1   *Revision 3*

2   **Grain-scale zircon Hf isotope heterogeneity inherited from sediment-**

3   **metasomatized mantle: Geochemical and Nd-Hf-Pb-O isotopic constrains on**

4   **Early Cretaceous intrusions in central Lhasa Terrane, Tibetan Plateau**

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21

22   **Abstract**

23   Clarifying the mechanism of recycling of pre-existing continental crustal materials into the

24   source of mantle-derived magma is a challenging effort that can be of great value to improve our

25   understanding of mantle processes and continental crust growth. This study presents an integrated

26   investigation of whole-rock and mineral geochemical and Nd-Hf-O-Pb isotopic data for dolerites

27   and diorites intruded in the central Lhasa Terrane of Tibetan Plateau at ca. 120 Ma (zircon U-Pb

28   ages). These intrusions have similar distributions of trace elements that are characterized by

29   depletion in Nb-Ta relative to Th, Ba and U, and moderately negative whole-rock $\varepsilon_{Nd}(t)$ (–5.0 to –

30   1.7) values. Magmatic zircon shows dramatically variable $\varepsilon_{Hf}(t)$ values (from -5.0 to +13.7 in the

31   same rock, including up to 12 epsilon unit variability in single grains). On the other hand, the

32   zircon $\delta^{18}O$ values are relatively uniform (+6.0‰ to +7.7‰). The constant $^{208}Pb/^{206}Pb$ values of

33   clinopyroxene crystallized at ca. 500–900 MPa suggest no contamination with lower continental

34   crust. The lack of covariation between Hf and O isotopes from the same grains, and the lack of

35   relationship between Hf isotopes and trace elements (e.g., Hf, Th/U and Yb/Gd) in the magmatic

36   zircons, together with the absence of ancient zircon xenocrysts, implies limited upper crustal

37   contamination. In combination with high whole-rock Th/La (>0.29) ratios, we interpret the zircon

38   Hf isotope heterogeneity as inherited from a depleted asthenospheric mantle with the addition of

39   1–4 % Hf from isotopically heterogeneous sediments. Our study therefore emphasizes the need for

40   caution when using complex Hf isotopic zonation in zircon as an argument for intracrustal

41   hybridization of two end-member magmas derived from distinct reservoirs. In addition, the high

42   Zr/Y ratios and no negative Zr-Hf anomalies of the Aruo intrusions imply a high surface

43   temperature of the downgoing slab that was able to fully dissolve zircons in the subducted

44   sediments. This requires a special geodynamic condition that was most likely related to the

45   steepening of flatly subducted Neo-Tethyan lithosphere at ca. 120 Ma according to a synthesis of

46   regional tectonic-magmatic-sedimentary records.

47

48   **Keywords:** Zircon Hf heterogeneity, sedimentary recycling, mantle metasomatism, magma

49   mixing, Lhasa Terrane

50

51   **Introduction**

52   Recycling of pre-existing continental crustal materials in magmatic arcs is a fundamental

53   process controlling the chemistry and evolution of the continental crust. The involvement of a

54   crustal component in arc-related magmas has been widely recognized and is typically manifested

55   by the enriched radiogenic (e.g., Sr-Nd-Hf) isotope composition. This can be achieved in two

56   profoundly different ways, either in the mantle through the addition of subducted material (mostly

57   sediments), or in the crust by assimilation and contamination of mantle-derived precursors (e.g.,

58   Hawkesworth et al. 1979; Gasparon et al. 1994). Distinguishing the two opposing mechanisms is

59   not easy unless unequivocal evidence, for example sediment-derived magmas intruding the

60   peridotite mantle wedge, is preserved (e.g., Zeng et al., 2016; Spencer et al., 2017), but is crucial

61   to determine the rates and volumes of continental growth and recycling.

62   Zircon is an ideal mineral to trace source complexity and evolution of the parent magma, as it

63   is chemically stable and has the potential to provide information on crystallization age (U–Pb),

64   radiogenic (Hf) and stable (O) isotopes, as well as on melt compositions (Valley et al., 2005;

65   Kemp et al., 2007; Grimes et al., 2015). Studies based on combined U–Pb, Hf and O isotopes

66   measured within single zircon grains have significantly advanced our capacity to understand the

67   role of mantle and sediment (or mature continent crust) components in the petrogenesis of

68   granitoid, as well as to enhance our insight into the timing of continental crust growth (e.g., Kemp

69   et al. 2007). Substantial variation of zircon $\varepsilon_{Hf}(t)$ values has been recognized in granitoid

70   worldwide and is commonly interpreted as evidence of mixing of magma derived from

71   isotopically different reservoirs (Kemp et al., 2007; Farina et al., 2018), non-equilibrium melting

72   of individual crustal sources (Tang et al., 2014) or due to local processes involving zircon

73   dissolution and recrystallization (Villaros et al., 2012, Farina et al., 2014, Finch et al., 2021).

74   However, subducted sediments may also play a significant role in the Hf budget of arc magmas, as

75   exemplified by the Banda arc where the mafic rocks underwent no or very limited crust

76   contamination but present substantial changes in Hf isotopes (ε_{Hf} = –3.8 to +12.5) with only little

77   variation in $\delta^{18}O$ of +5.8‰–+7.5‰ (Nebel et al. 2011). This phenomenon indicates that zircon Hf

78   isotope heterogeneity in granitoids may occur in mafic magmas that are free from any kind of

79   assimilation. However, in comparison with granitoids, large Hf isotopes variations at grain-scale

80   are rather rare in zircon from mafic rocks, probably due to higher crystallization temperature

81   and/or less abundance of zircons in mafic magma system.

82   In this paper, we present zircon U–Pb, Lu–Hf and O isotopic, whole-rock geochemical and

83   Sr–Nd isotopic, and in situ clinopyroxene elemental and Pb isotopic data, for some newly

84   identified Early Cretaceous dolerites and diorites in the Aruo area of central Lhasa Terrane,

85   Tibetan Plateau. The combined datasets lead us to contend that dramatic zircon Hf isotope

86   heterogeneities are not caused by intracrustal mixing of crust- and mantle-derived magma. We

87   argue instead that the isotopic variability is inherited from a depleted asthenospheric mantle

88   metasomatized by 1-4% sediment with heterogeneous Hf isotopes.

89

90   **Geological background and sample descriptions**

91   The Tibetan Plateau consists of four E-W-trending tectonic units, which from north to south

92   are the Songpan-Ganzi, Qiangtang, Lhasa and Himalaya terranes separated by three main suture

93   zones (Yin and Harrison, 2000). As the leading edge of the India-Asia collision, the Lhasa Terrane

94   is sandwiched between the Bangong-Nujiang suture zone (BNSZ) and the Indus-Yarlung Zangbo

95   suture zone (IYZSZ) (Fig. 1a). The BNSZ represents the remnants of the Meso-Tethys Ocean,

96   which was proposed to have closed in the Late Jurassic or the Early Cretaceous (Zeng et al.,

97   2021a). The Meso-Tethys Ocean slab has subducted northwards beneath the Qiangtang Terrane

98   since Early Jurassic (Li et al., 2014, Li et al., 2020), but it remains a controversy whether this

99   paleo-ocean slab has ever subducted towards south beneath the Lhasa Terrane (e.g., Zeng et al.,

100   2018; Li et al., 2018). The IYZSZ represents the remnants of the Neo-Tethyan lithosphere that

101   started to subduct beneath the Lhasa Terrane at Middle Triassic and closed at Early Paleogene

102   following the suturing of Indian and Asian continents (Kang et al., 2014; Kapp and DeCelles,

103   2019).

104   Based mainly on plentiful zircon ε_{Hf} data from intermediate-felsic igneous rocks and

105   sediment cover, the Lhasa Terrane can be divided into northern, central, and southern parts,

106   separated by the Shiquan River-Nam Tso Mélange Zone and the Luobadui-Milashan Fault,

107   respectively (Fig. 1; Zhu et al., 2011a). The northern Lhasa Terrane is covered by Middle Triassic–

108   Cretaceous sedimentary rocks and contains widespread Early Cretaceous magmatic rocks which

109   predominantly have positive zircon ε_{Hf} values. The magmatic rocks exposed in the central Lhasa

110   terrane are dominantly Early Cretaceous granitoid and volcanic rocks. The zircon ε_{Hf} values of

111   these rocks are rather negative, which, in combination with the Cambrian-Paleozoic sediment

112   cover, likely suggests the presence of ancient basement materials. The southern Lhasa terrane is

113   mainly composed of Jurassic-Paleogene Gangdese batholith and associated volcanic successions

114   (Ji et al., 2009; Kang et al., 2014). The overall positive zircon ε_{Hf} values of the Mesozoic

115   magmatic rocks of this subterrane indicate a juvenile nature of the continental crust, although

116   Precambrian basement has been locally identified in eastern and western parts (Hou et al., 2015).

117   Samples in the present study were collected from the northeast bank of the Kering Lake, ~70

118   km NW of Xainza Town (Fig. 1), which tectonically belongs to the central Lhasa Terrane. In

119   addition to Quaternary deposits, the strata exposed in the study area are mainly composed of

120   Devonian Chaguolama Formation limestone; Permian Xiala Formation limestone, Angjie

121   Formation shale, mudstone and siltstone; and Lower Cretaceous Duoni Formation sandstone,

122   siltstone and mudstone. The dolerites and diorites occur as intrusions with irregular map patterns,

123   with diameters hundreds of meters, which intruded the siltstones of the Lower Permian Angjie

124   Formation (Fig. 1b) in the Early Cretaceous, based on previous K-Ar dating (134 Ma; Wang et al.,

125   2002). In addition, some Early Cretaceous basalts-basaltic andesites are exposed to the south of

126   the Aruo intrusions (Fig. 1b; Kang, 2009).

127   The Aruo dolerites and diorite are medium-fine-grained and are mainly composed of euhedral

128   plagioclase, subhedral to euhedral clinopyroxene and amphibole with accessory minerals such as

129   titanite, zircon, ilmenite, and apatite. In addition, some pyroxene and plagioclase grains have been

130   converted to epidote by post-emplacement alteration (Fig. 2). It is noteworthy that some of the

131   euhedral plagioclase grains are wrapped by clinopyroxene and amphibole (Figs. 2b-2c) suggesting

132   the plagioclase crystallized before clinopyroxenes and amphiboles.

133

Analytical methods

Zircon U-Pb, O and Lu-Hf isotope analyses

136   Zircons were separated by standard density and magnetic separation techniques and

137   individually selected by hand-picking under a binocular microscope. Zircon oxygen isotope

138   analyses were carried out before U-Pb and Lu-Hf isotope determinations. Zircon oxygen isotopes

139   were measured using the SHRIMP IIe-MC equipped with a Cs ion source at the Beijing SHRIMP

140   Center. During the analysis, a 3 nA beam of Cs^+ was focused into a spot 20 μm elliptical on the

141   target surface. The detailed analytical procedures were similar to those described by Ickert et al.

142   (2008). The measured oxygen isotopic data were corrected for instrumental mass fractionation

143   using the Qinghu zircon ($\delta^{18}O_{VSMOW}$ = +5.3 ‰) as the external standard (Li et al., 2013). Eleven

144   measurements of the internal standard PengLai zircon grains during the course of this study yield

145   a weighted mean of $\delta^{18}O = +5.30 \pm 0.19$ ‰, which is identical within errors to the reported value

146   of $+5.31 \pm 0.10$ ‰ (Li et al., 2010). Measured $^{18}O/^{16}O$ is normalized to VSMOW, and then

147   corrected for the instrumental mass fractionation factor (IMF) as follows: $(\delta^{18}O)_M = ((^{18}O/^{16}O)_M /$

148    $0.0020052 - 1) \times 1000$; IMF $= (\delta^{18}O)_{M(standard)} - (\delta^{18}O)_{VSMOW}$; $\delta^{18}O_{sample} = (\delta^{18}O)_M + IMF$.

149   Zircon U–Pb isotopic analyses were conducted in five analytical sessions by using laser

150   ablation-inductively coupled plasma-mass spectrometry (LA–ICP–MS) at the Guilin University of

151   Technology (GUT). Cathodoluminescence images were used to select the appropriate zircon

152   grains for analysis. Laser sampling was performed using a GeoLasHD LA system. An Agilent

153   7900 ICP-MS instrument was used to acquire ion-signal intensities. A detailed description of the

154   laboratory   and   sample   preparation   methods,   following   the   guidelines   in

155   http://www.plasmage.org/recommendations and recommended in Horstwood et al. (2016) are

156   available as metadata (Table S1). Prior to sample analysis, an additional nitrogen gas flow (4

157   mL/min) was used to improve the instrumental sensitivity. Helium and Ar were the carrier and the

158   make-up gas, respectively, and were mixed via a T-connector before entering the ICP. Following

159   about 20 s period of background analysis, samples were spot ablated for 60 s at a 6 Hz repetition

160   rate with a spot size of 32 μm beam and laser energy of 10 J/cm^2 at the sample surface. Plesovice

161   zircon ($^{206}Pb/^{238}U$ age = 337 Ma; Sláma et al., 2008) and NIST 610 were analyzed as U-Pb and

162   trace element external standards, respectively. GJ-1 standard zircon was measured as unknown

163   samples to the quality of monitor trace elements. During the course of this study, the analyzed GJ-

164   1 standard zircon grains yielded a weighted $^{206}Pb/^{238}U$ age of 605 ± 1 Ma (1σ, MSWD = 0.3, n =

165   40; Table S2), which is consistent with the recommended U–Pb age in allowed precision

166   ($^{206}Pb/^{238}U$ age = 608.5 ± 0.4 Ma; Jackson et al., 2004). In addition, the trace element

167   concentrations of GJ-1 standard zircon were also consistent with the reference values (Table S3).

168   The Lu–Hf isotopic compositions of dated zircons were determined using an MC–ICP–MS

169   coupled with the laser ablation system at GUT. Analyses were conducted using a beam diameter of

170   44 μm and a laser pulse frequency of 10 Hz. The GJ-1 zircon was used as an external standard to

171   monitor data quality. The ratios of $^{176}Lu/^{175}Lu = 0.02655$, $^{172}Yb/^{176}Yb = 0.5887$, βYb =

172    $0.8725\times$βHf (βHf and βYb refer to the mass bias of Hf and Yb, respectively) obtained during our

173   analysis were used in the isobaric interference correction, consistent with the reference value (Wu

174 et al., 2006). The stable $^{178}Hf/^{177}Hf$ and $^{180}Hf/^{177}Hf$ ratios of GJ-1 zircon grains analyzed in this

175 study were 1.46715 ± 0.00023 and 1.88689 ± 0.00024 (2σ, n = 48), respectively, which are within

176 200 ppm of known values based upon atomic masses and abundances as recommended by Spencer

177 et al. (2020). The $^{176}Hf/^{177}Hf$ ratios of GJ-1 zircon grains during the course of this study range

178 from 0.281970 ± 16 to 0.282048 ± 20 with a mean value of 0.282009 ± 0.000016 (2σ; n = 48),

179 consistent with the reference value (0.282015 ± 0.000019, 2σ; Elhlou et al., 2006) within allowed

180 analytical errors.

181

Whole-rock major and trace elements analyses

183 Major and trace element analyses were conducted using an XRF-1500 X-ray fluorescence

184 analyzer and a double focusing high resolution inductively coupled plasma-mass spectrometer

185 (ICP-MS) at the Institute of Geology and Geophysics, Chinese Academy of Sciences. The loss on

186 ignition (LOI) values were measured firstly. About 2.0 g powdered samples were baked in the

187 muffle furnace for more than 2 hours at 915 °C. Then about 0.6 g baked samples were fluxed with

188 $Li_2B_4O_7$ (6g) at ~1200°C to make homogeneous glass disks for major elements determination.

189 During analysis, the USGS standard reference materials (GSR-1, GSR-2 and GSR-3) were used to

190 monitor the data quality. The analytical precision was better than 5%.

191 For trace elements analysis, 40 mg sample powders were dissolved in Teflon bakers using a

192 hybrid acid (1mL 22 M HF, 0.5mL 8 M HNO_3) at 120 °C. Then the solutions were dried and the

193 residues were dissolved using the hybrid acid (1.5mL 22 M HF, 0.5mL 8 M HNO_3) again and

194 moved in the high-pressure bombs at 200 °C for 5 days. After that, the sample solutions were

195 dried again and dissolved with 2.0 mL 8 M HNO_3 in the bombs at 150 °C for 24 hours. After

196 repeating the previous steps, solutions were transferred into PE bottles with 50 g 1% HNO_3

197 containing 10 ppt In. During the procedure, the USGS standard reference materials (GSR-1, GSR-

198 2, and GSR-3) and two blank samples were taken as the external calibration standards. The

199 analytical precision was better than 10%.

200

Whole-rock Sr-Nd isotope analyses

202 Powdered samples (~100 mg) for Sr and Nd isotopic analyses were dissolved first using a 1:3

203 mixture of HF and HNO_3 for 7 days at 120 °C in PFA beakers. The solutions were evaporated to

204   approximatively dry at 120 °C, and the residues were dissolved in 16 M HNO_3 and then

205   evaporated again. Subsequently, the residues were re-dissolved in 1 mL 16 M HNO_3, and insulated

206   at 120 °C for over 4 hours. The solutions were re-evaporated at 120 °C, and the residues were

207   dissolved in 2 mL 2M HNO_3 at 120 °C for over 2 hours, which were centrifuged and prepared to

208   separation of the target elements. Strontium and rare earth elements (REE) were separated using a

209   cationic resin. The REE solutions was then processed through HDEHP resin to obtain purified Nd.

210   Strontium and neodymium isotopes were measured by Neptune Plus multi-collector (MC)–ICP–

211   MS at GUT. The Sr and Nd isotopic data were obtained on MC-ICP-MS and normalized to

212    $^{86}Sr/^{88}Sr = 0.1194$ and $^{146}Nd/^{144}Nd = 0.7219$, respectively. $^{87}Rb/^{86}Sr$ and $^{147}Sm/^{144}Nd$ ratios were

213   calculated from abundances of these trace elements measured by ICP-MS. The USGS standard

214   reference materials (BHVO-2, GSR-1, and GSR-3) were used to monitor the data quality. The Sr

215   and Nd isotopes of BHVO-2 ($^{87}Sr/^{86}Sr = 0.703542 \pm 6$, $^{143}Nd/^{144}Nd = 0.512959 \pm 5$), GSR-1

216   ($^{87}Sr/^{86}Sr = 0.738340 \pm 7$, $^{143}Nd/^{144}Nd = 0.512217 \pm 6$), and GSR-3 ($^{87}Sr/^{86}Sr = 0.704131 \pm 6$,

217    $^{143}Nd/^{144}Nd = 0.512892 \pm 5$) are consistent with the reference values (Weis et al., 2006; Bao et al.,

218   2018).

219

In situ elemental and Pb isotopic compositions of minerals

221   Major element compositions of clinopyroxene and amphibole were determined using a JOEL

222   JXA 8230 electron microprobe at the Shandong Analysis Center of China Metallurgical Geology

223   Bureau. Analytical conditions for the electron microprobe were 15 kV accelerating voltage, 20 nA

224   beam current, 10–20 s counting time and 10 μm electron beam diameter. Natural and synthetic

225   materials were used as standards.

226   Major and trace elements of clinopyroxene were measured by using the LA–ICP–MS at the

227   GUT. Laser ablation was undertaken at a constant energy 10 J/cm^2 and at 10 Hz with a spot

228   diameter of 46 μm. Trace element concentrations were calibrated by using international standards

229   including NIST 610, BCR-2G, BIR-1G and BHVO-2G. Detailed analytical methods and the

230   operating conditions for the laser ablation system are given by Liu et al. (2008).

231   In situ analysis of Pb isotopes in clinopyroxene used MC-ICP-MS (Thermo Scientific)

232   equipped with a 193 nm laser ablation system (Resonetics) at the Guangzhou Institute of

233   Geochemistry, Chinese Academy of Sciences. An X-skimmer cone and additional nitrogen gas

234 flow (2 mL/min) were used to improve the instrumental sensitivity. Isotope signals were detected

235 with ion counters in the static mode. During the first 28s, the gas blank of the system was

236 monitored. In the following 30s, the signals of laser ablation on clinopyroxene were collected. The

237 analytical procedures were described in detail by Zhang et al. (2014). Repeated analyses of the

238 international basaltic glass BHVO-2G yielded mean values of $^{208}Pb/^{206}Pb = 2.054 \pm 0.004$ (2σ,

239 n = 9) and $^{207}Pb/^{206}Pb = 0.833 \pm 0.001$ (2σ, n = 9), which are consistent with the reported values

240 (2.048 and 0.832, respectively; Elburg et al., 2005; Zhang et al., 2014).

241

Results

243 Analytical results from zircon (O and Lu–Hf isotopes, U–Pb ages, and trace elements), whole

244 rocks (Sr and Sm-Nd isotopes, major and trace elements), and clinopyroxenes (trace elements and

245 Pb isotopes) are listed in Table S2–S9.

Zircon U-Pb ages and trace elements

247 Zircons separated from two dolerite samples (18AR-01 and 19AR-03) and one diorite sample

248 (19AR-01) were analyzed for U–Pb isotopes and trace elements (Tables S2-S3). All zircon grains

249 were colorless and subhedral to euhedral, have variable sizes (~50–300 um) with length-to-width

250 ratios of 1:1–3:1, display high Th/U ratios (1.4–2.8), and present clear oscillatory or banded

251 zoning in cathodoluminescence images (Fig. 3).

252 A total of 65 zircons from sample 18AR-01 were analyzed, seven of which did not yield

253 concordant results, probably because of Pb loss, although they have similar $^{206}Pb/^{238}U$ ages to

254 those of the other zircons. The remaining 58 spots yielded similar concordant zircon $^{206}Pb/^{238}U$

255 ages varying from 125.3 ± 2 to 117.4 ± 2 Ma with a weighted mean of 119.7 ± 0.5 Ma ($\pm 1\sigma$,

256 MSWD = 0.43; Fig. 4a). Similarly, 107 of 115 analytical spots on zircons from sample 19AR-03

257 yielded concordant zircon $^{206}Pb/^{238}U$ ages varying from 124.7 ± 1 to 117.0 ± 3 Ma with a weighted

258 mean of 119.9 ± 0.3 Ma ($\pm 1\sigma$, MSWD = 0.75, Fig. 4b). Nineteen of twenty-six analytical spots on

259 zircon grains from sample 19AR-01 yielded concordant zircon $^{206}Pb/^{238}U$ ages varying from 120.4

260 ± 2 to 117.0 ± 1 Ma with a weighted mean age of 118.6 ± 0.7 Ma ($\pm 1\sigma$, MSWD = 0.72 Fig. 4c).

261 Our new zircon geochronological data imply that the Aruo diorites and dolerites were coevally

262 formed at ca.120 Ma instead of 134 Ma as suggested by previous K-Ar dating (Wang et al., 2002).

263 The chondrite normalized rare earth element (REE) patterns of the analyzed zircon grains are

264   similar and are characterized by positive Ce and negative Eu anomalies, strong depletions in

265   light(L)REE relative to heavy(H)REE (Fig. 4d).

266

Zircon Lu-Hf and O isotopes

268   The $(^{176}Hf/^{177}Hf)_i$ of zircons from dolerites (a total of 291 analyses on zircon grains of sample

269   18AR-01 and 19AR-03) exhibit a wide range of 0.282557–0.283084 with corresponding $\varepsilon_{Hf}(t)$

270   values of –5.0 to + 13.7 and T_{DM-Hf} ages (Hf depleted mantle model ages; Griffin et al., 2000) of

271   258-1224 Ma (Fig. 5; Table S4). The $\varepsilon_{Hf}(t)$ heterogeneity is also observed in some individual

272   grains with up to 12 epsilon units in the same individual grain (Figs. 3 and 5). Besides, the $\varepsilon_{Hf}(t)$

273   values vary irregularly. For example, as shown in Fig. 3, low $\varepsilon_{Hf}(t)$ core with high $\varepsilon_{Hf}(t)$ rim, high

274    $\varepsilon_{Hf}(t)$ rim with low $\varepsilon_{Hf}(t)$ core, are both developed. In addition, repeatedly varied $\varepsilon_{Hf}(t)$ values are

275   observed in the same individual grain (Fig. 3c). Thirteen zircon grains from diorite sample (19AR-

276   01) also exhibit heterogeneous $(^{176}Hf/^{177}Hf)_i$ values of 0.282908–0.283141, with corresponding

277    $\varepsilon_{Hf}(t)$ values of +6.3 to +15.1 and T_{DM-Hf} ages of 190-768Ma.

278   The zircons of dolerite (sample 19AR-03) have restricted variability in oxygen isotopes with

279    $\delta^{18}O$ values of +6.0‰ to +7.2‰ (Fig. 3; Table S4) and a mean value of +6.8‰ ± 0.21‰ (±2σ; n =

280   42), which are slightly higher than those (+5.3 ± 0.6‰; 2σ) of igneous zircons in equilibrium with

281   mantle-derived magmas (Valley, 2003).

282

Whole-rock major and trace elements

284   The petrographic observations (Fig. 2) and moderate LOI values (2.3–3.9 wt. %) suggest that

285   the investigated rocks were altered after emplacement. However, based on the following

286   evidences, we contend post-emplacement alteration played a negligible role in modifying the

287   whole-rock chemical makeup. There is no correlation between LOI and fluid-mobile major

288   elements (e.g., K_2O and Na_2O; not shown). Instead, the two elements are in good correlation with

289   TiO_2 and Al_2O_3 (Fig. S1) that are relatively immobile during alteration (Dilek and Furnes, 2011;

290   Pearce, 2014). In terms of trace elements, all samples display coherent chondrite-normalized REE

291   and primitive mantle-normalized multi-element pattern (with the notable exception of Sr),

292   suggesting that these elements were relatively immobile during metamorphism. We further test the

293   mobility of target elements by their relationships with the most fluid-immobile elements, i.e., Nb

294   (e.g., Kurtz et al., 2000; Hastie et al., 2007). For example, the good correlations between Nb and

295   elements such as Y, La, Eu, Ba, and Ce, this suggests that these elements survived alteration,

296   whereas Sr underwent significant change.

297   Sixteen representative Aruo intrusions were analyzed for whole-rock major and trace

298   elements (Table S5). In the following descriptions and diagrams, the contents of all major

299   elements were recalculated on a normalized anhydrous basis. Consistent with the petrography, the

300   majority of the samples plot in the gabbro, gabbroic diorite and diorite fields on the SiO_2-

301    $Na_2O + K_2O$ diagram (Fig. 6a). With increasing SiO_2 content, the major oxides show roughly

302   negative (e.g., MgO, $Fe_2O_3^T$, TiO_2, and CaO) or positive (Al_2O_3, K_2O) relationship (Fig. 7). On

303   the SiO_2 vs FeOt/MgO plot, the Aruo intrusions show an evolutionary trend from low-K tholeiitic

304   to calk-alkaline (Fig. 6b).

305   All samples show low contents of compatible elements with (Ni = 2.26–21.9 ppm, Cr = 8.22–

306   76.2 ppm). The primitive mantle-normalized trace-element distributions exhibit enrichment in

307   large ion lithophile elements (LILE; e.g., Rb, U), depletion in high field strength elements (e. g.,

308   Nb, Ta), and no negative Zr and Hf anomalies (Fig. 8). The Aruo intrusions show moderate

309   enrichment in LREE relative to HREE on the chondrite-normalized patterns (($La/Yb)_N$ = 3.74–

310   4.97) with negligible Eu anomalies (δEu = 0.85–1.02, $\delta Eu = Eu_N/(Sm_N \times Gd_N)^{1/2}$).

311

Whole-rock Sr-Nd isotopes

313   Ten samples were analyzed for Sr–Nd isotopes (Table S6). The initial Sr–Nd isotopes were

314   corrected to 120 Ma based on zircon U–Pb dating. The age-corrected ($^{87}Sr/^{86}Sr)_i$ values of the

315   Aruo intrusions are variable (0.7064 to 0.7105), probably because the original Sr contents and

316   isotopic signature of these rocks have been disturbed by post-emplacement alteration as suggested

317   above. In comparison, the ($^{143}Nd/^{144}Nd)_i$ values are relatively uniform (0.512239 to 0.512409)

318   with corresponding to $\varepsilon_{Nd}(t)$ values of -1.5 and -4.8 and T_{DM-Nd} of 1295–2017 Ma (Fig. 9).

319

Mineral chemistry

321   This and our previous studies (Zeng et al., 2021a, b) demonstrate that the major elements of

322   clinopyroxene obtained by the two independent techniques (LA-ICP-MS and EPMA) are identical

323   within analytical errors; therefore, we refer to both in the following descriptions.

324   The clinopyroxenes in dolerites are augitic ($Wo_{36-47}En_{37-43}Fs_{15-24}$; Fig. 10) and exhibit

325   moderately varied CaO, MgO, and FeO contents (17.14–22.28 wt. %, 12.71–15.27 wt. % and

326   8.95–14.18 wt. % respectively; Table S7). Clinopyroxenes from the diorites are similar to those in

327   the dolerites and show strong compositional variations (CaO = 16.09–21.73 wt. %, MgO = 11.50–

328   16.95 wt. %, and FeO = 6.81–16.33 wt. %).

329   The amphiboles from one dolerite sample (19AR-05) are magnesio-hornblende and

330   tschermakitic pargasite, and have major element contents as follows: SiO_2 (40.02–44.25 wt.%),

331   CaO (10.39–12.33 wt.%), MgO (11.48–12.55 wt.%), TiO_2 (2.55–3.74 wt.%), FeO (14.66–16.58

332   wt.%), and Al_2O_3 (9.09–10.76 wt.%) and contain minor amounts of Na_2O (1.95–2.72 wt.%) and

333   K_2O (0.58–0.78 wt.%; Table S8).

334   The trace element composition of clinopyroxenes in dolerite and diorite are similar in

335   general. The chondrite-normalized REE pattern is characterized by depletion in LREE relative to

336   HREE ($(La/Sm)_N$ = 0.23–0.86; $(La/Yb)_N$ = 0.33–1.21, where the average is average 0.51). The

337   primitive mantle-normalized trace element patterns show Nb, Ta, Zr, Hf, Sr and Eu depletion

338   relative to the neighboring elements and variable Ba, Th and U enrichments (Fig. 10). The Sr

339   values are uniform or slightly decreasing with decreasing Mg#, which, together with the negligible

340   positive Eu anomalies and the petrographic observations, implies that clinopyroxene crystallized

341   simultaneous with and/or after plagioclase (Fig. 11).

342   The initial Pb isotopes of Clinopyroxene are corrected to 120 Ma following the methods of

343   Zhang et al. (2014). Clinopyroxene from two dolerite samples (19AR-04 and 19AR-05) exhibit

344   relatively homogeneous Pb isotopic compositions with $(^{208}Pb/^{206}Pb)_i$ and $(^{207}Pb/^{206}Pb)_i$ ratios

345   varying from 2.102 to 2.145 and from 0.844 to 0.863, respectively (Figs. 11c–11d; Table S9).

346

347   **Discussion**

348   **Physical condition during crystallization**

349   The chemistry of clinopyroxene is sensitive to melt temperature and pressure, but most

350   thermometers and barometers require an initial assumption of pressure and temperature,

351   respectively. The clinopyroxene-melt thermobarometer of Putirka et al. (2003) can simultaneously

352   estimate the crystallization pressure and temperature if the equilibrated liquid (e.g., whole-rock,

353   glass, or matrix) composition is known. However, the premise of using this thermobarometer is

354   that the clinopyroxene should be chemically in equilibrium with the melt (usually represented by

355   the glass, matrix, or whole-rock composition). The equilibrium can be tested by the Fe–Mg

356   exchange coefficient [$K_D(Fe–Mg)^{clinopyroxene–melt}$] between the clinopyroxene and the assumed melt,

357   which should be 0.28 ± 0.16 (±2σ; Putirka, 2008) if they are chemically equilibrated. As shown in

358   Fig. 10d, the clinopyroxene and whole-rock compositions are equilibrated, providing us access to

359   adequately use the clinopyroxene-melt thermobarometer. The clinopyroxenes of dolerite

360   crystallized under temperatures of 1149–1197 °C (average temperature is 1162 °C) and pressures

361   of 590–935 MPa (corresponding to a continental depth of 18 to 29 km). The clinopyroxenes of

362   diorite crystallized at lower temperatures of 999–1094 °C (average temperature is 1050 °C) and

363   pressures of 49–515 MPa (corresponding to a continental depth of 2 to 17 km; Table S7).

364   Amphibole compositions provide us another independent access to infer the physicochemical

365   conditions of melt crystallization. By using the methods of Ridolfi et al. (2010), the temperatures,

366   H_2O contents and pressures for magma in equilibrium with amphibole grains in the Aruo

367   intrusions, are 863–937 °C, 4.2–5.9 wt. %, and 190–280 MPa (corresponding to a continental

368   depth of 9 to 13 km), respectively (Table S8). Because the Aruo intrusions are not cumulate rocks,

369   the physicochemical estimations based on the mineral geothermobarometer are indicative of the

370   pressure, temperature, and water content occurring in the magma during the crystallization of the

371   target minerals. Thus, these results suggest that the parental magmas of the Aruo intrusions were

372   H_2O-rich and underwent a polybaric evolution with the crystallization of clinopyroxene at depth

373   (18-29 km) and that of amphibole at shallower levels (9–13 km).

374

375   **Nature of the source**

376   **Magma differentiation**

377   The MgO (3.7–5.9 wt. %), Mg# (42.1–47.2), Cr (8.22–71.0 ppm) and Ni (2.26–12.5 ppm)

378   values of the Aruo intrusions are much lower than those of primary mantle-derived melts

379   (Mg# = 68–76, Cr > 300 ppm, Ni > 200ppm; Roeder and Emslie, 1970), indicating a considerable

380   degree of differentiation of their parent magma. Fractional crystallization and assimilation of pre-

381   existing continental crust are two end-member mechanisms responsible for the observed chemical

382 variations in the Aruo intrusions. The heterogeneous zircon Hf isotopes plausibly suggest that

383 intracrustal assimilation or mixing might have played an important role during magmatic

384 evolution. However, as demonstrated by MELTS simulation and simple binary mixing modeling

385 of magma evolution, crustal assimilation fails to explain the initial increase in Al_2O_3

386 concentrations followed by the Al_2O_3 decrease and SiO_2 increase (Fig. 7b). Moreover, a

387 significant crustal assimilation is inconsistent with the variations in Th/Nb, Th/La and Nd isotopes

388 (Fig. 9). Therefore, we suggest that the chemical diversity of the Aruo intrusions are mainly

389 controlled by crystallization differentiation, and we will test the reason(s) of the zircon Hf isotope

390 heterogeneity in detail later. Simulation results and the negative correlations of SiO_2 with MgO,

391 $Fe_2O_3^T$, TiO_2, and CaO (Fig. 7) indicate that olivine, clinopyroxene, and Fe-Ti oxides were the

392 main fractionated phases (Fig. 2). The negligible to slightly positive Eu anomalies ($\delta Eu = 0.87$–

393 1.02; Fig. 8; Table S5), and the increasing in Al_2O_3 contents with increasing SiO_2 contents until 60

394 wt. % (Fig. 7b) suggest minimal or no fractionation of plagioclases in the mafic to intermediate

395 rocks.

396

Origin of the zircon Hf isotopes heterogeneity

398 There are several alternative ways to account for the zircon Hf isotope heterogeneity of the

399 Aruo rocks. First, we need to exclude that the Hf variability is related to analytical issues. An

400 inaccurate interference correction can lead to miscalculation of $^{176}Hf/^{177}Hf$ values, and analytical

401 inaccuracies are amplified proportionally to the $^{176}Yb/^{177}Hf$ of the analyzed crystals. For example,

402 a 0.1% inaccurate correction will lead to a miscalculation of $^{176}Hf/^{177}Hf$ values by about ~2 ε_{Hf}

403 units for zircon crystals with $^{176}Yb/^{177}Hf$ as low as 0.05 (Fisher et al., 2014; Farina et al., 2014).

404 Moreover, an inaccurate interference correction should give rise to a positive correlation between

405 $^{176}Yb/^{177}Hf$ and $^{176}Hf/^{177}Hf$ (Fisher et al. 2014). The great majority of zircons in the Aruo rocks

406 have $^{176}Yb/^{177}Hf$ ranging between 0.02 and 0.05, together with no positive correlation between

407 $^{176}Hf/^{177}Hf$ and $^{176}Yb/^{177}Hf$ (Fig. S2a), suggesting the Hf isotopic variability described reflects a

408 natural process rather being an analytical artifact.

409 The $\varepsilon_{Hf}(t)$ variability in zircon thus argues for the involvement of isotopically different

410 components. The Lhasa lithospheric mantle has suffered multiple stages of metasomatism via

411 oceanic or continental subduction since the Proterozoic (e.g., Yin and Harrison, 2000; Zhu et al.,

412   2011a; Hu et al., 2018; etc), which should have extremely enriched the Hf isotopic signature (Fig.

413   5a; Zhu et al., 2012; Hu et al., 2018). Besides, the central Lhasa Terrane is underlain by an ancient

414   basement as demonstrated by the local exposure of high-grade metamorphic rocks and the

415   negative zircon $\varepsilon_{Hf}(t)$ values of the Late Triassic-Jurassic granitoids (Zhu et al., 2011b; Fig. 5a).

416   Therefore, the depleted end-member can only be the asthenospheric mantle, which gains support

417   from the fact that the highest ε_{Hf} values in the Aruo zircons are similar to those observed in the

418   regional Jurassic MORB-type rocks ($\varepsilon_{Hf}(t)$ = +12.8 and +17.7; Tang et al., 2020).

419   These two components (enriched and depleted end-members) are both participating in the

420   petrogenesis of the Ario intrusion and four hypotheses can be formulated to account for the origin

421   of these rocks: (1) the enriched components come from the lithospheric mantle; (2) the enriched

422   component comes from the assimilation of an ancient basement (lower continental crust) with low

423    $\varepsilon_{Hf}(t)$ and $\varepsilon_{Nd}(t)$ but equilibrated with the mantle for oxygen; (3) the addition of an upper crustal

424   component at shallow level via assimilation of the country rocks; (4) partial melting of the

425   asthenosphere (depleted component) metasomatized by a crustal (sediment) component coming

426   from the oceanic slab. Below we test the four hypotheses in detail.

427   Firstly, the constant $^{208}Pb/^{206}Pb$ values of the Aruo clinopyroxene are inconsistent with

428   mixing between magmas derived from the asthenospheric and lithospheric mantle. This view is

429   further supported by the binary mixing model of trace element ratios (in particular Th/La; Fig. 9).

430   The basement of the Lhasa Terrane is directly sampled by recently discovered Triassic gneiss

431   exposed in the Rendui area (Fig. 1a; Zhou, 2017), yet, the Sr-Nd-Pb isotopic signature of this

432   gneiss remains unknown. The Early Cretaceous Cuoqen metaluminous dacites are interpreted as

433   melts of the Lhasa basement (Huang et al., 2017), as supported by their similar zircon ε_{Hf} values

434   with the metamorphic zircons of Rendui gneiss that are much higher than the S-type granites in

435   central Lhasa Terrane. It is therefore reasonable to use the Sr-Nd-Pb isotopes of the Cuoqen

436   dacites to represent the Lhasa basement. The constant $^{208}Pb/^{206}Pb$ values of clinopyroxene

437   crystallized at 590–935 MPa preclude any significant contamination with the lower continental

438   crust (Fig. 11).

439   Disentangling mantle source heterogeneity from shallow crustal contamination by using

440   radiogenic isotopes alone is not straightforward. Zircons of the Baingoin S-type granites nearby

441   the study area, which were formed by anatexis of Lhasa sediment-rich upper crustal assemblages,

442   display markedly varied ε_{Hf} values (-21.36 to -0.02; Hu et al., 2019). Asthenospheric melts

443   assimilating upper crustal rocks will, therefore, crystallize zircons with high ε_{Hf} core and low ε_{Hf}

444   rim, though the degree of ε_{Hf} variation depends on the Hf isotopic composition and the amount of

445   sediment assimilated. However, both low and high ε_{Hf} cores are developed in the Aruo zircons,

446   and the Hf isotope composition changes irregularly (Fig. 3). Thus, it is unlikely the Hf variability

447   in zircon of Aruo intrusions resulted from mixing of asthenospheric and crustal component.

448   Additional supports for this view include: (1) ancient xenocrystic zircons are absent in the Aruo

449   intrusions, yet our binary mixing modeling implies assimilation of a considerable amount (30–

450   50%) of upper crustal sediment would be needed to achieve the low $\varepsilon_{Nd}(t)$ values and Th/Nb and

451   Th/ La ratios of the Aruo rocks (Figs. 9c–9d). Such large-scale assimilation would undoubtedly

452   leave petrological traces that are not witnessed in these rocks. (2) The whole-rock $\varepsilon_{Nd}(t)$ values of

453   the Aruo intrusions remain constant or even slightly higher with increasing SiO_2 contents, at odds

454   with the trend of assimilation of an ancient crustal component (Fig. 9a). (3) There are no

455   correlations between the Hf isotopes and key trace element concentrations in zircon that are

456   sensitive to physicochemical variations, for example Th/U ratios and Hf contents for the degree of

457   magma differentiation and temperature, Yb/Gd for crystallization depth (Fig. S2; e.g., Kemp et al.,

458   2007; Barth et al., 2013; Cooper et al. 2014; Grimes et al., 2015).

459   The spread of data within Hf-O isotope space provides an independent means of assessing

460   mixtures of crustal and mantle reservoirs (Roberts et al., 2013). The zircon oxygen isotopes are

461   relatively uniform, albeit they are slightly higher than pristine mantle values and thus provide

462   further evidence of incorporation of supra-crustal materials into the petrogenetic process (e.g.,

463   Valley et al., 2005). Given that oxygen concentrations in sediments, crust and mantle are broadly

464   similar, the shape of mixing/assimilation curves on a Hf-O plot is largely controlled by the Hf

465   concentrations of the various components. The lack of covariation between Hf and O isotopes

466   from the same grains by the Aruo zircons (Fig. 5b) suggests the mixed components have

467   contrasting Hf concentrations (i.e., depleted mantle and sediment rather than basaltic melt and

468   sediment). The elevated $\delta^{18}O$ of high ε_{Hf} zircon suggests introduction of a high $\delta^{18}O$ component

469   relatively early on in magma differentiation (Valley et al., 2005; Kemp et al., 2007), that is, to the

470   mantle source region. The lower $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ isotopic values of the Aruo

471   clinopyroxenes than that of the Neo-Tethyan ophiolite further support the interpretation that

472 recycled sediment was incorporated into the depleted mantle (Fig. 11c). Besides, the high whole-

473 rock Th/La ratios (>0.29; Table S5) and their negative relationship with the Sm/La ratios (not

474 shown), are consistent with the results of sediment-mantle mixing (Plank, 2005). Our modeling

475 shows that partial melting of asthenosphere mantle with the addition of 1–4% terrigenous

476 sediment can explain well the Hf-O isotope variability of the Aruo zircons (Fig. 5b). We note,

477 however, that we must explain why the whole-rock trace element and Nd isotopes distributions, as

478 well as Pb isotopes of clinopyroxene of the Aruo intrusions are relatively uniform (Figs. 8, 9 and

479 11). These phenomena argue that the amounts of recycled sediments are probably not the primary

480 control of the Hf isotope variation. The Hf concentrations and isotopes of subducted terrestrial

481 sediments are primarily controlled by zircon (e.g., Carpentier et al., 2009; Nebel et al. 2011),

482 which themselves may be heterogeneous in Hf isotopes as regionally manifested by the Baingoin

483 S-type granites described above (Hu et al., 2019). Zircon, however, contributes little Nd and Pb to

484 the whole rock budget compared to Zr and Hf. Thus, we argue that zircons in subducted sediments

485 were fully dissolved, and the Hf isotopes of the dissolved crystals governed the Hf isotopic

486 signature of the metasomatized mantle with negligible influence on Nd-Pb isotopes and most other

487 trace elements. Although the true composition of subducted sediments in this area is unknown, the

488 Lhasa Terrane suffered several phases of oceanic slab subduction and continental collision prior to

489 and during its assembly with the Gondwana supercontinent. We suggest that this complex history

490 justifies the variability in Hf isotopes of detrital zircon from Paleozoic-Mesozoic sediments (e.g.,

491 Zhu et al., 2009). Accordingly, we propose that the Hf isotope heterogeneity in zircon of Aruo

492 intrusions most likely was inherited from a mantle source metasomatically enriched with Hf from

493 sedimentary sources like those from which the Baingoin S-type granites were formed.

494

Implications

496 Our study has two main major implications, one is related to the regional tectonic evolution,

497 the other is more general and concerns the use of zircon Hf isotope data to trace magmatic

498 evolution and the growth of continental crust.

Regional tectonic evolution

500 Prior to the India-Asia collision, the Andean-type subduction of Neo-Tethyan lithosphere

501 along the southern margin of Lhasa Terrane formed prolonged Mesozoic magmatism with several

502   phases of flare-up (Ji et al., 2009). The underplating of depleted mantle-derived materials during

503   the oceanic slab subduction led to the growth of large amounts of juvenile crust in the southern

504   Lhasa Terrane (e.g., Ma et al., 2013a; Zhu et al., 2017). However, the tectonomagmatic evolution

505   of the central-north Lhasa Terrane remains enigmatic, although widespread Early Cretaceous

506   magmatism with a flare-up at ca. 120–110 Ma has been identified (e.g., Zhang et al., 2012; Li et

507   al. 2018; Zeng et al., 2020). Previous studies focused on intermediate-felsic magmatic rocks with

508   relatively little attention to contemporaneous mantle-derived mafic rocks, given their rare

509   exposure in the central Lhasa Terrane.

510   Three competing hypotheses have been proposed to explain the Early Cretaceous

511   tectonomagmatic evolution of the central-north Lhasa Terrane: (1) intra-block underthrusting

512   and/or post-collision lithospheric delamination following the collision between Lhasa and

513   Qiangtang terranes (Hu et al., 2017); (2) southward subduction, roll-back and eventually break-off

514   of the Bangong-Nujiang Oceanic lithosphere (Wang et al., 2020); (3) northward subduction and

515   roll-back of the Neo-Tethyan lithosphere (e.g., Zhang et al., 2012; Ma et al., 2013b; Zeng et al.,

516   2020). The low-K tholeiitic to calk-alkaline evolutionary trend (Fig. 6b), the negative Nb-Ta

517   anomalies with enrichment in most LILEs (Fig. 8), and the presence of amphibole crystals and

518   high melt H_2O contents of the studied Aruo dolerites and diorites are characteristic of arc-type

519   rocks (Zhou et al., 2006), and accordingly are more in support of their formation in a subduction-

520   related, rather than post-collisional setting. Moreover, zircon Hf-O isotopes and whole-rock Th/La

521   ratios and Nd isotopes (Figs. 5 and 9) collectively suggest the recycling of terrigenous sediments

522   into the depleted mantle. The lack of negative Zr-Hf anomalies (Fig. 8), together with the

523   suprachondritic Zr/Hf (34.3–41.2) and Zr/Y (5.0–7.0) ratios of the Aruo intrusions, imply that the

524   zircons in the subducted sediments might have been completely destroyed.

525   The dissolution of zircon in a sediment-derived melt is governed by zircon saturation (i.e., Zr

526   solubility in the melt, which increases with temperature and possibly decreases with pressure;

527   Watson and Harrison, 1983; Rubatto and Hermann, 2007). Melting experiments using sedimentary

528   starting materials show that temperatures in excess of 925 °C are needed to fully dissolve zircon

529   (Hermann and Rubatto, 2009). This is why mafic arc rocks with high Zr/Y ratios and positive Zr-

530   Hf anomalies are rare and are mainly found in hot subduction zones (e.g., the high Zr/Y basalts

531   from the Sumisu Rift, Hirai et al., 2018). For the Aruo intrusions, a special tectonic environment

532   therefore is inferred to gain slab surface temperature high enough to dissolve zircon in subducted

533   sediments, regardless of whether the sediment is carried by a Neo-Tethyan slab or Bangong

534   Tethyan slab. Based on a synthesis of independent observations, we (Zeng et al., 2020) have

535   recently proposed a model of steepening and roll-back of flatly northwards subducted Neo-

536   Tethyan oceanic lithosphere at ca.120–110Ma to explain the observed late Early Cretaceous

537   tectonic-magmatic complexity (Fig. 13). These observation are (Fig. 12): (1) Jurassic-Early

538   Cretaceous magmatism migrated northward throughout the Lhasa terrane and finally migrated

539   southward at ~110 Ma; (2) rapid cooling of the Xiabie and Baingoin granites started at ~120 Ma,

540   (Kapp et al., 2007; Volkmer et al., 2014); (3) deposition in the Coqen Basin of limestone of the

541   Langshan Formation and limestone to clastic red beds of the Takena Formation took place at ca.

542   113–96 Ma and ca. 105–90 Ma, respectively (Fig. 12; Sun et al., 2015, Leier et al., 2007); (4) no

543   southward subduction-related accretionary complex is preserved in the northern Lhasa Terrane

544   (Hu et al., 2022). The Aruo intrusions provide new evidence of a change in the tectonic regime of

545   the central-northern Lhasa Terrane at ca. 120 Ma. We propose that the rising asthenosphere in

546   response to the change in the Neo-Tethyan subduction angle could provide the additional heat

547   required to dissolve zircon in the subducted sediments as manifested by the geochemistry of the

548   Aruo intrusions (Fig. 13).

549

The use of zircon Hf isotopes to track magmatic evolution and crust growth

551   Our study reports substantial Hf isotope variation in zircon crystallized from mantle-derived

552   magma that requires a source containing components of depleted asthenosphere and pre-existing

553   mature crust. Our combined clinopyroxene and whole-rock chemical and isotopic investigation

554   limits contamination or mixing of crust-derived melts with mantle-derived melts in the upper crust

555   as the cause of heterogeneous Hf isotopes. Instead, we suggest that the heterogeneity was inherited

556   through metasomatism of depleted asthenospheric mantle by terrestrial sediments. Magmas

557   derived from these sources may pass this Hf isotope heterogeneity on to granitoids either by

558   fractional crystallization or by partially melting, which may be misunderstood as evidence of

559   magma mixing or disequilibrium melting of the crust if only zircon Hf isotopes in granitoids are

560   studied. Therefore, we emphasize the necessity of isotopic investigations on minerals crystallizing

561   before zircons, and combined O isotopic and whole-rock data, to correctly resolve the mechanism

562 of recycling of pre-existing continental crust to the source of arc rocks as shown by complex

563 zircon Hf isotopes.

564   In addition, our study provides the first evidence to show that the increasing zircon Hf

565 isotope values of Early Cretaceous magmatic rocks at ca. 120 Ma (Fig. 12) was related to

566 underplating of depleted mantle-derived materials, namely juvenile continental crustal growth.

567 Because of the addition of terrestrial sediments to the wedge mantle, the Hf isotopic model ages

568 (258–1224 Ma) are much older than the formation of this juvenile crust. Importantly, although

569 regional geological and geochemical data do provide testimony of crustal growth (Fig. 5), even

570 some of the zircons in the Aruo dolerites and diorites with MORB-like depleted Hf isotopes will

571 probably be omitted as evidence of continental crustal growth following the landmark criterion

572 (δ^{18}O +5.9‰ in 2σ level) proposed by Kemp et al. (2007). We therefore hold a conservative view

573 on simply using detrital zircon Hf-O isotopes to infer the timing and rate of continental crustal

574 growth through time regionally and globally, which certainly is useful but should be accompanied

575 with other additional constraints either from zircons themselves (e.g., the Th/Nb ratios as a proxy

576 of melt SiO_2 content; Turner et al., 2020) or minerals crystallized before zircons.

577

578   **Acknowledgments**

590

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591

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870

871   **Figure Captions:**

872   Figure 1. (a) Simplified tectonic map of the Tibetan Plateau showing the distribution of Mesozoic

873   magmatic rocks of the Lhasa Terrane (after Zeng et al., 2020). (b) Geological map of the Aruo

874   region, central Lhasa Terrane. IYZSZ-Indus-Yarlung Zangbo Suture Zone; BSZ-Bangong Suture

875   Zone; SNMZ-Shiquanhe-NamuTso ophiolite mélange zone; LMF-Lobuodui-Milashan Fault. The

876   zircon age (zircon U–Pb) of the Xainza basalts-dacites is from Kang (2009).

877

878   Figure 2. Representative photomicrographs (cross-polarized illumination) showing the mineral

879   assemblages of the Aruo dolerites (a-c) and diorites (d). Pl-plagioclase; Cpx-clinopyroxene; Amp-

880   amphibole; Ilm-ilmenite; Ep-epidote.

881

882   Figure 3. Representative cathodoluminescence (CL) images showing the morphology, $^{206}Pb/^{238}U$

883   age (Ma), and Hf-O isotopes of the zircon crystals in the Aruo dolerites and diorites.

884

885   Figure 4. (a-c) Zircon U-Pb concordia diagrams of the Aruo dolerites and diorites, (d) chondrite-

886   normalized zircon rare earth element patterns for the Aruo dolerites and diorites (normalizing

887   values are from Sun and McDonough, 1989).

888

889   Figure 5. Plots of (a) zircon age vs $\varepsilon_{Hf}(t)$ and (b) zircon $\varepsilon_{Hf}(t)$ vs $\delta^{18}O$. In (b) the curves are binary

890   mixtures of putative mantle and crustal end-members (upper continental crust). The basalt was

891   represented by the Jurassic MORB-type rock (15T160) in the Ren Co region (Tang et al., 2020),

892   and the upper crust was represented by the Baingoin S-type granite (16BG50-5; Hu et al., 2019).

893   Mixing with upper continental crust with different $\delta^{18}O$ values is modeled given the substantially

894   varied zircon $\delta^{18}O$ values of the Bangoin S-type granites (Hu et al., 2019), and a mantle end-

895   member with moderately elevated $\delta^{18}O$ value is assumed and modeled because the zircons in the

896   Aruo intrusions with asthenosphere-like $\varepsilon_{Hf}(t)$ values also display $\delta^{18}O$ value higher than those in

897   equilibrium with mantle-derived magmas (e.g., Roberts et al., 2013).

898

899   Figure 6. (a) $Na_2O + K_2O$ versus SiO_2 (Middlemost, 1994); (b) SiO_2 vs. FeO_t/MgO diagram (after

900   Miyashiro, 1975) showing data of the Aruo intrusions. The data of the nearby Xainza basalts-

901   dacites are plotted here and in the following geochemical diagrams for comparison because they

902   were formed synchronously with the Aruo intrusions (Fig. 1; Kang, 2009).

903

904   Figure 7. (a-f) Harker diagrams showing CaO, Al_2O_3, TiO_2, $Fe_2O_3^T$, MgO and K_2O versus SiO_2 for

905   the Aruo intrusions. The dashed and solid lines represent the MELTS simulated fractional

906   crystallization trends of magma evolution (Ghiorso and Sack, 1995). The model assumes that the

907   initial magma composition is a dolerite sample 19AR-05, the water content is +3 wt. %, the

908   pressure is 200 MPa, and $fO_2=\Delta QFM \sim \Delta QFM +1$. The data of the Xainza basalts-dacites are from

909   Kang (2009).

910

911   Figure 8. (a) Primitive mantle-normalized trace element patterns and (b) chondrite-normalized

912   rare earth element data for the Aruo intrusions. Normalizing values are from Sun and McDonough

913   (1989). The data of the Xainza basalts-dacites are from Kang (2009).

914

915   Figure 9. Plots of (a) whole-rock SiO_2 vs $\varepsilon_{Nd}(t)$, (b) Th/Nb versus $\varepsilon_{Nd}(t)$, and (c) Th/La versus

916    $\varepsilon_{Nd}(t)$ for the Aruo intrusions. The AFC simulation was performed using the method described by

917   Ersoy and Helvacı (2010); "r" refers to the assimilation rate in AFC models. In Fig. b, the

918   assimilated end-member is represented by one sample of the Baingoin S-type granites (16BG49-2;

919   Hu et al., 2019), and the deleted end-member is the Jurassic Ren Co MORB-type rock (sample

920   15T269; Tang et al., 2020). The data of lithospheric mantle-derived gabbros are from Zhu et al.

921   (2012) and Hu et al. (2018), and of the Xainza basalts-dacites are from Kang (2009).

922

923   Figure 10. (a) $CaSiO_3$–$MgSiO_3$–$FeSiO_3$ diagram showing the compositions of pyroxene

924   (Morimoto et al., 1988), (b) plot of clinopyroxene $Mg^{\#}$ vs. whole-rock $Mg^{\#}$ values, showing the

925   melt–clinopyroxene equilibrium band, (c-d) primitive mantle-normalized multi-element patterns

926   and chondrite-normalized REE patterns for clinopyroxenes of the Aruo dolerites and diorites.

927

928   Figure 11. Plots of (a) Al_2O_3 versus Mg# values, (b) Sr versus Mg# values, (c) $(^{208}Pb/^{206}Pb)_i$

929   versus $(^{207}Pb/^{206}Pb)_i$, and (d) $(^{207}Pb/^{206}Pb)_i$ versus Mg# values for clinopyroxene from the Aruo

930   intrusions. The data of Neo-Tethyan ophiolite are from Xu and Castillo (2004) and Zhang et al.

931   (2005), and of the ancient basement are inferred from Huang et al. (2017). The field of sediment is

932   from Qian et al. (2017).

933

934   Figure 12. Temporal and spatial distribution of Mesozoic magmatic rocks in the Lhasa Terrane

935   (after Zeng et al., 2020 and references therein). The zircon $\varepsilon_{Hf}(t)$ values of magmatic rocks in the

936   central Lhasa Terrane notably increase at ca. 120 Ma. IYZSZ—Indus-Yarlung Zangbo Suture

937   Zone; BSZ—Bangong Suture Zone; SNMZ—ShiquanheNamuTso ophiolite mélange zone;

938   LMF—Lobuodui-Milashan Fault.

939

940   Figure 13. Schematic model of the genesis of the Aruo intrusions. The subduction-derived melts

941   incorporate Hf derived from heterogeneous detrital zircon components. Without effectively

942   mixing, these components crystallize magmatic zircon that captures this sedimentary

943   heterogeneity.

944
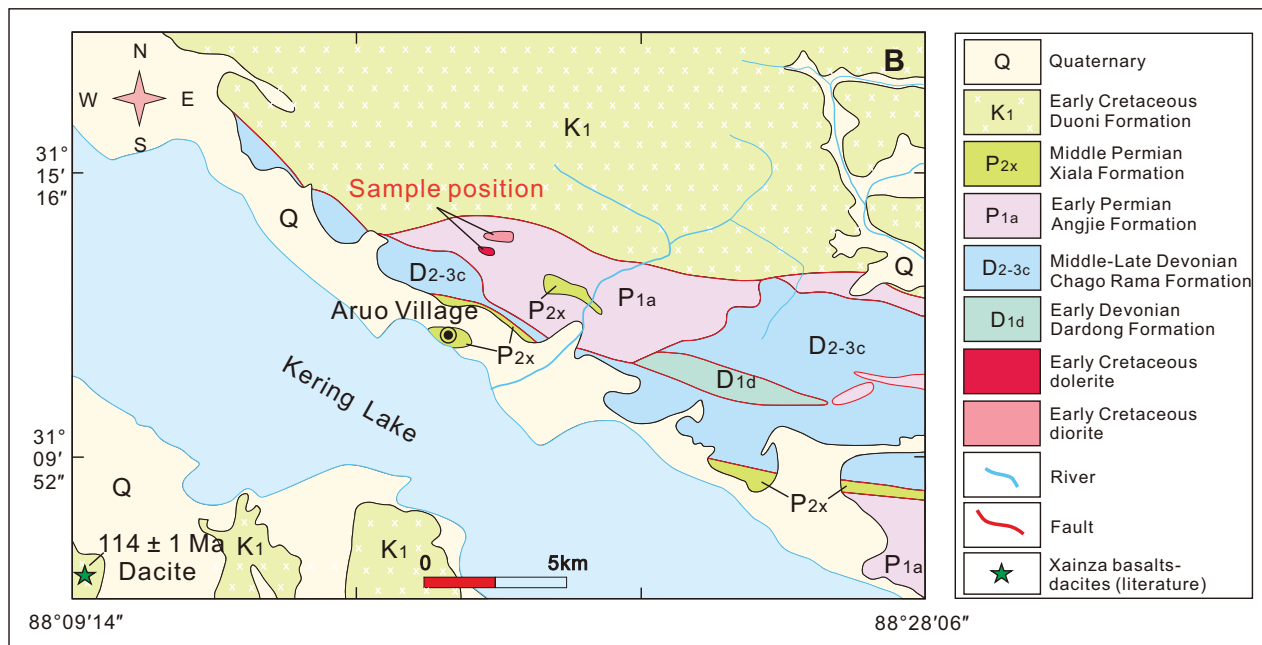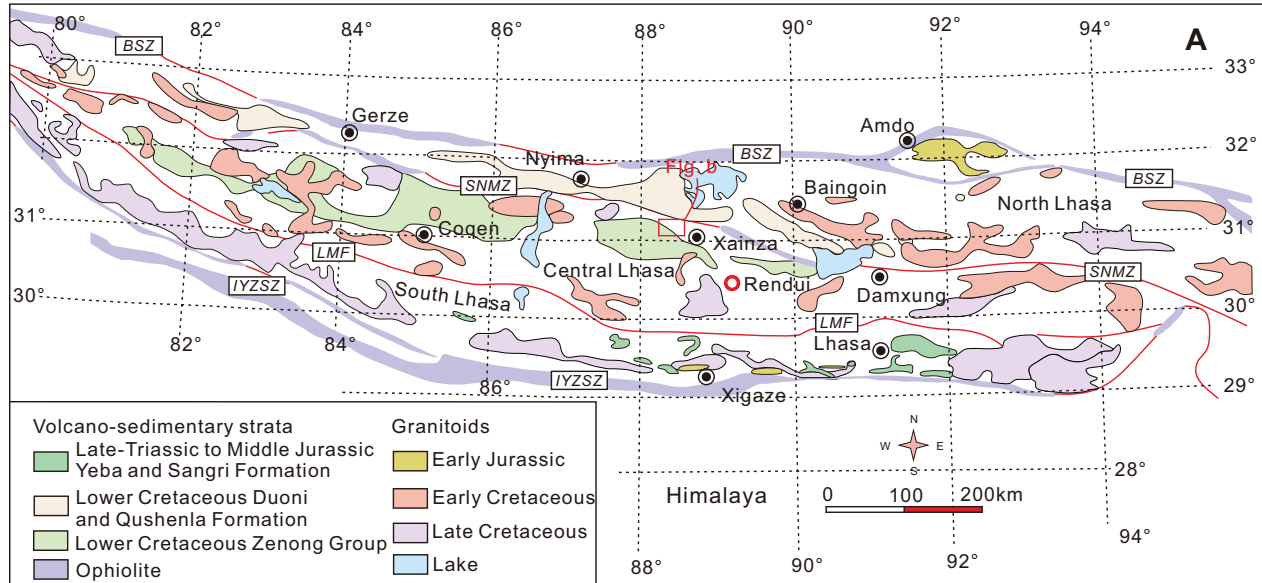
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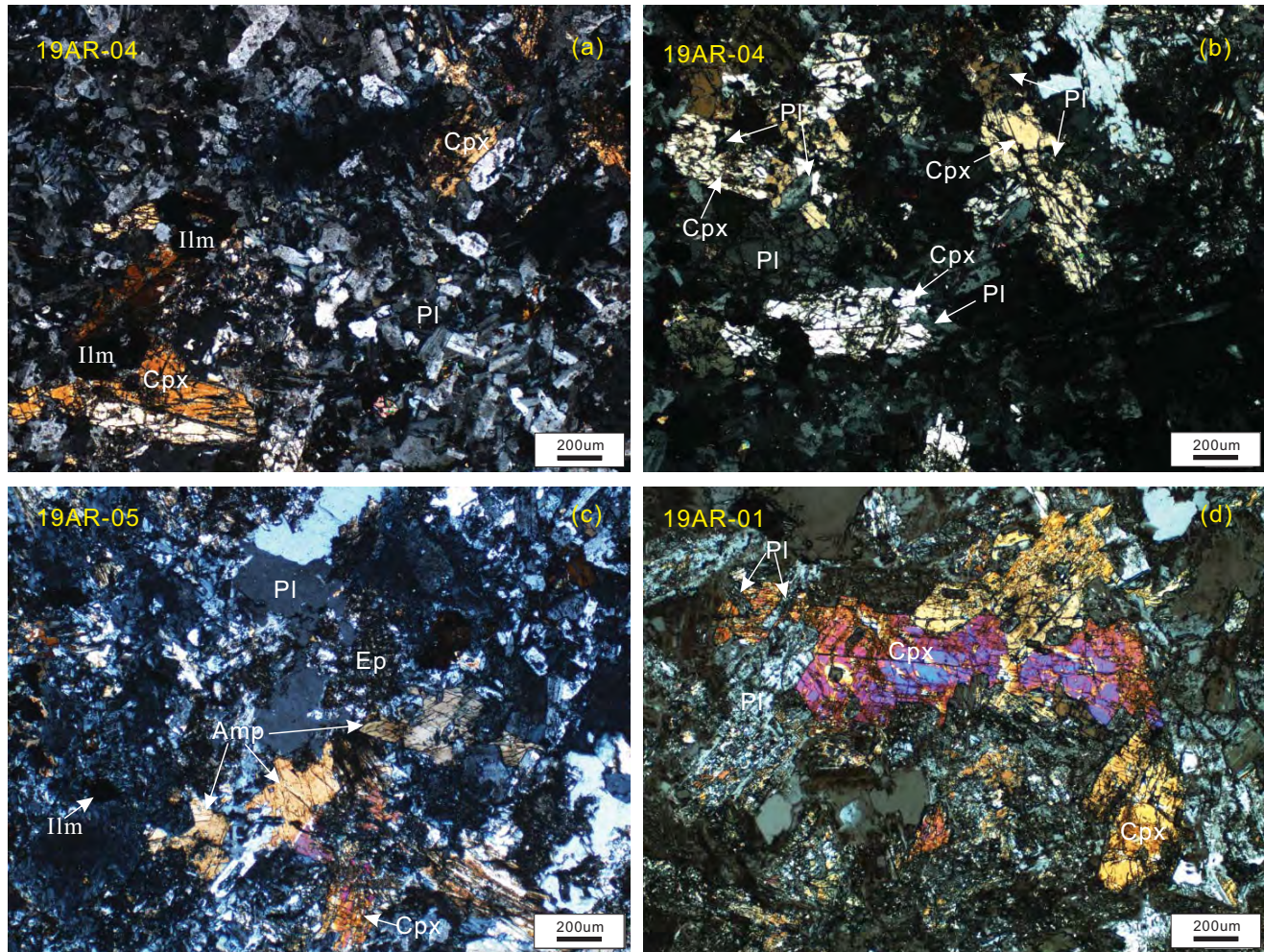
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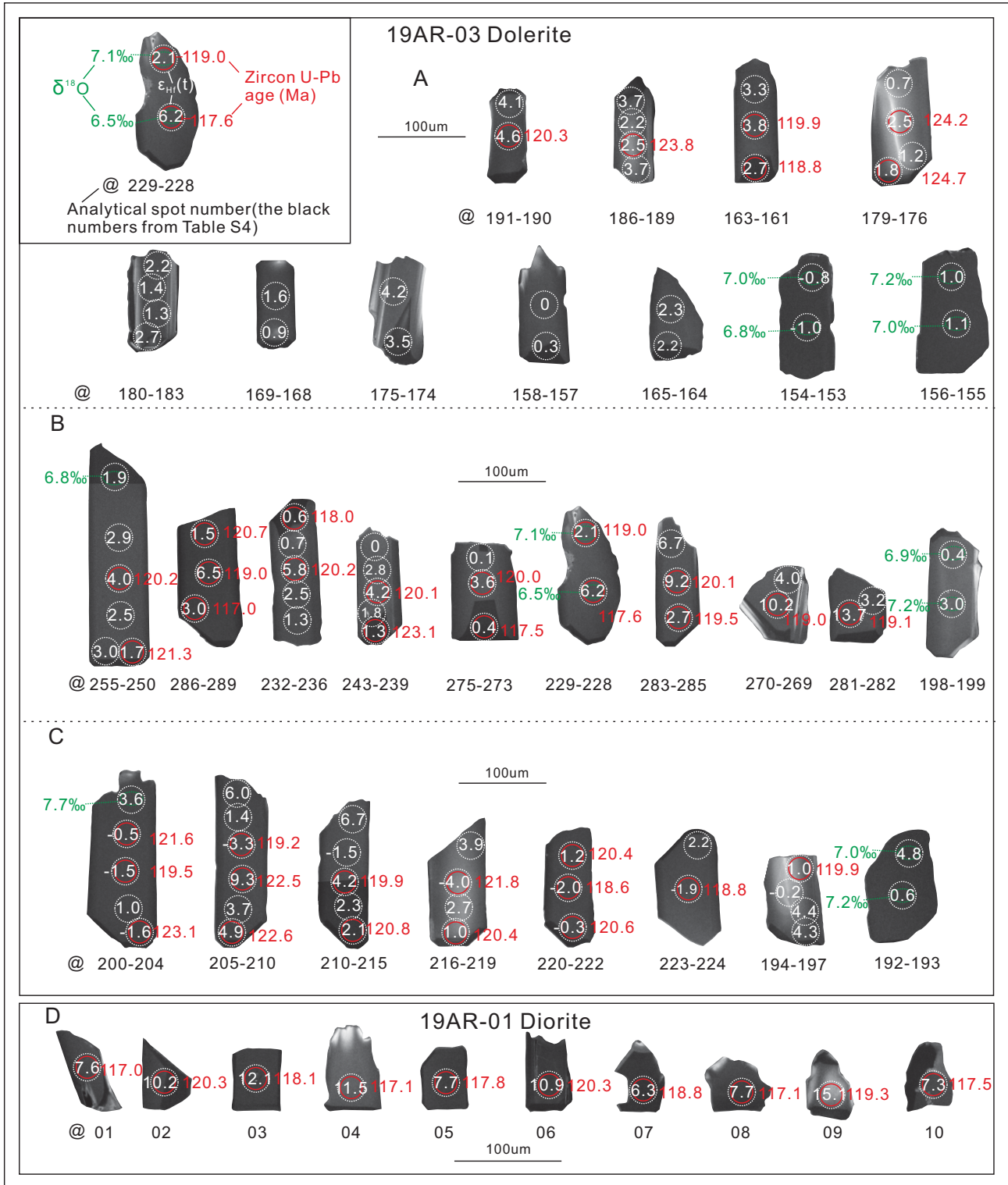
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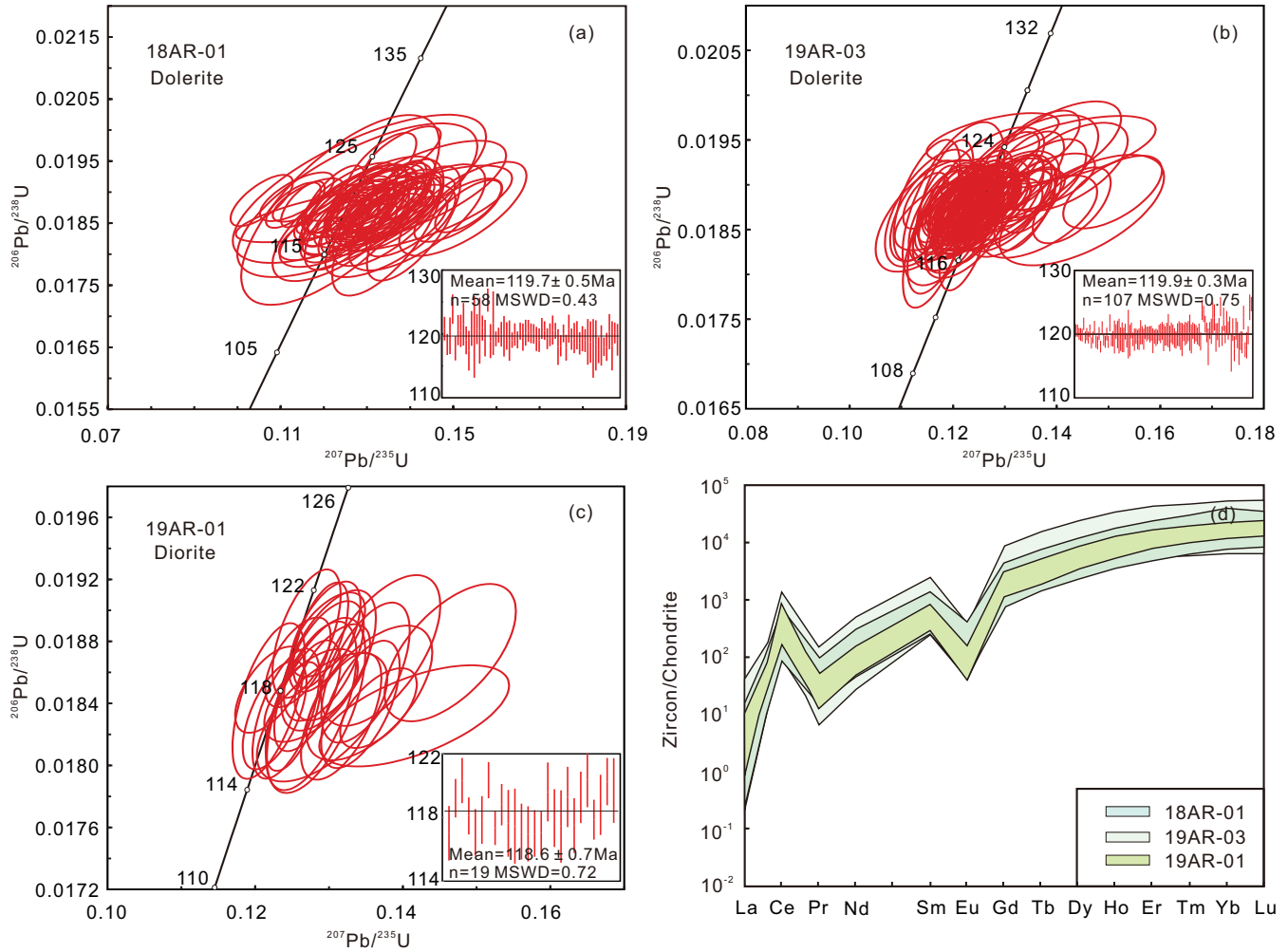
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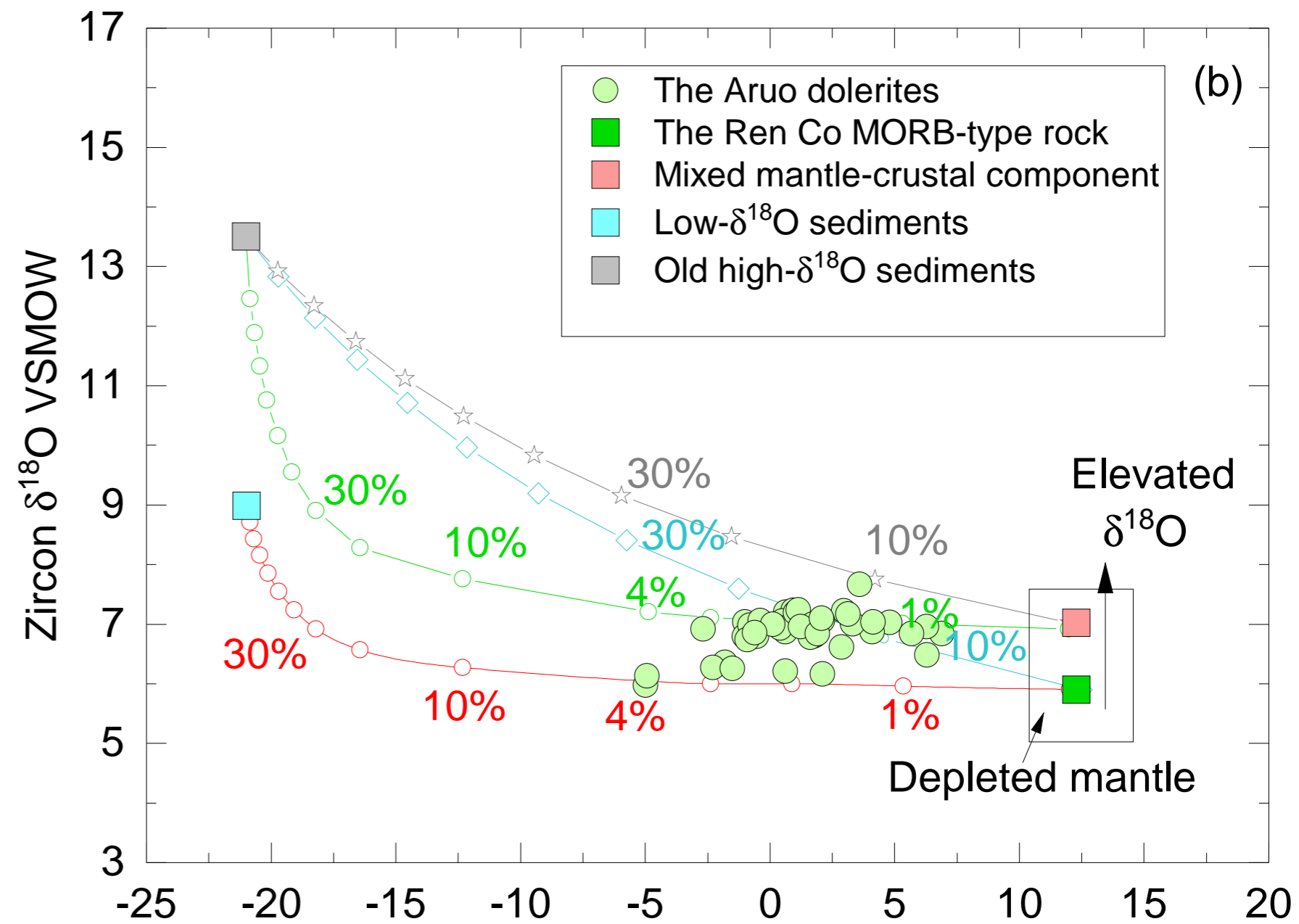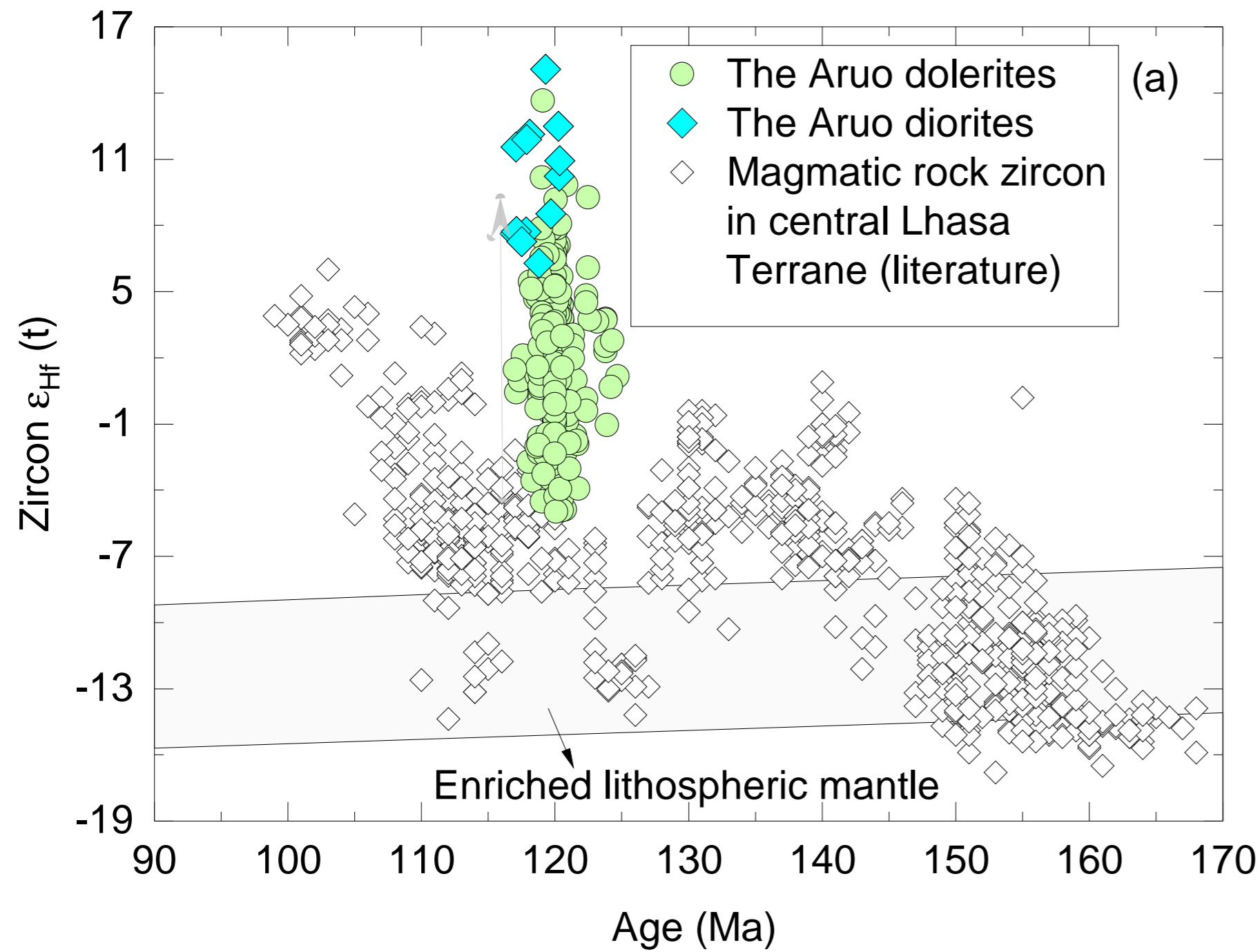
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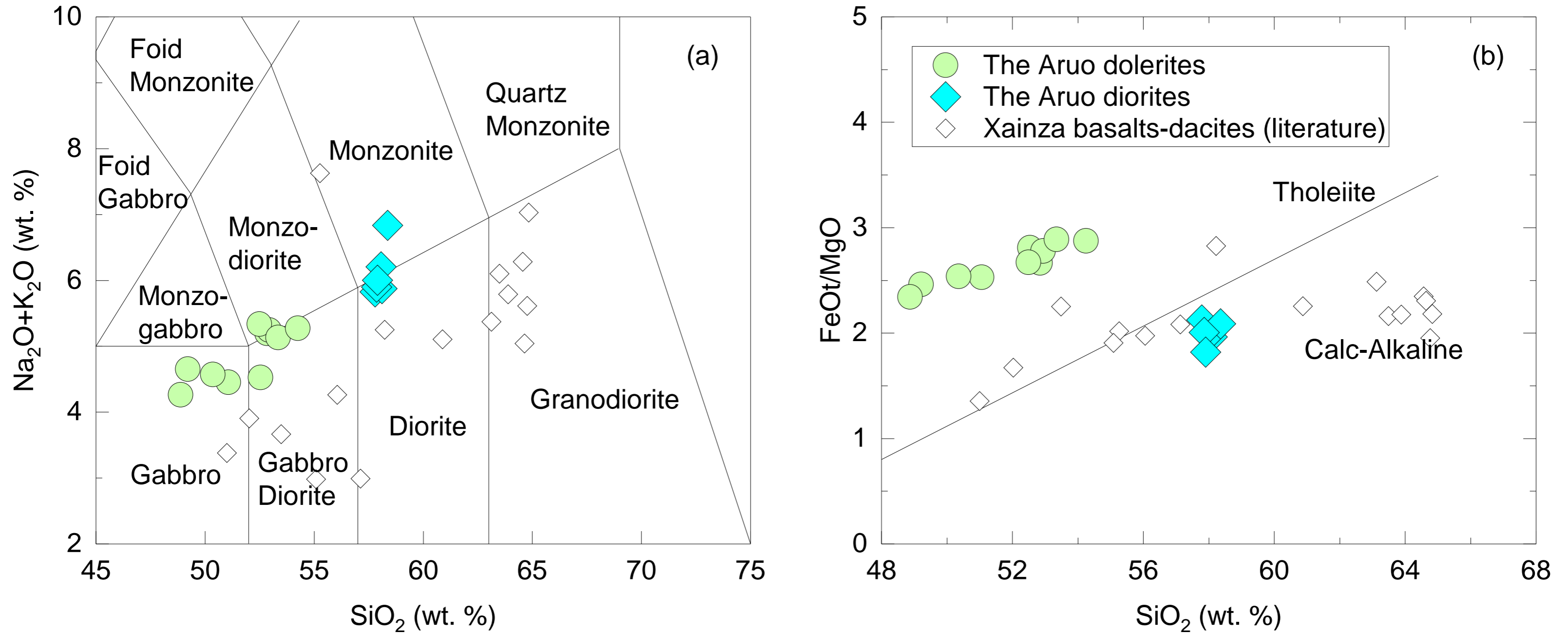
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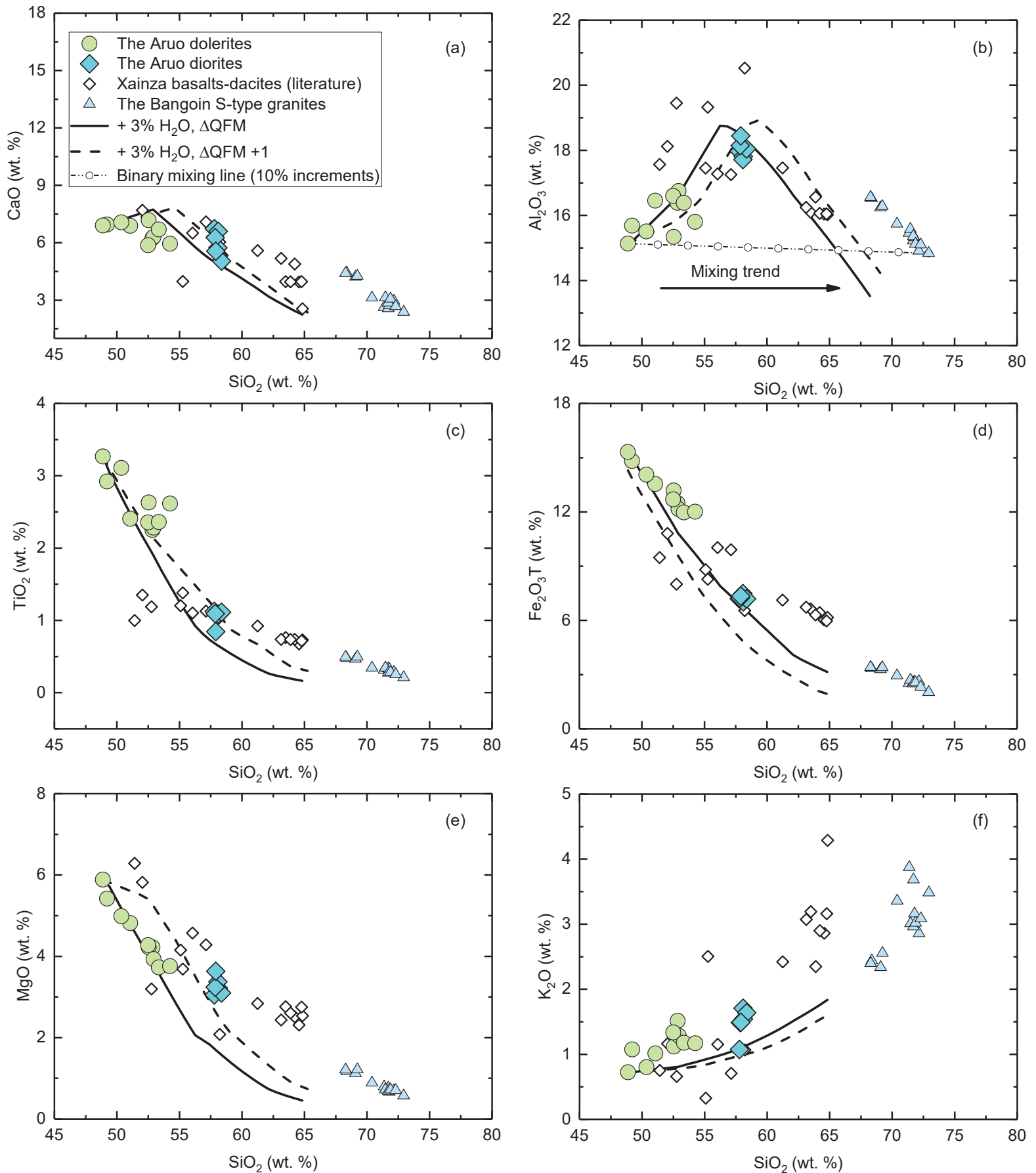
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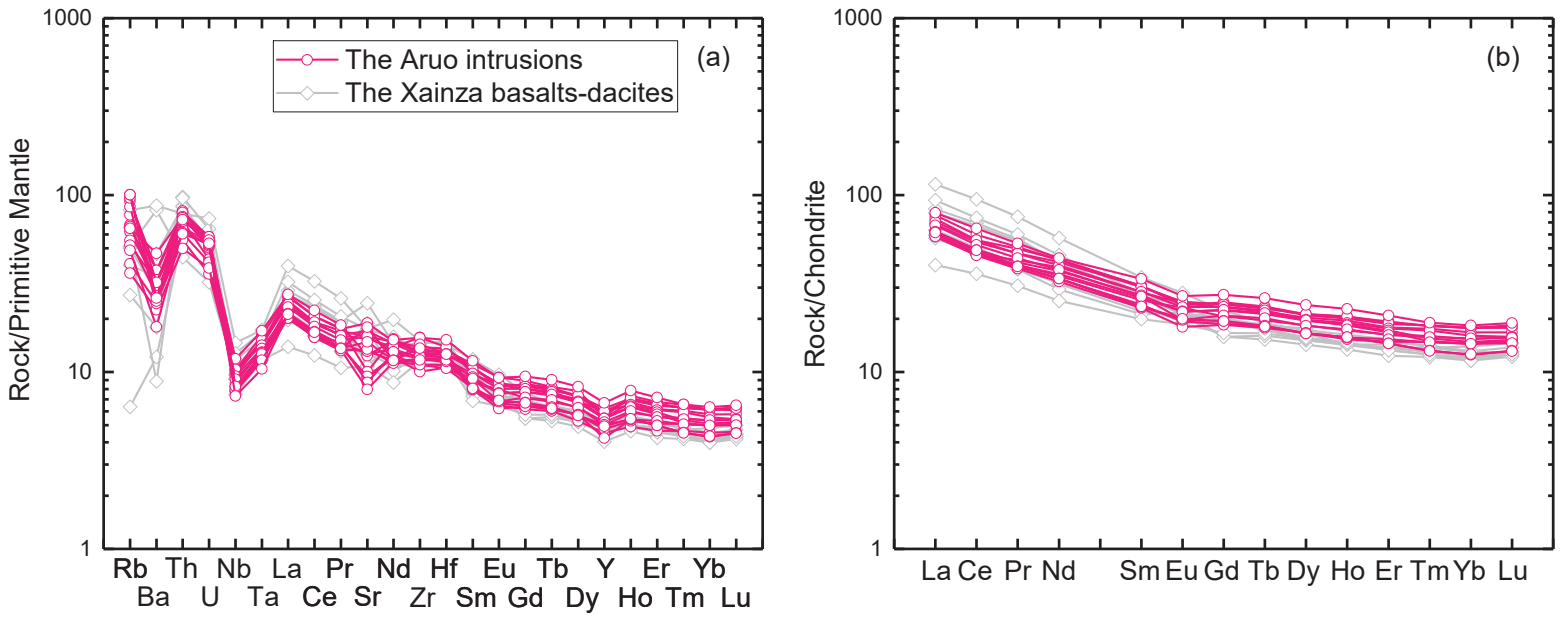
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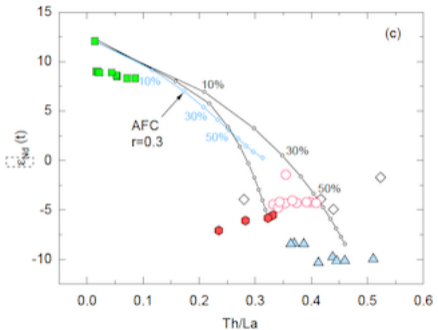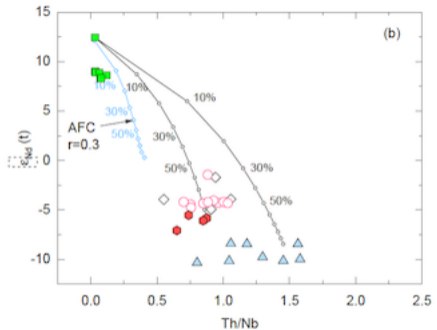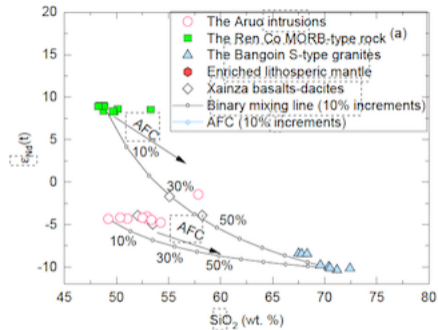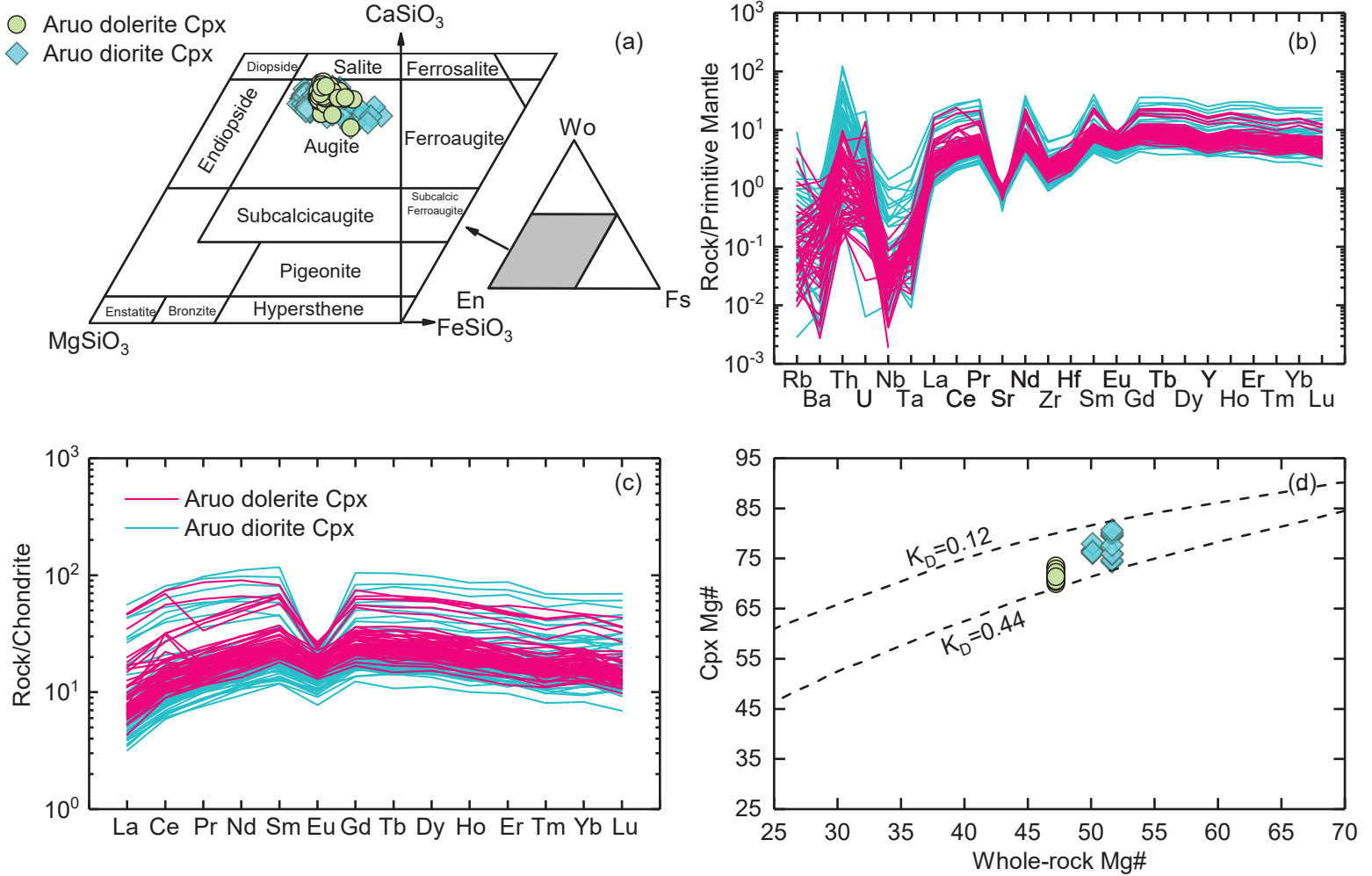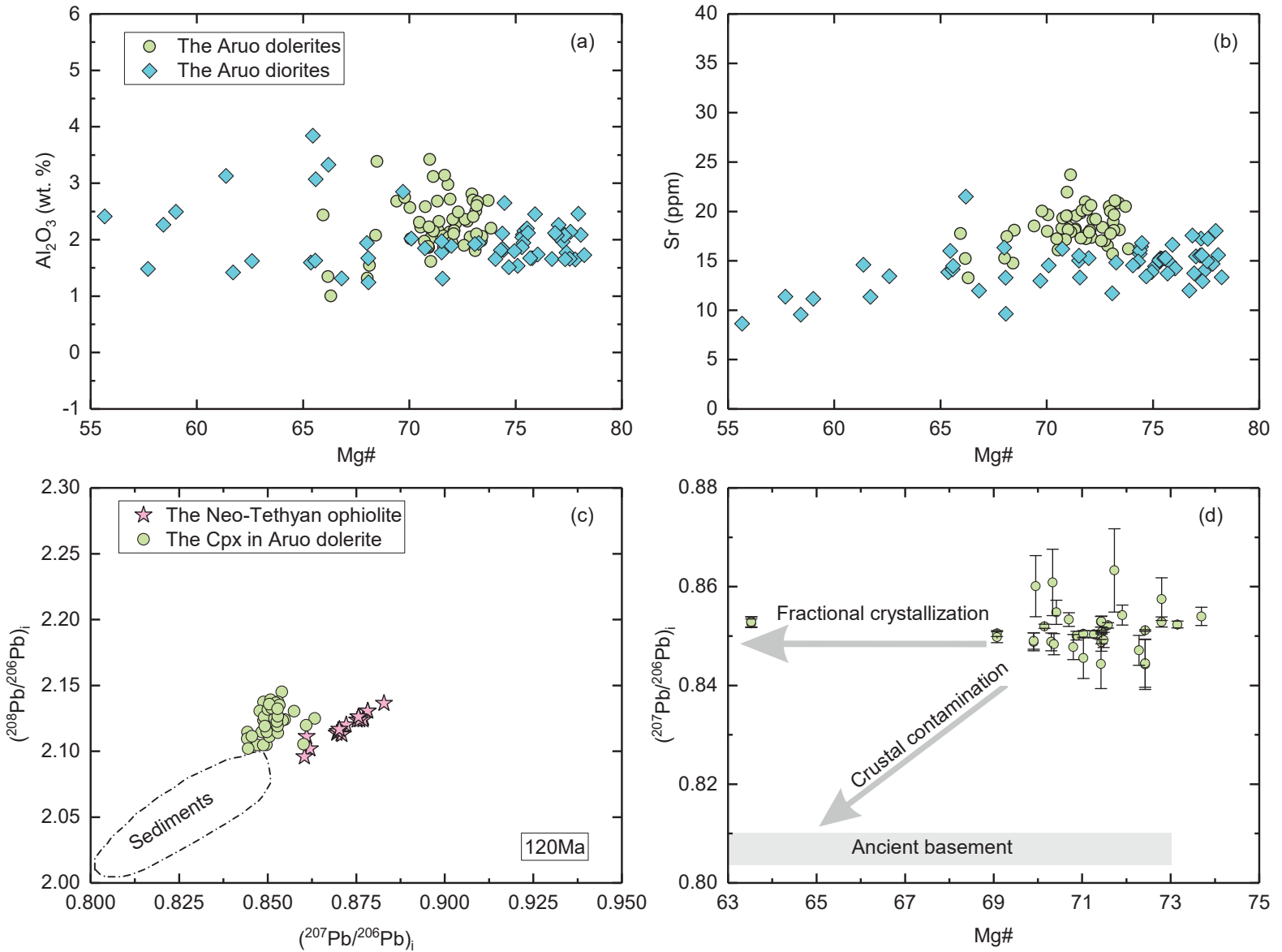
Figure 10

Figure 11

Figure 12

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