1 **Revision 2**

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3	Petrogenetic insights from chromite in ultramafic cumulates of the
4	Xiarihamu intrusion, northern Tibet Plateau, China
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32 Abstract

Chromite is one of the earliest crystallized minerals from mafic melts and has 33 been used as an important "petrogenetic indicator". Its composition may be modified 34 by interaction with intercumulate melt and adjacent minerals. Thus, chromite in 35 mafic-ultramafic rocks contains clues to the geochemical affinity, evolution and 36 37 mantle source of its parent magmas. The Devonian Xiarihamu intrusion, located in the East Kunlun Orogenic Belt in the northern Tibet Plateau, China, hosts a very large 38 disseminated Ni-Co sulfide deposit. This study focuses on geochemistry of the 39 chromite enclosed in olivine of ultramafic rocks of the intrusion. Enrichments in Mg 40 and Al in the rim of the chromite indicate only minor effects of alteration on the 41 compositions of the chromite. The chromites enclosed in the olivines with forsterite 42 percentage (Fo) lower than 87 are characterized by large variations in major and trace 43 elements, such as large ranges of Cr*100/(Cr+Al) (Cr# = 15-47), Mg*100/(Mg+Fe²⁺) 44 (Mg# = 41-65) and Al_2O_3 (=26-53 wt%) as well as 380-3100 ppm V, 70-380 ppm Ga 45 and 1100-16300 ppm Zn. The chromites display positive correlations between 46 Cr/(Cr+Al) and Ti, Mn, V, Ga and Sc, inconsistent with fractional crystallization but 47 indicative of interaction between the chromites, intercumulate melts and hosting 48 49 minerals. In contrast, chromites hosted in olivine with Fo > 87 in harzburgite have small variations in Cr# (ranging from 37 to 41), Mg# (48 to 51) and Al₂O₃ (30 to 35 50 wt%) as well as restricted variation in trace elements, indicating relatively weak 51 interaction with trapped liquid and adjacent phases; these compositions are close to 52 those of the most primitive, earliest crystallized chromites. The most primitive 53

54	chromite has similarities with chromite in MORB in TiO_2 and Al_2O_3 contents
55	(0.19-0.32 wt% and 27.9-36.3 wt%, respectively) and depletion of Sc and enrichment
56	of Ga and Zn relative to MORB chromite. The geochemistry of the chromite indicates
57	a partial melting of the asthenospheric mantle that was modified by melts derived
58	from the subduction slab at garnet-stable pressure.
59	Key words: chromite, mineral geochemistry, intercumulate melt, primary magma,
60	trapped liquid, magmatic sulfide, nickel, spinel
61	1. Introduction
62	It is difficult to reveal the geochemical nature of the parental magma of
63	ultramafic cumulate rocks. There is always uncertainty in parent magma compositions,
64	especially in the absence of demonstrably comagmatic basalts and in cases where the
65	ultramafic cumulates are predominately adcumulate in character. In these cases parent
66	magma characteristics must be inferred from compositions of the cumulus minerals.
67	Chromite is often among the first phases crystallized from mafic and ultramafic
68	magma and is relatively refractory and resistant to alteration compared with the other
69	early crystallizing minerals, such as olivine and pyroxene (e.g., Barnes and Roeder
70	2001; Kamenetsky et al. 2001). Although compositions of chromite can be changed
71	due to reaction with hosting minerals and trapped liquids (e.g., Henderson 1975;
72	Roeder and Campbell 1985; Scowen et al. 1991), important clues of parental magma
73	and magma evolution can be preserved (e.g., Kamenetsky et al. 2001; Pagé and
74	Barnes 2009). Thus, it has been used as an important "petrogenetic indicator" to
75	reveal the geochemical natures of primary magmas and mantle sources of mafic and

76	ultramafic rocks (e.g., Irvine 1967; Barnes and Roeder 2001; Kamenetsky et al. 2001;
77	Proenza et al. 2004; Ahmed et al. 2005; González-Jiménez et al. 2014) and as a
78	potential indicator for mineralized intrusions or lava flows (Barnes and Tang 1999;
79	Barnes and Kunilov 2000; Locmelis et al. 2013; Evans 2017; Locmelis et al. 2018).
80	Mafic-ultramafic intrusions hosting magmatic sulfide deposits have been
81	discovered in orogenic belts over the world, where contemporaneous basalts are
82	commonly absent, such as in the Central Asian Orogenic Belt in NW China (Song and
83	Li 2009, Song et al. 2011; Zhang et al. 2011; Qin et al. 2011; Xie et al. 2012, 2014;
84	Deng et al. 2014, 2015 and references there in), the Variscan collisional orogeny,
85	Spain (Piňa et al. 2008, 2010), the Selebi-Phikwe belt, Botswana (Maier et al. 2008)
86	and the Itabuna-Salvador-Curaca Belt, Brazil (Barnes et al. 2011, 2013). There are
87	still controversies on why and how a voluminous mantle-derived mafic magma,
88	sufficient for the formation of large magmatic sulfide deposits, was generated in the
89	orogenic belts. Thus, it is significant to recognize geochemical features of the primary
90	magma and then to reveal specific characteristics of the mantle source and conditions
91	for intensively partial melting to producing large amounts of mafic magma in
92	orogenic belts. Some geologists prefer to associate the mafic-ultramafic intrusion in
93	orogenic belt to mantle plumes (Qin et al. 2011), whereas others favour other
94	geodynamic mechanisms to generate extensive basaltic magmatism in convergent
95	settings (Song et al. 2011, 2013; Kohanpour et al. 2017).

The Xiarihamu Ni-Co sulfide deposit, hosted in an ultramafic intrusion dated at 406.1 ± 2.7 Ma, is located in the Eastern Kunlun Orogenic Belt, northern Tibet

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98	Plateau, China (Li et al. 2015; Song et al. 2016; Liu et al. 2018). Its large reserves,
99	~157 million tonnes sulfide ores with average grades of ~0.65 wt% Ni, places it
100	among the largest 30 individual Ni sulfide deposits in the world, containing over 1
101	million tonnes of contained Ni metal in sulfide. Studies of the early Devonian
102	volcanic sequence consisting of andesite, dacite and rhyolite (408.2 \pm 2.4 Ma) have
103	confirmed that the Eastern Kunlun Orogenic Belt was a convergent margin from
104	Silurian to early Devonian (Lu et al. 2010; Liu et al. 2012). There is no geological
105	evidence for Paleozoic mantle plume magmatism in the northern Tibet Plateau. Thus,
106	the Xiarihamu intrusion is a very good study site to address the key factors controlling
107	production of voluminous basaltic magma for large scale magmatic sulfide
108	mineralization in a convergent setting. Previous whole-rock geochemical studies have
109	concluded that the Xiarihamu intrusion was genetically associated with basaltic
110	magmas derived from a metasomatised mantle source during subduction or post
111	collision in the East Kunlun Orogenic Belt (Song et al. 2016; Li et al. 2015). However,
112	the nature of mantle source and the mechanism of partial melting are still unclear.
113	In this study, major and trace elements of fresh chromites in the Xiarihamu

ultramafic rocks have been measured by means of wavelength-dispersive X-ray 114 emission spectrometry, using an electron probe microanalyzer (EPMA) and 115 laser-ablation inductively-coupled spectrometry plasma-mass (LA-ICPMS), 116 respectively. The new data indicate that the most primitive chromites enclosed in 117 olivine with fosterite contents in Mol.% (Fo) between 87 and 89 in harzburgite are 118 high in Al₂O₃, enriched in Ga, Ti and Zn, and depleted in Sc relative to the chromite in 119

mid-ocean ridge basalt (MORB). These features of the chromite lead us to propose
that the primary magma was derived from partial melting of asthenosphere peridotite
modified by silica-rich melts generated from melting of subducted MORB at a depth
where garnet was stable.

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2. Geological Background

125 The Xiarihamu ultramafic intrusion is situated in the E-W trending East Kunlun Orogenic Belt in the northern Tibet Plateau, China (Fig. 1a). The East Kunlun 126 Orogenic Belt lies between the Qaidam block to the north and the Songpan-Ganzi 127 block to the south and bounded by the Altyn Tagh fault to the west and the Qinling 128 Orogenic Belt to the northeast (Xu et al. 2007; Dong et al. 2018a). It is subdivided 129 into the Northern, Middle and Southern zones by the northern and central Kunlun 130 131 faults as shown in Figure 1b (Song et al. 2016). The Middle Kunlun Zone mainly comprises the Proterozoic metamorphic strata and a number of large Paleozoic and 132 Mesozoic granitoid plutons (Fig. 1c). The Proterozoic metamorphic strata are 133 dominantly composed of felsic gneiss, feldspar quartz schist and marble and is 134 overlain by Ordovician low grade metamorphosed clastic rocks, which are in turn 135 unconformably overlain by the Early Devonian volcanic strata comprising of andesite, 136 dacite and rhyolite with basalt interlayers (Chen et al. 2006; Lu et al. 2010). U-Pb age 137 dating of zircons separated from the rhyolite indicated that the Devonian volcanic 138 strata formed at 405-425 Ma when the East Kunlun Ocean closed (Lu et al. 2010). 139 The early Devonian volcanics are unconformably overlain by Carboniferous and 140 Permian sedimentary and volcanic strata. Voluminous Paleozoic and Mesozoic 141

This is a preprint, the final version is subject to change, of the American Mineralogist (MSA) Cite as Authors (Year) Title. American Mineralogist, in press. DOI: https://doi.org/10.2138/am-2020-7222

Revised Text of Xiarihamu chromite by Song et al. for AM

granitoid plutons (390-440 Ma and 240-260 Ma, respectively) were emplaced into the 142 Proterozoic metamorphic rocks and the late Ordovician strata (Fig. 1c) (Mo et al. 143 2007; Xu et al. 2007; Gao and Li 2011; Cui et al. 2011; Liu et al. 2012; Huang et al. 144 2014 and references therein). 145 Several mafic-ultramafic intrusions containing magmatic sulfide mineralization 146 147 occur in the Middle East Kunlun Zone, including Xiarihamu, Shitoukende and Shuixiannan. Recent zircon U-Pb dating using LA-ICPMS or SHRIMP (Sensitive 148 High Resolution Ion Microprobe) methods indicated that these intrusions were 149 produced by late Silurian to early Devonian regional magmatism (400–430 Ma) (Fig. 150 1c, Zhou et al. 2015). Recent zircon U-Pb dating demonstrated that the Xiarihamu 151 intrusion was formed in Early Devonian (411 ± 2.4 Ma, Li et al. 2015; 406.1 Song et 152

153 al. 2016).

Early Paleozoic ophiolitic mélanges, comprising of serpentinite, gabbro, basalt 154 and chert, have been discovered along the North, Central and South East Kunlun 155 Faults (Fig. 1b). Dong et al. (2018a) proposed that the ophiolitic mélanges along the 156 South East Kunlun Fault are the remnant of Kunlun Ocean, whereas the ophiolitic 157 mélanges along the North East Kunlun Fault represent the relic of a back-arc oceanic 158 159 basin to the south of the Qiadam Block, resulting from northward subduction of the Kunlun Ocean. The Northern, Middle and Southern East Kunlun zones have 160 comparable Proterozoic metamorphic basements (Dong et al. 2018a). A series of 161 eclogite blocks have been discovered recently along the Northern East Kunlun Fault 162 and one of them occurs near the Xiarihamu intrusion (Fig. 1, Song et al. 2018). 163

164 **3. Petrology of the Xiarihamu intrusion**

The Xiarihamu intrusion is NE-SW extending and oval shaped, ~2500 m long 165 and ~ 2300 m wide and up to 500m thick. Its northeastern part is exposed at the 166 surface over an outcrop area ~1400 m long and ~900 m wide. The intrusion is 167 composed of a gabbroic portion and an ultramafic portion (Fig. 2a, Song et al. 2016). 168 169 Zircons from the websterite of the ultramafic portion and the gabbronorite of the gabbroic portion to the north display the same U-Pb ages within error (406.1 \pm 170 2.7Ma and 405.5 \pm 2.7Ma, respectively, Song et al. 2016). This is consistent with 171 the observations of no chilled margins and no mineral size variation between the 172 gabbroic and the ultramafic portions. The gabbronorite comprises 50-60 vol.% 173 plagioclase (vol.% means volume percentage of a mineral in rock, expressed as % 174 herafter), 25-35 % orthopyroxene, 10-20 % clinopyroxene, and interstitial 175 hornblende and biotite (< 5 %). 176

177 The ultramafic portion is aligned NE-SW and dips shallowly to the west as shown in the 3D geological model in Figure 2b. Medium- and fine-grained olivine 178 orthopyroxenite, orthopyroxenite and websterite are the principal rocks of the 179 ultramafic portion as described in detail by Song et al. (2016). The common 180 occurrence of marble as footwall rock suggests that the Xiarihamu intrusion is an 181 irregular sill conformably intruded into the folded Jinshuikou Formation (Fig. 2c. d). 182 The distribution and geometry features of massive ores, net-textured and patchy 183 net-textured ores (terminology of Barnes et al. 2017) in the main ore pods and 184 sublayers show that the Xiarihamu deposit occurs in a magma chamber within a 185

magma plumbing system (Song et al. 2016; Barnes et al. 2016).

orthopyroxenite 187 The consists of cumulus medium-grained euhedral orthopyroxene crystals (60-90 vol.%) and fine-grained intercumulus grains of 188 plagioclase (<10%), clinopyroxene (<10%) and minor hornblende and phlogopite. 189 Fine-grained olivine (<10%) is enclosed in the orthopyroxene, which commonly 190 191 displays complex zoning in its Cr content (Wang et al., 2019). It comprises the middle part of the ultramafic portion, occupies about 50 vol.% of the ultramafic portion, and 192 grades gradually into the websterite at higher stratigraphic levels to the east of the 193 portion with increasing clinopyroxene. The websterite contains 10-20% 194 clinopyroxene and ~80% orthopyroxene and <10% plagioclase, hornblende and 195 phlogopite. The olivine orthopyroxenite, which only occurs in the western part of the 196 197 ultramafic portion, was emplaced within or above the orthopyroxenite as a planar body and formed a sharp contact with the orthopyroxenite. 198

The olivine orthopyroxenite consists of 10–40% olivine, 50–80% poikilitic 199 200 orthopyroxene, <10% clinopyroxene and plagioclase, with minor hornblende and phlogopite. It grades into harzburgite where olivine increases to more than 40% in a 201 few locations as layers, such as in ZK1903 (Fig. 2d). Although the olivine grains may 202 be in mutual contact in the harzburgite, they are commonly enclosed in poikilitic 203 orthopyroxene in the olivine orthopyroxenite, whereas clinopyroxene, plagioclase, 204 hornblende and phlogopite are interstitial between olivine and orthopyroxene. In the 205 western part of the ultramafic portion, sharp contacts between the olivine 206 orthopyroxenite and orthopyroxenite and harzburgite. The olivine orthopyroxenite is 207

above the orthopyroxenite and there is an interlayer of gabbronorite between them as
observed in the drill holes ZK2309 (Fig. 2c) (Song et al. 2016). The disseminated
sulfide ores, commonly containing 5-15 vol.% sulfides, are dominantly hosted in the
olivine orthopyroxenite and the western part of the orthopyroxenite (Song et al. 2016;
Peng et al. 2016; Zhang et al. 2017).

213 In both the olivine orthopyroxenite and harzburgite, chromite is present at < 0.5vol.% and is commonly enclosed in olivine and rarely in pyroxene or as interstitial 214 crystals (Fig. 3). Chromite forms fine rounded grains (commonly $< 60 \mu m$, up to 215 100µm) and is finer-grained in the pyroxene than in the olivine. Although most of the 216 ultramafic rocks in the Xiarihamu intrusion have experienced extensive alteration, 217 mafic minerals are well preserved and quite fresh in some samples. In the studied 218 219 samples, the olivine that has chromite inclusions has been variably serpentinized along fractures, but the chromite is commonly optically and homogeneous as shown 220 by backscattered electron images (Figs. 3b, d). Tiny high-reflective crystals, probably 221 magnetite or ilmenite formed by solid-state exsolution, may be seen in some of the 222 chromites from the olivine orthopyroxenite. The bright narrow rims (less than 10µm) 223 or very thin microveins along fracture seen in some chromite crystals in backscattered 224 225 electron images (Figs. 3b, d) probably are overgrowths of magnetite formed during serpentinization of olivine. These textural features indicate that the serpentinization 226 has had little effect on the composition of the chromite, which therefore preserves 227 magmatic signatures (Barnes 2000). 228

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In contrast, the chromites enclosed in orthopyroxene may show slight enrichment

of the core in Cr (Fig. 3f). These chromites commonly contains crystallographically
oriented tiny high-reflective crystals, which is probably magnetite or ilmenite formed
by solid exsolution during cooling, and had been variably modified in composition
(Fig. 3f) (Colás et al. 2016). Thus, this study focuses on the chromite enclosed in
olivine.

235 **4. Analytical methodology**

Samples in this study were collected from drill cores ZK1903 and ZK2309 in the western part of the ultramafic portion (Fig. 2). To avoid the effect of alteration on compositions of the olivine and chromite, 13 relatively fresh samples have been employed for this mineral geochemistry study. They are one orthopyroxenite, six olivine orthopyroxenites and six harzburgites, which were selected from 31 samples from the drill holes ZK2309 and ZK1903.

Ten harzburgites, thirteen olivine orthopyroxenites and eight orthopyroxenites 242 were collected from the drill holes ZK2309 and ZK1903 for whole-rock analysis. 243 Before whole-rock analysis, entire samples were crushed with steel jaws to -10 mesh, 244 and then ~ 200 g of this fraction were ground to -200 mesh powder using a tungsten 245 carbide ring mill. The powder was mixed completely with Li₂B₄O₇-LiBO₂ flux and 246 then fused to a glass bead at 1050–1100°C in an automatic melting instrument. Major 247 oxides and Cr of the glass beads were determined using a PANalytical Axios X-ray 248 fluorescence spectrometer (XRF) on fused glass beads at the ALS Chemex 249 (Guangzhou) Co., Ltd. Analytical uncertainties of the major and trace elements are 250 less than 5%. Loss on ignition (LOI) was obtained from 1 g powdered sample that 251

252	was heated up to 1100 °C for 1 h. Whole-rock Ni and Cu contents of samples
253	containing more than 2 vol.% sulfides were measured using Perkin Elmer Elan 9000
254	inductively coupled plasma mass spectrometer (ICP-MS). Whole-rock S contents
255	were measured using a gravimetric method and IR absorption with the detection limits
256	~1000 ppm.

257 Major (Mg, Al, Cr, Fe and Si) and trace (Ti, Mn, Zn and Ni) element contents of the chromite enclosed in olivine have been measured using wavelength-dispersive 258 X-ray emission spectrometry at 15 kV and 20 nA with beam diameter of 5 µm using 259 EPMA-1600 electron probe microanalyzer (Shimadzu Corporation, Japan) at Institute 260 of Geochemistry, Chinese Academy of Sciences. SPI (STRUCTURE PROBE Inc., 261 Canada) mineral standards were used for calibration. Contents of FeO and Fe₂O₃ of 262 263 the chromite were calculated from the microprobe analyses assuming a stoichiometric spinel structure (Barnes 1998). Major oxide contents of the olivine hosting the 264 chromite were analyzed as well to test for compositional correlations between olivine 265 and chromite. The analytical conditions for major elements of the olivine were 15 kV, 266 20 nA beam current, 5 µm beam size, and peak-counting time of 20 s. Minor elements, 267 such as Ni, Cr, Ca and Al, in the olivine was analyzed using a beam current of 100 nA 268 and 60 seconds. 269

LA-ICP-MS has lower limits of detection than EPMA and thus can analyze a more comprehensive suite of elements present in chromite (Ga, Ti, Ni, Zn, Co, Mn, V and Sc) (Dare et al. 2009; Pagé and Barnes 2009; González-Jiménez et al. 2011, 2013, 2014; Aldanmaz 2012). Trace element compositions of the chromite grains with

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274	diameter larger than 40 μ m were determined in situ by LA-ICP-MS at Institute of
275	Geochemistry, Chinese Academy of Sciences and CSIRO in Perth, Australia.
276	At Institute of Geochemistry, Chinese Academy of Sciences Agilent, the 7700x
277	quadrupole ICP-MS is coupled to a GeoLasPro 193 nm laser ablation system. The
278	operating conditions and procedures were described in detail by Pagé and Barnes
279	(2009). The plasma conditions were optimized by NIST 610 before analyzing to
280	obtain highest counts with oxide production (248 ThO/ 232 Th) remaining below 0.4%.
281	For this study, analysis of the chromite cores focused on the following masses: ⁴⁵ Sc,
282	⁴⁷ Ti, ⁵¹ V, ⁵⁵ Mn, ⁵⁹ Co, ⁶⁰ Ni, ⁶⁶ Zn, and ⁷¹ Ga. The isotopes ²⁹ Si and ⁴² Ca were monitored
283	to check for the presence of silicate inclusions. The analyses were conducted using a
284	40 μm beam diameter, 6 Hz frequency, and 0.032 - 0.105 mJ/pulse power, during 90
285	second analysis (30 s laser off for blank measurement and 60 s laser on for analysis of
286	chromite). Multiple-reference materials (GSD-1G, BCR-2G and BHVO-2G) were
287	used to calibrate element contents (Dare et al., 2012). A natural magnetite (BC-28)
288	from the Bushveld Complex was adopted to evaluate the accurate and precise of the
289	data ($<5\%$ for major elements and $<10\%$ for trace elements).

At CSIRO, the trace elements were collected using a Photonmachines, ATLex 300si-x Excite 193nm Excimer ArF laser connected to an Agilent 7700 ICP-MS located in the Australian Resources Research Centre in Perth. The plasma conditions were optimized daily, to obtain highest counts with oxide production (248 ThO/ 232 Th) remaining below 0.4%. The laser was masked with a 40 µm circular spot with a repetition rate of 9 Hz with 30 seconds of chromite and 30 second of blank

296	measurements in each analysis. The following isotopes were measured in this study:
297	Mg ²⁵ , Al ²⁷ , Si ²⁹ , P ³¹ , K ³⁹ , Ca ⁴³ , Sc ⁴⁵ , Ti ⁴⁹ , V ⁵¹ , Cr ⁵² , Mn ⁵⁵ , Fe ⁵⁷ , Co ⁵⁹ , Ni ⁶⁰ , Ni ⁶¹ , Ni ⁶² ,
298	Cu ⁶³ , Cu ⁶⁵ , Zn ⁶⁶ , Zn ⁶⁸ , Ga ⁷¹ , Ge ⁷² , Rb ⁸⁵ , Sr ⁸⁷ , Y ⁸⁹ , Zr ⁹⁰ , Zr ⁹² , Nb ⁹³ , Mo ⁹⁵ , Ru ⁹⁹ , Ru ¹⁰¹
299	Ru ¹⁰² , Rh ¹⁰³ , Pd ¹⁰⁵ , Pd ¹⁰⁸ , Ag ¹⁰⁹ , Cd ¹¹¹ , In ¹¹⁵ , Sn ¹¹⁸ , Sb ¹²¹ , Te ¹²⁵ , Ba ¹³⁷ , Hf ¹⁷⁸ , Ta ¹⁸¹
300	W ¹⁸² , Os ¹⁸⁹ , Ir ¹⁹³ , Pt ¹⁹⁵ , Tl ²⁰⁵ , Pb ²⁰⁶ , Bi ²⁰⁹ , Th ²³² , U ²³⁸ . The primary reference material
301	was Université du Québec à Chicoutimi (UQAC) nickel-copper-iron sulfide pressed
302	powder FeS-1 for the platinum group elements and USGS basaltic glass GSD-2g for
303	all other elements with Fe ⁵⁷ used as the internal standard value. The pure Co, Ni and
304	Cu metals were measured to correct for metal + argide interferences on the
305	platinum-group element (PGE). The quality of the data was examined using USGS
306	MASS-1 (iron sulfide) as well as the UQAC BC-28 magnetite (Barnes et. al 2004)
307	and an in-house chromite from Coobina. The values for all the secondary standards
308	were illustrated in supplementary Fig. s5. However, PGEs in the chromite are lower
309	than detection limits. This is consistent with extremely low PGE contents in the
310	sulfide ores reported by Song et al. (2016). Data was reduced using Iolite software
311	(Paton et al. 2011).

For both methods, pure He was used as the carrier gas to pass the ablation point within the cell, and mixed with Ar for improving efficiency for aerosol transport. The ablation points are focused on the cores of the chromite grains and kept away from fractures in the chromite crystals to avoid possible effects of hydrothermal alteration (Fig. 3b, d). The instrument was calibrated against the NIST 610 silicate glass (National Institute Standards and Technology, Gaithersburg, USA) (Norman et al.

318	1996). Iron values obtained by electron microprobe were used as the internal standard.
319	Therefore, the EPMA point in the large enough chromite grain was overlapped by
320	LA-ICPMS measurement (Fig. 3b, d). Supplemental Figure s1 shows that the
321	LA-ICPMS data of most trace elements in the chromites are consistent with the
322	EPMA data, with the exception of Al and Ti, although the LA-ICPMS data are slightly
323	lower in Cr and higher in Mn than the EPMA data. The differences of Al content data
324	of the chromite between EPMA and LA-ICPMS probably are due to compositional
325	zonation of some grains and much larger LA-ICPMS beam sizes. The high Ti contents
326	of a few of LA-ICPMS spots relative to EPMA data probably are due to
327	micro-exsolution of Ti-rich phases that are included in the LA-ICPMS spots. Thus,
328	the EPMA data of Cr, Al and Ti and LA-ICPMS data of Mn, Sc, V, Co, Ga of the
329	chromite are used in the discussions below.

5. Results

331 5.1 Whole-rock compositions

Whole rock analyses of the ultramafic rocks containing sulfides were recalculated 332 on a volatile- and sulfide-free basis. The harzburgite from drill hole ZK1905 is 333 characterized by high MgO (34.2-43.4 wt%), relatively low Al₂O₃ and Cr (1.6-5.7 wt%) 334 and 330-1040 ppm, respectively), MgO decreases along with increasing of Al₂O₃ 335 upwards gradually (supplementary Table s1, Figs. 4 a-c). In drill hole ZK2309, the 336 orthopyroxenite beneath the gabbronorite appears lower in MgO (21.1-28.4 wt%) and 337 slightly higher in Al₂O₃ (3.2-8.1 wt%) and intermediate in Cr (1200-1900 ppm) 338 relative to the olivine orthopyroxenite (25.8-40.6 wt% MgO, 2.0-6.8 wt% Al₂O₃ and 339

340	1400-3800 ppm Cr, respectively) above the gabbronorite (Figs. 4 h-j). The olivine
341	orthopyroxenites display gradual increase in Al ₂ O ₃ and decrease in MgO toward the
342	contact with the gabbronorite (Figs. 4 h-j). Such compositional variations imply that
343	the gabbronorite was assimilated by the new pulse of magma during the formation of
344	the olivine orthopyroxenite (Song et al. 2016). The evidently higher Cr (>1400 ppm
345	and up to 3800 ppm) in the orthopyroxenites than in the harzburgite (Cr <1040 ppm)
346	(Figs. 5c, j) reflects that Cr is dominantly contained in the orthopyroxene, which
347	commonly has 1000-4000 ppm Cr (up to 6000 ppm) (Yi 2016; Wang et al. 2019).
348	The binary plots of MgO versus FeO, Al ₂ O ₃ and CaO of the rocks normalized to
349	100% silicate indicate that these rocks are olivine and pyroxene mesocumulates with
350	minor chromite (Fig. 5). This is consistent with our observations described above.

to variation in proportions of olivine and orthopyroxene and clinopyroxene within therock units.

However, the harzburgite and olivine orthopyroxenite have overlapping chemistry due

354 *5.2 Olivine:*

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Olivine crystals of the Xiarihamu intrusion have a large variation in forsterite percentage and Ni content (Fo=82.4-88.7 mol. % and Ni=1200-2560 ppm, respectively) and very low CaO (less than 0.2 wt%) and moderate MnO contents (0.1 - 0.2 wt%) (Table s2). The recent study of Li et al. (2015) indicated that the Xiarihamu olivine may have Fo up to 89.9 and Ni contents up to 4600 ppm. In the diagram of Ni versus Fo, most of the Xiarihamu olivine are plotted in the field of the Stillwater Complex (America) and the Bushveld Complex (South Africa) (Teigler and

362	Eales 1996; Raedeke 1982) (Fig. 6). The Ni concentrations of the Xiarihamu olivine
363	are higher than those of the olivine from the Nova intrusion and slightly lower than
364	those of the olivine from the Santa Rita zone within the Fazenda Mirabela intrusion,
365	which are magmatic sulfide-associated intrusions in a Mesoproterozoic orogenic belt,
366	Western Australia and a Paleoproterozoic orogenic belt, Brazil, respectively (Barnes et
367	al. 2011; Maier et al. 2016). Our data show that Fo of the olivine decrease from
368	83.9-88.7 in the harzburgite to 82.4-87.0 and 82.9-85.1 in the olivine orthopyroxenite
369	and the orthopyroxenite, respectively (Table s2). As shown in Figure 4d, Fo of the
370	olivine in the harzburgite from the drill hole ZK1903 decreases regularly upward,
371	indicating normal fractionation.

In contrast, Fo of the olivine in the orthopyroxenites in the drill hole ZK2309 increases upwards accompanied by decrease of whole-rock Al₂O₃ content (Figs. 4k, i). Similar phenomena have also been observed by Li et al. (2015). Hence, the change in Fo content over this interval can be confidently interpreted as indicating progressive influx of hotter magma with time. This is consistent with the observation that the chromites of the orthopyroxenites from the drill hole ZK2309 show a slight increase in Cr# upwards (Fig. 4l).

- 379 *5.3 Chromite*
- 380 *5.3.1 Major oxides:*

EPMA data listed in Table s2 show that the Xiarihamu chromites have moderate Mg# (Mg*100/(Mg+Fe²⁺) = 40.1 to 65.5, atomic ratio) and low Cr# (Cr*100/(Cr+Al) = 18.6 to 46.7, atomic ratio) relative to chromites from boninites (Cr# > than 70,

384	Barnes and Roeder, 2001) (Fig. 7a). They are also characterized by extremely low
385	TiO_2 (0.06-0.78 wt%, with exception of some chromites in the olivine orthopyroxenite)
386	(Fig. 7c) compared with those from the mafic-ultramafic intrusions hosting world
387	class Ni-Cu-(PGE) sulfide deposits genetically linked with mantle plume and
388	continental rifting, such as Jinchuan, Pechenga, Noril'sk and Talnakh (Abzalov 1998;
389	Barnes and Tang 1999; Barnes and Kunilov 2000). Most of the chromites in the
390	Jinchuan and Noril'sk and Talnakh intrusions contain more than 0.5 wt% TiO ₂ ,
391	ranging up to 12 wt% (Barnes and Kunilov 2000). The chromites from both the
392	harzburgite and orthopyroxenites have similar Cr# (17.8-46.7 and 20.1-41.9,
393	respectively), and the chromites in the harzburgite have slightly higher Mg#, lower
394	MnO and V_2O_5 contents than those in the orthopyroxenites and similar TiO ₂ contents
395	at comparable Cr# (Fig. 7). In contrast, the chromites in the orthopyroxenite are
396	characterized by low Cr# (14.5-41.1) and high ZnO (0.25-1.55 wt%) relative to those
397	in the harzburgite and olivine orthopyroxenite (Fig. 7d). Decrease of Cr# of the
398	chromites in these rocks is coupled by decreases of MnO, TiO_2 and $\mathrm{V_2O_5}$ and
399	increases of Mg# and ZnO (Fig. 7).

It is noteworthy that cores of the chromites in both the harzburgite and orthopyroxenites are slightly higher in Cr# and lower in Mg# due to increase of Al_2O_3 and MgO and decrease of Cr_2O_3 from cores to rims (Table s2, Figs. 4, 7a). The differences in Cr# between the cores and rims of the chromites are mostly less than 4.0 and but range up to 15.0 for a few grains (Table s2, Fig. 7a). The increase of Mg# and decrease of Cr# as well as enrichment of Al toward to the rims of the chromite

grains (Fig. 4) are coupled by slightly reduction of MnO and V_2O_5 (Table s2, Figs. 7b
d). The core/rim texture of the Xiarihamu chromites demonstrate an incomplete
chemical diffusion and exchange between the chromites and hosting olivine due to
409 relatively fast cooling. In contrast, in large layered intrusions, chromites are

411 cooling and sufficient re-equilibration (e.g., Wilson 1982; Roeder and Campbell 1985;

compositionally homogeneous and show no signs of internal zoning due to slow

412 Vogt et al. 2015).

410

Plotting of the Cr-Al-Fe³⁺ triangular diagram shows that the Xiarihamu chromites plot in a restricted field and are very low in Fe³⁺ relative to those of the Noril'sk and Jinchuan intrusions, in which Fe^{3+} increases during equilibration with evolving trapped liquid (Fig. 8) (Barnes and Kunilov 2000). In the Xiarihamu intrusion, the chromites are lower in Fe³⁺ and Al and relatively higher in Cr in the harzburgite than those in the orthopyroxenite.

419 *5.3.2 Trace element:*

LA-ICPMS measurements (Table s3) indicate that decreasing Cr goes along with 420 decreasing V, Mn, Sc and Ga and increasing Zn in the chromite (Fig. 9). Thus, the 421 chromites from the orthopyroxenite are lower in V, Mn and Sc and slightly higher in 422 Zn than those from the harzburgite (Fig. 9). In both harzburgite and olivine 423 orthopyroxenites, the chromites hosted in olivine are evidently higher in Cr, V and Ga 424 than those hosted in orthopyroxene or interstitial chromites, and similar in Mn and Sc 425 (Fig. 9). Particularly, the chromites hosted in the olivine with Fo > 87 in the 426 harzburgite not only have relatively constant Cr#, Mg# and oxides (Figs. 4, 7), but 427

also restricted ranges of Cr, Sc, Ga, V and Mn concentrations, indicated by the green
circles in Figure 9. Whereas, the chromites hosted in the olivines with Fo < 87 in both
the harzburgite and orthopyroxenites have much larger ranges of oxide and trace
element concentrations (Figs. 4, 7, 9).

Relative to the chromite from MORB, all of the Xiarihamu chromites are 432 433 distinctly enriched in Ga, Zn, Co and Mn and strongly depleted in Sc (Fig. 10). This is evidently different from the chromite in boninite, which is characterized by 434 conspicuous depletion of Al, Ga and Ti and enrichment of Sc relative to the MORB 435 chromite (Pagé and Barnes 2009), as well as having remarkably high Cr# (Crawford 436 et al. 1989). Due to the limitations of analyzing small grains, it is not clear if there are 437 differences in trace element concentrations between the cores and rims of the 438 439 Xirihamu chromites.

440

5.4 Correlation between olivine and chromite

Compositional variations of the chromite are well correlated with Fo of the 441 hosting olivine. Firstly, as Fo of the olivine decrease, Cr₂O₃ of the chromites tend to 442 decrease slightly and ZnO to increase (Fig. 11). In the harzburgite, it is noteworthy 443 that the chromites hosted in the olivine with Fo > 87, at depths of 333 to 369 m (Fig. 444 445 4), not only have restricted compositions (marked by green circles), but also show positive correlation between Fo of the olivine and Mg# of the chromite (Fig. 11). In 446 contrast, the chromites displaying large variations in oxides and Mg# value are hosted 447 in olivine with Fo < 87 in both the harzburgite and orthopyroxenites (Figs. 4, 11). The 448 highest TiO₂ contents of a few of chromites in the olivine orthopyroxenites are 449

450	probably associated with tiny exsolution lamellae of ilmenite (Figs. 3f, 11e). The large
451	compositional variations of the chromites are roughly correlated with elevation of
452	whole-rock Al ₂ O ₃ (Fig. 4), implying that the whole-rock Al ₂ O ₃ content can be taken as
453	an index of increasing trapped liquid content.

454 6. Discussion

455 There are differences on scales, grades of Ni, Cu and PGE, formation mechanism of the magmatic sulfide deposits formed in different tectonic settings. Many world 456 class magmatic sulfide deposits were formed in large igneous provinces due to 457 anomalously high magma fluxes and high degree partial melting, whereas the deposits 458 discovered in orogenic belts are commonly smaller and have lower metal grades (e.g., 459 Song et al. 2013; Lesher 2017, 2019; Lu et al. 2019). Compositions of primary 460 461 chromite, as one of the earliest crystallized mineral from mantle derived magma, are potentially very useful for estimation of the nature of the mantle source and tectonic 462 setting of the intrusion (Barnes and Roeder 2001; Kamenetsky et al. 2001; Ahmed et 463 al. 2005; González Jiménez et al. 2014). However, the composition of chromite may 464 vary due to postcumulus modification, such as equilibrium with hosting minerals and 465 intercumulus melt during slow solidification (Roeder and Campbell 1985; Barnes 466 1986; Barnes and Roeder 2001). Although chromite is relatively refractory and 467 resistant to alteration compared with high temperature igneous minerals such as 468 olivine and pyroxene, hydrothermal alteration could still result in compositional 469 changes of chromite (Colás et al. 2014). Thus, it is very important to identify the 470 compositional variations of the chromite resulted from reaction with hosting mineral 471

and trapped liquid as well as alteration before discussion on nature of primary magmafrom which the chromite crystallized and their mantle source.

In the Xiarihamu intrusion, the common observation of chromite enclosed in 474 olivine Fo with higher than 87 implies that the chromite was a high-temperature 475 liquidus phase (Table s2, Fig. 3), and hence that its composition is likely to be a useful 476 477 indicator of magma provenance. However, composition of the chromite in intrusive rocks may be modified both by interaction with trapped intercumulus liquid during 478 solidification and by subsolidus re-equilibration with hosting minerals (e.g., Roeder 479 and Campbell 1985; Scowen et al. 1991). Thus, it is first necessary to estimate effects 480 of alteration, postcumulus trapped liquid reaction and subsolidus equilibration, to 481 allow us to recognize the most primitive chemical features of the chromite before 482 483 discussion of the nature of primary magma and mantle source of the Xiarihamu intrusion. 484

485 **6.1 Limited alteration of the chromite**

Studies of hydrothermal alteration and metamorphism of chromite in ophiolites 486 and komatiites indicate that Al and Mg can be released from the chromite to form 487 secondary minerals, like chlorite (Colás et al. 2014). Metamorphism can result in an 488 optical rim enriched in FeO and Cr₂O₃ and depleted in MgO and Al₂O₃ surrounding a 489 primary core, which grades outwards from chromite strongly enriched in Fe³⁺ (ferrian 490 chromite) to ferritchromit or magnetite (Barnes 2000; González Jiménez et al. 2014; 491 Merlini et al. 2009; Mukherjee et al. 2010; Gervilla et al. 2012; Grieco and Merlini 492 2012). Therefore, compared with the primary core of chromite crystal, the altered or 493

494	metamorphosed rim is relatively high in Cr# and low in Mg# due to loss of Al ₂ O ₃ and
495	MgO to chlorite (Colás et al. 2014). However, this is not the case for the chromites in
496	the Xiarihamu intrusion. The rims of the chromite grains have lower Cr# and higher
497	Mg# values than the cores and the rims of most chromite grains have higher Al_2O_3
498	and MgO contents than the cores (Figs. 4, 7, Table s2). Thus, the decrease of Cr# and
499	increase of Mg# from cores to rims for the Xiarihamu chromite grains is interpreted as
500	the result of magmatic processes rather than hydrothermal alteration. Consistently,
501	small variation in ZnO and MnO contents between the cores and rims of the chromite
502	grains also indicate that alteration effects are only minor (Table s2).

503 **6.2 Effects of intercumulus liquid and host mineral on chromite compositions**

According to textural relationships in the olivine-rich Xiarihamu ultramafic rocks 504 505 (Fig. 3), it is clear that both chromite and olivine crystallized as cumulus phases followed by peritectic reaction of olivine with the magma to form orthopyroxene. 506 Crystallization of intercumulus liquid is indicated by local occurrences of the 507 interstitial fine grained assemblage of clinopyroxene, hornblende and plagioclase 508 (Figs. 3g, h). Furthermore, chromite is minor accessory phase in the rocks and is 509 therefore susceptible to having its composition modified by equilibration with 510 intercumulus liquid and host silicates (e.g., Cameron 1975; Clark 1978; Wilson 1982; 511 Roeder and Campbell 1985; Scowen et al. 1991; Evans 2018). Thus, it is necessary to 512 evaluate the effect of fractionation and reaction with intercumulus liquid on the 513 compositions of chromite, and also the effects of subsolidus reactions with enclosing 514 silicates due to changing equilibrium constants for exchange reactions, before 515

516 discussion of petrogenetic significance of the Xiarihamu chromite.

517 6.2.1 Effect on major elements of the chromite

518 The Mg/Fe ratios of coexisting chromite and olivine are governed by the 519 exchange equilibrium:

520
$$FeO_{chromite} + MgO_{olivine} = FeO_{olivine} + MgO_{chromite}$$
 (1)

521 The equilibrium constant for this reaction changes with temperature to favor the increasing partition of FeO into chromite and MgO into olivine with falling 522 temperature (Irvine 1967; Roeder et al. 1979). Hence this exchange reaction is driven 523 to the left with falling temperature both during supra-solidus crystallization and 524 during sub-solidus equilibration. This process resulted in decrease of MgO and 525 increase of FeO towards the rims of chromites enclosed in olivine in the Kabanga 526 intrusion, Tanzania (Evans 2018). However, this is not the case for the Xiarihamu 527 chromites, whose rims are commonly lower in Cr# and higher in Mg# than cores 528 (Table s2, Figs. 4, 7a). 529

During solidification of the trapped liquid, the chromite is also capable of 530 exchanging Cr and Al with the trapped liquid or intercumulus pyroxene. Owing to the 531 strong non-ideality of the spinel solid solution series (Sack and Ghiorso, 1991), the 532 533 effect of decreasing Mg/Fe in the chromite (driven by equilibration with silicates as the temperature falls) is to drive the Cr/Al exchange between chromite and melt in the 534 direction of increasing Cr/Al in the chromite. As in the case of the well-documented 535 "Rum Trend" (Henderson 1975), reaction between chromite and intercumulate melt 536 can drive chromite compositions along a trend of increasing Cr/Al with decreasing 537

538	Mg/Fe, the opposite relationship to that expected during simple crystallization (Allan
539	et al. 1989; Naldrett et al. 2009). The extent of this trend will be limited by
540	simultaneous equilibration with Cr and Al bearing pyroxene. Consequently, trapped
541	liquid reaction trends for chromite can be complex and unpredictable. As shown by
542	Roeder and Campbell (1985) and Scowen et al. (1991) trapped liquid reaction can still
543	take place to some extent even in the case of chromite enclosed within cumulus
544	olivine, as a result of diffusion through micro fractures or through the olivine lattice,
545	although the effect is smaller for enclosed chromites than for those directly exposed to
546	the intercumulus liquid (e.g., Barnes and Tang, 1999).

With this complexity in mind, we consider the chromite composition trends in 547 Figures 7 and 11. In each individual sample, there is a strong negative correlation 548 549 between Mg# and Cr# (Fig. 7a), which is the expected consequence of reaction with 550 trapped liquid for small modal abundance of chromite. However, the trends between 551 the harzburgite and the orthopyroxenites are offset, with the harzburgite sample having higher Cr# in comparable Mg# (Fig. 7a). This is consistent with the 552 harzburgite having crystallized from a more primitive magma with a higher Cr/Al 553 (Roeder and Reynolds 1991). However, in both orthopyroxenites, the reduction of 554 555 Cr₂O₃ contents of the chromite along with decrease of Fo of the olivine can be 556 attributed to crystallization of the orthopyroxene (Fig. 11a). The lack of consistent correlation between the Mg# of the chromite and Fo of the enclosing olivine (Fig. 11d) 557 reflects the wide variability in the Cr#, which substantially effects the K_d for the 558 Fe-Mg exchange reaction at the scale of individual grains as described above. The 559

variability in Cr# is most likely inherited from original liquidus compositions,
reflecting the composition of the parent liquid, possibly combined with disequilibrium
between chromite and melt at the time it became trapped in olivine.

In conclusion, the compositional relationship between the chromite and the hosting olivine in the harzburgites and orthopyroxenites is primarily due to liquidus composition variations, with only minor modification by reaction with trapped intercumulus liquid. The within-sample variability in Cr# is most likely a primary effect, possibly related to mechanical accumulation of chromite and olivine grains from incompletely homogenized mixed or contaminated parent magmas.

569 6.2.2 Effect on minor and trace elements in chromite

Although interstitial minerals including plagioclase and hornblende are present in 570 proportions less than 5.0 vol.% in the harzburgite, compositional exchange of 571 chromite with intercumulus liquid is not negligible for the minor and trace elements. 572 The effects are largest where chromite competes with these minerals for trace 573 components. Furthermore, subsolidus exchange of more rapidly diffusing divalent 574 ions (Zn, Mn and Ni) between chromite and olivine or orthopyroxene, by equivalent 575 exchange reactions to equation (1) above, also play a role in controlling the 576 abundances of these elements in chromite (Barnes 1998). 577

Titanium is moderately incompatible in chromite and anhydrous silicate minerals and compatible in hornblende ($D_{Ti}^{Hb/melt}$ is up to 8, Hb = hornblende) (Table s4). The lack of enrichment of TiO₂ in chromite with decreasing Fo of olivine (Fig. 11e) may in part be explained by the reaction of chromite with the interstitial hornblende (Fig.

582	3g). Similarly, Sc is moderately compatible in pyroxenes and hornblende $(D_{Sc}^{Py/melt})$
583	=0.81-2.28, $D_{Sc}^{Hb/melt}$ =2.18, Matsui et al. 1977) and incompatible to chromite (Table
584	s4). Therefore, large variation of Sc content of the chromite could reflect equilibration
585	with hornblende and clinopyroxene, either during crystallization of in the
586	intercumulus liquid or by subsequent subsolidus reaction (Fig. 9c). However, given
587	that the proportion of interstitial hornblende is very small, we conclude that the
588	extensive negative Sc anomalies relative to MORB chromite (Fig. 10) of the
589	chromites are unlikely to be explicable by this effect, particularly for those chromites
590	that are enclosed within the olivine with $Fo > 87$. No apatite was observed in any of
591	the sections studied, so redistribution of Sc or any other trace elements between
592	apatite and chromite can be disregarded.

593 Zinc is moderately compatible in chromite, moderately compatible in in olivine, pyroxene and hornblende and incompatible in plagioclase (Table s4). Zinc in chromite 594 commonly shows a moderate negative correlation with Fo in associated olivine (Fig. 595 11d), primarily due to an exchange reaction between Zn and Fe in chromite and 596 olivine and intercumulate melt; Zn partitions into chromite as temperature falls 597 (Barnes 1998). Nickel is also subject to temperature-dependent divalent ion exchange 598 with Fe in olivine, complicated by the tendency of Ni to partition more strongly into 599 ferric-iron enriched spinel (Barnes 1998). Figure s2 shows that Ni, Mn and V variably 600 increase with ferric iron while Co remains constant, although the Xiarihamu chromite 601 has a limited variation in ferric iron (Fig. 8). 602

603 Fractional crystallization and local trapped liquid equilibration leads to variable

enrichments in Ga in the chromite owing to the overall moderate incompatibility of
Ga in the crystallizing silicate assemblage (Table s4). This is the case of the chromite
in the Xiarihamu harzburgite and orthopyroxenites shown in Figure 9e.

Under normal magmatic redox conditions, V and Mn are moderately and weakly compatible in chromite as well as clinopyroxene and hornblende, respectively (Table 509 s4). Thus, although crystallization of pyroxene and hornblende reduces the concentrations of V and Mn in the intercumulus liquid, the trends of V and Mn against Cr/(Cr+Al) of the chromite from the orthopyroxenites are only weakly affected by reaction with the silicates (Figs. 9a and b).

In conclusion, the divalent elements Ni, Mn and Zn are likely to be slightly 613 controlled by subsolidus equilibration with host olivine, while the non-divalent 614 615 elements Sc, Ti, V and Ga in the chromite may moderately modified by crystallization of hornblende and clinopyroxene during trapped liquid crystallization. Scandium, Ti 616 and V became relatively depleted in chromite compared with its liquidus composition 617 while Ga became enriched. However, the relatively small variation of Sc and Ga in 618 most of the chromite enclosed by the olivine with Fo > 87 in the harzburgite indicates 619 that they have undergone less modification during reaction with the intercumulus 620 liquid than those enclosed by the olivine with Fo < 87 (Fig. 9). Hence, the 621 compositions of the chromites hosted in the olivine with Fo > 87 in the harzburgites 622 can be taken as good indications of the original liquidus chromite composition and 623 624 used to give information about magma compositions and sources.

625 **6.3 Effect of crustal contamination**

626	Recent studies confirmed the contribution of crustal contamination to sulfide
627	segregation in the Xiarihamu deposit (Song et al. 2016). According to Sr-Nd isotope
628	data of the sulfide mineralized samples of the Xiarihamu deposit, Zhang et al. (2017)
629	estimated a large range of crustal contamination (5-30%) and a moderate enriched
630	primary magma with \mathcal{E}_{Nd} close to -1.8 and $({}^{87}Sr/{}^{86}Sr)_i$ close to 0.705. New data by Yi
631	(2016) indicated that the harzburgites containing olivine with Fo values up to 88.8
632	have positive \mathcal{E}_{Nd} values of +1.6-+2.1, whereas the sulfide mineralized rocks have \mathcal{E}_{Nd}
633	values ranging from -0.4 to -4.0 (Fig. s3). Thus, it is reasonable to infer that degrees
634	of crustal contamination are highly variable between different pulses of magma.
635	Crustal contamination commonly results in increasing SiO ₂ in the mafic magma,
636	while associated crystallization leads to decrease of Fo values of the crystallizing
637	olivine. The high Fo values of the olivine (up to 88.8, Table s2) in the harzburgite
638	from the ZK1903 suggest that the parental magma to this rock experienced the least
639	crustal contamination and fractionation. Thus, it is concluded that the chromites
640	enclosed in the olivines of the harzburgite are the most primitive chromites in the
641	Xiarihamu intrusion. In particular, those chromites in the harzburgite with lowest Cr#
642	(bearing in mind that Cr# increases with trapped liquid reaction for reasons given
643	above) and low Ti are the best approaches to primary liquidus chromite in the most
644	primitive magmas.

645 **6.4 Compositional features of the most primary chromite**

646 The original Fo values of the chromite-hosting olivine in the harzburgite should647 be ~89-91, allowing for a small trapped liquid shift if proportion of the trapped liquid

was only ~10% (Fig. s4) (Barnes 1986). Thus, it is again reasonable to conclude that
the chromite in the harzburgite was crystallized from the most primitive magma in the
Xiarihamu intrusion.

The most primitive chromite hosted in the olivine with Fo > 87 is characterized 651 by relatively low Cr# (=37-42) and contains 0.3-0.4 wt% TiO₂, 0.3-0.5 wt% ZnO, 652 653 1500-1770 ppm Mn, 860-1000 ppm V, 630-1500 ppm Ni, 90-150 ppm Ga and 0.5-1.8 ppm Sc (Figs. 7 and 9) and show enrichments in Ga and Zn and strongly depleted in 654 Sc relative to the chromite in MORB (Fig. 10) (Pagé and Barnes 2009). These 655 features are distinctly different from those of typical boninite chromite. The calculated 656 parental magma of the Xiarihamu intrusive rocks according to the chromite 657 composition and partition coefficients (Pagé and Barnes 2009) is characterized by 658 659 enrichments of Zn, Ga and Co and depletions of Sc relative to MORB (Fig. 12). This cannot be explained by crustal contamination of boninitic magma or MORB. 660

661 **6**

6.5 Nature of the mantle source

Kamentsky et al. (2001) demonstrated that Al₂O₃ and TiO₂ contents of Cr-spinel 662 in volcanic rocks are well correlated with melt composition. In the binary diagrams of 663 TiO₂ versus Al₂O₃ and Ti versus Cr# of Cr-spinel proposed by Kamentsky et al. 664 665 (2001), the most primitive chromites hosted in the olivine with Fo > 87 in the harzburgite plots within the MORB area (Fig. 13). However, the enrichments of Ga, 666 Ti and Zn and depletion of Sc of the chromite relative to the chromite in MORB 667 clearly show the difference between the Xiarihamu primary magma and MORB (Fig. 668 12). Previous studies have illustrated that Sc is incompatible in spinel 669

670	$(D_{Sc}^{Spinel/melt}=0.18-0.48)$ and compatible in garnet and pyroxenes $(D_{Sc}^{Garnet/melt}=5.98)$,
671	$D_{Sc}^{Opx/melt} = 1.01 - 1.64$, $D_{Sc}^{Cpx/melt} = 0.81 - 2.28$); whereas Ga is compatible in spinel
672	$(D_{Ga}^{Spinel/melt}=1.83\sim3.28)$ and incompatible in garnet and pyroxenes $(D_{Ga}^{Garnet/melt}=0.39)$
673	$D_{Sa}^{Opx/melt}=0.18\sim0.23$, $D_{Ga}^{Cpx/melt}=0.3\sim1.06$, Table s4) (Horn et al. 1994; Laubier et al.
674	2014). Thus, the Ga enrichment and Sc depletion of the parental magma equilibrium
675	with the most primitive chromite relative to MORB indicate that spinel is absent in
676	the mantle source during partial melting and pyroxenes and garnet should be present
677	as residual solid phases (Fig. 12). Although primitive mantle normalized trace element
678	patterns of the Xiarihamu rocks show moderate enrichments in LREE (light rare-earth
679	elements) relative to HREE (heavy REE) (Li et al. 2015), it can be speculated that the
680	parental magmas were evidently more enriched in LREE relative to HREE. This is
681	because HREE are moderately incompatible and LREE are highly incompatible in the
682	orthopyroxene (Bédard 1994), which is the dominant mineral in the Xiarihamu
683	cumulate rocks and are depleted in LREE and moderately enriched in HREE (Wang et
684	al. 2019).

685

7. Implications for magma generation of the Xiarihamu intrusion

Recent geodynamic numerical modeling shows that subducting oceanic slab break-off from continental crust due to their different densities may occur at ~100 km depth soon after continent-continent collision (Kohanpour et al. 2017; Davies and von Blanckenburg 1995). Arc volcanic rocks in pre-collision are thought to be derived by partial melting of a combination of the subducting oceanic crust and the metasomatised mantle wedge, whereas following collision and slab delamination

692	basaltic	magmatism	is	attributable	to	upwelling	and	decompression	melting	of
693	asthenos	sphere throug	h th	e resulting "s	slab	window".				

Unusual high Ni/Cu ratios (~4-18) of the Xiarihamu sulfide ores led Song et al. 694 (2016) to propose that the associated basaltic magma was generated by melting of a 695 pyroxenite mantle triggered by upwelling of asthenosphere from a slab-window in 696 697 Early Devonian. This model is supported by studies of the eclogites along the east and west sections of the North East Kunlun Fault. Rims of the zircons separated from the 698 eclogites yielded concordia U-Pb age of ~428 Ma (Song et al. 2018). Trace elements 699 700 of the eclogites have the geochemical characteristics of normal or enriched mid-ocean ridge basalts. Calculations based on compositions of garnet demonstrated that the 701 eclogites experienced ultrahigh-pressure metamorphism at 29-30 kbar and 610-675° 702

C (Song et al. 2018), whereas a few zircon grains gave a younger mean age of $410.5 \pm$

2.0 Ma, which may represent the amphibolite facies retrograde metamorphism of the
eclogites. Song et al. (2018) proposed that the eclogites were exhumed from depths of
100–120 km, which is consistent with garnet being a residual phase in the mantle
source of the Xiarihamu magma, revealed from chromite composition in this study.
The exhumation of the eclogites implies closure of the back-arc basin between the
Qiadam Block and the Middle East Kunlun terrane (Fig. 1b) (Dong et al. 2018a).

According to the above insights on the nature of the mantle source of the Xiarihamu intrusion, a model for generation of the magma is proposed as follows (Fig. 14). In the early Devonian, subduction slab break-off occurred at a depth of ~100 km following collision between the middle and southern East Kunlun microcontinental

714	blocks (Yang et al. 1999, 2000; Chen et al. 2002; Song et al. 2016) and formed a slab
715	window. This is evidenced by the 430-410 Ma eclogites along the Northern East
716	Kunlun Fault (Fig. 1b), which were considered as the exhumed blocks of subducted
717	oceanic crust that experienced ultrahigh-pressure metamorphism at 29-30 kbar and
718	610-675°C (Song et al. 2018). The exhumation of these eclogites indicates
719	delamination of subduction oceanic crust and upwelling of asthenosphere following
720	closure of the oceanic basin represented by the ophiolites along the Northern East
721	Kunlun Fault (Dong et al. 2018a). Melts produced due to metasomatic reaction of a
722	low degree partial melt from an eclogite, derived by metamorphism of subducted
723	oceanic slab, mixed into the asthenosphere and reacted with peridotite generating
724	pyroxenite (Sobolev et al. 2007; Song et al. 2011, 2013; Lu et al. 2019). Partial
725	melting of a pyroxenite source may occur at lower temperature and a wider range of
726	pressure than for a peridotite source (McKenzie and Bickle 1988; Hirose and
727	Kawamoto 1995; Kogiso et al. 2013). The enrichments of Zn, Mn and Co in the
728	primary magma can be attributed to minor sedimentary oxides or clays contained in
729	the subduction slab (Fig. 12).

Recent studies proposed a linkage between the formation of Paleozoic magmatic Ni-Cu sulfide deposits and upwelling of asthenosphere at the southern margin of the Central Asian Orogenic Belt (e.g. Song and Li 2009; Song et al. 2011; Tang et al. 2011; Xie et al. 2012; Gao and Zhou 2013; Yao et al., 2018). The significance of decompression melting of asthenospheric mantle for magmatic sulfide mineralization has also been discussed in previous publications (e.g., Lesher 1989; Lehser and Stone

736	1996, Lesher and Keays 2002; Maier et al. 2008; Yao et al., 2018). On the basis of
737	compositions of chromite and clinopyroxene, Song et al. (2013) inferred the
738	importance of syn- or post-collision slab break-off and upwelling and melting of the
739	asthenosphere through the slab window for the formation of the early Permian
740	mafic-ultramafic intrusions hosting the Ni-Cu sulfide deposits along the
741	Huangshan-Jingerquan belt, NW China. The above discussion again indicates that
742	partial melting of asthenosphere is very important for the formation of the giant
743	Xiarihamu deposit in the Eastern Kunlun Orogenic Belt. Increase of pyroxene in the
744	peridotite due to reaction with siliceous melt from a subduction slab can reduce the
745	partial melting temperature of the asthenosphere and result in greater melt
746	productivities in a more dynamic system, which has been shown to be a critical
747	component in making Ni-Cu-PGE deposits (Lesher et al. 1984; Keays 1995). In
748	summary, asthenosphere upwelling is very important for the formation of magmatic
749	sulfide deposits in convergent settings. The ascending asthenospheric mantle could
750	not only result in decompression melting, but also lead to heating and melting of the
751	ambient metasomatized mantle in a convergent setting. This process may produce a
752	large amount of basaltic magma associated with the formation of Ni-Cu sulfide
753	deposits. Variable degrees of crustal contamination, particularly extraction of sulfur
754	from crustal rocks, commonly played a significant role for sulfide immiscibility in the
755	ascending magma and the formation of magmatic sulfide Ni-Cu deposits (Lesher 2019;
756	Lu et al. 2019; Song 2019).

757 Acknowledgment

758	We thank Wen-Qin Zheng and Zhi-Hui Dai for high quality EPMA and
759	LA-ICPMS analytical data. This study is supported by NSFC (grant numbers
760	41630316, 41473050 and 41772067) to Song XY.; The National Key Research and
761	Development Program of China (grant number 2016YFC0600503); and a Research
762	Fund from SKLODG (grant number SKLODG-ZY125-06, 201201).

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1148

1149 **Figure Captions**

1150 Fig. 1 Major tectonic units of China (a), tectonic divisions in the northern part of Tibet Plateau (b)

- and regional geological map of East Kunlun orogenic belt (c) (modified after Song et al. 2016,
- 1152 locations of the ecologies are from Song et al. 2018)
- Fig. 2 Simplified geological map of the Xiarihamu intrusion (a), three dimension model showing
 distribution of the ultramafic phases and ore bodies of the Xiarihamu Ni-Co sulfide deposit (b),
 cross sections of exploration line L23 and L19 (c, d).
- 1156 Fig. 3 Plane-polarized microscope photos of the olivine and orthopyroxene hosting chromite (a, c
- and **e**) and backscattered electron images of the enclosed chromite in square areas (**b**, **d** and **f**).
- 1158 Cross-polarized microscope photos (g and h) show interstitial hornblende and plagioclase in the1159 harzburgite, respectively.
- Fig. 4 Chemostratigraphic columns of whole-rock, olivine and hosted chromite. Diagrams a, b, c 1160 1161 and **h**, **i**, **j** show stratigraphic variations of whole-rock MgO, Al₂O₃ and Cr contents for the drill 1162 holes ZK1903 and ZK2309, respectively. Whole-rock oxides of the samples containing sulfides were recalculated on a volatile- and sulfide-free basis. Diagrams d, e, f and g display stratigraphic 1163 variations of Fo of olivine and Cr#, Mg# and Al₂O₃ concentrations of chromite hosted in the 1164 1165 olivine, respectively, in ZK1903. Diagrams k, l, m and n are stratigraphic variations of Fo of 1166 olivine and Cr#, Mg# and Al₂O₃ concentrations of chromite hosted in the olivine, respectively, in ZK2309. It is noticeable that the chromites enclosed in the olivine with Fo > 87 in the harzburgite 1167 1168 from ZK1903 have restricted Cr#, Mg# and Al₂O₃ contents.

Fig. 5 Binary plots of MgO versus FeO (a), Al₂O₃ (b) and CaO (c) of the ultramafic rocks 1169 1170 normalized to 100% silicate. The black and dark grey solid segments show compositions of 1171 olivine and chromite data of electron microprobe listed in Table s2. The solid and dotted line 1172 circles show compositions of orthopyroxene and clinopyroxene measured by electron microprobe 1173 after Yi (2016). Asterisk and cross are calculated compositions of olivine (Ol) and orthopyroxene 1174 (Opx) according to their formulas. The calculated Opx is obviously high in En (enstatite) percentage due to neglected up to 5.0 wt% Al₂O₃ in the Opx at Xiarihamu according to electron 1175 microprobe data by Yi (2016). Abbreviations: Ol, olivine; Chr, chromite; Opx, orthopyroxene; 1176 1177 Cpx, clinopyroxene.

- Fig. 6 Binary plots of Ni versus Fo of the Xiarihamu olivine (data of Li et al. (2015) are plotted in
 shaded area). Data of global basic-ultrabasic lavas are from Sobolev et al. (2011), data of
 Bushveld Complex are from Teigler and Eales (1996), data of Stillwater Complex are from
 Raedeke (1982). The data of the Santa Rita (Brazil) and Nova (Western Australia) intrusions
 hosting Ni-Cu sulfide deposits are from Barnes et al. (2011) and Maier et al. (2016), respectively.
- Fig. 7 Binary correlations of Cr/(Cr+Al) versus Mg/(Mg+Fe²⁺) (**a**), ZnO (**b**), TiO₂ (**c**), V₂O₅ (**d**) and ZnO (**e**) of core and rim of the Xiarihamu chromite (EPMA data) enclosed in olivine in different type of rocks. The chromites enclosed in the olivine with Fo > 87 in the harzburgite from drill hole ZK1903 as shown in Figs. 4**d-g** are plotted within the green circles, showing relatively restricted compositions.

Fig. 8 Cr-Al-Fe³⁺ triangle diagram plots of the Xiarihamu chromite. The areas of the chromites
from layered intrusions (90th), the Noril'sk-Talnakh intrusions and Jinchuan intrusion are after
(Barnes and Kunilov, 2000).

Fig. 9 Binary diagrams of Cr/(Cr+Al) versus V (a), Mn (b), Sc (c), Zn (d) and Ga (e) of core of the Xiarihamu chromites. Ratios of Cr/(Cr+Al) of chromite were calculated using EPMA data, and concentration of V, Mn, Sc, Zn and Ga of chromite were LA-ICPMS data. The chromites enclosed in the olivine with Fo > 87 in the harzburgite are plotted within the green circles, showing relatively small compositional variation. Label of (in/out of olivine) means the data of the chromites enclosed in olivine or pyroxenes.

- Fig. 10 MORB chromite-normalized trace element patterns of the Xiarihamu chromite (data of Cr,
 Al and Ti are from EPMA). Data of the chromite in MORB and boninite are from Page and Barnes
 (2009).
- Fig. 11 Correlations of forsterite percentage of olivine versus Cr/(Cr+Al) (a), $Mg/(Mg+Fe^{2+})$ (b), Cr₂O₃ (c), Al₂O₃ (d), TiO₂ (e) and ZnO (f) of the enclosed chromite, showing different fractionation tendencies of the chromite in different rocks and effect of reaction with intercumulus trapped liquid. The chromites enclosed in the olivine with Fo > 87 in the harzburgite are plotted within the green circles.

Fig. 12 MORB normalized trace element patterns of the parental magmas equilibrium with the chromites enclosed in the olivine with Fo > 87 in the harzburgite. Partition coefficients of the elements between chromite and basaltic melt are from Page and Barnes (2001), trace element concentrations of MORB and crust averages are after www.petdb.org

Fig. 13 Binary plots of TiO_2 vs. Al_2O_3 (a) (after Kamenetsky et al. 2001) and Ti vs Cr# (=Cr/(Cr+Al)) (b) (Page and Barnes 2009) of the most primitive chromite enclosed in olivine with Fo > 87 from the harzburgite.

1212 Fig. 14 Model for generation of primitive magma at Xiarihamu. Northward subduction of Kunlun 1213 Ocean in early Paleozoic induced Middle Kunlun block separated from Qiadam Block and the 1214 formation of a back-arc basin (after Dong et al. 2018) (a). Subduction of the back-arc basin 1215 southward during ~460-420 Ma resulted in metasomatism of the mantle wedge and the formation 1216 of pyroxenite in some locations (b). Collision between Middle Kunlun and Qiadam blocks 1217 triggered break-off of subduction oceanic slab and ascending of asthenosphere from the slab 1218 window, as well as partial melting of both pyroxenite and asthenosphere at depth where garnet is 1219 stable (c).

1220 Supplementary materials:

Fig. s1 Correlation diagrams for selected elements in the chromite measured by EPMA andLA-ICPMS

Fig. s2 Binary plots of $100*Fe^{3+}/(Fe^{3+}+Cr+Al)$ vs. Ni (a), Mn (b), V (c), Co (d) and Zn (e) in the chromite

Fig. s3 Correlation of Nd–Sr isotopes for the Xiarihamu ultramafic rocks and the gneiss surrounding the intrusion (data from Yi 2016).

1227 Fig. s4 Correlations between fosterite percentage and concentrations of Ni (a) and Mn (b) of the

- 1228 olivine, as well as correlation of 100*Mn/Fe vs. Ni/(Mg/Fe)/1000 of the olivine (Ni in ppm)
- 1229 Fig. s5 A comparison between the measured values and the preferred/working values for the

secondary standards. The USGS FeS pressed powder MASS-1 (n=8), the UQAC magnetite BC-28

1231 (n=4) from the Bushveld Complex with working values from Barnes et al. (2004) and Dare et al.

1232 (2012) and an inhouse chromite standard from the Coobina Chromitite.

Table s1 Contents of major oxides (in wt%) and selected trace elements (in ppm) of the rocks fromthe drill holes ZK1903 and ZK2309

Table s2 Electron microprobe results (in wt%) of core and rim of chromite crystal and hosting olivine from drill holes ZK2309 and ZK1903

- 1237 Table s3 Trace element concentrations (in ppm) of core of chromite hosted in olivine from bore
- 1238 holes ZK2309 and ZK1903 by LA-ICPMS

1239 Table s4 Mineral/melt partition coefficients







Exploration line

Drill core

-124

0.2309



Olivine orthopyroxenite

Orthopyroxenite

Websterite

Massive ore

Sulfide ore NI<1 w1%

Sulfide ore Ni> 1wt%











OZK2309 Orthopyroxenite (in olivine)

•ZK2309 Olivine orthopyroxenite (in olivine)

△ZK1903 Harzburgite (in olivine)

+ ZK2309 Orthopyroxenite (in/out of olivine)

×ZK2309 Olivine orthopyroxenite (in/out of olivine)

+ ZK1903 Harzburgite (in/out of olivine)

XZK2309 Olivine orthopyroxenite (in Opx)

XZK1903 Harzburgite (in Opx)

Boninite

Fig. 10

ZK1903 Harzburgite ZK2309 Olivine orthopyroxenite ZK2309 Orthopyroxenite

ZK1903 Harzburgite (Fo > 87)

