalerite, galena, tennantite-(Zn), pentlandite, tetrahedrite-

254   (Zn), heazlewoodite, antimonite, spionkopite, millerite, and chalcocite inclusions

255   were observed in the Luobusa chromitites (Fig. 3; Figs. S1-S5). Most pyrite

256   inclusions (Fig. 3) have stoichiometric compositions and Fe/S ratios between 0.52

257   and 0.54 (Table S1). Minor amounts of other elements such as Cd, Cu, Pb were

258   above the detection limits in some inclusions. Arsenopyrite (Table S1) shows

259   variations in the atomic (Fe+Mo)/(S+As) ratios from 0.52 to 0.56 and

260   stoichiometric composition. Some analyses also contain minor amounts of Bi and

261   Ag. Sphalerite shows an atomic (Zn+Fe)/S variation of 0.97 to 1.09 with a

262   stoichiometric composition of $Zn_{0.9}S$. Cadmium, Cu, and Ge are present in trace

263   quantities. Galena, tennantite-(Zn), and tetrahedrite-(Zn) occur as inclusions in

264   the pyrite grains. Galena has an atomic (Pb+Fe)/S ratio of 1.07 to 1.11 with a

265   composition of $Pb_{1.09}S$. Tennantite-(Zn) shows variations in the atomic

266   (Fe+Ni+Zn)/S ratios from 0.71 to 1.19 and a composition formulated as

267    $Cu_{10.6}Zn_{1.8}Fe_{0.6}As_{3.4}S_{13}$. Tetrahedrite-(Zn) shows an atomic (Fe+Ni+Zn)/S variation

268   of 1.13 to 1.24 and a composition of $Cu_{10}Zn_{1.9}Fe_{0.5}Sb_{3}S_{13}$. Heazlewoodite inclusions

269   ($Ni_{1.4}Fe_{1.2}S_{2}$) show an atomic (Fe+Ni+Zn)/S of 1.32 (Table S1). Antimonite shows

270   an atomic (Sb+Cu)/S variation of 0.66 to 0.67 and has a stoichiometric

271   composition of $Sb_{2}Cu_{0.1}S_{3}$. Millerite shows an atomic (Ni+Fe)/S variation between

272   1.09 and 1.12. In addition, micron-sized augite, microcline and phlogopite were

273   observed in the large pyrite inclusions (Fig. 4; Table S2).

274   Abundant particles of PGM and alloys were observed in the examined thin

275   sections. PGM inclusions in the Luobusa chromitites include RuS_{2} (laurite), (Ru, Os,

276   Ir)S_{2}, cuproiridsite, and ruthenarsenite (Ru, Os, Ir)S_{2}. These inclusions are

9

277   commonly < 100 μm in size and locally intergrown with amphibole and phlogopite.

278   Most of the PGM inclusions (< 10 μm-across) are euhedral crystals, while the BMS

279   inclusions are subhedral to anhedral grains. Some of monomineralic and

280   composite grains are PGM and are associated with silicate minerals (Fig. S1, Table

281   S3). The (Ru, Os, Ir) S_2 series minerals exhibit a wide range of these three elements

282   (Table S3, Fig. 5), for example 5.97 to 41.10 wt% Ru, 5.44 to 63.49 wt% Os, and up

283   to 23.36 wt% Ir. RuS_2 was also observed as euhedral inclusions ($Ru_{0.27}S_2$ to $Ru_{0.96}S_2$)

284   in Luobusa chromitite (Fig. 5, Table S3). The associated Ru-Os-Ir phases may

285   indicate that some RuS_2 inclusions (5-80 μm) are of late metasomatic origin. The

286   RuS_2 inclusions have variable S contents ranging from 37.5 to 42.6 wt% and Ru

287   contents ranging from 29.9 to 60.39 wt%, as well as a small amount of Ir and As.

288   In addition to the PGE alloys, a large number of PGM were observed in the Luobusa

289   chromitites, including Pt-Fe-Ir, Pt-Fe-Rh, Pt-Pd-Fe, Os-Ru-Ir, Pt-Pd-Fe-Ir, Pt-Pd-Fe-

290   Cu, and Os-Ir (Figs. 5, 6, Figs. S1, S5; Table S4). The Os-Ir alloy occurs commonly as

291   small particles (1-3 μm) and is considered the most abundant alloy in the studied

292   samples. Other inclusions with Ir- and Cu-sulfide compositions exhibit

293   idiomorphic habits (Fig. 5, Figs. S6, S7).

294   The EPMA data indicate that the compositions of Os-Ir-Ru alloys vary widely,

295   with or without traces of Fe and Ni. The Os content ranges from 14.68 to 48.46

296   wt%, Ir from 20.94 to 76.11 wt%, and Ru from 1.02 to 56.72 wt%, indicating an

297   osmium-rich iridium sulfide. In contrast, Fe-Ni alloys most commonly occur in

298   fractures or as interstitial fillings (Fig. 5) between chromite grains. Their

299   compositions are uniform (Table S5) and are formulated as $Ni_{2.4}Fe_{0.8}$, typical of

300   awaruite (ideally Ni_3Fe).

301

302   *Trace element abundances in pyrite inclusions*

303   LA-ICP-MS analysis of pyrite inclusions shows a proportional covariance between

304   Co-Ni, Cu-Zn, and Cu-Pb. Ag contents are also positively correlated with Au, Te, Sb,

305 and Cu contents. The Cu and As contents in the analyzed pyrite range from < 1 to

306 6038 ppm and from 1 to 16964 ppm, respectively. The average values are 258 ppm

307 (Cu) and 3113 ppm (As). The Pb content in the pyrite ranges from 0.1 to 11846

308 ppm, with an average value of 236 ppm. Gold values in the analyzed pyrite

309 inclusions are remarkably high, ranging from 13.4 to 150 ppm, with an average

310 value of 119 ppm. In general, the Pb and Zn contents in the pyrite were positively

311 correlated, as are Au and Ag (Fig. 7). The As content ranges from 0 to 13644 ppm

312 (Table S6).

313

314 *Bulk rock PGE concentrations*

315 Whole-rock PGE concentrations in 50 samples of Luobusa podiform chromitites

316 and peridotites show significantly high total PGE contents in chromitites (40.2-

317 333.6 ppb), with an average value of 146.3 ppb (Table S7). Total PGE contents in

318 peridotites range from 0.3 to 63.4 ppb, with an average value of 33.7 ppb (Table

319 S7). The studied chromitites feature high $(Ru/Pt)_N$ ratios (8.1 to 90.1). The

320 chromitites have extremely low $(Pd/Ir)_N$ values (0.01-0.30) compared to the

321 chondrite value of 1.01 (McDonough and Sun 1995; Fig. 9).

322

323

324 **DISCUSSION**

325 *Sulfide inclusions*

326 The presence of unusual mineral phases (e.g., arsenotučekite ($Ni_{18}Sb_3AsS_{16}$),

327 eliopoulosite (V_7S_8), tsikourasite ($Mo_3Ni_2P_{1+x}$) and grammatikopoulosite (NiVP))

328 in podiform chromitites may reflect their formation under specific conditions

329 related to variations in oxygen fugacity (fO_2) and sulfur fugacity (fS_2) (e.g., Bindi et

330 al. 2020a, b). In the Luobusa peridotites and chromitites, intergrowths of sulfide

331 and platinum group mineral phases may also be related to variations in fO_2 and fS_2

11

332   (e.g., Klein and Bach 2009; Zhu and Zhu 2019).

333   The trace element geochemistry of pyrite in Luobusa chromitite highlights a

334   possible contrast between crustal contamination and mantle source influences.

335   While some areas of individual pyrite inclusions exhibit enrichments in Ni, Co, Ag

336   and Au, other portions reveal elevated levels of As, Cu, Zn, Pb, Cd, Sn, and W,

337   suggesting contamination from crustal materials. The presence of silicate

338   inclusions, such as pyroxene, and potentially other sulfide phases within the pyrite

339   inclusions, likely contributed to the observed heterogeneity in their trace element

340   composition. Figure 7 shows a notably positive correlation between Pb and Zn, as

341   well as Au and Ag, suggesting hydrothermal or metasomatic process may have

342   been active during pyrite formation (Zhu and Zhu 2019).

343

344   *PGM inclusions*

345   The mineralogy and textures of PGM are commonly utilized to understand the

346   origin and evolution of chromitites, as well as to determine parameters such as

347   temperature, oxygen fugacity (fO_2), and sulfur fugacity (fS_2) of the parent melt

348   (Garuti et al. 1999; González-Jiménez et al. 2012, 2014; Xiong et al. 2021). In the

349   Luobusa chromitite, PGM inclusions intergrown with amphibole and phlogopite

350   likely represent primary composite inclusions derived from a hydrous melt (see

351   Fig. 3 and Fig. S1). The observed chemical variations, irregular morphologies, and

352   alteration rims may be attributed to post-crystallization alteration or

353   metamorphism processes (Stockman and Hlava 1984; Proenza et al. 2008).

354   Within the Luobusa chromitites, the PGM are disseminated and occur as

355   single-phase (Fig. 5a-c), two-phase, or polyphase hypidiomorphic or xenomorphic

356   inclusions (Fig. 5d-i). In some examples, symplectite textures with microscopic

357   multiphase (Fe-Ni, Ir-Fe) pseudo-structures in Os-Ir, Os-Ir-As, Lrt, Os-Al, and Ir-Al

358   matrices (Fig. 5d, e, f) may suggest decompression under high-pressure/high-

359   temperature conditions during the evolution of the Luobusa chromitites through

360   recycling in deeper mantle levels (e.g., White and Powell 2011; Groppo et al. 2015).

The identified PGM exhibit compositions rich in Ir-Os-Ru or Pt-Rh-Os (Fig. 6a, b), with their diversity in PGE mineralogy closely aligning with the immiscibility gap, which is believed to narrow at elevated temperatures. This diversity in PGE mineralogy, coupled with the chemical composition variations observed in chromite grains within the Luobusa chromitites, indicates potential differences in the parent magma(s). Such distinctions may arise from varying degrees of partial melting and melt-rock interactions (e.g., Economou-Eliopoulos et al., 2017). Consequently, these features likely reflect fluctuations in temperature, oxidation state, and/or sulfur fugacity (fS_2) (e.g., Garuti et al., 1999; Ahmed and Arai, 2002).

The formation and stability of platinum group minerals in chromitites are predominantly controlled by the sulfur fugacity and temperature of the melt (Augé 1988). Luguet and Reisberg (2016) proposed that Ir-Pt-(Os) alloys in mantle peridotite associated with BMS likely originated from PGE-bearing monosufide solid solution (MSS) that underwent desulfurization during high-temperature partial melting. Ir-Pt-(Os) release from BMS could also result from mantle redox changes or interactions with S-undersaturated melt/fluids. On the other hand, experiments and observations in the Fe-Cu-S-(Pt+As) system and in PGE ores in the Bushveld Complex suggest that Pt-rich nanoparticles may form before MSS crystallization (Helmy et al. 2013; Wirth et al. 2013; Junge et al. 2015). Finnigan et al. (2008) provided evidence for the direct crystallization of Ir-Pt nanocrystals, approximately 700 nm in diameter, from basaltic melt. The experimental work was conducted under the relevant conditions for chromite deposit formation in the upper mantle, specifically at approximately 0.5 GPa, temperatures of around 1300–1400°C, and with fO_2 at fayalite-magnetite-quartz (FMQ) buffer (Stockman and Hlava 1984; Johnson et al. 1992; Zhu and Zhu 2019).

Some PGM can be incorporated into chromitites through reactions between the melt and peridotite. Multiple episodes of melt extraction can lead to a gradual decrease in melt fS_2, resulting in the decomposition of PGE sulfides and the formation of Ru-osmite and Os-Ir alloys while chromite crystallizes. This mechanism can account for the variable mineralogical characteristics of PGM and the wide range of Re-depletion model ages (TRD) (up to ~ 234 Ma) and magmatic events observed in chromitites, which are distinct from the age of the Luobusa ophiolite (177 ± 33 Ma; Zhou et al. 2002; Shi et al. 2007; McGowan et al. 2015).

13

394   The Os-Ir alloys tend to form at earlier paragenetic stages and under higher

395   temperature and pressures compared to sulfides and sulfarsenides (e.g., laurite-

396   erlichmanite series) (Augé 1988; Andrews and Brenan 2002a,b; Gonzalez-Jimenez

397   et al., 2012, 2014; Uysal et al. 2015). This implies that the fS_2 was initially low and

398   increased significantly at lower temperatures, allowing the crystallization of

399   laurite and erlichmanite. Experimental research has demonstrated that

400   erlichmanite can coexist in equilibrium with Os-Ir alloys at fS_2 values ranging from

401   -1 to +2 (Barnes et al. 2001). Based on the PGE mineralogy group include laurite,

402   Os-Ir-Ru alloys, erlichmanite, and PGE-BM sulfides and theoretical meta-sulfide

403   equilibrium curves for Ru, Ir, Os, and Ni as a function of fS_2 and temperature (see

404   Fig. S8), it is suggested that the Luobusa chromitites and their associated PGM

405   formed under low fS_2 conditions, within a temperature range of 1300°C to 1050°C,

406   indicating upper mantle origins (Zhu and Zhu 2019 and references therein).

407

PGE geochemistry

409   The geochemical characteristics of PGE are frequently employed to elucidate

410   various petrological processes involved in the formation of podiform chromitite

411   (Barnes et al. 1985). The behavior of PGE during the crystallization of host

412   chromitite is significantly influenced by two key mechanisms: the degree of mantle

413   partial melting to form the source magma and the extent of crystal fractionation

414   during the magma evolution (e.g., Stockman and Hlava 1984; Legendre and Augé

415   1986). These mechanisms are central to understanding the distribution and

416   behavior of PGE within the chromitite. The chondrite-normalized PGE patterns of

417   mantle peridotite xenoliths and Alpine peridotites typically exhibit flat profiles (Xu

418   and Liu 2019; Xu et al. 2020; Liu et al. 2020). The Pd/Ir ratio, a measure of

419   PPGE/IPGE fractionation, can be used as an indicator of melt depletion and

420   metasomatism (e.g., Liu et al. 2010; Pearson et al. 2004). The continental and

421   oceanic basalts exhibit steeply sloping positive patterns with Pd/Ir ratios ≤100,

422   reflecting more pronounced PGE differentiation in the parental magmas (Xu and

423   Liu 2019; Xu et al. 2020; Liu et al. 2020).

424   Leblanc (1991) suggested that the PGE concentrations in ophiolite-associated

425   podiform chromitites increase as Pd/Ir ratios decrease. Chondrite-normalized

426 PGE patterns of ophiolites typically display a positive slope and variable Pd/Ir

427 ratios, whereas associated chromitites exhibit a negative slope and Pd/Ir ratios

428 typically around 0.1 (Zhou and Robinson 1994). The total PGE (ΣPGE)

429 concentrations in both the Luobusa high-Cr and high-Al podiform chromitites

430 range from tens to thousands of ppb (Fig. 8). In the chondrite-normalized PGE

431 patterns, the high-Cr chromitites from Luobusa consistently exhibit significant

432 depletion of PPGE relative to the IPGE (Fig. 8). The geochemical characteristics of

433 Luobusa dunite are similar to those of the high-Al chromitite (Fig. 8b), showing

434 diverse behavior with varying degrees of incompatibility or preferential varying

435 removal, typically in the order of rhenium (Re) > Pd > Pt > Ir > Ru > Os (Pearson et

436 al. 2004).

437 The absolute abundances of the PGE and their normalized patterns, as well

438 as the chemical composition of Cr spinel in podiform chromitites, are sensitive

439 indicators of mantle partial melting, primary melt composition, and melt

440 saturation with volatile components (Zaccarini et al. 2005). The Luobusa

441 chromitites commonly described as high-Cr chromitite (Xiong et al. 2015), which

442 are enriched in the Ir subgroup (IPGE = Os, Ir, Ru) relative to the Pd subgroup

443 (PPGE = Rh, Pt, Pd) and display negatively-leaning chondrite-normalized PGE

444 patterns typical of ophiolitic high-Cr chromitites elsewhere (Melcher and Meisel

445 2004; Fig. 8). Dunite and harzburgite of Luobusa ophiolite have variable $(Ru/Pt)_N$

446 ratios ranging from 0.25 to 8.37, 0.12 to 6.55, respectively. These variations

447 indicate the percolation of sulfide-rich melts enriched in PGE (Figs. 3 and 4).

448 Similar to most high- Cr chromitites, the Luobusa chromitite samples show a

449 consistent relationship between $(Pt/Ir)_N$ and $(Pd/Ir)_N$ ratios. The majority of data

450 points align along a common fractional melting curve in the $(Pt/Ir)_N$ versus

451 $(Pd/Ir)_N$ space (Fig. 9a). In contrast to abyssal peridotites, forearc peridotites

452 exhibit depletion of osmium (Os) and low Os/Ir ratios. The higher Os/Ir ratio (e.g.,

453 5.9 in the chromitite sample 11Y910-4) can indicate processes like partial melting

454 or fractional crystallization of peridotites (Table S7). In contrast, the lower Os/Ir

455 ratio (e.g., 1.108 in the dunite sample 11Y-1107) suggests a different geological

456 history, possibly influenced by sulfide segregation or other processes (Fig. 9c). The

457 studied chromitites have undergone a significant loss of Os during sulfide

458 dissolution (e.g., Liu et al. 2010).

459   The Luobusa chromitites have Cr#s = 76-83 (Xiong et al. 2015). Compared to
460   other high-Cr and high-Al chromitites, the Luobusa chromitites display contrasting
461   fractionation trends (Fig. 9b), while dunite samples show a similar trend to abyssal
462   peridotites. Os, present in highly siderophile element (HSE) mineral phases in the
463   mantle wedge, can be mobilized by sulfide-undersaturated melt or fluid
464   components, such as those with high oxygen fugacity (fO_2) and chloride-rich
465   compositions derived from subducted slabs (Liu et al. 2010; Foustoukos 2019; Xu
466   et al. 2020). This mobilization of Os relative to Ir, Ru, and Pt leads to significant
467   fractionation, resulting in low Os/Ir ratios and overall abundances of total PGE in
468   peridotites (Liu et al. 2010) (Figs. 5, 6; Fig S1) as observed in the Luobusa ophiolite.
469   In contrast, the chromitites have Os/Ir ratios higher than the peridotites (Fig. 9c).
470

471   *Genesis of the PGM and BMS inclusions*

472   The formation of Luobusa podiform chromitites is believed to result from multiple
473   stages of partial melting and mantle metasomatism, include the MOR and SSZ
474   processes (Bai et al. 2000; Robinson et al. 2004; Xiong et al. 2015, 2022a, b). The
475   heterogeneity of PGE composition and Os isotopes reflects the mixing of melts
476   derived from different mantle source regions during multiphase metamorphism
477   or migration of chromitite to different mantle levels. According to Zhou et al.
478   (1996), the multi-stage crystallization and re-equilibrium of chromite may have
479   led to PGM dissolution and reprecipitation, suggesting that some PGM inclusions
480   are secondary. Nonetheless, euhedral PGM inclusions and silicate exsolutions in
481   chromite may reflect deeper mantle depths (e.g., Yang et al. 2007).
482   The PGE-bearing sulfides and sulfarsenides occur as euhedral inclusions,
483   while BMS typically form anhedral or amoeboid particles interstitially between
484   chromitite grains. In some cases, micro-diamond inclusions in Os-Ir alloys were
485   reported in the Luobusa chromitite (Yang et al. 2007). Laurite and Os–Ir–Ru alloys
486   commonly form during the early stages of magma crystallization at high
487   temperatures (~1300°C) and low sulfur fugacity (fS_2) (e.g., Zaccarini et al. 2005;
488   Fig. S8). IPGE partition favorably into BMS when they crystallize from S-rich melts,
489   which tend to retain Ag, and possibly Au and semi-metals (As, Sb, Pb, Bi)
490   fractionate into the co-existing sulfur-rich melt.

16

491   During mantle upwelling, the BMS and laurite-erlichmanite solid–solution

492   crystallize at lower temperatures and higher levels of $f\mathrm{S}_2$ (Fig. 10) (Klein and Bach

493   2009). The abundant BMS inclusions, including millerite and heazlewoodite, in

494   Luobusa chromite may indicate high $f\mathrm{S}_2$ conditions (~-11). Also, the coexistence

495   of pentlandite and heazlewoodite has been explained by the establishment of

496   highly reducing conditions under low temperatures (Klein and Bach 2009).

497   Millerite inclusions in chromite may imply that crystallization occurred below the

498   heazlewoodite-millerite buffer and continued to low temperatures (< 1000°C) and

499   high $f\mathrm{S}_2$ conditions (Fig. 10). Sulfides with low PGE contents can be explained by

500   the late paragenesis of some of these sulfides. This is similar to the Bacuri complex

501   in Amapa of northeastern Brazil (Prichard et al. 2001; Zhu and Zhu 2019), in which

502   the PGE were concentrated in chromitite and sulfide-bearing serpentinite, where

503   pentlandite is partially altered to millerite, maucherite, and gersdorffite. The

504   irarsite formed at the final stage of hydrothermal processes with the introduction

505   of As, Ir, and Os, which are associated with serpentine, chlorite, and base metal

506   minerals (Fig. 11).

507   The genesis of the Luobusa chromitite unveiled a complex interplay of

508   geological processes (Fig. 11). Euhedral PGE-bearing sulfides and sulfarsenides

509   contrasted with anhedral or amoeboid BMS particles, indicating distinct

510   crystallization environments. Early magma crystallization at high temperatures

511   gave rise to laurite and Os-Ir-Ru alloys, with subsequent BMS crystallization

512   occurring under lower temperatures and higher sulfur fugacity. The presence of

513   millerite and heazlewoodite suggested highly reducing conditions, while the late-

514   stage introduction of As, Ir, and Os was indicative of hydrothermal processes. This

515   dynamic evolution, marked by interactions between mantle upwelling,

516   suprasubduction processes, and fluid intrusion, resulted in the formation of

517   chromian spinels, PGE-bearing sulfides, and PGM at high temperatures. The

518   integration of geochemical data and isotopic characteristics further supported a

519   multifaceted genesis involving mantle partial melting, melt-peridotite interaction,

520   and slow spreading ridge dynamics, ultimately shaping the unique composition

521   and structure of the Luobusa chromitite.

522

523 **IMPLICATIONS**

524 The mineralogical and geochemical characteristics of the Luobusa chromitite and

525 ophiolite suggest an intricate geological history shaped by a dynamic interplay of

526 factors. Abundant occurrences of PGM and BMS inclusions underscore a multi-

527 stage genesis involving processes like partial mantle melting, melt-rock

528 interactions, and fluctuations in oxygen and sulfur fugacity. The geochemical

529 fingerprints, encompassing the patterns of PGE, provide substantial evidence for

530 the transformation of the Luobusa ophiolite from a sub-ridge environment to a

531 supra-subduction zone setting.

532 The bulk-rock geochemical attributes of the Luobusa chromitite align closely

533 with those of forearc supra-subduction zone (SSZ) settings, implying a

534 compositional influence from both mantle and crustal melts. Additionally, the

535 trace element characteristics of pyrite inclusions bear striking resemblances to

536 sulfides found in MORB and OIB rocks, accentuating the complex intermingling of

537 various magmatic sources.

538 The observed diversity in PGM and BMS inclusions, coupled with the

539 geochemical features of the chromitite, advocate for an intricate genesis

540 characterized by mantle partial melting, assimilation of crustal components, and

541 extensive melt fractionation. The coexistence of well-formed Os-Ir alloys alongside

542 interstitial BMS implies conditions of elevated temperatures and low fS_2 during

543 earlier stages. The presence of sulfarsenides as small inclusions in pyrite point

544 towards lower temperatures and higher fS_2 values during later paragenetic phases.

545 This multi-faceted geological history exemplifies the intricate nature of chromitite

546 formation in the Luobusa ophiolite.

547

548 **Acknowledgments**

18

559

560 **Appendix A. Supplementary Material**

561

562 This section provides a brief overview of the supplementary files associated with

563 this paper, offering readers additional information to complement the main text.

564 In Fig. S1, backscattered electron (BSE) images offer insights into the morphology,

565 texture, and mineral assemblage of the discovered PGM, BMS, and silicates within

566 the Luobusa chromitite in Tibet. Fig. S2 to Fig. S6 feature detailed mapping of

567 various mineral intergrowths and inclusions, shedding light on their spatial

568 distribution within the chromitite. Additionally, Fig. S7 showcases the different

569 PGE-bearing alloy inclusions in the chromitite. Finally, Fig. S8 places the

570 crystallization conditions for melt-sulfide into a thermodynamic context, as

571 referenced from Uysal et al. 2015.

572 Furthermore, this appendix contains essential supplementary tables,

573 including Table S1 to Table S7, which present representative compositions of

574 various minerals, inclusions, and alloy phases within the Luobusa chromitite,

575 along with trace element data. This supplementary material is indispensable for a

576 comprehensive understanding of the research presented in this paper.

577

578

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Figure captions

Figure 1. Geological map of the Luobusa ophiolite, modified from Xiong et al. (2015). Cumulate rocks include grabbros, wehrlite, and pyroxenite.

Figure 2. Field photographs illustrating the occurrence of chromitite in the Luobusa ophiolite. (a) Disseminated chromitite in the dunite unit. (b) Disseminated chromite in dunite and small nodules of massive chromitite. (c) Well-developed nodular chromitite in contact with dunite containing thin layers of disseminated chromite. (d) Sharp contact between massive chromitite and dunite.

Figure 3. Petrographic features of the BMS inclusions in the Luobusa chromitites. (a) Reflected light microphotograph of pyrite inclusions. (b)-(0) Backscattered electron (BSE) images of (b) Galena inclusion in chromite. (c) Millerite inclusion. (d) Auwrite inclusion. (e) Irregular millerite inclusions. (f) Pyrite intergrowth with millerite. (g) Amphibole inclusions in pentlandite. (h) Serpentine and Ni in NiS inclusion. (i) Chalcocite inclusion. (j)-(k) Millerite and serpentine intergrowths. (l) Millerite intergrown with clinopyroxene. (m) Quartz and chalcocite inclusions in pyrite. (n) Awaruite intergrown with millerite as inclusions in chromite. (o) Intergrown Co-Fe-Ni-S and millerite in chromite. Py = pyrite, Gn = galena, Mlr = millerite, Pn = pentlandite, Amp = amphibole, Serp = serpentine, Cc = chalcocite, Cpx = clinopyroxene, Qz = quartz, Awr = awaruite, Chr = chromite.

Figure 4. BSE images of pyrite and other mineral inclusions in the Luobusa chromitites. Sh = schuetteite, Py = pyrite, Srp = serpentine, Sph = sphalerite, Tnt-Zn = tennantite-(Zn), Gn = galena, Ttr-Zn = tetrahedrite-(Zn).

Figure 5. BSE images of different PGM inclusions and alloys in chromite from the Luobusa ophiolite. (a-c) PGM inclusions are dispersed as single-phase, (d-i) Polyphase hypidiomorphic and xenomorphic PGE alloys. In some examples, symplectite textures with microscopic multiphase (Fe-Ni, Ir-Fe) pseudostructures in Os-Ir, Os-Ir-As, Lrt, Os-Al, and Ir-Al matrices (d, e, f). Lrt = laurite, Chr = chromite.

Figure 6. Triangular diagram illustrating the compositional variation of PGM and alloys in the Luobusa chromitite. (a) Classification of PGM in the Luobusa

1027　　　chromitites based on Ru, Os, etc., compared with data from Cabri (2002) and

1028　　　Bai et al. (2004). (b) Ternary diagram depicting the compositional variation

1029　　　of Fe–Ni–Pt alloys in the Luobusa chromitites in terms of Pt, Rh, Os – Ni, Co,

1030　　　Cu – Fe.

1031　Figure 7. Binary plots of pyrite trace element compositions. Measurements below

1032　　　the minimum detection limit were excluded.

1033　Figure 8. Chondrite-normalized (Naldrett and Duke 1980) PGE patterns for

1034　　　chromitite (a) and dunite (b) in the Luobusa ophiolite, compared with other

1035　　　podiform chromitites (data sources: Uysal et al. 2007; Rui et al. 2022).

1036　Figure 9. Plots of whole-rock compositions: (a) $(Pt/Ir)_N$ versus $(Pd/Ir)_N$; (b) 1/Os

1037　　　versus Pd/Ir; (c) 1/Os versus Os/Ir (based on Xu et al. 2020, 2021). The melt

1038　　　extraction curve was calculated using nonmodal fractional melting, assuming

1039　　　a primitive mantle source with 300 ppm S and extracted melts with a sulfur

1040　　　concentration of 1000 ppm. The primitive upper mantle (PUM) values of

1041　　　these ratios are from Meisel et al. (2001). The plot includes abyssal

1042　　　peridotites (Parkinson et al. 1998; Liu et al. 2008; Day et al. 2017; Xu and Liu

1043　　　2019; Scott et al. 2019), massif peridotites (data from Becker et al., 2006;

1044　　　Fischer-Gödde et al., 2011; Lorand et al., 2009; Luguet et al., 2007; Rampone

1045　　　et al., 1995; etc.), and forearc peridotites (SSZ; data from Haller et al., 2021;

1046　　　O'Driscoll et al., 2012, 2015, 2018; Prelević et al., 2015; Uysal et al., 2012; etc.).

1047　　　The high-Cr and high-Al chromitite data are from Rui et al. (2022).

1048　Figure 10. (a) Metal-sulfide equilibrium curves for Ru, Os, Ir, and Ni as a function

1049　　　of sulfur fugacity ($\log f_{S2}$) and temperature (T) based on Stockman and Hlava

1050　　　(1984) and Garuti et al. (1990). (b) Oxygen fugacity (f_{O2}) versus sulfur

1051　　　fugacity (f_{S2}) diagram in the Fe–Ni–Cu–O–S system at 300 °C and 50 MPa,

1052　　　providing insights into the evolution of Fe–Ni minerals in the matrix of

1053　　　Luobusa chromitite (adapted from Zhu and Zhu 2019).

1054　Figure 11. Conceptual genetic model of the formation of PGM and BMS inclusions

1055　　　in the Luobusa chromitites. (a) Early-stage melt interaction with harzburgitic

1056　　　peridotite. (b) Accumulation of chromite spinel in layers or pockets and

1057   encapsulation of BMS, PGE-bearing sulfides, and PGM inclusions during

1058   chromite crystallization at high temperatures. (c) Serpentinization and melt

1059   percolation during subduction, leading to the appearance of amphibole and

1060   other hydrous minerals, alteration of PGE-bearing minerals and Ni-sulfides,

1061   and precipitation of pyrite, as well as Ag, Pb, Bi-sulfide phases. Ol = olivine,

1062   Opx = orthopyroxene, Spl = spinel, Pn = pentlandite, Lrt = laurite, Chr =

1063   chromite.

1064 **Supplementary Material**

1065 Fig. S1 Backscattered electron (BSE) images showing morphology, texture and

1066 mineral assemblage of the discovered PGM, BMS and silicates in Luobusa

1067 chromitites.

1068 Fig. S2 Mapping of millerite intergrown with serpentine in chromite (Fig. 3j).

1069 Fig. S3 Millerite intergrowth with serpentine in chromite (Fig. 3k).

1070 Fig. S4 Awaruite intergrowth with millerite in chromite (Fig. 3n).

1071 Fig. S5 Co-Fe-Ni-S intergrowths with millerite in chromite (Fig. 3o).

1072 Fig. S6 Millerite intergrowth with PGM in the chromite.

1073 Fig. S7 PGE-bearing alloys in chromite (Fig. 5f).

1074 Fig. S8 The thermodynamic context of crystallization conditions for melt-sulfide

1075 (after Uysal et al. 2015)

1076

1077 **Table captions:**

1078 **Table S1.** Representative compositions of sulfides in Luobusa chromitite (wt%).

1079 **Table S2.** Compositions of sulfide inclusions in the Luobusa chromitite (wt%).

1080 **Table S3.** Compositions of the PGM alloys in the Luobusa chromitite (wt%).

1081 **Table S4.** Compositions of PGM alloys in the Luobusa chromitite (wt%).

1082 **Table S5.** Compositions of Fe-Ni phases in the Luobusa chromitite (wt%).

1083 **Table S6.** Trace element analysis of pyrite in the Luobusa chromitite.

1084 **Table S7.** PGE contents of chromitites in the Luobusa ophiolite.

1085
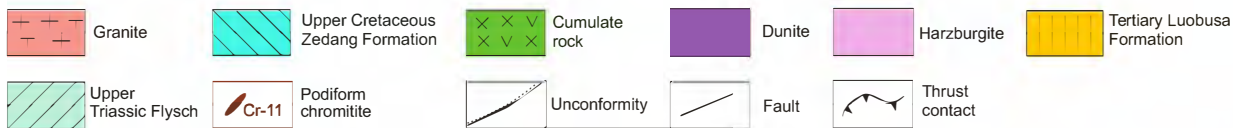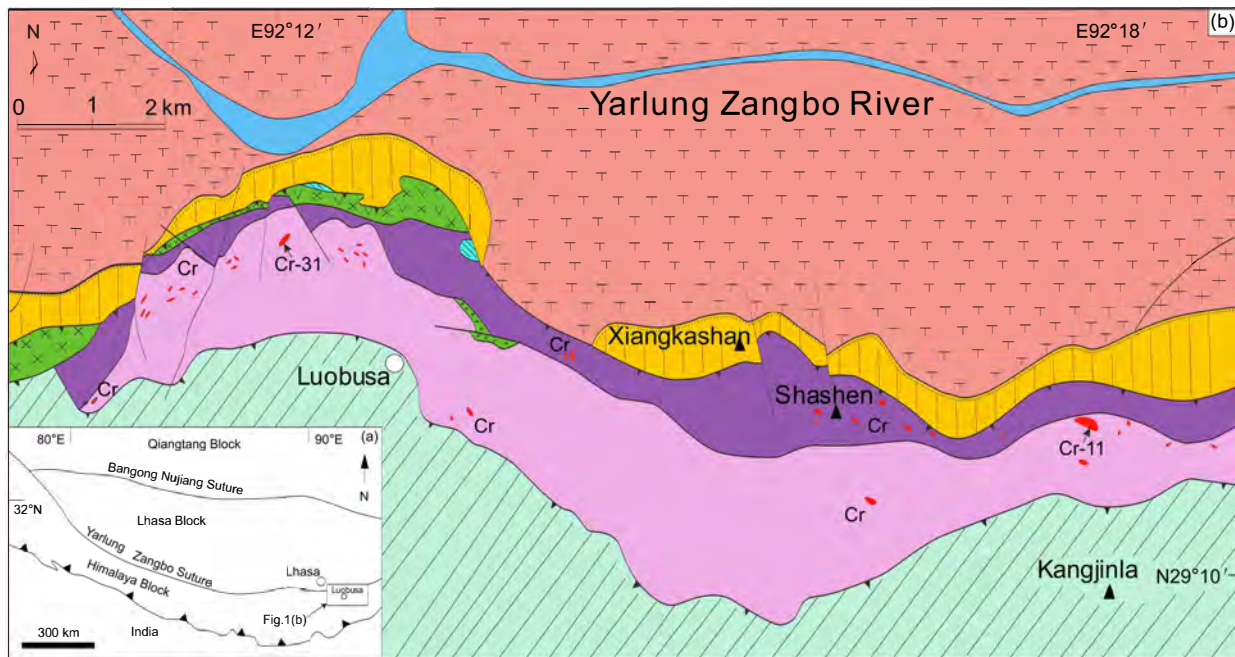
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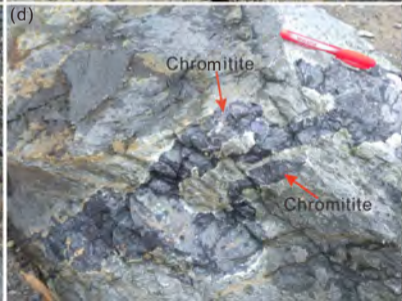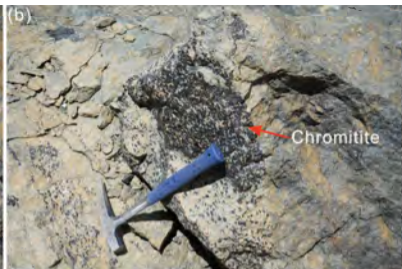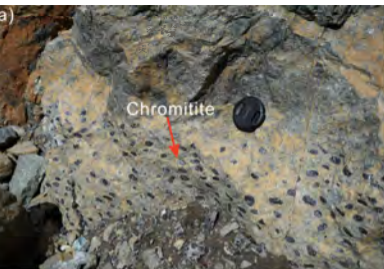
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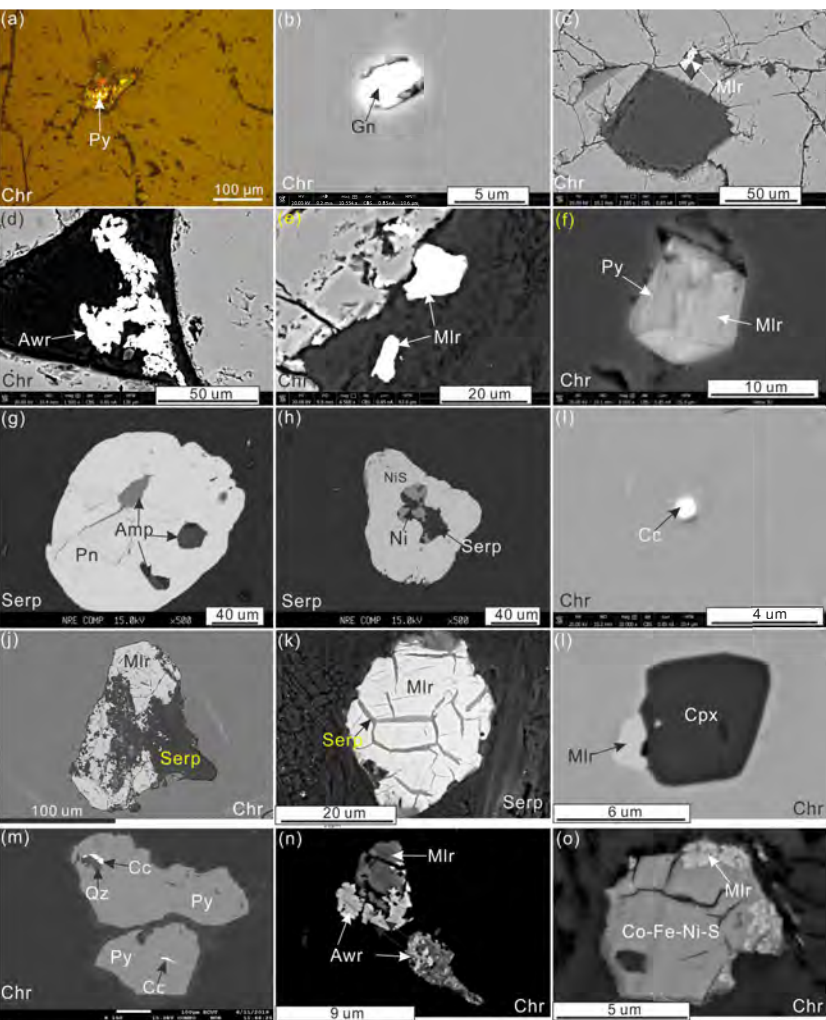
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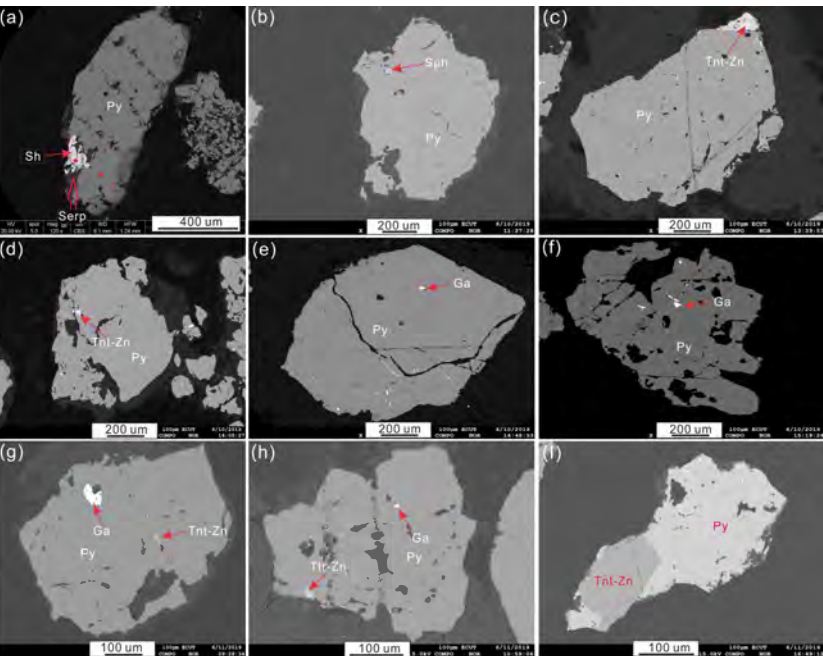
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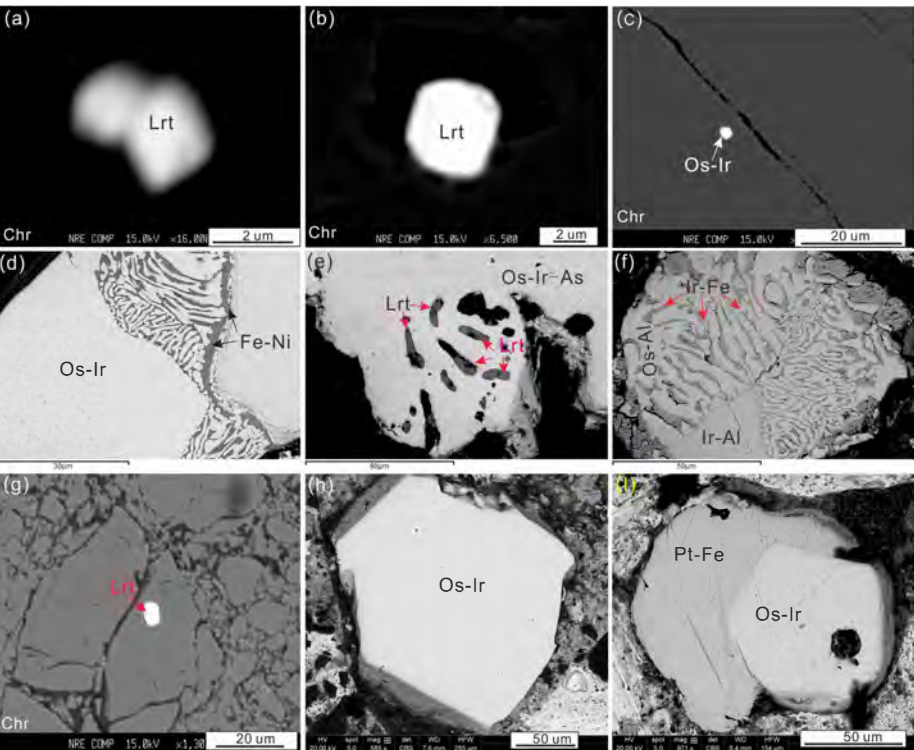
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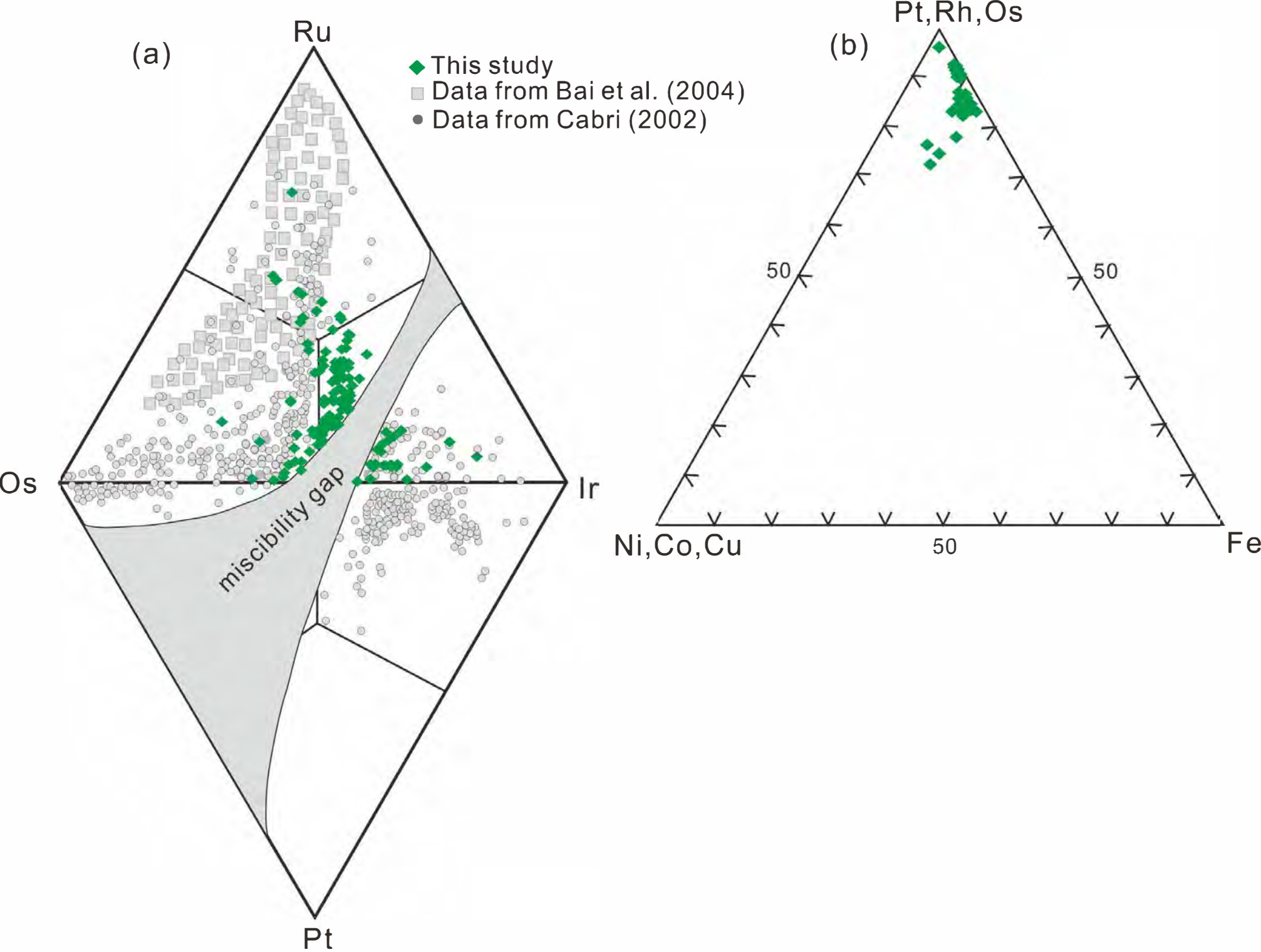
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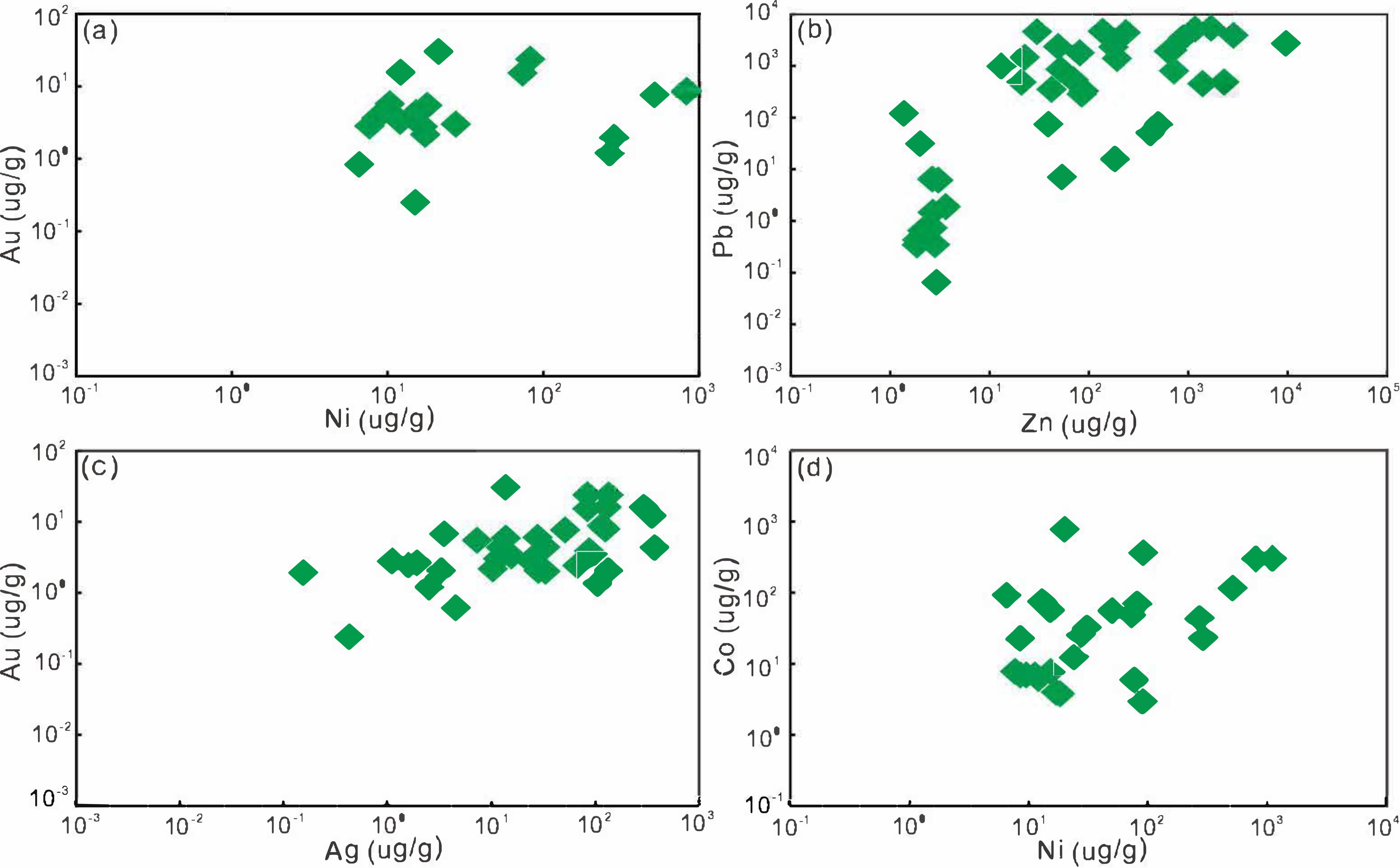
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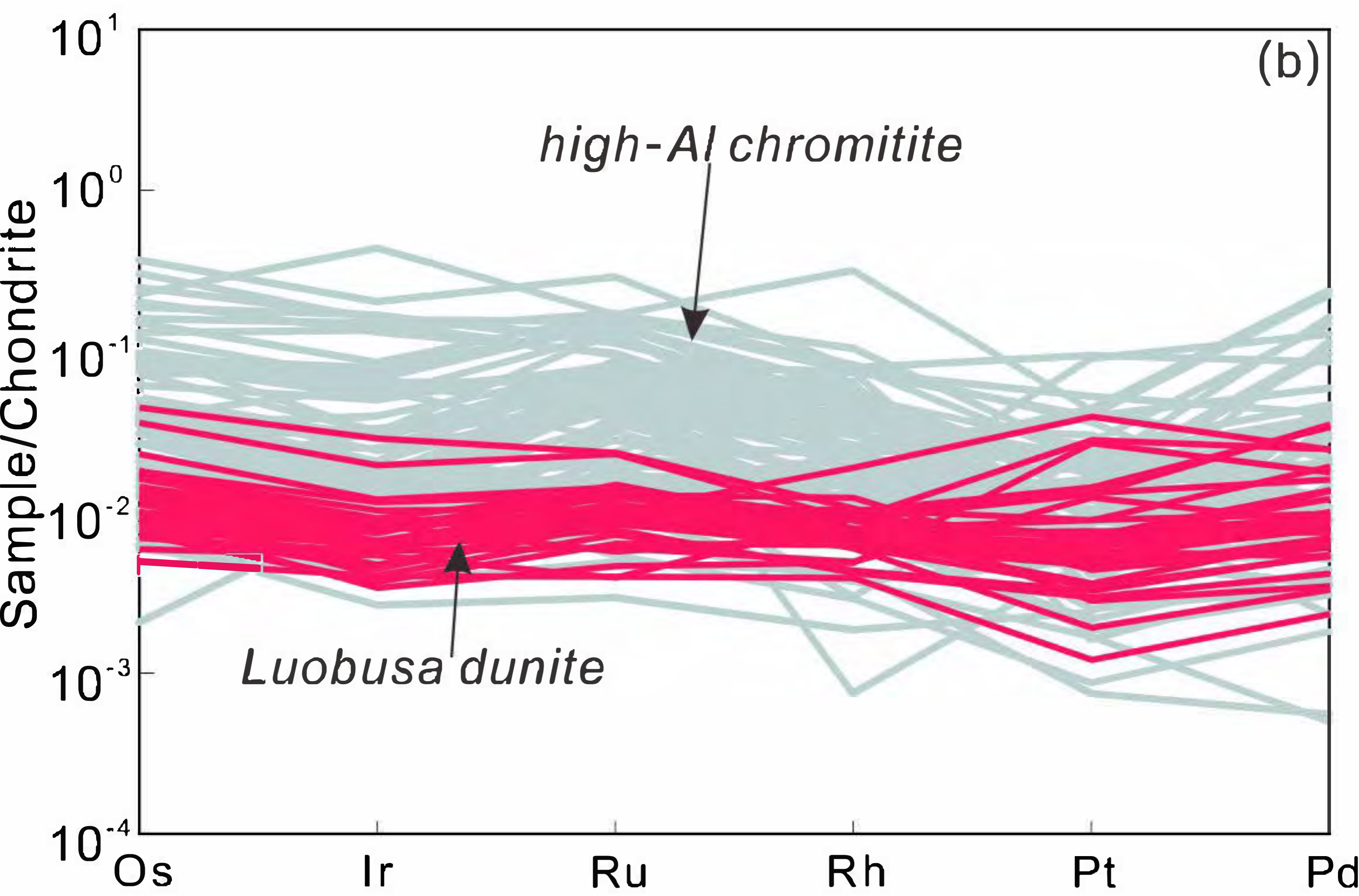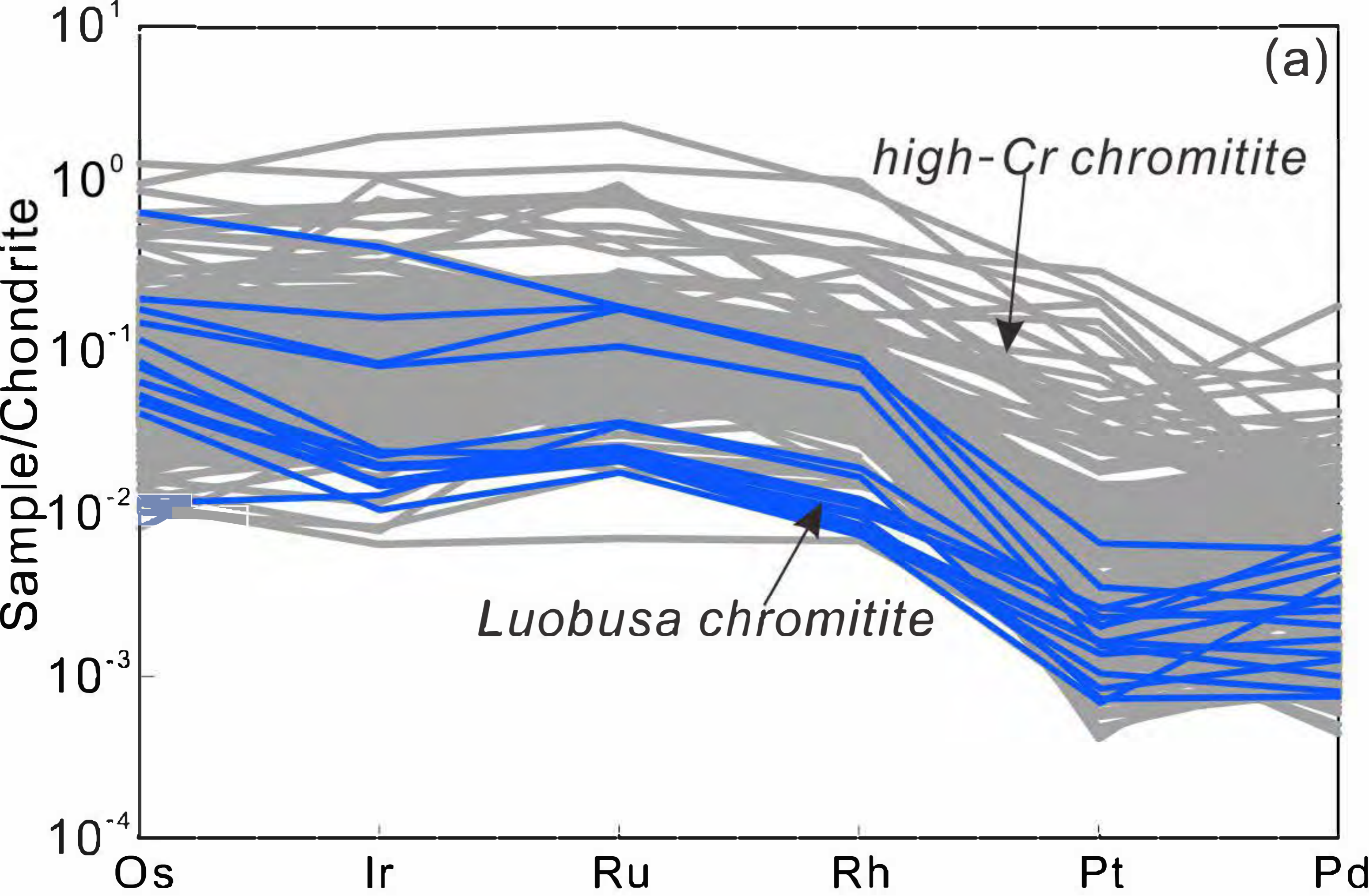Figure 8
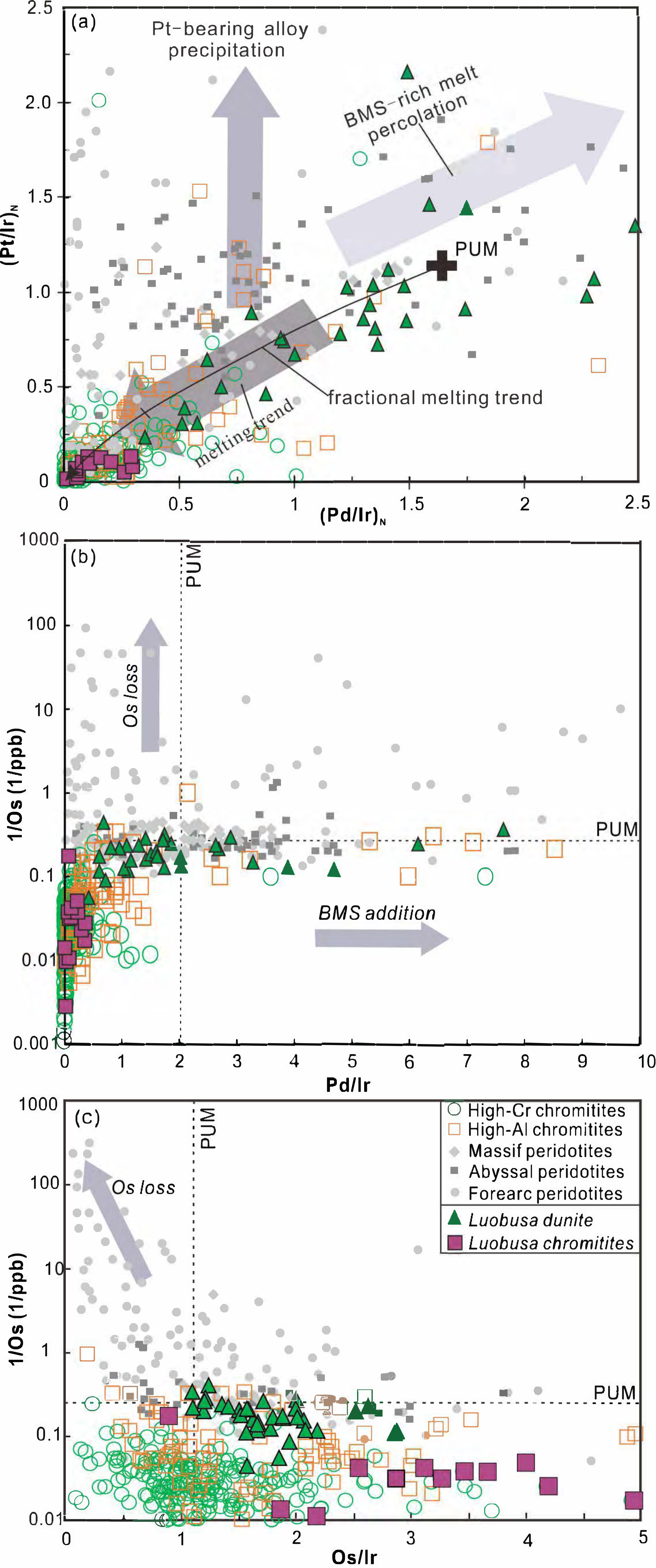
Figure 9

Figure 10

Figure 11