

1 **Nb/Ta intracrustal differentiation during granulite-facies metamorphism**

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10 **Abstract**

11 Both continental crust and depleted mantle are characterized by subchondritic
12 Nb/Ta, leading to a mass imbalance compared to the bulk Earth. Even though several
13 potential high Nb/Ta reservoirs in Earth's core and mantle have been proposed to
14 solve this "missing Nb paradox", little attention has been given to those in the crust.
15 Here we present bulk-rock and rutile geochemical data for samples of lower crustal
16 pelitic granulite from the North China Craton, which exhibit systematic variation in
17 Nb and Ta contents. Melt-depleted residua from high-temperature granulite (<900 °C)
18 are characterized by Nb/Ta values close to chondritic, restite from ultra-high
19 temperature (UHT) granulites (>900 °C) have subchondritic Nb/Ta, and leucosome
20 from UHT granulite has suprachondritic Nb/Ta. These variations are best explained
21 via competition for Nb and Ta between biotite and rutile during metamorphism,
22 although initial bulk-rock Nb/Ta values also have an effect. Biotite dehydration
23 melting generates a high-Nb/Ta residue and low-Nb/Ta melt because biotite
24 preferentially incorporates Nb over Ta. However, geochemical modeling suggests that
25 once biotite is depleted, the Nb/Ta ratio of the system is instead controlled by rutile
26 growth, which promotes formation of a lower Nb/Ta residue and higher Nb/Ta melt.
27 Anhydrous melt generation without involving Nb- or Ta-rich mineral fractionation or
28 contamination from other low Nb/Ta source would then retain a high Nb/Ta. We
29 propose that *in-situ* to in-source leucosome and leucocratic veins in UHT terranes
30 may be a high Nb/Ta reservoir, and the residuum of high-temperature granulite makes
31 much less of a contribution to the crustal Nb budget.

32 **1 Introduction**

33 Although the continental crust constitutes only ~0.6% mass of the silicate
34 Earth, it contains a very large proportion of incompatible elements, which makes it a
35 critical reservoir for mass-balance calculations of the Earth as a whole (Sclater et al.,
36 1980). Previous studies show that both continental crust and depleted mantle have
37 subchondritic Nb/Ta (Rudnick, 1995; Kelemen, 1995; Rudnick et al., 2000), which is
38 problematic for chondritic Earth models since no complementary suprachondritic Nb/
39 Ta reservoirs are known. Several such domains within the Earth have been proposed,
40 including the core, the lower mantle, carbonatite-metasomatized lithospheric mantle,
41 refractory eclogite formed during slab melting, and deeply crystalized high-Nb
42 cumulates (Münker et al., 2003; Pfänder et al., 2012; Rudnick et al., 2000; Tang et al.,
43 2019; Wade and Wood, 2001; Willbold and Stracke, 2006), but only a few workers
44 have considered the crust. In particular, Stepanov and Hermann (2013) suggested that
45 a potential high Nb and Nb/Ta reservoir was restitic lower crust that had experienced
46 significant biotite-dehydration melting, owing to experimental data showing that
47 biotite preferentially incorporates Nb over Ta. However, rutile is another major host of
48 Nb and Ta in metamorphic rocks, and both minerals have distinctly different partition
49 coefficients for Nb and Ta that vary as a function of pressure and temperature
50 conditions (Klemme et al., 2005; Stepanov and Hermann, 2013). So, what role does
51 granulite-facies metamorphism and anatexis play in Nb/Ta intracrustal differentiation?

52 High grade metamorphism and crustal melting are ubiquitous in the lower
53 crust, and melts that subsequently ascend through the overlying rock column and

54 crystallize as granite at shallower depths represent a key mass-transfer process that
55 leads to geochemical differentiation (Brown et al., 2011). During burial, metapelites
56 experience continuous partial melting as temperature increases, and biotite undergoes
57 incongruent melting to form peritectic garnet and rutile (Sawyer et al., 2011). As a
58 restitic phase, rutile is a major host for high field strength elements (HFSE) and may
59 hold the vast majority of Nb and Ta in a rock (Meinhold, 2010). In such cases, a
60 detailed study of bulk-rock and rutile geochemistry in pelitic granulites would be
61 highly informative in deciphering the evolution of the continental crust and
62 developing reservoir models.

63 In this study, a series of high-temperature (HT) to ultrahigh temperature
64 (UHT) pelitic granulites from the northwestern part of the North China Craton (NCC)
65 were selected for a bulk rock and rutile geochemical study (see analytical methods in
66 Text S1). These data are compared to the results of petrological forward modeling
67 combining phase diagram analysis, accessory mineral solubility modeling and trace
68 element modeling conducted for a typical metapelite (see modeling methods in Text
69 S2). Integration of geochemical data from the study area with data for granulites
70 preserved globally then allowed the impact of intracrustal differentiation during
71 granulite facies metamorphism on Nb/Ta ratios to be further understood.

72

73 **2 Geological setting**

74 In this work, we considered rocks from the northern part of the Trans-North
75 China Orogen (TNCO) and eastern part of the Khondalite Belt (KB), NCC, both of

76 which are Late Paleoproterozoic orogens (*c.* 1.85–1.95 Ga) (Fig. 1). The north TNCO
77 is dominated by *c.* 2.5 Ga tonalite–trondhjemite–granodiorite (TTG) gneisses that
78 contain mafic and metapelitic enclaves recording peak metamorphic pressure–
79 temperature (*P–T*) conditions of 1.1–1.2 GPa at <900 °C, followed by decompression
80 cooling to the solidus (Huang et al., 2018; Wu et al., 2016).

81 The eastern KB is dominated by aluminous gneiss, garnet orthogneiss, and
82 minor gabbro and norite. Ultrahigh temperature (UHT) granulites with the diagnostic
83 mineral assemblage sapphirine + quartz record an initial stage of isobaric cooling
84 from peak *P–T* conditions of 0.8–0.9 GPa at >960 °C, followed by decompression
85 cooling to the solidus (Huang et al., 2019; Li and Wei, 2018).

86

87 **3 Sample description**

88 **3.1 HT-HP granulite from Trans-North China Orogen (TNCO)**

89 Pelitic granulites from northern Trans-North China Orogen show evidence of
90 minor partial melting (Fig. 2a, b). Ten samples were collected from Manjinggou area
91 (Fig. 1b), which contain mineral assemblage of K-feldspar, quartz, plagioclase,
92 garnet, sillimanite, biotite, rutile and minor kyanite (Fig. 3). A detailed petrological
93 work was conducted on sample 18MJ09.

94 Garnet in sample 18MJ09 is usually coarse-grained and poikiloblastic, with a
95 lot of tiny inclusions of quartz, and/or biotite in the garnet core (Fig. 3c). These
96 inclusions along with garnet core were suggested to represent prograde mineral
97 assemblage (M_0). Kyanite occasionally occur as inclusions in garnet rim (Fig. 3a, b),

98 but not in the matrix. Therefore, kyanite, biotite, as well as K-feldspar, plagioclase,
99 quartz and rutile in garnet rim should belong to peak pressure mineral assemblage
100 (M_1). Rutile inclusions could be further divided into single inclusions and combined
101 inclusions with quartz, and/or biotite (Fig. 3c, d). In the matrix, K-feldspar,
102 plagioclase, quartz, sillimanite, garnet, biotite and rutile represent decompressed
103 mineral assemblage (M_2). Rutile could occur either next to felsic minerals or garnet,
104 sillimanite (Fig. 3).

105

106 **3.2 UHT granulite from east Khondalite Belt**

107 Ultra-high temperature granulites from east Khondalite belt experienced high
108 degree of partial melting (Fig. 2d). Fourteen felsic granulite samples were collected
109 from Tuiguiwula area. The protolith of these samples vary from greywacke to pelite.
110 Sample 16TG53 was a felsic paragneiss, whose protolith should be greywacke (Fig.
111 2c). Mineral assemblage in sample 16TG53 consists of plagioclase, quartz, K-
112 feldspar, garnet, sillimanite, biotite and rutile (Fig. 4).

113 Considering sillimanite only occur as inclusions in garnet but not in matrix
114 (Fig. 4a, c), garnet inclusions stable with sillimanite were suggested to be prograde
115 mineral assemblage (M_0), which should contain plagioclase, K-feldspar, quartz,
116 garnet, sillimanite, biotite and rutile. Rutile inclusions could be further divided into
117 single rutile inclusion and combined rutile inclusion with quartz, and/or biotite (Fig.
118 4). In the matrix, coarse-grained garnet (>3 mm diameter) along with plagioclase, K-
119 feldspar, quartz and rutile were suggested to be peak assemblage (M_1). Rutile grains

120 occur both next to felsic minerals and garnet (Fig. 4b, c, d). In places in the matrix,
121 coarse-grained garnet and K-feldspar are separate by fine- to medium- grained biotite
122 as well as rutile (Fig. 4c, d). Therefore, mineral assemblage of plagioclase, K-feldspar,
123 quartz, garnet, biotite and rutile was suggested to be retrograde mineral assemblage
124 (M_2).

125

126 **4 Results**

127 **4.1 Bulk rock composition of granulites and leucosomes**

128 Granulites collected from the northern TNCO have mostly similar bulk rock
129 compositions. Niobium concentrations range from 11.90 to 16.50 ppm and Nb/Ta
130 ratios vary from 17.18 to 23.91, with a mean of 20.95, which are slightly higher than
131 the chondritic value of 19.90 (Fig. 5). By comparison, restites collected from the
132 eastern KB show variable bulk compositions and inconsistent concentrations of heat-
133 producing elements (HPEs), which is inferred to be a result of various degree of
134 partial melting. These UHT restites have Nb contents in the range 3.70–26.82 ppm
135 and Nb/Ta ratios of 12.84–19.26, with a mean value of 15.58, and are so mostly
136 subchondritic (Fig. 5). Conversely, leucosomes within these granulites, which
137 represent anhydrous melts generated *in-situ* during UHT metamorphism, have a
138 distinct bulk rock composition from the restites. These leucosomes have Nb contents
139 of 0.36 ppm to 11.41 ppm, and Nb/Ta ratios that are mostly suprachondritic in the
140 range of 17.15 to 42.51, and a mean of 23.46 (Fig. 6a).

141

142 **4.2 Modeling results**

143 If melt produced during metamorphism is allowed to leave its host rock when
144 it reaches a critical melt connectivity transition (MCT) of 7 vol. % (Rosenberg and
145 Handy, 2005), petrological modeling of equilibrium phase assemblages must take into
146 account repeated melt loss and the production of an increasingly refractory residuum.
147 Isobaric heating at 0.8 GPa from subsolidus conditions (700 °C) to UHT conditions
148 (1000 °C) was used to simulate thermal metamorphism of the lower continental crust.
149 At these conditions, five ‘melt loss events’ (MLE) would occur at 754 °C, 780 °C, 808
150 °C, 828 °C, and 913 °C (Fig. 6b, 7). The first melt produced during metamorphism
151 forms via muscovite dehydration melting, whereas melt generated in MLE 2 to 4 is
152 produced by biotite dehydration melting. Melt lost in MLE 5 formed via anhydrous
153 breakdown of feldspar and quartz. All melt fractions show a progressive increase in
154 ASI [molar $\text{Al}_2\text{O}_3/(\text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O})$] with increasing temperature, and MLE 1–3
155 show increasing but subchondritic Nb/Ta ratios of 7–15. MLE 4 has a Nb/Ta ratio of
156 20, which is close to chondritic, whereas the last two melts have suprachondritic
157 Nb/Ta ratios of 25, which are most consistent with bulk compositions recorded by
158 natural granulites (Fig. 6b).

159

160 **4.3 Rutile trace elements**

161 Rutile grains in HT granulite sample 18MJ09 has Zr concentrations mostly
162 ranging from 644 ppm to 2924 ppm (Fig. 8a). Based on the results of pressure-
163 corrected Zr-in-rutile thermometry (Tomkins et al., 2007), sample 18MJ09 registered

164 a peak temperature of 883 °C at 1.1 GPa, as suggested by Wu et al. (2016). Niobium
165 concentrations in rutile are mostly 1321–2257 ppm, and Nb/Ta ratios are 16.32–32.62
166 with a mean value of 20.67; thus, are slightly supra-chondritic (Fig. 8a). In a plot of
167 Zr against Nb/Ta, rutile grains with high Nb/Ta have high Zr concentrations –
168 indicating that they formed at higher temperatures – and so are inferred to have
169 formed from a biotite-breakdown peritectic reaction (Fig. 8a).

170 Rutile in UHT granulite sample 16TG53 has Zr concentrations ranging from
171 314 to 6708 ppm (Fig. 8b), corresponding to temperatures of 645–980 °C at using 0.8
172 GPa (cf. Huang et al., 2019). Niobium concentrations in rutile are 607–3962 ppm with
173 a mean of 2782 ppm, and Nb/Ta ratios range from 10.32 to 34.99 with a mean value
174 of 17.54. Some individual rutile inclusions within garnet have much lower Nb
175 concentrations but higher Nb/Ta, and some retrograde rutile grains around garnet have
176 lower Zr concentration but higher Nb/Ta ratios (Fig. 8b).

177

178 **5 Discussion**

179 **5.1 Intracrustal differentiation during granulite-facies metamorphism**

180 Several episodes of partial melting are known to occur in felsic metamorphic
181 rock types during prograde metamorphism, driven by the breakdown of various
182 hydrous and anhydrous constituent minerals (Brown, 2013; Sawyer et al., 2011). Loss
183 of melt from these host rocks and vertical ascent to shallower levels induces
184 intracrustal differentiation, which is the main form of mass transfer that controls the
185 evolution of the continental crust (Brown et al., 2011).

186 At metamorphic conditions below the granulite facies, fluid-present melting
187 may occur in pelitic rocks at temperatures as low as ~650 °C by simultaneously
188 consuming grain-boundary fluid and anhydrous felsic minerals (Sawyer et al., 2011).
189 However, as neither biotite nor rutile are involved in these reactions, the Nb/Ta ratio
190 of the lithological system would not show any significant change, despite a small
191 reduction in net volume (Clemens and Droop, 1998). Once free fluids are exhausted,
192 incongruent melting of hydrous minerals occurs at higher temperatures, with these
193 breakdown reactions considered as the primary mechanism for generating crustal (S-
194 type) granite (Brown, 2013). Partial melt in the source rock may generate substantial
195 overpressure, leading to fracturing and so aiding subsequent melt loss (Clemens and
196 Droop, 1998). Our modeling of this process indicates that metapelites can lose up to
197 27 vol. % of their original mass via extraction of five discrete melt fractions (Fig. 6b,
198 7). The heat-producing element (U, Th and K) budget of the source would be
199 progressively depleted, as these elements are incompatible and will readily enter the
200 silicate melt prior to its extraction (Ewing et al., 2014). The bulk rock Nb/Ta ratio at
201 this stage is thus controlled by biotite stability, since biotite is ubiquitous but peritectic
202 rutile is scarce. As biotite preferentially incorporates Nb over Ta (Stepanov and
203 Hermann, 2013), anatexis would produce a restite with a high Nb/Ta ratio and melt
204 with a low Nb/Ta ratio (Fig. 6b, 9). The Nb/Ta ratio in rutile also shows an increase
205 during heating (Fig. 8a). With biotite totally consumed at about 840 °C, the restites
206 are usually characterized by a strongly depletion of heat-producing elements (e.g.
207 Ewing et al., 2014). Even though the rate of melt production would dramatically

208 decrease (Sawyer et al., 2011), the process of partial melting would generate a low
209 Nb/Ta restite and high Nb/Ta melt (Fig. 6, 9), since rutile is the main host mineral and
210 preferentially incorporates Ta over Nb (Klemme et al., 2005).

211 In UHT granulite sample 16TG53, most rutile is characterized by
212 subchondritic Nb/Ta values, which controls the bulk-rock Nb/Ta ratio (Fig. 8b).
213 However, several rutile inclusions and retrograde rutile grains around garnet have
214 much higher Nb/Ta. Among these, high Nb/Ta rutile inclusions are most likely
215 inherited rutile derived from mafic rocks (Meinhold, 2010). Since Nb and Ta are
216 strongly incompatible in garnet, Nb and Ta concentrations within garnet could be well
217 preserved even during UHT metamorphism (Fulmer et al., 2010). High Nb/Ta
218 retrograde rutile grains are most likely to have been produced with garnet and K-
219 feldspar during UHT melting, although this is expected to only produce small
220 absolute volumes of leucosome.

221

222 **5.2 Potential high Nb/Ta reservoirs**

223 HT and UHT granulites in this study show distinctly different suprachondritic
224 and subchondritic Nb/Ta ratios, respectively, which is a pattern that is consistent with
225 most other granulites around the world. For example, Fig. 10 summarizes the Nb/Ta
226 frequency of typical HT and UHT granulites, including well-studied examples from
227 the Ivrea Verbano zone, north Italy, and Napier complex, Antarctic (Ewing et al.,
228 2014; Grew et al., 2006). The residua of HT granulite have previously been proposed
229 as a high Nb/Ta reservoir (Stepanov and Hermann, 2013); however, we find that the

230 Nb/Ta of the residue would be a function of the amount and type of Nb and Ta host
231 mineral in the residual as well as the initial bulk rock Nb/Ta. Given an average pelitic
232 bulk rock composition (Nb/Ta = 11) (Taylor and McLennan, 1985), our modeling
233 results show that the Nb/Ta of the residue would always be subchondritic (Fig. 9).

234 Leucosomes in UHT terranes are thought to be crystallized melt derived from
235 *in-situ* anatexis of their host rocks, and usually have a suprachondritic Nb/Ta ratio.
236 However, a summary of 82 S-type granite samples from the GEOROC database
237 (<http://georoc.mpch-mainz.gwdg.de/georoc/>), which were derived from segregation,
238 ascent and emplacement of relatively low temperature melt generated by biotite
239 dehydration (Clemens and Stevens, 2016), show Nb/Ta values in the range 2.10–
240 26.50 (mean = 7.82), with most being subchondritic (Fig. 6a). These data are
241 consistent with our modeling results (Fig. 6b) and indicate that the different Nb/Ta
242 ratios of restites and leucosomes produced during UHT metamorphism can facilitate
243 strong fractionation of these elements throughout the continental crust, if this melt is
244 allowed to migrate away.

245 As such, it is reasonable to consider that A-type granite, which is derived from
246 anatexis of dry refractory granulite residue (Collins et al., 1982), would have a
247 suprachondritic Nb/Ta ratio and so represent a relatively high Nb/Ta reservoir within
248 the continental crust. Nonetheless from protolith, ascent, mixing, and emplacement
249 (Clemens, 2012; Brown, 2013). High Nb/Ta minerals such as biotite may fractionate
250 during any of these processes, leading to a decrease of Nb/Ta in the melt (Stepanov et
251 al., 2014). In addition, magma mixing and contamination from other low Nb/Ta

252 source would also induce a chemical change in the melt during its ascent (e.g.
253 Korhonen et al., 2015). Then, anhydrous melt – if unfractionated and lacking
254 contamination – might keep a high Nb/Ta ratio, which means *in-situ* leucosome, in-
255 source leucosome and some leucocratic veins in UHT terranes would be a potential
256 high Nb/Ta reservoir., most A-type granites are characterized by high Nb but scattered
257 Nb/Ta ratios (e.g. Auwera et al., 2003; Xue et al., 2018). This can be accounted for by
258 granite generation caused by episodic melt production, extraction

259 We find that *in-situ* to in-source leucosome and leucocratic veins in UHT
260 terranes are a potentially high Nb/Ta reservoir within the continental crust, but the
261 residua of HT granulite are not. As UHT granulites occur at extensional tectonic
262 settings or in overriding plates above Precambrian subduction zone (Sizova et al.,
263 2014), these places would be suitable to find high Nb/Ta reservoirs. Thus, it is
264 possible that these domains are severely under-represented in estimates of the
265 elemental composition of the continental crust.

266

267 **Acknowledgements**

268 This work is financially supported by National Natural Science Foundation of
269 China (grant No. 41890832, 41902057) and China Postdoctoral Science Foundation
270 (2019M650834).

271

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409

410 **Figure captions**

411 **Figure 1**(a) Tectonic subdivision of the North China Craton (modified after Zhao and
412 Zhai, 2013). EB: Eastern Block; YB: Yinshan Block; OB: Ordos Block;
413 TNCO: Trans–North China Orogen; KB: Khondalite Belt; the red star
414 represents location of Fig. 1b. (b) Geological sketch map of the study area
415 (modified after Guo et al., 2002).

416 **Figure 2** Field photographs of HT-HP and UHT granulite from study area. (a) shows
417 typical HT-HP pelitic granulite sample 18MJ09 collected from Manjingou
418 area, northern TNCO. (b) shows minor degree of partial melting in HT-HP
419 granulite. (c) UHT sample 16TG53 from Tuguiwula area, east Khondalite
420 belt. (d) shows UHT granulite experienced intensive partial melting process.

421 **Figure 3** Photomicrographs of HT-HP granulite, 18MJ09. (a) and (b) kyanite occur
422 as inclusion within garnet rim, but absent in matrix. (c) shows inclusions
423 within poikiloblastic garnet core represent prograde mineral assemblage
424 (M0), and rutile occur as single inclusion in garnet (M1) and in matrix (M2).
425 (d) rutile and biotite inclusions within garnet (M1) and rutile in matrix.

426 **Figure 4** Sample photomicrographs of UHT granulite, 16TG53. (a) single rutile
427 inclusion along with quartz and sillimanite within garnet, representing

428 prograde stage rutile (M0). (b) single rutile inclusion within garnet (M0) and
429 rutile in matrix next to felsic mineral (M1). (c) shows combined inclusion in
430 garnet with biotite (M0), rutile in matrix next to felsic minerals and garnet
431 (M1). (d) shows rutile in matrix next to felsic minerals (M1) and tiny
432 retrograde rutile along with biotite surrounded garnet (M2).

433 **Figure 5** Bulk rock composition plots of Nb/Ta vs Nb of granulite sample from study
434 area and other granulite terranes. Geochemical data summarized from:
435 Trivandrum Block, South India (Nandakumar and Harley, 2019); Ivrea
436 Verbano Zone, north Italy (Ewing et al., 2014); Ulashan-Daqingshan terrane,
437 Khondalite Belt (Cai, 2014); Bhopalpatnam granulite belt, central India
438 (Vansutre et al., 2013); Napier complex, Antarctic (Grew et al., 2006); Altay,
439 northwestern China (Yang et al., 2015); Highland complex, Sri Lanka (Table
440 S3).

441 **Figure 6(a)** Plots of bulk-rock Nb/Ta vs A/CNK from statistical S-type granite and
442 leucosomes within UHT granulites. (b) Plots of bulk-rock Nb/Ta vs A/CNK
443 from modeling results.

444 **Figure 7** P–T mosaic pseudosection panels of metapelite at open system along
445 isobaric heating at 0.8 GPa, contoured with isopleth of mol. % melt. The
446 mineral proportion against temperature is shown below.

447 **Figure 8** Plots of Nb/Ta vs Zr of rutile in granulite sample 18MJ09 and 16TG53.

448 **Figure 9** Nb/Ta against temperature diagrams of modeling results.

449 **Figure 10** Statistical Nb/Ta frequency of typical pelitic granulite.