

1 Contamination of heterogeneous lower crust in Hannuoba tholeiite: evidence from in
2 situ trace elements and strontium isotopes of plagioclase Word Count:6669 Revision 1

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13 **Abstract**

14 The Hannuoba basalt, located in the northern margin of the North China Craton,
15 is a typical intracontinental basalt with ocean island basalt-like geochemical features
16 and has been extensively studied. However, its origin and deep processes, such as
17 magma mixing and crystallization conditions, are still unclear. To further understand
18 the mechanisms leading to the compositional heterogeneity and magmatic processes
19 of Hannuoba basalt at crustal and/or mantle depth, in-situ major element, trace
20 element, and ⁸⁷Sr/⁸⁶Sr compositional heterogeneity of four representative plagioclase
21 crystals in three Hannuoba tholeiite samples, as well as whole-rock major and trace
22 elements data, are reported. According to the petrographic characteristics, the basalts

23 are divided into fine-grained and coarse-grained groups. The anorthite content in
24 plagioclase of samples varies in a small range (56-64%), but the content of trace
25 elements in plagioclase from the coarse-grained samples is generally higher than that
26 of the fine-grained samples. Clinopyroxene-melt equilibrium thermobarometer and
27 plagioclase-clinopyroxene magnesium and rare earth element exchange thermometer
28 show that the magma for the two types of basalt was stored and crystallized at a
29 similar depth, and crystallized within a 20 °C (fine-grained basalt) and 50°C
30 (coarse-grained basalt) temperature window, which may be a reason for the grain size
31 differences between the two types of basalts. We found that $^{87}\text{Sr}/^{86}\text{Sr}$ of all the studied
32 plagioclase crystals varied from 0.70333 ± 0.00018 (2SE) to 0.70556 ± 0.00031 (2SE),
33 a much large range than the whole rock of Hannuoba basalts reported previously and
34 consistent with that of Cenozoic basalts in North China. Therefore, at least two kinds
35 of melts with significant differences in isotope and minor heterogeneity in major and
36 trace elements are injected into each magma plumbing system. The content of trace
37 elements in the Hanuoba tholeiite is between the Hanuoba alkaline basalt and the
38 lower crust, which can be explained by the mixing of the alkaline basalt and the lower
39 crust, but the low $^{87}\text{Sr}/^{86}\text{Sr}$ (<0.704) characteristics of plagioclase cannot come from
40 alkaline basalt, because trace element in the plagioclase is not in equilibrium with the
41 alkaline basalt. Therefore, we believe that the compositional heterogeneity of
42 Hannuoba tholeiitic basalt is mainly caused by the mixing of heterogeneous lower
43 crust rather than different mantle-derived melts, which indicates the contribution of
44 the continental lower crust to the continental basalt is more complicated than

45 previously recognized.

46

47 **Keywords:** Basalt, plagioclase, strontium isotope, magmatic process, lower
48 continental crust, Hannuoba

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Introduction

53 A large number of studies show that the lower continental crust plays an
54 important role in the geochemical diversity of continental basalts (Glazner et al., 1991;
55 Jiang and Zhi, 2010; Liu et al., 2008b; Lustrino, 2005; Xu et al., 2017; Zeng et al.,
56 2011). Many studies suggest that the signals of the lower continental crust imply that
57 it is a source of material for basalt through the recycling of crustal materials. (Liu et
58 al., 2008b; Xu et al., 2017; Zeng et al., 2011). Others, however have suggested that
59 the contamination of the lower continental crust during the ascent of basaltic magma
60 may be an important reason for the compositional diversity of continental basalts, but
61 this contamination is usually very difficult to identify (Glazner and Farmer, 1992;
62 Glazner et al., 1991; Jiang and Zhi, 2010) because the lower continental crust is
63 mainly composed of mafic rocks with heterogeneous compositions (Rudnick and
64 Fountain, 1995). Moreover, the heterogeneity becomes more significant through
65 partial melting of the lower crust, crystallization differentiation, and mixing various
66 mantle-derived magmas.

67 Hannuoba basalt is a typical continental ocean island type basalt in that its rock
68 types, petrography, and geochemistry have significant diversity. Although there is a
69 long history of research, there is still controversy about how the lower continental
70 crust affects the diversity of Hannuoba basalts (Basu et al., 1991; Guo et al., 2016;
71 Jiang and Zhi, 2010; Liu et al., 1994; Qian et al., 2015; Song et al., 1990; Xu et al.,
72 2017; Yang et al., 2016; Zhi et al., 1990). The systematic whole rock data of major
73 and trace elements, as well as Sr, Nd, and Pb isotopes, seem to indicate that the
74 ancient lithospheric mantle (Basu et al., 1991; Liu et al., 1994; Song et al., 1990) or
75 recycled ancient oceanic crust and sediments (Qian et al., 2015; Xu et al., 2017) may
76 be the source of Enrich Mantle I (EMI) end components in Hannuoba tholeiite.
77 However, recent Os isotopic data show that the very high Os isotopic ratios,
78 especially tholeiites, can only be explained by significant crustal contamination (Jiang
79 and Zhi, 2010) rather than recycled products of the lower continental crust (Liu et al.,
80 2008b). However, the details of crustal contamination are still unclear. One of the
81 critical reasons may be that previous studies usually focus on the geochemical
82 characteristics of the whole rock scale. The complex processes of crustal
83 contamination and magmatic mixing are more commonly recorded in mineral growth
84 zoning, but whole-rock compositions that average the signals of various minerals and
85 glasses will weaken or eliminate the information recorded in minerals. Recent studies
86 on melt inclusions in olivine have shown that melt inclusions with very heterogeneous
87 isotopes but relatively homogeneous major and trace elements contribute to Hannuoba
88 tholeiite (Qian et al., 2015). However, these melt inclusions probably only record a

89 particular crystallization stage in the magma evolutionary history, and it may be
90 homogenized during the crystallization process. This suggests that more diverse
91 materials may be involved in the Hannuoba basalt magmatic system.

92 Plagioclase is an ideal material for strontium isotope analysis because of its high
93 strontium content and low rubidium content, and it is a widely existing rock-forming
94 mineral in igneous rocks. In the past few decades, with the development of analytical
95 techniques, a large number of studies have begun to use in-situ Sr isotopes of
96 plagioclase to reveal the complex evolutionary history of the magmatic system
97 (Davidson et al., 2001; Davidson and Tepley, 1997; Ramos et al., 2004; Ramos et al.,
98 2005; Tepley et al., 2000; Yan et al., 2020). Many researchers have found that the
99 strontium isotope heterogeneity of plagioclase is far more significant than that of
100 whole-rock scale (Edwards et al., 2019; Lange et al., 2013), and there is a significant
101 strontium isotopic disequilibria between phenocrysts and matrix, as well as within a
102 single grain, which is considered to be significant evidence of magma mixing,
103 contamination and contribution of multiple source materials (Davidson et al., 2001).
104 When the isotopic, minor, and trace elements data from the core to the rim of
105 plagioclase are combined with the geochemical characteristics of the whole rock, the
106 magma evolution history from crystal nucleation to eruption can be understood, and
107 the crustal contamination and mixing of different mantle-derived magmas can be
108 effectively distinguished, although some of the information might be lost due to the
109 dissolution of plagioclase (Ginibre and Davidson, 2014; Hagen-Peter et al., 2019;
110 Ramos and Reid, 2005).

111 In this study, the in-situ strontium isotope, major and trace elements of
112 plagioclase, combined with the major and trace elements of the whole rock, are used
113 to clarify or explain the contribution of the lower continental crust during the
114 formation of Hannuoba tholeiite. We find that the range of strontium isotope recorded
115 by plagioclase is much more extensive than that recorded by Hannuoba alkaline and
116 tholeiitic whole-rock compositions, but it is consistent with the observed range
117 Cenozoic basalts in the North China Craton. Although the mixing of the alkaline
118 basalt and the lower crust can explain that the content of trace elements in the
119 Hannuoba tholeiite is between the Hannuoba alkaline basalt and the lower crust, the low
120 $^{87}\text{Sr}/^{86}\text{Sr}$ (<0.704) characteristics of plagioclase cannot come from alkaline basalt,
121 because trace element in the plagioclase is not in equilibrium with alkaline basalt.
122 Therefore, we believe that the compositional heterogeneity of Hannuoba tholeiitic
123 basalt is caused by mixing heterogeneous lower crust rather than the mixing of
124 different mantle-derived melts, which indicates the contribution of continental lower
125 crust to continental basalt is more complicated than previously recognized.

126

127 **Geological setting and samples**

128 The North China Craton is one of the three largest cratons in China (Fig.1A),
129 which has attracted international attention due to its relatively complete record of
130 geological events from Archean to Cenozoic (Zhao et al., 2001). Cenozoic continental
131 intraplate basalts with geochemical characteristics of oceanic island basalts are widely
132 distributed in the North China Craton (Chu et al., 2017; Fan and Hooper, 1991; Yang

133 and Zhou, 2013; Zou, 2000). The Hannuoba basaltic area, located 200 Km
134 northwest of Beijing (Fig.1A), is an important occurrence because of its large
135 area ($> 1700 \text{ Km}^2$), compositional diversity in intercalated tholeiitic and
136 alkaline lavas, and abundant mantle xenoliths in the alkaline basalts (Zhi et al.,
137 1990). Previous studies suggest that Hannuoba volcanism was controlled by
138 deep fractures and ranges in age from Miocene in the east to Pliocene in the
139 west (Fig.1B) (Zhi et al., 1990). Geochemical analysis of whole-rock and
140 olivine hosted melt inclusions has indicated that Hannuoba tholeiitic basalts
141 have relatively homogeneous characteristics of major and trace elements but
142 vary significantly in radiogenic isotopes compared with the alkaline basalt
143 (Jiang and Zhi, 2010; Qian et al., 2015; Song et al., 1990). Source
144 heterogeneity, magma mixing, high-pressure fractionation, and crustal
145 contamination have been suggested to account for compositional diversity in
146 Hannuoba basalts (Jiang and Zhi, 2010; Qian et al., 2015; Song et al., 1990;
147 Yang et al., 2016; Zhi et al., 1990). Three tholeiitic basalts samples studied in
148 this paper were collected from the eastern part because they are well exposed,
149 and the stratigraphy is clear (Fig.1B). Their spatial relationship is shown in Fig.
150 1C. Samples SQB03 and JSB04 are collected from the lavas erupted before the
151 eruption of alkaline basalt, and JSB12 is collected from relatively late lavas
152 after the eruption of alkaline basalt. (Fig. 1C).

153

154

Analytical methods

155 Petrography and mineral major element analysis

156 Thin sections (30 μm thickness) were prepared from representative billets cut
157 from the three samples, and optical and electrical microscopy was carried out on all
158 thin sections to document petrographical features. High-resolution backscattered
159 electron images were obtained at the Research Center of Genetic Mineralogy, China
160 University of Geosciences (Beijing), using a field emission MIRA3 XMU scanning
161 electron microscope. Electron-probe micro-analysis (EPMA) of individual plagioclase
162 and clinopyroxene phenocrysts was carried out at Wuhan Sample Solution Analytical
163 Technology Co., Ltd. using a JXA-8230 electron probe micro-analyzer (EPMA) under
164 operating conditions of 20 nA, an accelerating voltage of 15 keV, and beam size of 1
165 μm in diameter for clinopyroxene and 3 μm for plagioclase. Peak and background
166 counting times were 10 and 5 s for Si, Ca, Na, Al, Fe, and Mg, 20 and 10 s for Ti, Mn,
167 Cr, K, Ni, and P. Analytical results were corrected using the ZAF correction routines.
168 The standards used were SPI standard mineral and were analyzed as internal standards
169 to monitor data quality. Analyses are accurate to 1–2% for major elements (>10 wt%)
170 and 2–10% relative for minor elements (0.5–10% wt%). (Supplementary Tables 1,2
171 and 3)

172 In-situ mineral trace element analysis

173 In situ trace element analyses of plagioclase and clinopyroxene were carried out
174 using laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) in
175 the mineral laser microprobe analysis laboratory (Milma laboratory) of China

176 University of Geosciences (Beijing). The Agilent 7900 ICP-MS instrument was
177 coupled to a resonetic 193 nm excimer laser ablation system. Single spot ablation was
178 adopted with a laser beam of 50 μm , and ablation frequency was 8 Hz. Helium gas
179 was used as a carrier gas. The detailed parameter settings of the instrument and
180 experimental process can be found in (Zhang et al., 2019). NIST610 was used as an
181 external standard, while ^{43}Ca was selected as the internal standard to correct the
182 element composition. The Ca content in the samples is from the data obtained by
183 EPMA. The precise value of Ca content in NIST 610 can be seen in (Jochum et al.,
184 2011). The trace elements data were processed using the Iolite software based on the
185 Igor platform (Paton et al., 2011). We analyzed the USGS reference glass BCR-2G as
186 an unknown sample, and the results were within the error range of the recommended
187 values (Supplementary Tables 4 and 5) which may be referred to (Jochum et al., 2005).
188 The analytical uncertainty is better than 10% (relative).

189 **In-situ Sr isotope analysis**

190 In-situ Sr isotope measurements were performed on a Neptune Plus MC-ICP-MS
191 (Thermo Fisher Scientific, Germany) in combination with a J-200 343 nm
192 femtosecond laser ablation system (Applied Spectra, USA) housed at the National
193 Research Center for Geoanalysis, Chinese Academy of Geological Sciences (CAGS),
194 Beijing, China. The JET sample and X skimmer cones were used along with the guard
195 electrode (GE), and all measurements were conducted under the low resolution and
196 static mode. At the beginning of every analytical session, the fs-LA-ICP-MS system
197 was optimized using NIST 612 to achieve maximum signal intensity and low oxide

198 rates. Samples were ablated inline mode with a spot size of 30 μm , line length of 20
199 μm , stage movement speed of 0.65 $\mu\text{m/s}$, laser repetition rate of 8 Hz, and beam
200 energy density of 1.5 J/cm^2 . The instrumental mass bias for Sr isotopes was corrected
201 using an exponential law function based on $^{86}\text{Sr}/^{88}\text{Sr}$ value of 0.1194. Correction of
202 interferences of Kr isotopes on mass 84 and 86 was accomplished by background
203 subtraction. The interferences of doubly charged ions of $^{168}\text{Er}^{2+}$ on ^{84}Sr , $^{170}\text{Er}^{2+}$ and
204 $^{170}\text{Yb}^{2+}$ on ^{85}Rb , $^{172}\text{Yb}^{2+}$ on ^{86}Sr , and $^{174}\text{Yb}^{2+}$ on ^{87}Sr were corrected based on the
205 measured signal intensities of $^{167}\text{Er}^{2+}$ (m/z 83.5) and $^{173}\text{Yb}^{2+}$ (m/z 86.5) and the natural
206 isotopic composition of Rb, Er and Yb (Li et al., 2018). The determined $^{87}\text{Sr}/^{86}\text{Sr}$ of
207 Durango standard was 0.70675 ± 0.00012 (2SD, n = 9) (Supplementary Table 6),
208 which is slightly higher than the results determined by previous studies with $^{87}\text{Sr}/^{86}\text{Sr}$
209 of 0.70629 ± 0.00002 to 0.70638 ± 0.00013 (2SD, n = 8) (Yang et al., 2014) and
210 references therein; however, $^{87}\text{Sr}/^{86}\text{Sr}$ for every single point ranges from $0.70670 \pm$
211 0.00027 to 0.70689 ± 0.00033 (2SE) which is consistent with the literature results
212 within the error range. $^{84}\text{Sr}/^{86}\text{Sr}$ was also monitored during analyses, and the average
213 values are 0.05596 ± 0.00034 (2SD, n = 9) for Durango standard and $0.05667 \pm$
214 0.0028 for all in-situ plagioclase points, which agree well with the “true” value of
215 0.0565 (2SD, n = 27). Further analysis of the reliability of the Sr isotope data will be
216 presented in the discussion section.

217 **Whole-rock major and trace element analysis**

218 Quantitative analyses of major elements contents in whole rocks were conducted
219 using X-ray Fluorescence Spectrometer (XRF) at the Ore Deposit Geochemistry

220 Microanalysis Laboratory, affiliated to the State Key Laboratory of Geological
221 Processes and Mineral Resources, China University of Geosciences (Beijing). The
222 laboratory is equipped with Shimadzu's wavelength dispersive X-ray fluorescence
223 spectrometer. The fused beads method was used in the preparation of test specimens.
224 Mixed weighed amounts of sample and flux and placed the mixture in a clean
225 platinum/gold crucible. A 0.7 g of sample (200 mesh) plus 7.0 g of mixed lithium
226 borate fluxes were used in this study. The addition of a small amount (100 mg) of a
227 halide, such as LiBr, acted as a release agent when using platinum/gold molds. The
228 mixture was heated in a high-frequency fusion machine at a fixed temperature,
229 usually from 950 °C to 1100 °C, until thoroughly melted. Then the fused beads were
230 loaded into the XRF instrument for determination; the fusion time was about 30 min.
231 The measurement procedure and data quality were monitored by repeat analysis of
232 international basalt standards BCR-2 and BHVO-2. The analytical precision (RSD,
233 relative standard deviation) and accuracy (RE, relative error between measured and
234 recommended values) are better than 5% for major elements, with many elements
235 agreeing to within 2% of the reference values (Supplementary Table 7).

236 All whole-rock trace element compositions were measured by ICP-MS (Agilent
237 7500a with a shielded torch) at the State Key Laboratory of Geological Processes and
238 Mineral Resources, China University of Geosciences (Wuhan). The detailed
239 sample-digestion procedure for ICP-MS analyses has been described by (Liu et al.,
240 2008a). The reproducibility is better than 5% (RSD) for all elements. The accuracy,
241 determined relative to reference values of BHVO-2 and BCR-2, is better than 5% (RE)

242 (Supplementary Table 7).

243

244

Results

245 Petrography

246 Representative thin-section photographs of the samples are presented in Figure 2.

247 The petrographic characteristics of the basalts may be divided into a fine-grained

248 group (samples SQB03A, SQB03B, and JSB04) and a coarse-grained group (sample

249 JSB12). For the fine-grained group, the most common phenocryst phase is plagioclase

250 feldspar (~10-15 vol%), followed by clinopyroxene (~5-10 vol%) with trace amounts

251 of olivine (<5 vol%). The plagioclase phenocrysts are euhedral and tabular, with sizes

252 (length) ranging from ~400 μm to ~3 mm, and crystals of all sizes exhibit concentric

253 zoning. Clinopyroxene phenocrysts are euhedral to rounded and commonly form

254 glomerocrysts with plagioclase and olivine (Figure 2). Simple twinning of

255 clinopyroxene is common, and phenocrysts range up to ~1 mm in size. Equant olivine

256 phenocrysts (0.2–2 mm) are present. All phenocrysts are set in fine-grained

257 plagioclase, clinopyroxene, olivine, and Fe-Ti oxide matrix. Although the overall

258 grain size of the coarse-grained group is larger than that of the fine-grained group, its

259 phenocrysts and matrix minerals are not easy to distinguish (sample JSB12 in Fig. 2).

260 For the coarse-grained group, the major rock-forming minerals are plagioclase (~45

261 vol%) and clinopyroxene (~25 vol%), followed by olivine (~15 vol%) and K-feldspar

262 (~10 vol%) with small amounts of Fe-Ti oxide (~5 vol%). Plagioclase phenocrysts are

263 typically tabular-shaped in all the samples. The most significant difference between

264 the fine-grained and coarse-grained samples is that the former has an overall smaller
265 grain size but has some larger plagioclase phenocrysts (Fig. 2).

266 **Whole-rock major and trace element**

267 Whole-rock major and trace element data are presented in Supplementary Table 7.
268 All the samples are classified as basalt (Supplementary Fig. 1) and plot within the
269 range of Hannuoba tholeiitic basalt samples collected previously (Zhi et al., 1990).
270 The visible freshness of the lavas chosen for this study is confirmed by low loss on
271 ignition (LOI) values that range from 0.15 to 3.01 wt%, and the minerals in the
272 samples are fresh and have not undergone alteration. Compared with the fine-grained
273 samples, the coarse-grained samples have higher TiO₂, FeO, K₂O, and P₂O₅ content.
274 Other oxides contents are very similar in the two types of samples.

275 All the samples have a smooth chondrite-normalized rare earth element (REE)
276 pattern with a slight positive Eu anomaly (Supplementary Fig. 2,3). LREE and MREE
277 contents in the coarse-grained samples are higher than those in the fine-grained
278 samples, but their HREE contents are similar. Abundances of trace elements,
279 normalized to values for the primitive mantle (McDonough and Sun, 1995), show
280 typical continental oceanic island basalt-like pattern with marked depletion in Pb, Th,
281 and U, and enrichments in Ba, Nb, Ta, and Sr (Fig. 3), which is consistent with data
282 reported previously (Qian et al., 2015; Zhi et al., 1990). All the incompatible element
283 contents and Ce/Pb, Nb/U, and Nb/La ratios in the coarse-grained samples are higher
284 than those in the fine-grained samples (Supplementary Table 7).

285 **Mineral major and trace element**

286 The major and trace element data of all minerals are provided in Supplementary
287 Tables 1-5. From the backscattered electron (BSE) images, it can be observed from
288 Fig.4 that compositional zoning in plagioclase within a given sample generally
289 consists of only oscillatory zoning type, and zoning is consistent between crystals.
290 Representative backscattered electron images of four individual plagioclase crystals
291 from the three samples and variation trends diagrams for An, Mg, Ti, and Sr from the
292 interior to the rim are shown in Fig. 4. Given that the spot size for in-situ trace
293 element measurement is $\sim 50\mu\text{m}$, which is significantly wider than the width for a
294 single zoning, detailed EPMA data with data point distance ranging from $5\mu\text{m}$ to
295 $40\mu\text{m}$ are shown in Fig. 5 and Supplementary Table 2.

296 Plagioclase phenocrysts in the fine-grained samples (SQB03A, SQB03B, and
297 JSB04) are oscillatory zoned, with occasional dissolution surfaces. The An content
298 fluctuates between 57 and 64 when the distance from the rim is more than $100\mu\text{m}$
299 (Figs 4 and 5) and decreases significantly to 32~56 in the rim as displayed by EMPA
300 profiles (Fig. 5). Mg and Sr are positively correlated with An content, while other
301 trace elements are scattered at a given An content (Fig. 6). Partially resorbed zones are
302 richer in Ca (An=62~64), Sr (800~900 ppm) and Mg (>1000 ppm) than other zones
303 (Fig. 4), which is interpreted as reflecting a more mafic recharge event by a Ca and Sr
304 richer magma.

305 Plagioclase phenocrysts in the coarse-grained sample JSB12 are also oscillatory
306 zoned but generally more sodic than those in the fine-grained samples. Two different

307 oscillatory zones may be identified, inner calcic zones with An fluctuate between 54
308 and 62 and outer sodic zones with An fluctuate between 48 and 57 (Fig. 5). The zone
309 with dissolution surface mainly occurs in the inner zones and has a thickness ranging
310 from 5 to 20 μ m. Partially resorbed zones with the embayed surface are richer in Ca
311 (An=60~62) and Sr (1150~1300ppm) than other zones (Fig. 4). Sr is positively
312 correlated with An content, while other trace elements are scattered at a given An
313 content, and Ti, Sr, Ba, La, and Eu contents are generally higher than those in the
314 fine-grained samples (Fig. 6).

315 The chondrite-normalized diagram (supplementary Fig. 3) drawn according to the
316 in-situ trace element data of plagioclase indicates that plagioclase phenocrysts from
317 all the studied samples show enrichment in light rare earth elements and strong Eu
318 positive anomalies. Variation of Eu anomalies ranges from 1.52 to 9.94. Except for a
319 few individual data, the abundance of rare earth elements in plagioclase phenocrysts
320 from the coarse-grained sample (JSB12) is higher than that in the other three
321 fine-grained samples. The clinopyroxene phenocrysts in the two groups have
322 comparable major element characteristics and range from En₄₃ Fs₁₂ Wo₄₄ to En₄₆ Fs₁₂
323 Wo₄₁ (Supplementary Table 3). All the clinopyroxene REE data show a similar pattern
324 with enriched MREE compared with LREE and HREE (Figure 7), and the
325 coarse-grained sample has overall higher LREE and lower HREE concentrations than
326 those in the fine-grained samples.

327 **Sr isotope in plagioclase**

328 The $^{87}\text{Sr}/^{86}\text{Sr}$ Sr ratios of plagioclase phenocrysts are presented in Supplementary
329 Table 6. All four plagioclase phenocrysts show large variation (>0.001) in $^{87}\text{Sr}/^{86}\text{Sr}$ Sr
330 ratio, with the lowest ratio of 0.70333 ± 0.00018 (2SE) in sample JSB12 and the
331 highest ratio of 0.70556 ± 0.00031 (2SE) in sample SQB03A (Figure 8). The average
332 $^{87}\text{Sr}/^{86}\text{Sr}$ of the fine-grained samples are 0.70434 ± 0.00112 (2SD) (SQB03A),
333 0.70471 ± 0.00102 (2SD) (SQB03B) and 0.70455 ± 0.00074 (2SD) (JSB04), which
334 all higher than the coarse-grained sample JSB12-1 of 0.70425 ± 0.00109 (2SD). The
335 variation of strontium isotope recorded by plagioclase is much larger than that
336 recorded by Hannuoba alkaline and tholeiitic whole rocks (Song et al., 1990), but it is
337 consistent with the variation range of Cenozoic basalts in North China Craton (Fig. 9).
338 Partially resorbed zones with higher An contents may have either higher or lower
339 $^{87}\text{Sr}/^{86}\text{Sr}$ Sr ratios than other zones in the fine-grained samples, the variation of Sr
340 isotope value may be characteristic of the dissolution zone or may be caused by the
341 adjacent bands (Fig. 9). Sample SQB03A show similar correlation trends between An
342 and $^{87}\text{Sr}/^{86}\text{Sr}$ Sr ratios with sample SQB03B, except the two rim points (Fig. 9),
343 suggesting a similar crystallization process occurred for different individual crystals,
344 which is consistent with their comparable zoning pattern as displayed by BSE images
345 (Figure 8). For sample JSB04, rim and core have a higher $^{87}\text{Sr}/^{86}\text{Sr}$ Sr ratio than other
346 zones. In contrast, partially resorbed zones in sample JSB12 are accompanied by a
347 significant decrease in the $^{87}\text{Sr}/^{86}\text{Sr}$ Sr ratio and an abrupt increase in the rim zones.

348 **Thermobarometers**

349 The REE in plagioclase-clinopyroxene geothermometer (Sun and Liang, 2017)
350 gives crystallization temperatures of 1088~1156°C for the fine-grained samples and
351 1113~1129°C for the coarse-grained sample (Supplementary Table 8.). Temperatures
352 calculated from the Mg-Ca coupled exchange thermometer of plagioclase and
353 clinopyroxene (Sun and Lissenberg, 2018) are 1078~1134°C for the fine-grained
354 samples and 1076~1079°C for the coarse-grained samples (Supplementary Table 8.).
355 Given that the two different thermometers were applied to the same samples, the
356 systematic differences between T_{Mg} and T_{REE} (Figure 10) are most likely due to the
357 differential diffusive responses of Mg and REE to changes in temperature after
358 crystallization of the cumulus minerals. The average difference between T_{Mg} and T_{REE}
359 (Figure 10) indicates that crystallization occurs within the range of 20°C (fine-grained
360 basalt) and 50°C (coarse-grained basalt). The crystallization of these samples are
361 complete within 20 to 50°C before eruption, which may be a reason for the textural
362 differences between the two types of basalts. The inverted initial temperatures
363 (1120-1140 °C) by Mg-REE coupled geothermometer are very close to T_{REE}
364 (1088-1156 °C), indicating a small diffusive redistribution of REE in Hannuoba basalt.
365 Cooling rates calculated by Mg-REE coupled geospeedometer (Sun and Lissenberg,
366 2018) suggest that the coarse-grained sample generally (0.004 °C/yr) cooled slower
367 than the fine-grained samples (0.0004–0.05 °C/yr), although a small number of
368 results, possibly due to the large error, are abnormal. (Supplementary Table 8.).
369 Clinopyroxene-melt equilibrium thermometer Eq.33 of (Putirka, 2008) and

370 barometer of (Neave and Putirka, 2017) have been suggested as the most accurate and
371 precise thermobarometer. Crystallization temperatures and pressures calculated from
372 the thermobarometer are 1129~1189 °C and 0.7 ~4.4 Kbar for the fine-grained
373 samples, and 1149~1159°C and 3.9 ~4.3 Kbar for the coarse-grained samples. The
374 temperatures are slightly higher than T_{REE} but agree within error (Supplementary
375 Table 8.). The temperature obtained by the rare earth element thermometers may be
376 regarded as the crystallization temperature of the phenocrysts because 1) the error on
377 the rare earth element exchange thermometers of plagioclase and pyroxene is smaller
378 than that of the pyroxene thermometers and 2) the rare earth element thermometers
379 are less affected by pressure and water concentrations (Sun and Liang, 2017).

380 Because the plagioclase mineral-melt equilibrium thermobarometer is
381 significantly affected by the water content in the melt (Putirka, 2008) and the initial
382 water content in Hannuoba tholeiitic basalt is unknown, this thermobarometer was not
383 considered in this study.

384

385 **Discussion**

386 **Reliability of in situ Sr isotope data**

387 Possible interference with in situ Sr isotope analysis includes Kr and Rb and other
388 elements (see Analytical methods section and Li et al. (2018)). Trace amounts of Kr
389 may exist as impurities in trace amounts of argon, which may affect the accuracy of
390 the data, because ^{84}Kr and ^{86}Kr interfere with ^{84}Sr and ^{86}Sr signals, respectively. This
391 interference was corrected by measuring the Kr blank of the gas, and based on the

392 measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratio at the known apatite standard, the effect proved to be valid.
393 ^{87}Rb is a significant interference for Sr isotopic analysis. Generally, an analysis of
394 ^{85}Rb is needed for corrections. The content of Rb in the plagioclase samples from
395 Hannuoba basalts is relatively low (<1ppm), which indicates that ^{87}Rb has a minimal
396 interference to the Sr isotope (<0.00001).

397 All the LA-ICPMS Sr isotope data in this study have more significant analytical
398 error (0.00024 of average 2σ error) than micro-drilling data (Davidson et al., 2001;
399 Ginibre and Davidson, 2014) mainly due to the spot size of $30\mu\text{m}$. The accuracy of
400 the data may be ensured because the data for the Durango standard analyzed in this
401 study agree well with the recommended values and within the analytical error. On the
402 other hand, the Sr isotope data for the compositional zones of plagioclase with a
403 thickness less than $50\mu\text{m}$ are very close or nearly indistinguishable (Figure 9),
404 suggesting that the precision of the Sr isotope is high enough to capture the isotopic
405 heterogeneities within individual plagioclase crystal.

406 **Magma mixing recorded in compositional and isotopic zoning**

407 There are at least three mechanisms to explain the variation in the An content of the
408 plagioclase: a hotter magma underplating increased the temperature of the magma,
409 leading to an increase in An content of plagioclase; reductions in the water content of
410 the melt through degassing or removing volatile fluids; changes in the composition of
411 the melt caused by magma recharge. We may distinguish these processes using
412 abundances and patterns of slow diffusing trace elements in plagioclase phenocrysts
413 because each process should result in different trace element responses and patterns.

414 The An values of Hannuoba samples varied between 56 and 64. We assume that
415 the melt's composition (not including volatiles) remains the same, and the changes of
416 the An values may be caused by a modest increase in temperature or by the reduction
417 of H₂O content in the melt. Although temperature does affect the distribution of Sr
418 between plagioclase and siliceous melt, this effect does not affect the relationship
419 between An value and Sr (Bindeman et al., 1998). Using the partition coefficient of
420 Bindeman et al. (1998), to change the temperature by 100°C as suggested by the
421 plagioclase REE thermometer would only change the concentration of Sr in
422 plagioclase 10-15ppm. Moreover, at a given temperature, when the An content varies
423 from 56 to 64, the Sr partition coefficient could only change 20%. However, the Sr
424 variation in the plagioclase phenocrysts in Hannuoba basalts is far beyond this range
425 (Figure 6). Therefore, changes in the concentration of trace elements and An in
426 plagioclase are not caused by changes in temperature alone. Blundy and Wood (1991)
427 found that there is almost no difference in the distribution coefficient of Sr between
428 hydrothermal and silicate melts, which means that H₂O has only a weak effect on the
429 distribution coefficient of Sr. Therefore, zoning patterns in plagioclase is most likely
430 attributable to changes in melt composition, and changes in melt chemistry that may
431 accompany changes in melt temperature and H₂O content.

432 The dissolution surface and the oscillatory changes in An content (Fig. 5) suggest
433 compositionally distinct pulses of melt. Also, the intracrystalline Sr isotope data suggest
434 some episodes of magma recharge are occurring.

435 Thus, magma mixing could be responsible for the compositional and isotopic

436 diversity in the plagioclase phenocrysts. The relatively large size of the LC-ICPMS
437 laser spot compared to the width of the individual oscillatory zones results in the
438 simultaneous analysis of multiple zones, providing an averaged composition, so the
439 observed variation should be considered a minimum. Even with the limited spatial
440 resolution of the laser-ablation system, our data demonstrate that some plagioclase
441 phenocrysts in Hannuoba basalts preserve significant isotopic variation and that the Sr
442 isotope compositions of these crystals are commonly higher or lower than bulk-rock
443 $^{87}\text{Sr}/^{86}\text{Sr}$ in Hannuoba basalts (Figure 9). To preserve intracrystalline Sr isotopic
444 disequilibria on the scale of 30 μm or less, plagioclase phenocrysts cannot have been
445 kept at magmatic temperatures ($\sim 1000^\circ\text{C}$) for significant periods (100 years) because
446 this would result in diffusive re-equilibration (Davidson et al., 2001; Ramos and Reid,
447 2005). This is consistent with the fast cooling rate obtained from
448 plagioclase-clinopyroxene Mg-REE coupled geospeedometer (Sun and Lissenberg,
449 2018) and a small diffusive redistribution of REE in Hannuoba basalt. The
450 intracrystalline disequilibria were most likely generated during episodic crystal
451 growth through interaction with isotopically distinct melts at crustal pressures,
452 possibly within the middle-crust magma chamber or plumbing system as recorded by
453 crystallization pressures from the barometer of clinopyroxene.

454 **Contamination of heterogeneous continental lower crust**

455 Two hypotheses have been proposed to account for compositional and isotope
456 variations in Hannuoba basalts. The first attributes them to the mixing of basaltic
457 melts generated from compositionally and isotopically distinct mantle sources with

458 negligible crustal contamination based on trace elements and Sr-Nd-Pb isotopic
459 characteristics (Qian et al., 2015; Song et al., 1990; Zhi et al., 1990). The second
460 suggests that the compositional characteristics result from mantle melts assimilating
461 crustal components (Jiang and Zhi, 2010) based on very high $^{187}\text{Os}/^{188}\text{Os}$. Although
462 the Nb/U ratio in Hannuoba alkaline and tholeiitic basalts are significantly higher than
463 average continental crust (Qian et al., 2015), lower crust contamination cannot be
464 excluded because the Nb/U ratio in the lower continental crust has been suggested to
465 be very heterogeneous (Gao et al., 1998; Huang et al., 2004; Rudnick and Fountain,
466 1995), and mafic lower crustal materials can have Nb/U up to 50 (Rudnick and
467 Fountain, 1995), which is very close to the values documented in the Hannuoba
468 tholeiitic basalts (Nb/U=46~51). In addition, the $^{187}\text{Os}/^{188}\text{Os}$ of the Hannuoba
469 tholeiites (0.15882 - 0.61607) are mostly higher than those of the transitional basalts
470 (0.23425-0.35779) and alkaline basalts (0.14869- 0.29528) (Jiang and Zhi, 2010), and
471 they are significantly higher than those of the lithospheric mantle, suggesting that
472 crustal contamination is widespread in the Hannuoba basalts. Therefore, the best
473 explanation for the EMI characteristics of the enriched radiogenic strontium isotopes
474 of the Hannuoba tholeiites seems to be contamination by the lower continental crust.

475 The significant change of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is likely due to assimilation of Hannuoba
476 alkaline basalts by lower continental crust because the former has significantly lower
477 $^{87}\text{Sr}/^{86}\text{Sr}$. However, trace element contents in the melt that are in equilibrium with the
478 plagioclase, calculated using the Bindeman et al. (1998) model, show minimal
479 variation (Figure 11, Supplementary Table 9), and Sr and Ba contents similar to that in

480 tholeiitic basalts, but significantly lower than those in alkaline basalts, suggesting that,
481 at least, Hannuoba alkaline basaltic melts were not directly contributing to the
482 intracrystalline Sr isotopic disequilibria. Other trace elements such as La and Eu are
483 either lower or higher than that in tholeiitic basalts (Figure 11), which is possibly due
484 to model uncertainty as shown by many other experimental and theoretical studies
485 (Bedard, 2006; Bindeman et al., 1998). Given that trace element content in Hannuoba
486 tholeiite is between Hannuoba alkaline basalts and lower continental crust (Figure 3),
487 mixing of Hannuoba alkaline basalts and the lower continental crust can explain the
488 bulk rock compositional characteristic of Hannuoba tholeiite, which is also consistent
489 with intercalated tholeiitic and alkaline lavas. However, in this case, the low
490 $^{87}\text{Sr}/^{86}\text{Sr}$ (<0.704) signature in plagioclase cannot be from alkaline basaltic melts. It is
491 likely that the alkali basalt interacted with the lower continental crust and formed a
492 mixed magma. The trace element abundances and ratios in this magma are similar to
493 the Hannuoba tholeiite and relatively homogeneous, but the isotopes are
494 heterogeneous. The crystalline plagioclase will inherit the decoupled isotopic and
495 trace element characteristics when the magma rises and injects into the magma
496 chamber. The Hannuoba tholeiite crystallized within a 20 °C (fine-grained basalt) and
497 50°C (coarse-grained basalt) temperature window also suggest that compositionally
498 (major and trace element) similar magmas were mixed with each other before the
499 eruption. This understanding is similar to that obtained from the study of melt
500 inclusions: the Pb isotope variation range of melt inclusions entrapped in olivine in a
501 Hannuoba tholeiite vary significantly and extend toward an EM1-type isotopic

502 composition, similar to that of all tholeiites, but the variation range of major and
503 minor elements is relatively tiny (Qian et al., 2015).

504 The potential problem is that the major and trace elements and isotopes in the
505 lower crust are quite heterogeneous. If significant isotope heterogeneity is recorded in
506 plagioclases in the two types of tholeiitic basalts in Hannuoba, then it would be
507 expected that the strontium isotope ratios would be lower than those for the average
508 lower continental crust. This may be related to the highly compositional heterogeneity
509 and multi-stage evolution of the lower crust in the North China Craton (Jiang et al.,
510 2013; Liu et al., 2004). Previous studies have shown that strontium isotopes in the
511 lower crust of the North China Craton vary from 0.704 to 0.746, and strontium
512 contents vary from < 100ppm to > 1500ppm (Jiang et al., 2013; Liu et al., 2004). The
513 major and trace elements in the lower crust (probably local) may be relatively
514 homogeneous due to the long-term and multi-stage magmatic underplating, but the
515 isotopic heterogeneity is still significant. In addition, it should be noted that the
516 contents of trace elements in the whole rock and plagioclase of the two types of
517 Hannuoba basalts are different, which indicates that there are differences in the parent
518 magma itself. This difference may be caused by the heterogeneity of local lower crust
519 materials, or it may be the product of magma evolution by crystallization and
520 differentiation after mixing alkaline basaltic rocks and different proportions of the
521 lower crust. Our current research does not distinguish between these possibilities. In
522 general, the contribution of lower crustal contamination to the formation of Hannuoba
523 tholeiite is more complex than previously thought. Identifying the existence of lower

524 crust contamination on the basis of whole-rock geochemical characteristics and the
525 average composition of the lower crust is challenging. However, in-situ trace elements
526 and isotopes of plagioclase may help solve these problems.

527 **Implication**

528 Although the plagioclase phenocryst zoning in Hannuoba tholeiite is oscillatory
529 and normal, and the content of anorthite and trace elements in each plagioclase grain
530 does not vary significantly, which is similar to the oscillatory zoning formed by a
531 closed system, the strontium isotope in the plagioclase micro area shows significant
532 isotope disequilibrium. Moreover, the range of strontium isotope ratios is far more
533 than that of all Hannuoba basalts, which has not been found in previous studies.
534 Although the error on strontium isotope ratios is significant, we have detected greater
535 than expected isotopic variation. If the beam spot increases, the error will decrease,
536 but the observed isotope heterogeneity will be reduced or eliminated.

537 Because the isotopic heterogeneity of plagioclase zoning occurs in the
538 crystallization process at the depths of the middle crust, both the lower crust and the
539 upper mantle of the continent may contribute to the heterogeneity of magma, and the
540 contamination and magma mixing may mainly occur in the lower crust and/or the
541 upper mantle. Our study suggests that the contamination by the lower continental
542 crust is more critical because the addition of material from the lower continental crust
543 can explain the very high osmium isotope, the typical EMI type Sr-Nd-Pb isotope,
544 and the lower incompatible element content of tholeiitic basalts compared with
545 alkaline basalts. This phenomenon may be shared in Cenozoic basalts in eastern

546 China; However, previous studies have attributed the compositional characteristics of
547 EMI endmembers to the continental lithosphere, recycled continental or oceanic crust
548 materials. An important reason for this phenomenon is that previous studies usually
549 exclude contamination of the lower crust by comparing the whole rock composition
550 characteristics with the global average continental lower crust composition, but this
551 practice has a substantial uncertainty because the continental lower crust has
552 significant compositional heterogeneity (Jiang et al., 2013; Liu et al., 2004; Rudnick
553 and Fountain, 1995). The mafic lower crust may have a high degree of partial melting,
554 mixing, and evolution, and then injected into the magma chamber of the shallow crust,
555 resulting in heterogeneous geochemical characteristics and even leading to the
556 decoupling of isotopes and trace elements, similar to those recorded in the plagioclase
557 micro zonation in this study.

558 Another important finding of this study is that there are significant differences in
559 petrography and trace element contents in mineral micro areas between the two types
560 of tholeiites with different whole-rock compositions, which indicates that the magma
561 with different initial compositions has undergone different crystallization history. The
562 crystallization process of the two tholeiites might occur in an independent magma
563 chamber or plumbing system. The in-situ study of mineral composition and isotope at
564 the micro-regional scale, and the selection of basalts with significant differences in
565 petrography, will help to reveal the complex evolutionary process of the magmatic
566 system and is also of great significance for a comprehensive understanding of the
567 origin and genesis of continental basaltic magma.

568

569

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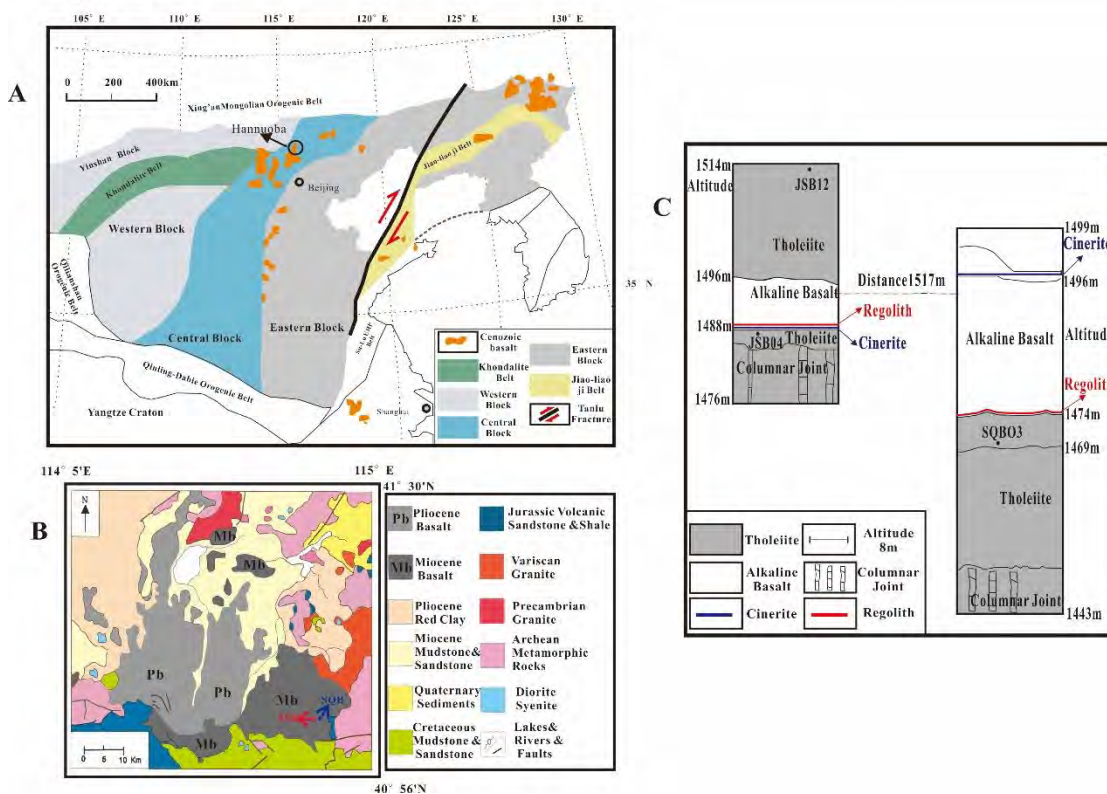
746 melting with linear variations in mineral/melt distribution coefficients.

747 *Geochimica Et Cosmochimica Acta*, 64(6): 1095-1102.

748 **Figures**

749

Figure 1



750

751 **Figure 1.** (a). Sketch map of the North China Craton (modified according to (Liu

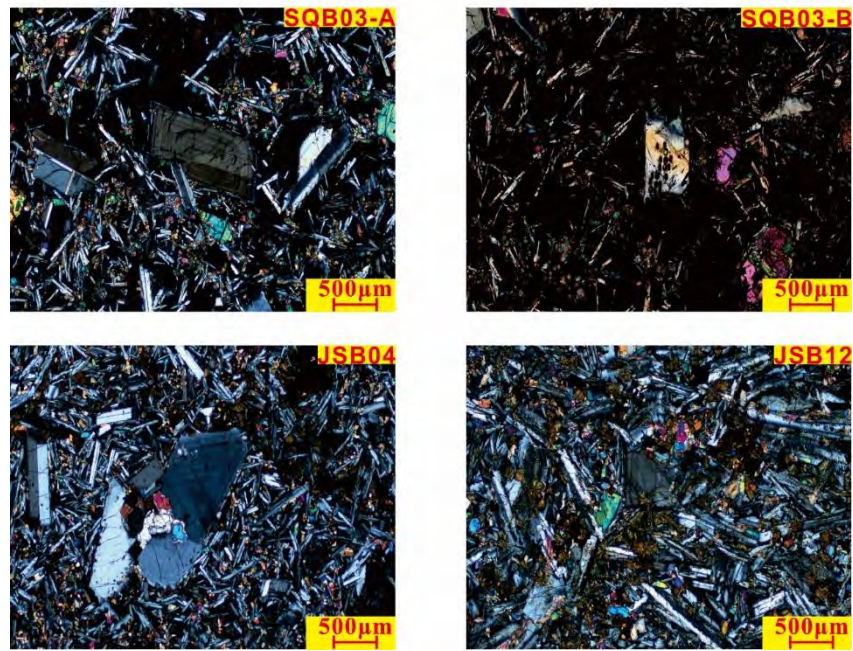
752 et al., 2019; Xu et al., 2005) (b). Geological map of Hannuoba (modified according to

753 (Zhi et al., 1990) (c). Spatial distribution of basalt in the sampling area.

754

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Figure 2



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757

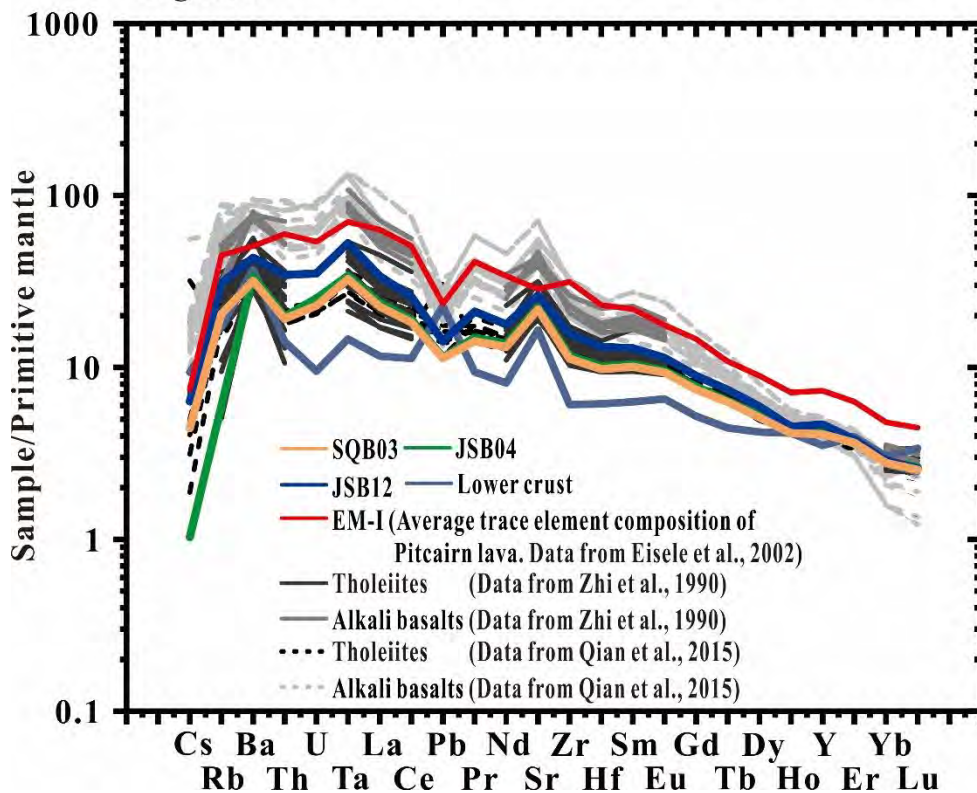
Figure 2. Representative micro-textural characteristics of the Hannuoba tholeiite

758

under crossed polars

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Figure 3



760

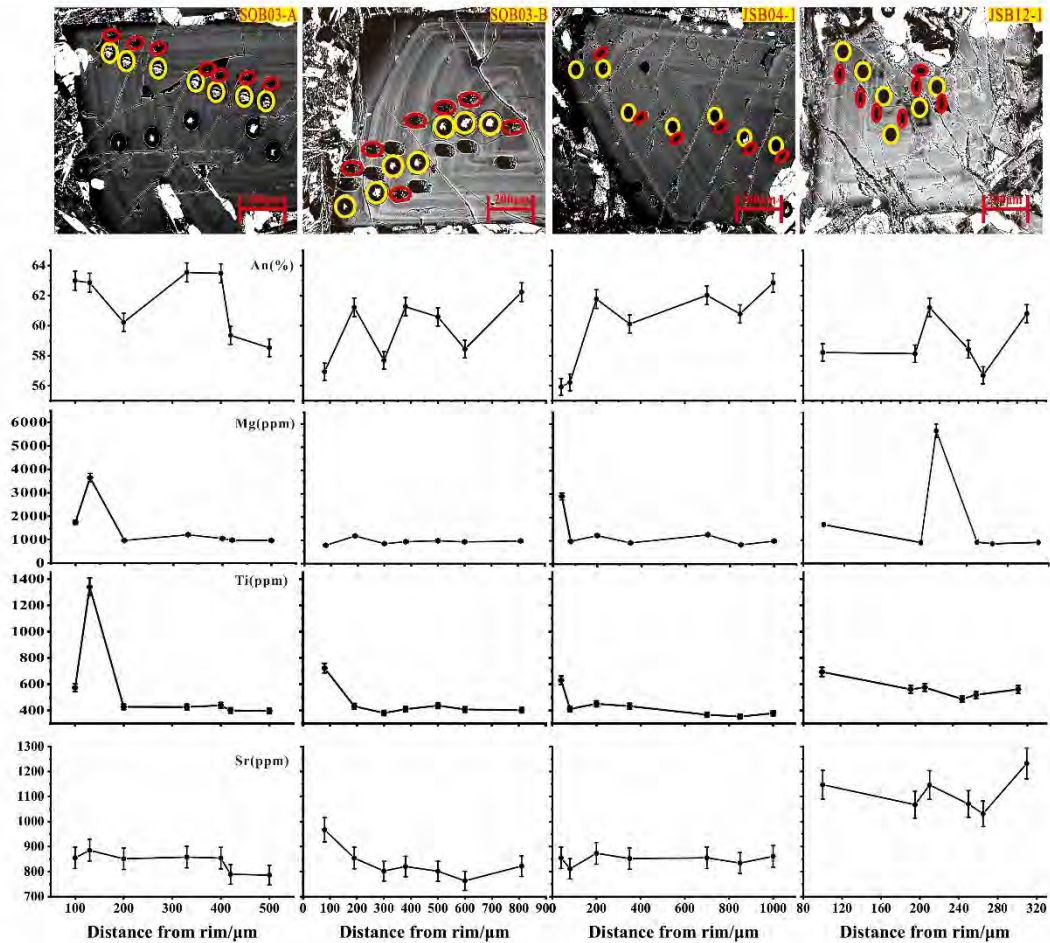
761

Figure 3. Primitive mantle (McDonough and Sun, 1995) normalized trace

762 element pattern for Hannuoba basalts and the average lower continental crust.

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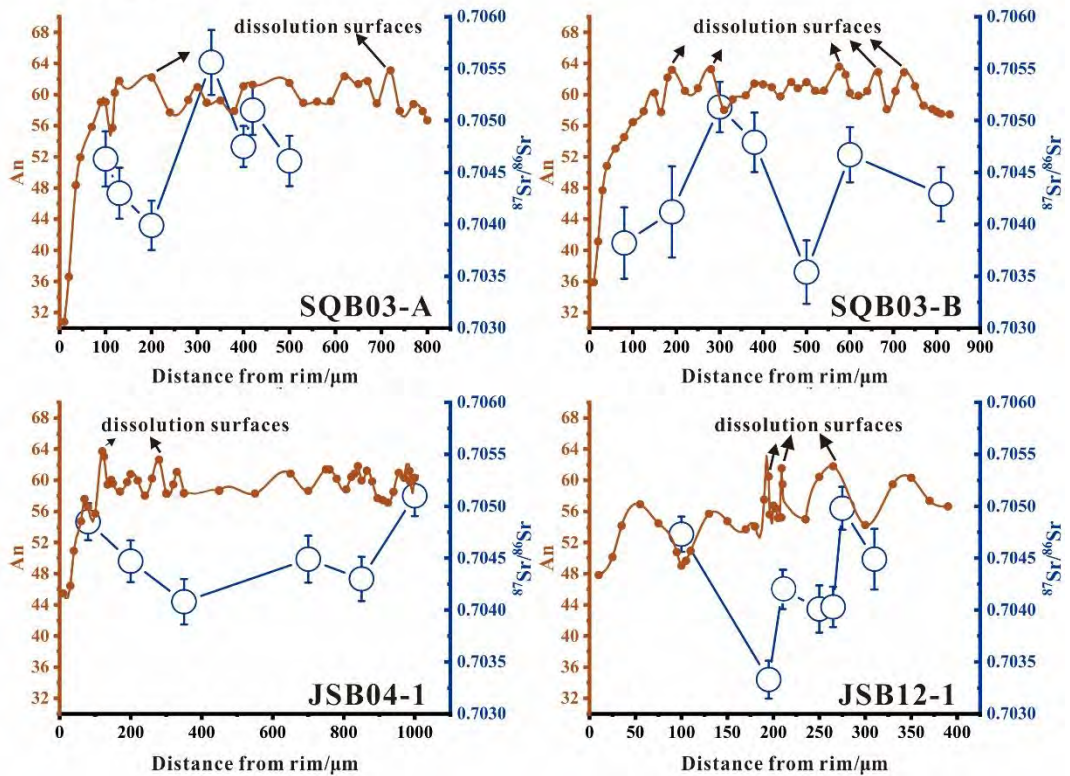
Figure 4



764

765 **Figure 4.** The relationship between An value and Mg, Ti, Sr element. (The
766 circles in yellow represent the in-situ trace element analyses, and the circles in red
767 represent the in-situ strontium isotope analyses. Error bars are 2σ SE)

Figure 5



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769

Figure 5. Variation diagram of Sr isotope, An, and distance in plagioclase.

770

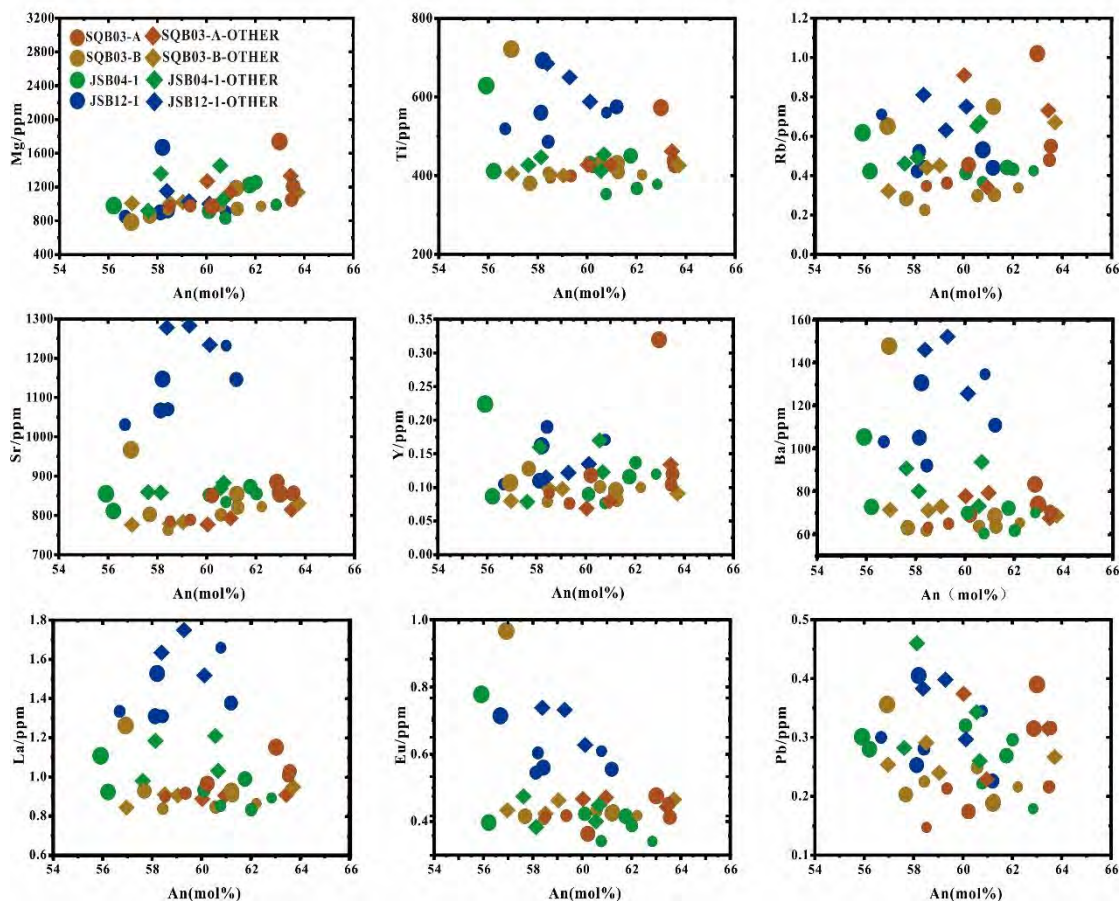
The arrow indicates dissolution surfaces and major Sr isotope variation trends.

771

Error bars are 2σ SE.

772

Figure 6



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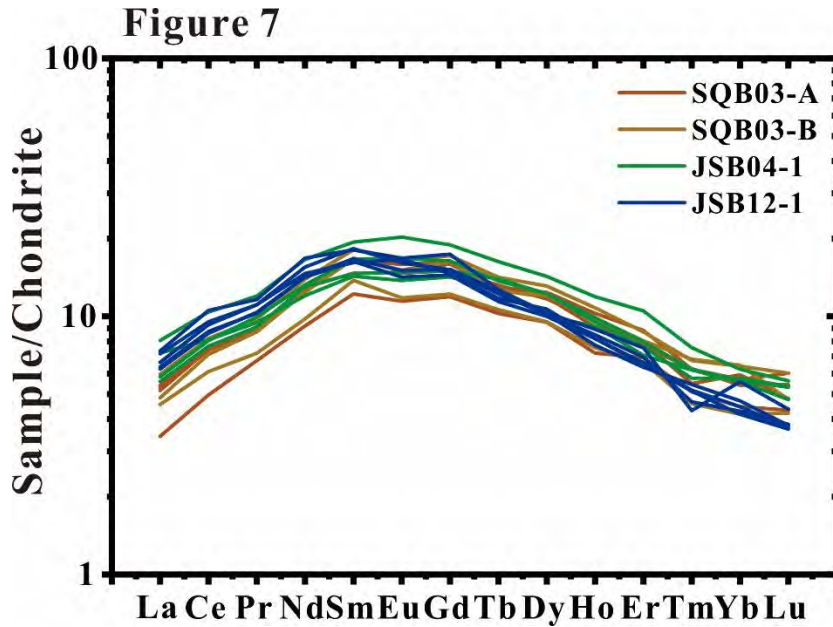
774

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Figure 6. Relationship between trace element concentration and An content in plagioclase. The circle data measured isotopes simultaneously, and the diamond data are other plagioclase crystals randomly selected from each sample. Colored circles from large to small represent from rim to core.



778

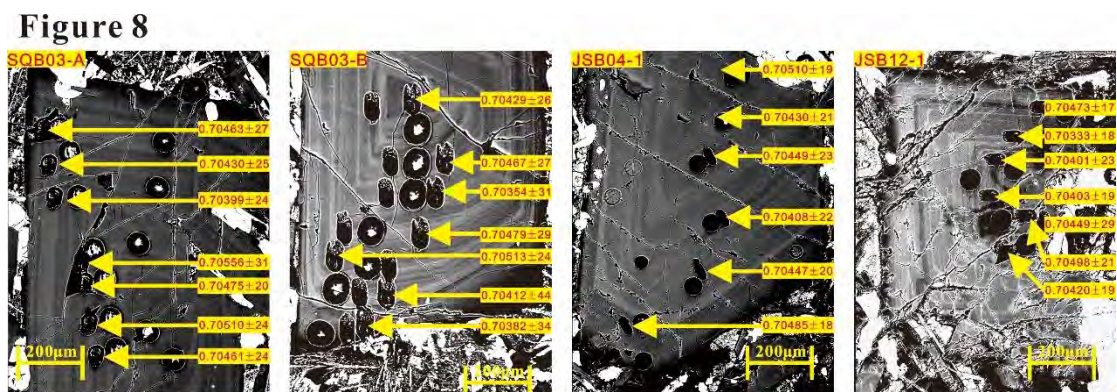
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Figure 7. Chondrite-normalized (McDonough and Sun, 1995) REE

780

concentration patterns for clinopyroxenes from Hannuoba tholeiite

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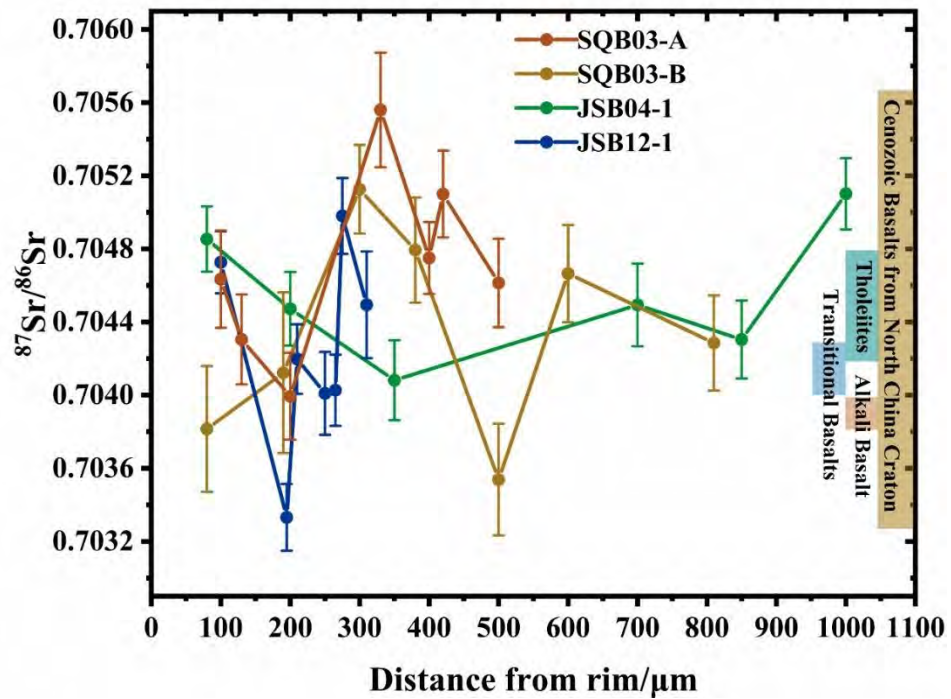
782

783

Figure 8. BSE image of the plagioclase sample with strontium isotope indicated.

784

Figure 9



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786 **Figure 9.** In situ $^{87}\text{Sr}/^{86}\text{Sr}$ variations of plagioclase samples from Hannuoba

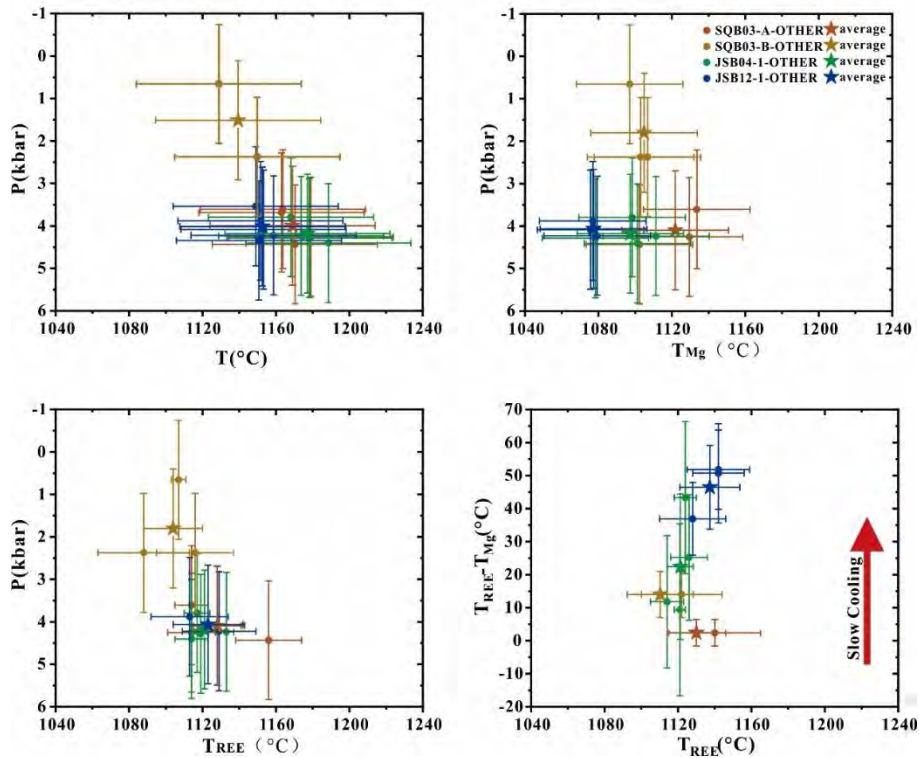
787 basalts. Sr isotope data of whole-rock Hannuoba alkaline, transitional and tholeiitic

788 basalts are from (Song et al., 1990); Cenozoic basalts from North China Craton after

789 (Liu et al., 2008b). The error bar indicates 2σ SE.

790

Figure 10



791

792

793 **Figure 10.** Relationship between crystallization temperature and pressure of

794 tholeiite in Hannuoba. (a) Crystallization pressure and temperature of clinopyroxene-

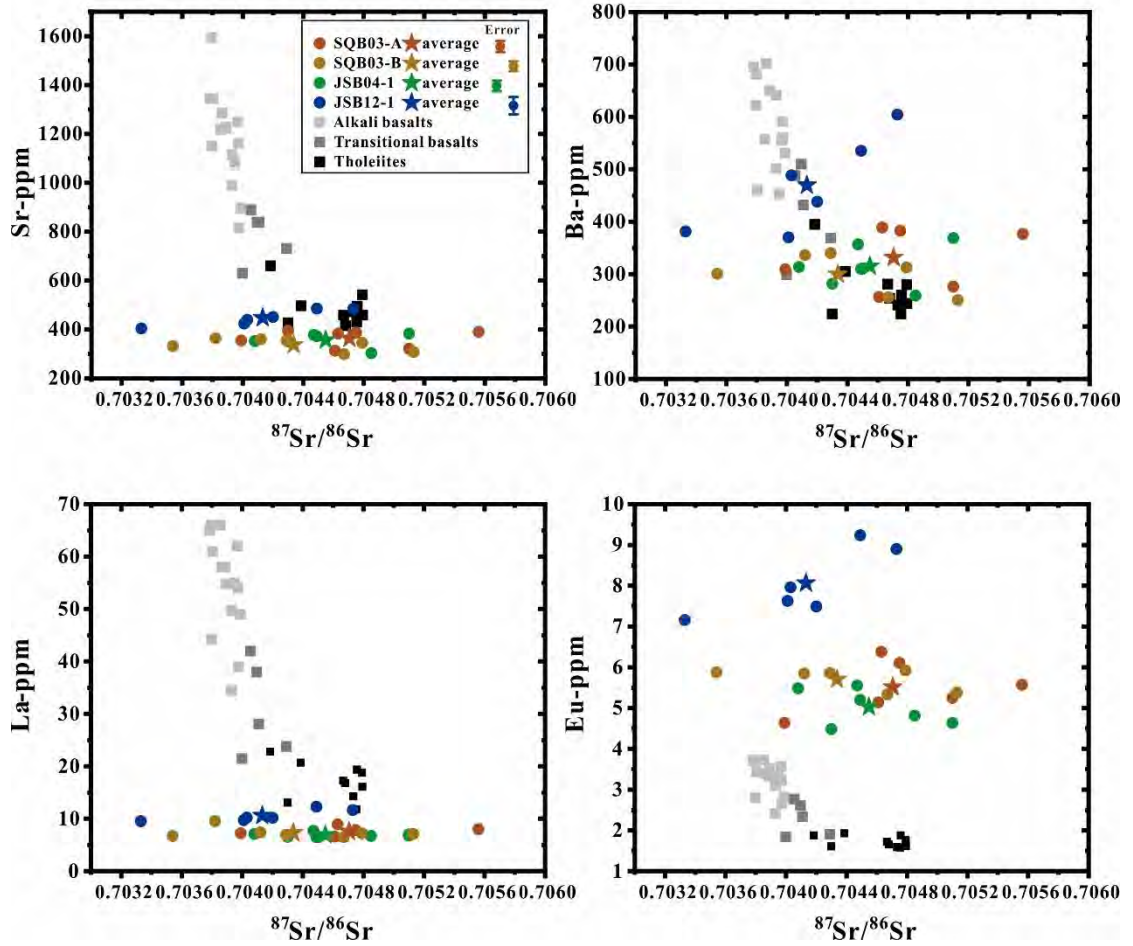
795 melt. (b) Closure temperature obtained by pyroxene-plagioclase magnesium

796 thermometer. (c) Closure the temperature obtained by pyroxene-plagioclase trace

797 element thermometer. (d) The temperature difference between T_{Mg} and T_{REE} . The error

798 bar indicates 2σ SE.

Figure 11



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Figure 11. Sr isotope versus trace element content for Hannuoba basalts and the calculated melts in equilibrium with plagioclase. Hannuoba basalts from Zhi et al., 1990; Song et al., 1990. Error bars smaller than the symbol are not shown.