

Title : Geochemical implication of Eu isotope ratio in the anorthosite: a new evidence of Eu isotope fractionation by feldspar crystallization ”.

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1           **Geochemical implication of Eu isotope ratio in the anorthosite: a new**  
2           **evidence of Eu isotope fractionation by feldspar crystallization**

3  
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13           Rare earth element geochemistry is important for understanding the evolution of the crust-  
14           mantle system. Europium (Eu) exists in divalent and trivalent states, and Eu<sup>2+</sup> can be substituted  
15           for Ca<sup>2+</sup> during plagioclase feldspar crystallization in reducing magmas. This leads to positive  
16           Eu anomaly in Ca-plagioclase-rich anorthosite derived from the mantle and negative Eu  
17           anomalies in fractionated silica-rich crustal rocks. But while Eu anomalies are well known, Eu  
18           has only two stable isotopes (<sup>151</sup>Eu and <sup>153</sup>Eu), Eu isotope ratios have not been compared with  
19           Eu anomalies in igneous rocks. Here we report a systematic variation of the Eu isotope ratio  
20           ( $\delta^{153/151}\text{Eu}$ ) from igneous rocks including anorthosite. This study finds a linear relationship  
21           between Eu anomalies and Eu isotope ratios in igneous rocks, with rhyolites and highly  
22           fractionated granites having large negative Eu anomalies and negative  $\delta^{153/151}\text{Eu}$  values but

23 anorthosites having large positive Eu anomalies and positive  $\delta^{153/151}\text{Eu}$  values. Particularly, in  
24 the area of the highly fractionated igneous rocks with negative Eu anomaly, the Eu isotope  
25 fractionation proceeds with different slope according to the degree of magmatic differentiation  
26 in extrusive (volcanic) and intrusive (plutonic) rocks. Our finding reveals that Eu isotope  
27 fractionation in igneous rocks will provide new information related to magmatic differentiation  
28 and plagioclase feldspar fractional crystallization including anorthosite formation in the crust-  
29 mantle.

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31 Key words: Eu isotope fractionation, Eu anomaly, magmatic differentiation, feldspar  
32 crystallization, anorthosite

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## INTRODUCTION

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Rare earth elements (REEs) and their radiogenic isotope geochemistry (especially the  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  and  $^{138}\text{La}$ - $^{138}\text{Ce}$  system) have provided abundant information for interpreting the geochemical evolution of Earth and extra-terrestrial materials as a result of their similar chemical behavior and continuously varying atomic masses of REEs. In particular, the geochemistry of chondrite-normalized REEs provides valuable petrogenetic information during the magma evolution processes such as partial melting from the mantle-derived rocks or crystallization from the magma (Coryell et al., 1963; Fowler and Doig, 1983; Masuda, 1962; Shearer and Papike, 1989; Weill and Drake, 1973). Most REEs have a stable (+3) state in natural systems; however, Eu can exist in both divalent and trivalent state under magmatic redox conditions, which indicates that the behavior of Eu during magmatic differentiation depends on the oxygen fugacity and crystallization of minerals (Burnham et al., 2015). Fractional crystallization is considered the dominant mechanism of magmatic differentiation and isolates crystallized-minerals from magma step by step (Bowen, 1928). Positive or negative Eu anomalies from the igneous rocks are produced by feldspar (particularly plagioclase) fractional crystallization with removal or accumulation, respectively, of plagioclase during magma evolution and have been interpreted as indicating the degree of differentiation of the source magma (Fowler and Doig, 1983; Shearer and Papike, 1989; Weill and Drake, 1973). For example, extremely large positive Eu anomaly in the anorthosite is due to concentration of Eu due to be substituted into the Ca site in plagioclase feldspar because the  $\text{Ca}^{2+}$  site in feldspar readily accepts  $\text{Eu}^{2+}$ . However, highly fractionated granite and high-silica rhyolite shows extremely large negative Eu anomalies.

59 Eu has only two isotopes,  $^{151}\text{Eu}$  (47.81% ) and  $^{153}\text{Eu}$  (52.19%) (Rossman and Taylor, 1998).  
60 Though  $^{151}\text{Eu}$  decayed to  $^{147}\text{Pm}$  by  $\alpha$  decay with the half-life  $T_{1/2}=5\times 10^{18}\text{yr}$  (Belli et al., 2007),  
61  $^{151}\text{Eu}$  can be considered as a stable isotope in earth and solar system. In addition, recently, Lee  
62 and Tanaka (2021a) reported Eu isotope fractionation due to light Eu isotope enrichment ( $^{151}\text{Eu}$ )  
63 in highly fractionated granite and high-silica rhyolite with large negative Eu anomalies. The  
64 authors proposed that the heavier Eu isotope ( $^{153}\text{Eu}$ ) might be enriched in anorthosite with large  
65 Eu positive anomaly due to Ca-feldspar crystallization.

66 At present, there have been no report on Eu anomaly and Eu isotope ratio in anorthosite  
67 including gabbro as well as the volcanic rocks such as andesite and trachyte. Therefore, here,  
68 we report the Eu isotope ratio and Eu anomaly among plutonic (intrusive) rocks such as  
69 anorthosites, gabbro and volcanic (extrusive) rocks such as andesite and trachyte, and compare  
70 the data of Eu isotope ratio from the igneous rocks such as basalt, rhyolite and granitoids.

71 The objective in this article is to find a possibility of a new tracer for studying the  
72 relationship between Eu anomaly in the chondrite-normalized REE pattern and the Eu isotope  
73 fractionation by comparing the magnitude of Eu anomaly and the degree of Eu isotope  
74 fractionation in various kinds of igneous rocks such as the extrusive rocks and intrusive rocks  
75 including anorthosite.

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## **SAMPLES AND EXPERIMENTAL METHODS**

78 *Samples*

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80 In order to measure Eu isotope ratio of various kinds of igneous rocks, 49 igneous rock  
81 samples were used for Eu isotope ratio and REE abundance determination, of which 25 samples  
82 were geochemical reference materials purchased from the United States Geological Survey  
83 (USGS) and the Geological Survey of Japan (GSJ), while the others were anorthosites,  
84 granitoids and trachytes from Korea and Antarctica. The 25 geochemical reference materials  
85 in this study were as follows; Seven basalts (BCR2, BHVO2 and BIR1a purchased from the  
86 USGS; JB1a, JB1b, JB2 and JB3 from the GSJ), four andesites (AGV2 from USGS; JA1, JA2  
87 and JA3 from GSJ), four rhyolites (RGM2 from USGS; JR1, JR2 and JR3 from GSJ), one  
88 diabase (W-2a from USGS), one dolerite (DNC1a from USGS), two gabbros (JGb1, JGb2 from  
89 GSJ), one syenite (STM2 from USGS), and five granites (G2 and GSP2 from USGS; JG1a,  
90 JG2 and JG3 from GSJ). The 24 rock samples from Korea and Antarctica are as follows;  
91 fourteen granites and five anorthosites from Korea, and five trachytes from Antarctica. Because  
92 there was no SRM trachyte, we collected five trachytes from Antarctica.

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#### 94 *Sample digestion and determination of REE concentrations*

95 The sample digestion procedure was based on the approach of Lee et al (2016).  
96 Approximately 100~200 mg of each sample powder was decomposed by a 2:1 mixture of 2~4  
97 mL of concentrated HF (29M) and 1~2 mL of concentrated HNO<sub>3</sub> (16M) at ca. 160 °C for more  
98 than 72 hours in 15 mL Savillex vial. After the addition of 0.1~0.2 mL of concentrated HClO<sub>4</sub>,  
99 the decomposed sample solution was heated to dryness at ca. 180 °C for more than 1 day. The  
100 cakes were re-dissolved by a mixture of 1 mL of concentrated HCl and 0.5 mL of concentrated  
101 HNO<sub>3</sub> at ca. 160 °C for 1 day. Each sample solution was dried again, and diluted in 10 ml of 6  
102 M HCl as a stock solution. Of this 10 ml stock solution, 0.5~1 ml stock solution was used to

103 determine the rare earth element (REE) concentrations, and the remainder was used for  
104 determination of Eu isotope ratio.

105 Before Eu purification, we analyzed REE concentration of the sample using inductively  
106 coupled plasma mass spectrometry (ICP-MS, NexION350, Perkin Elmer) at KIGAM.  
107 Although Eu anomalies of the 25 geochemical reference rocks (USGS, GSJ) have previously  
108 been characterized, we also reanalyzed their REE abundances for comparison. The analyzed  
109 REE data from the geochemical reference sample powders (SRM) agreed with the  
110 recommended values within 5~10%.

111

#### 112 *Experimental procedures for determination of Eu isotope ratio*

113 Recently, Lee and Tanaka (2019, 2021b) developed a method for determining highly  
114 precise and accurate Eu isotope ratio using Sm spike as an internal standard in combination  
115 with standard-sample-standard bracketing mass bias correction (C-SSBIN). In addition, Lee  
116 and Tanaka (2021a, 2021b) also showed that incomplete Eu purification from the geological  
117 material lead to change in the Eu isotope ratio, that is, pseudo-fractionation of Eu isotope ratio.

118 In this study, Eu was separated from the obtained REE fraction using 0.12 M 2-  
119 hydroxyisobutyric acid (HIBA) with the pH adjusted to ~4.60 (Lee and Tanaka, 2019, 2021b).  
120 To minimize isobaric interference because we used Sm which was prepared from ultrapure  
121 Sm<sub>2</sub>O<sub>3</sub> produced by Alfa Aesar as a spike for normalization of Eu isotopes, we always checked  
122 for tailing of both Gd and Sm.

123 Eu isotope ratios were measured using multicollector inductively coupled plasma mass  
124 spectrometry (MC-ICP-MS; Neptune Plus, Thermo Fisher Scientific Ltd.) in static mode with

125 nine Faraday cups at KIGAM. The instrument was tuned to achieve high sensitivity while  
126 maintaining flattened square peaks and stable signals enough to ensure accurate measurements.  
127 The gain on each Faraday cup was monitored daily to ensure normalization of its efficiency.  
128 Sample dilution for Eu isotope measurement by MC-ICP-MS was performed with 2% HNO<sub>3</sub>  
129 which was prepared from 60% ultrapure HNO<sub>3</sub> (Merck, Darmstadt, Germany) and DIW (Milli-  
130 Q system, Millipore, Milford, USA). We used a diluted solution of NIST 3117a (10,000 µg/mL,  
131 Lot No. 120705) as an in-house standard solution for comparison of the Eu isotope ratios.

132 The isotopes <sup>147</sup>Sm(L4), <sup>149</sup>Sm(L3), <sup>150</sup>Sm(L2), <sup>151</sup>Eu(L1), <sup>152</sup>Sm(C), <sup>153</sup>Eu(H1),  
133 <sup>154</sup>Sm(H2), <sup>155</sup>Gd(H3), and <sup>157</sup>Gd(H4) were monitored simultaneously using nine Faraday cups  
134 for Sm normalization and Gd interference correction by the Gd matrix (Lee and Tanaka, 2021b).  
135 Data acquisition consisted of 1 block of 50 cycles with an integration time of 4.194 seconds  
136 and a sample aspiration rate of 80-100 µL/min. Peak centering was performed at the beginning  
137 of each analysis and 250 s of washout time was used between sample measurements. Blanks  
138 were checked during, before, and after each sample measurement. Operating conditions and  
139 data acquisition parameters, including cup configuration, are the same as Lee and Tanaka  
140 (2021a, 2021b). In determination of Eu isotope ratio, we used <sup>147</sup>Sm/<sup>149</sup>Sm (1.0868, Dubois et  
141 al., 1992) for normalization to obtain an optimum value of Eu isotope fractionation from the  
142 natural materials because of isobar matrix problem by <sup>154</sup>Gd due to incomplete separation from  
143 the geological rock during <sup>150</sup>Sm/<sup>154</sup>Sm normalization (Lee and Tanaka, 2021a, 2021b). Eu  
144 isotope fractionation is represented in standard δ-notation in per mil relative to the NIST3117a  
145 Eu standard solution as follows:  $\delta^{153/151}\text{Eu} = 1,000 \times [(\text{}^{153}\text{Eu}/\text{}^{151}\text{Eu}_{\text{sample}})/(\text{}^{153}\text{Eu}/\text{}^{151}\text{Eu}_{\text{NIST3117a}})$   
146  $- 1]$ .

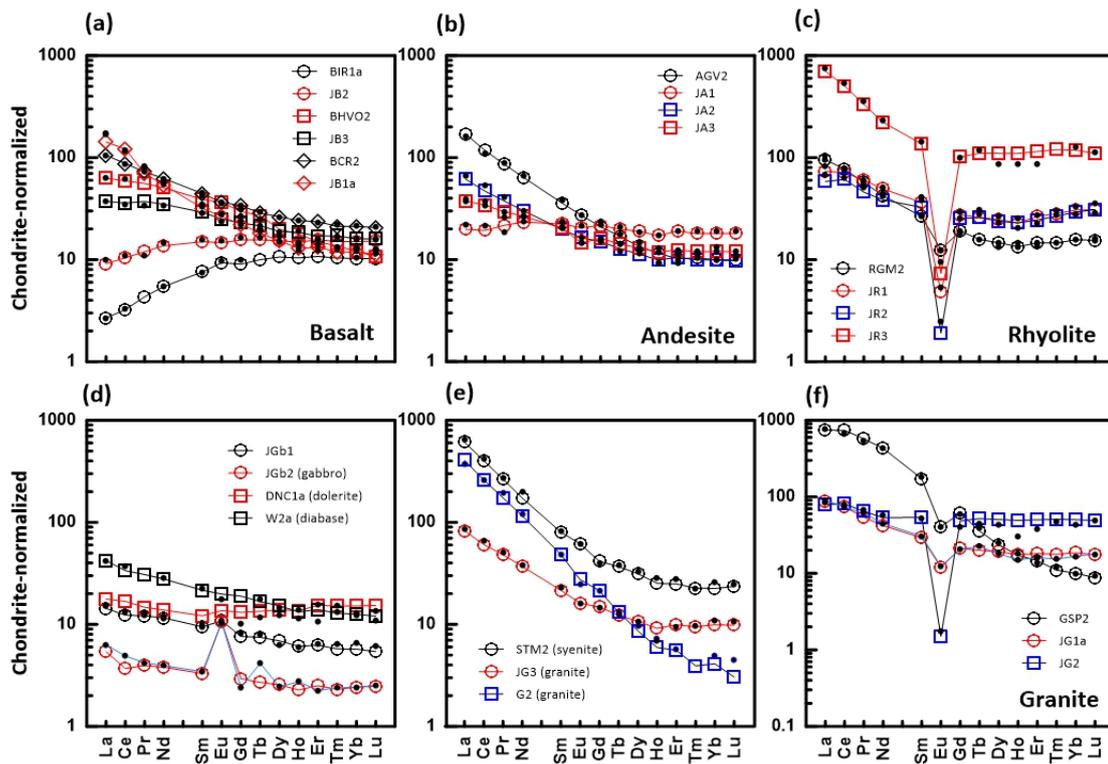
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## RESULTS

149 *Rare Earth Element concentration in the extrusive rocks and intrusive rocks*

150 In order to validate the accuracy of the REE data in this study, we determined REE  
 151 concentrations of 25 geochemical standard reference materials (SRM) produced by USGS and  
 152 GSJ. REE abundances of the geochemical reference materials from the United States of  
 153 Geological Survey (USGS) and Geological Survey of Japan (GSJ) measured in this study are  
 154 reported in Supplementary Table S1. In addition, REE concentrations of the local igneous rocks  
 155 in Korea and Antarctica including anorthosite are presented in Table 1.

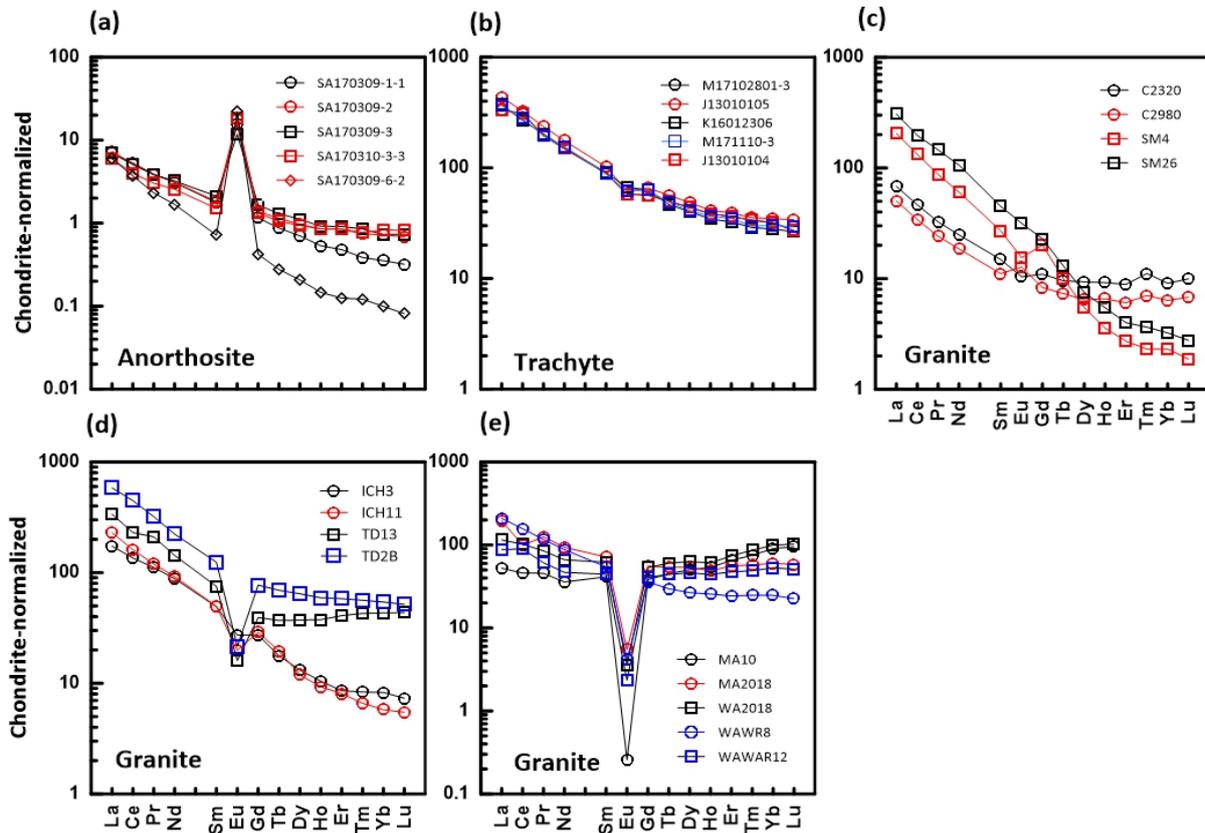


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157 **Fig. 1.** Chondrite (McDonough and Sun, 1995)-normalized REE pattern of standard reference materials (SRMs)  
 158 of USGS and GSJ. (a), (b) and (c) are SRMs for volcanic (extrusive) rocks whereas (d), (e) and (f) are SRMs for  
 159 intrusive rocks such as gabbro, diabase and granitoids. In this paper, we classified dolerite and diabase as plutonic  
 160 rocks rather than volcanic rocks. The REE abundances of all SRMs were re-measured from this study (see Table  
 161 S1). REE patterns by solid black dots were drawn by recommended values for each SRM from USGS and GSJ.

162

163 Figures 1 and 2 are chondrite-normalized REE patterns for SRMs and Korea and Antarctic  
 164 igneous rocks, respectively. In Fig. 1, REE patterns from the volcanic rocks such as basalt,  
 165 andesite and rhyolite were drawn in Fig. 1a~1c whereas those from the plutonic rocks such as  
 166 gabbro and granitoids were drawn in Fig. 1d~1f. The chondrite-normalized REE patterns from  
 167 various kinds of igneous rocks in Figs. 1 and 2 clearly show variation of the magnitude of Eu  
 168 anomaly due to feldspar crystallization during magmatic differentiation even though they are  
 169 not cogenetic igneous rocks. Particularly, the anorthosites in Fig. 2a has strikingly large Eu  
 170 positive anomaly.



171  
 172 Fig. 2. Chondrite (McDonough and Sun, 1995)-normalized REE pattern of Korean and Antarctic igneous rocks.  
 173 Except (b) trachyte, the others are all collected from Korea. Trachyte was collected from Antarctica. The REE  
 174 abundances of Korean anorthosites and Antarctic trachytes were re-measured from this study. REE patterns of  
 175 Korean granites are from Lee et al. (2004, 2006, 2008, 2013).

176

177 *Eu isotope ratio in in the extrusive rocks and intrusive rocks*

178 Eu isotope ratio and magnitude of Eu anomaly from various kinds of the igneous rocks in  
179 this study are presented in Table 2. For comparison, in Table 2, we divided the samples into  
180 SRM and local igneous rocks. The  $^{153/151}\text{Eu}$  value of anorthosite all are positive, indicating that  
181 heavier Eu isotope ( $^{153}\text{Eu}$ ) was enriched compared to lighter Eu isotope ( $^{151}\text{Eu}$ ).

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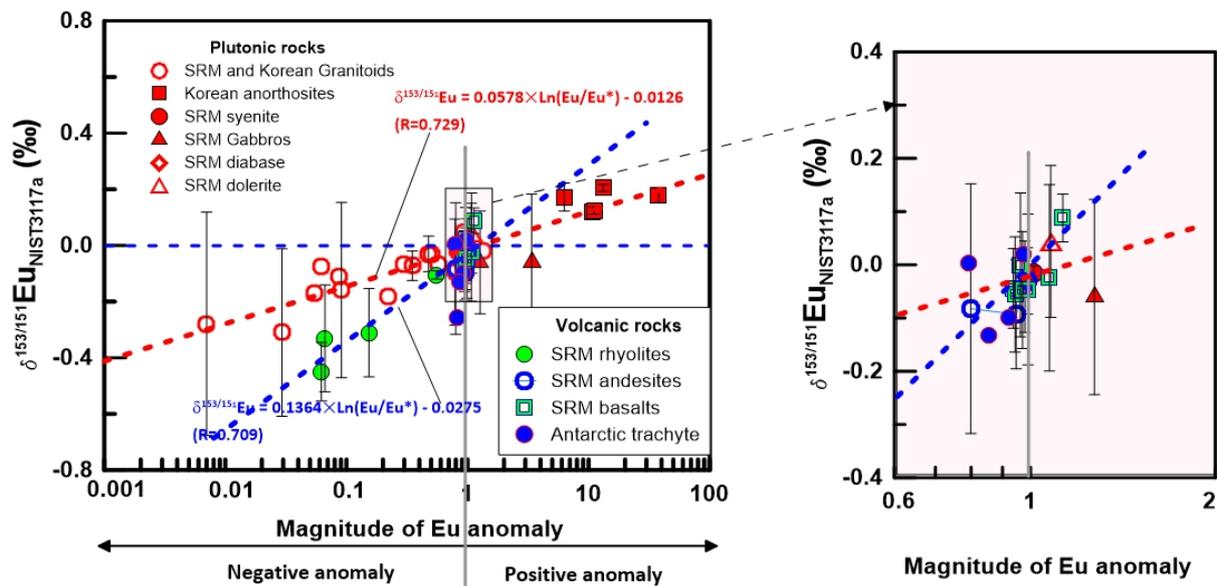
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## DISCUSSION

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185 A major objective of this study is to examine the relationship between the magnitude of  
186 Eu anomaly due to feldspar crystallization during magmatic differentiation and Eu isotope  
187 differentiation. Therefore, we first compared the magnitudes of Eu anomalies with the  
188  $\delta^{153/151}\text{Eu}$  values of the 49 samples to investigate the possibility that Eu isotope fractionation  
189 had occurred during magma differentiation (Fig. 3). Figure 3 illustrates **three** geochemical  
190 characteristics of Eu isotope fractionation as follows; 1) The  $\delta^{153/151}\text{Eu}$  values of the highly  
191 fractionated granites and rhyolites with extremely large negative Eu anomaly are negative,  
192 whereas the anorthosites with extremely large positive Eu anomaly show relatively large  
193 positive  $\delta^{153/151}\text{Eu}$  values. This contrast indicates that the highly fractionated igneous rocks,  
194 which are emplaced from felsic magma in an upper crustal environment, were enriched in the  
195 lighter Eu isotope ( $^{151}\text{Eu}$ ), whereas Ca-plagioclase-rich anorthosite, which are derived from the  
196 mafic magma in a lower crustal environment, were enriched in the heavier Eu isotope ( $^{153}\text{Eu}$ ).  
197 2) The  $\delta^{153/151}\text{Eu}$  value in the igneous rock varies systematically with magnitude of the Eu  
198 anomaly. 3) Another interesting feature of the Eu fractionation trends in the volcanic and

199 plutonic rocks is that intrusive rocks (red symbols in Fig. 3) and extrusive rocks (green and  
 200 blue symbols in Fig. 3) are distributed with different slopes.



201  
 202 **Fig. 3.** Variation of Eu isotope ratio according to magnitudes of Eu anomalies from igneous rocks. The  
 203 error bars represent uncertainties (2SD) of the average  $\delta^{153/151}\text{Eu}$  values from some of igneous rock  
 204 samples.

205  
 206 Ismail et al. (1998) employed cation exchange chromatography and concluded that an  
 207 isotope effect in the  $\text{Eu}^{2+}/\text{Eu}^{3+}$  exchange reaction may occur in aqueous solutions; specifically,  
 208 they found isotope effects in which the heavier isotope  $^{153}\text{Eu}$  is enriched in  $\text{Eu}^{2+}$  in the  
 209  $\text{Eu}^{2+}/\text{Eu}^{3+}$  electron exchange system. The positive Eu anomaly in anorthosite can easily be  
 210 explained by the substitution of  $\text{Eu}^{2+}$  for  $\text{Ca}^{2+}$  in plagioclase during differentiation of the  
 211 anorthositic (primary) magma either in the upper mantle or at the lower crust. Therefore, we  
 212 can suggest that the enrichment of heavier isotope  $^{153}\text{Eu}$  in the anorthosites with large positive  
 213 Eu anomaly should be explained due to isotope effects in  $\text{Eu}^{2+}/\text{Eu}^{3+}$  electron exchange system  
 214 during Ca-plagioclase accumulation in the anorthositic magma. Moreover, the systematic

215 correlation between Eu isotope fractionation and the magnitudes of Eu anomaly from the  
216 fractionated igneous rocks and anorthosites indicates that Eu isotope fractionation was closely  
217 related to magmatic differentiation processes such as feldspar fractional crystallization.

218 Another interesting finding in Figure 3 is that the trend of Eu isotope fractionation  
219 between the extrusive volcanic rocks and intrusive plutonic rocks seems to be different.  
220 Although more in-depth studies are needed in the future, the existence of such different slope  
221 can be interpreted as indicating the possibility that the Eu isotope fractionation in intrusive and  
222 extrusive rocks occurred by different mechanism or geochemical environment. The behavior  
223 of Eu is known to be determined by temperature and oxygen fugacity (Weill and Drake, 1973),  
224 and Philpotts and Schnetler (1968) and Philpotts (1970) suggested that Eu anomalies are  
225 controlled by crystal chemistry and magmatic oxidation potential. Dauphas et al. (2014)  
226 showed that equilibrium iron isotope fractionation is controlled mainly by the redox and  
227 structural conditions in magma and suggested that magmatic differentiation is the main driver  
228 of Fe isotope fractionation in felsic magmas. In addition, Dauphas et al. (2014) proposed that  
229 stable isotopes from heterovalent elements, including Eu, may show isotopic variations in bulk  
230 rocks controlled by the redox and structural conditions in the magma. Therefore, we also may  
231 be able to consider a possibility that a slight difference of the  $\delta^{153/151}\text{Eu}$  values in the plutonic  
232 rocks and volcanic rocks may be due to the oxidation potential in the magma. Further study is  
233 needed to clarify the relationship between Eu isotope fractionation and oxidation potential in  
234 an intrusive magmatic system.

235 Besides Eu isotope fractionation, recently, several research groups reported isotope  
236 fractionation study of REEs like Ce, Nd, Sm, Dy, Er and Yb (Moynier et al., 2006; Nakada et  
237 al., 2013; Shollenberger and Brebbecka, 2020; Hu et al., 2021). This means that combined

238 information using chondrite-normalized REE pattern, radiogenic isotope geochemistry and  
239  $\delta$ REE will provide new constraints for understanding more clearly the processes in our Earth  
240 and Planetary system such as redox conditions in the mantle and/or crust, early earth formation,  
241 and crust/mantle differentiation.

242

## 243 CONCLUSION

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245 We compared the magnitude of Eu anomaly in the chondrite-normalized REE pattern from  
246 various kinds of igneous rocks such as the extrusive rocks and intrusive rocks including  
247 anorthosite and their Eu isotope ratio from the fractionation. The anorthosites having large  
248 positive Eu anomalies show a geochemical characteristic of a heavier Eu isotope ( $^{153}\text{Eu}$ )  
249 enrichment (i.e., positive  $\delta^{153/151}\text{Eu}$  value) whereas the rhyolites and highly fractionated  
250 granites having large negative Eu anomalies show a geochemical characteristic of a lighter Eu  
251 isotope ( $^{151}\text{Eu}$ ) enrichment (i.e., negative  $\delta^{153/151}\text{Eu}$  value). Particularly, our results clearly  
252 showed that variation of the magnitude of Eu anomaly and Eu isotope fractionation in igneous  
253 rocks has systematic correlation, suggesting that Eu isotope fractionation in igneous rocks  
254 should be produced by feldspar crystallization during magma evolution. In addition, the Eu  
255 isotope fractionation in the highly fractionated volcanic and plutonic rocks was proceeded with  
256 different trend, implying that the Eu isotope fractionation from the intrusive and extrusive  
257 magma in the crustal environment may occur under different mechanism or geochemical  
258 environment.

259

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359

## Figure Captions

360

361

362 Fig. 1. Chondrite (McDonough and Sun, 1995)-normalized REE pattern of standard reference  
363 materials (SRMs) of USGS and GSJ. (a), (b) and (c) are SRMs for volcanic (extrusive) rocks  
364 whereas (d), (e) and (f) are SRMs for intrusive rocks such as gabbro, diabase and granitoids.  
365 In this paper, we classified dolerite and diabase as plutonic rocks rather than volcanic rocks.  
366 The REE abundances of all SRMs were re-measured from this study (see Table S1). REE  
367 patterns by solid black dots were drawn by recommended values for each SRM from USGS  
368 and GSJ.

369

370 Fig. 2. Chondrite (McDonough and Sun, 1995)-normalized REE pattern of Korean and  
371 Antarctic igneous rocks. Except (b) trachyte, the others are all collected from Korea. Trachyte  
372 was collected from Antarctica. The REE abundances of Korean anorthosites and Antarctic  
373 trachytes were re-measured from this study. REE patterns of Korean granites are from Lee et  
374 al. (2004, 2006, 2008, 2013).

375

376 Fig. 3. Variation of Eu isotope ratio according to magnitudes of Eu anomalies from igneous  
377 rocks. The error bars represent uncertainties (2SD) of the average  $\delta^{153/151}\text{Eu}$  values from some  
378 of igneous rock samples.

379

380

Table 1 . Rare earth element concentrations of igneous rocks

Rock Type	Area	Sample Name	La (ppm)	Ce (ppm)	Pr (ppm)	Nd (ppm)	Sm (ppm)	Eu (ppm)	Gd (ppm)	Tb (ppm)	Dy (ppm)
Trachyte	Mt. Melbourne (Antarctica)	M17102801-3	78.42	190.5	18.12	68.07	13.17	3.26	11.29	1.80	10.81
		J13010105	103.3	198.7	21.92	80.38	15.09	3.46	13.09	2.04	11.92
		K16012306	87.78	164.8	18.74	70.02	13.35	3.76	12.51	1.68	9.89
		M171110-03	86.93	172.4	18.10	70.93	12.98	3.49	12.50	1.77	10.11
		J13010104	78.42	190.5	18.12	68.07	13.17	3.26	11.29	1.80	10.81
Anorthosite	Sancheong (Korea)	SA20170309 1-1	1.64	3.25	0.36	1.41	0.26	0.87	0.23	0.03	0.17
		SA20170309 -2	1.73	3.18	0.36	1.43	0.27	0.99	0.28	0.04	0.24
		SA20170309-3	1.65	3.12	0.35	1.47	0.31	0.66	0.33	0.05	0.27
		SA20170309-6-2	1.42	2.27	0.22	0.77	0.11	1.24	0.08	0.01	0.05
		SA20170310 3-3	1.41	2.45	0.28	1.17	0.22	1.04	0.26	0.04	0.23
Granitoid	Muamsa-Weolaksan (Korea)	MA10	12.42	28.12	4.23	16.35	6.12	0.01	7.61	1.62	12.39
		WAWR12	20.97	54.60	5.67	21.40	6.56	0.13	8.13	1.59	11.35
		WAWR8	49.60	96.30	10.80	40.20	8.07	0.24	7.15	1.07	6.56
		MA2018	46.39	60.86	11.64	42.91	10.59	0.31	11.08	1.93	13.23
		WA2018	27.30	62.63	7.88	30.31	9.17	0.20	10.63	2.17	15.58
	Seokmodo (Korea)	SM4	48.70	81.50	8.11	27.70	4.00	0.87	4.00	0.36	1.35
		SM26	72.80	120.80	13.50	48.50	6.66	1.78	4.55	0.47	1.85
	Icheon (Korea)	ICH3	41.20	83.00	10.32	40.28	7.29	1.54	5.42	0.64	3.24
		ICH11	54.50	98.70	11.10	42.10	7.34	1.12	5.75	0.71	2.92
	Pohang (Korea)	C2320	16.15	28.50	3.03	11.35	2.20	0.59	2.18	0.35	2.30
		C2980	11.95	20.93	2.24	8.49	1.64	0.72	1.65	0.27	1.58
	Taedo (Korea)	TD13	79.97	141.0	19.52	65.33	11.11	0.90	7.83	1.33	9.16
		TD2B	138.3	273.9	29.76	101.8	18.36	1.21	15.36	2.50	15.84

Ho (ppm)	Er (ppm)	Tm (ppm)	Yb (ppm)	Lu (ppm)	Reference
2.10	5.79	0.83	5.14	0.66	This study
2.26	6.25	0.88	5.57	0.83	
1.88	5.19	0.71	4.50	0.65	
1.99	5.58	0.73	4.88	0.72	
2.10	5.79	0.83	5.14	0.66	
0.03	0.08	0.01	0.06	0.01	
0.05	0.13	0.02	0.12	0.02	
0.05	0.15	0.02	0.12	0.02	
0.01	0.02	0.00	0.02	0.002	
0.05	0.14	0.02	0.13	0.02	
2.92	10.46	1.86	14.55	2.34	
2.41	7.57	1.21	8.54	1.25	
1.40	3.88	0.61	3.97	0.56	
2.66	8.95	1.42	9.67	1.43	
3.31	11.85	2.14	16.01	2.51	
0.20	0.44	0.06	0.37	0.05	Lee et al. (2006)
0.30	0.64	0.09	0.52	0.07	
0.57	1.37	0.21	1.33	0.18	Lee et al. (2004)
0.51	1.29	0.16	0.93	0.13	
0.50	1.42	0.28	1.46	0.25	Lee et al. (2008)
0.36	0.97	0.17	1.03	0.17	
2.05	6.54	1.06	6.96	1.07	This study
3.23	9.31	1.39	8.73	1.27	

Table 2. Eu isotope ratio from SRM and local igneous rocks in this study

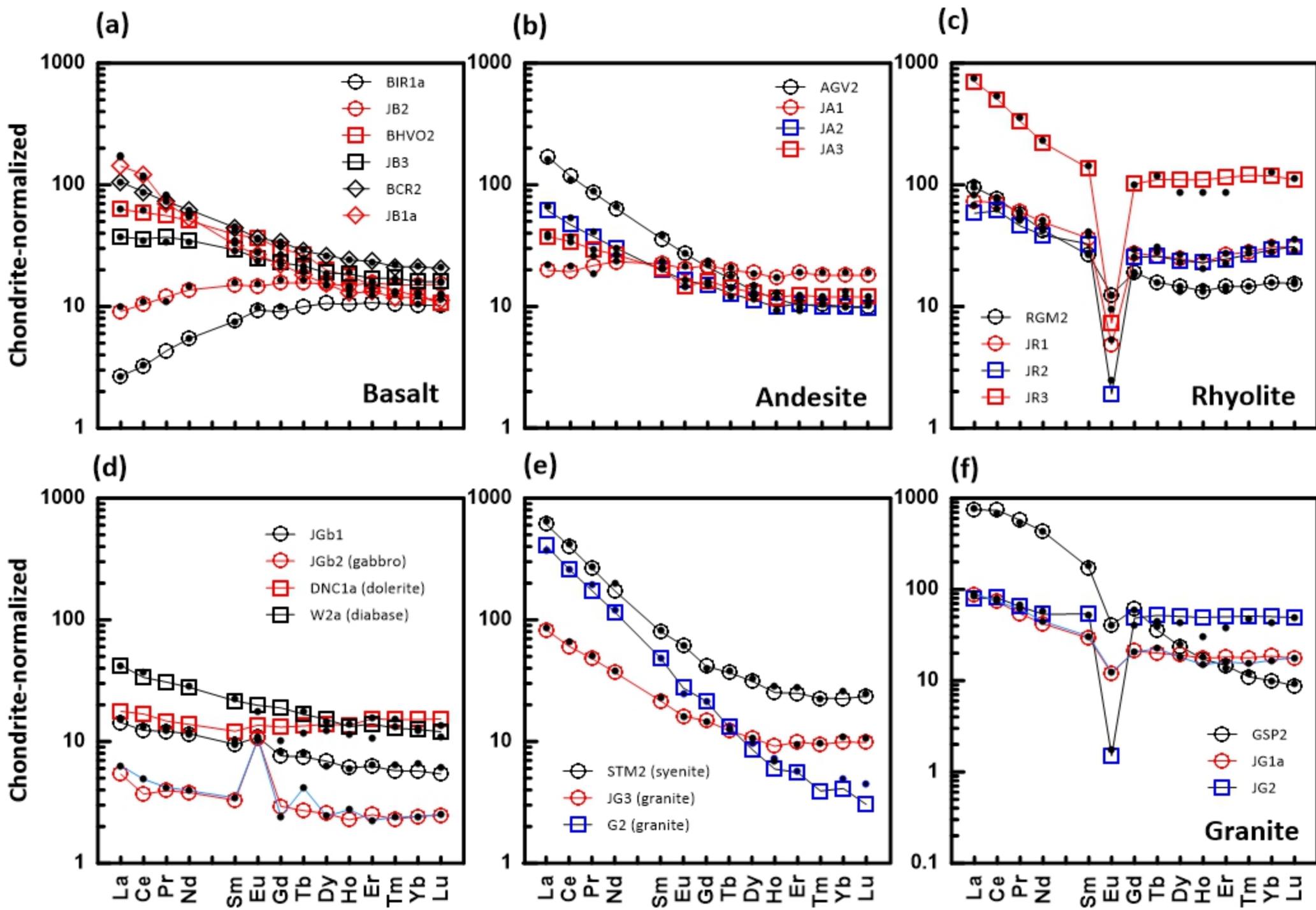
Extrusive (volcanic) rocks																								
Rock type	Basalt (SRM)							Andesite (SRM)				Trachyte (Antarctica)					Rhyolite (SRM)				SRM		Gabbro (SRM)	
Sample name	<b>BCR2<sup>1)</sup></b>	<b>JB2</b>	<b>JB1a</b>	<b>JB1b</b>	<b>JB3</b>	<b>BHVO2</b>	<b>BIR1a</b>	<b>JA1</b>	<b>JA2</b>	<b>JA3</b>	<b>AGV2</b>	<b>M1710</b>	<b>J1301</b>	<b>K1601</b>	<b>M1711</b>	<b>J1301</b>	<b>JR2</b>	<b>JR1</b>	<b>RGM2</b>	<b>JR3</b>	<b>W2a</b>	<b>DNC1a</b>	<b>JGb2</b>	<b>JGb1</b>
Eu/Eu* <sup>2)</sup>	0.94	0.96	0.99	0.94	0.97	1.07	1.13	0.93	0.95	0.80	0.99	0.24	0.16	0.94	1.01	0.25	0.07	0.15	0.56	0.06	0.98	1.08	3.38	1.27
$\delta^{153/151}\text{Eu}^{3)}$	-0.08	-0.02	-0.05	-0.06	-0.05	-0.02	0.09	-0.13	-0.09	-0.08	-0.03	-0.27	-0.41	-0.07	-0.05	-0.54	-0.37	-0.28	-0.10	-0.50	-0.03	0.04	0.08	-0.07
2SD	0.04 (n=12)	0.14 (n=10)	0.14 (n=8)	0.11 (n=2)	0.10 (n=14)	0.17 (n=4)	0.05 (n=7)	0.20 (n=7)	0.09 (n=7)	0.23 (n=7)	0.12 (n=3)	-	-	-	-	-	-	0.08 (n=2)	0.02 (n=3)	0.27 (n=4)	0.04 (n=4)	0.14 (n=6)	0.23 (n=11)	0.19 (n=7)
Intrusive (plutonic) rocks																								
Rock type	Anorthosite					Granitoids													Granitoids (SRM)					
Sample name	SA1703 09-1-1	SA1703 09-2	SA1703 09-3	SA1703 09-6-2	SA1703 10-3-3	MA10	<b>MA<sup>4)</sup></b> <b>2018</b>	<b>WAWR</b> <b>2018</b>	<b>WAWR</b> <b>12</b>	WAWR 8	<b>SM4</b>	<b>SM26</b>	<b>ICH11</b>	<b>ICH3</b>	<b>C2320</b>	<b>C2980</b>	TD13	TD2B	<b>STM2</b>	<b>JG1a</b>	<b>G2</b>	<b>JG3</b>	<b>GSP2</b>	<b>JG2</b>
Eu/Eu*	37.71	12.28	6.29	12.98	9.77	0.01	<b>0.09</b>	<b>0.06</b>	<b>0.06</b>	0.09	<b>0.66</b>	<b>0.93</b>	<b>0.51</b>	<b>0.72</b>	<b>0.81</b>	<b>1.32</b>	0.30	0.21	1.01	0.48	0.79	0.89	0.35	0.03
$\delta^{153/151}\text{Eu}$	0.118	0.125	0.172	0.184	0.244	-0.28	<b>-0.11</b>	<b>-0.07</b>	<b>-0.17</b>	-0.16	<b>0.05</b>	<b>0.00</b>	<b>-0.03</b>	<b>-0.07</b>	<b>-0.02</b>	<b>-0.02</b>	-0.07	-0.18	0.00	-0.03	-0.10	-0.01	-0.07	-0.31
2SD	-	0.01 (n=2)	0.05 (n=3)	0.03 (n=3)	-	0.40 (n=2)	-	-	-	0.31 (n=4)	-	-	-	-	-	-	-	-	0.06 (n=5)	0.06 (n=13)	0.19 (n=4)	0.04 (n=10)	0.05 (n=17)	0.30 (n=6)

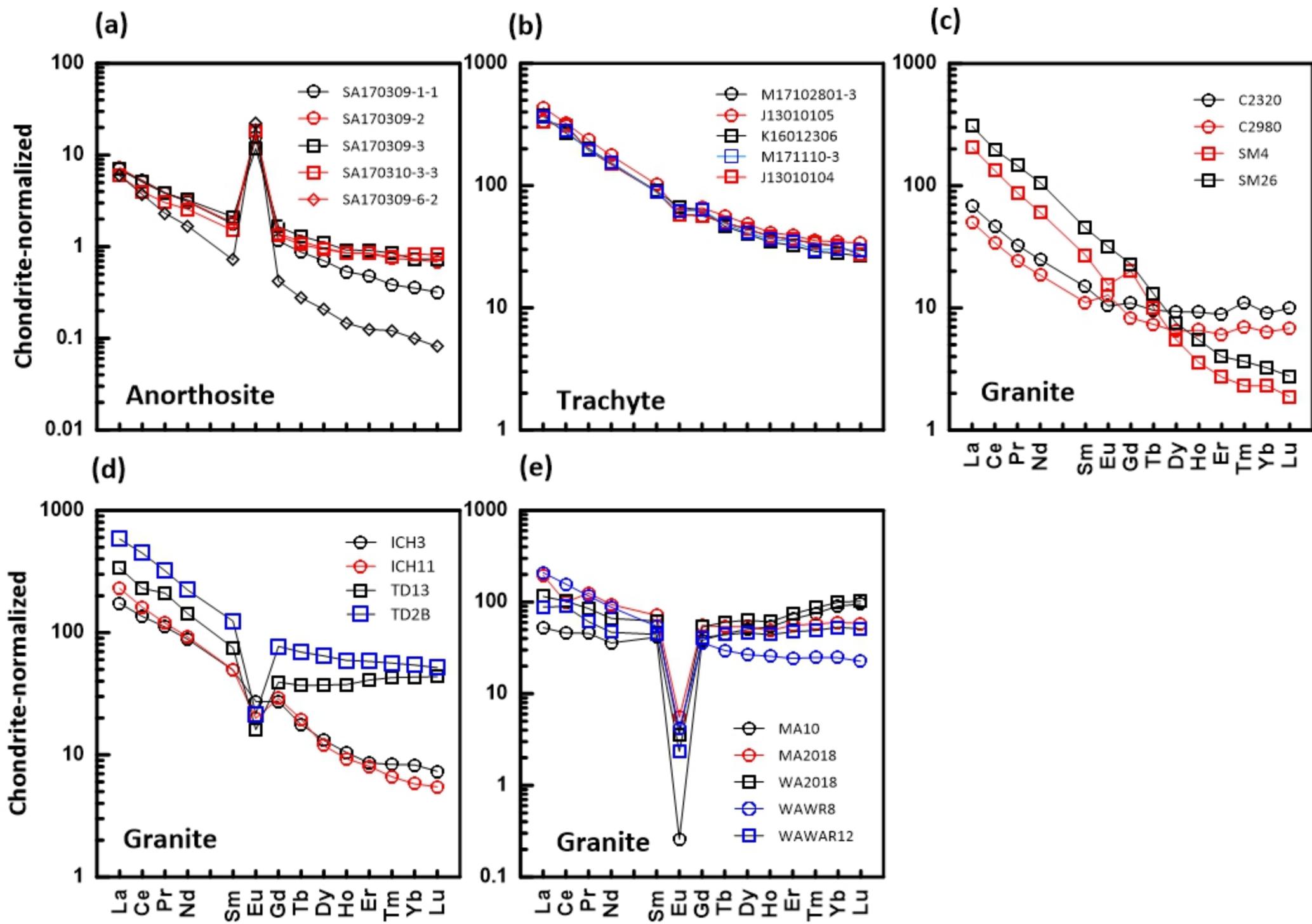
1) The samples of Bold character are SRMs of USGS and GSJ.

2) The magnitude of Eu anomaly is defined as the ratio  $\text{Eu}_N/\text{Eu}^*$  where  $\text{Eu}^*$  is  $\text{SQRT}(\text{Sm}_N \times \text{Gd}_N)$ .

3) Eu isotope ratio normalized by  $^{147}\text{Sm}$ - $^{149}\text{Sm}$  isotope pair (Lee and Tanaka, 2021a, 2021b).

4) Eu isotope data (Bold Italic numbers) are from Lee and Tanaka (2021a)





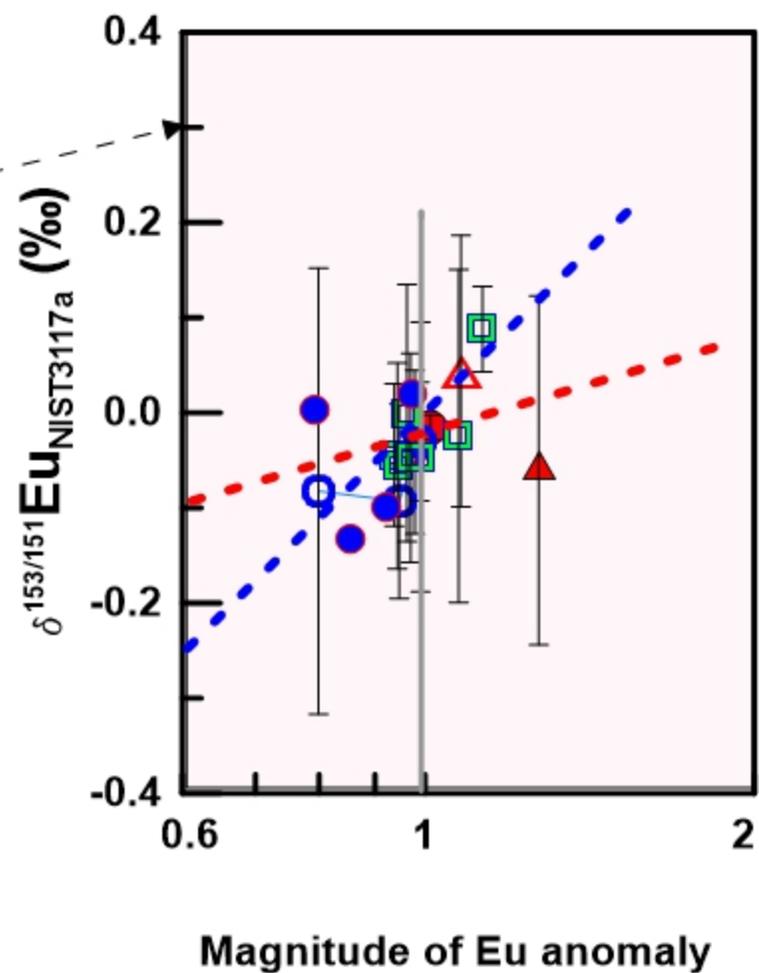
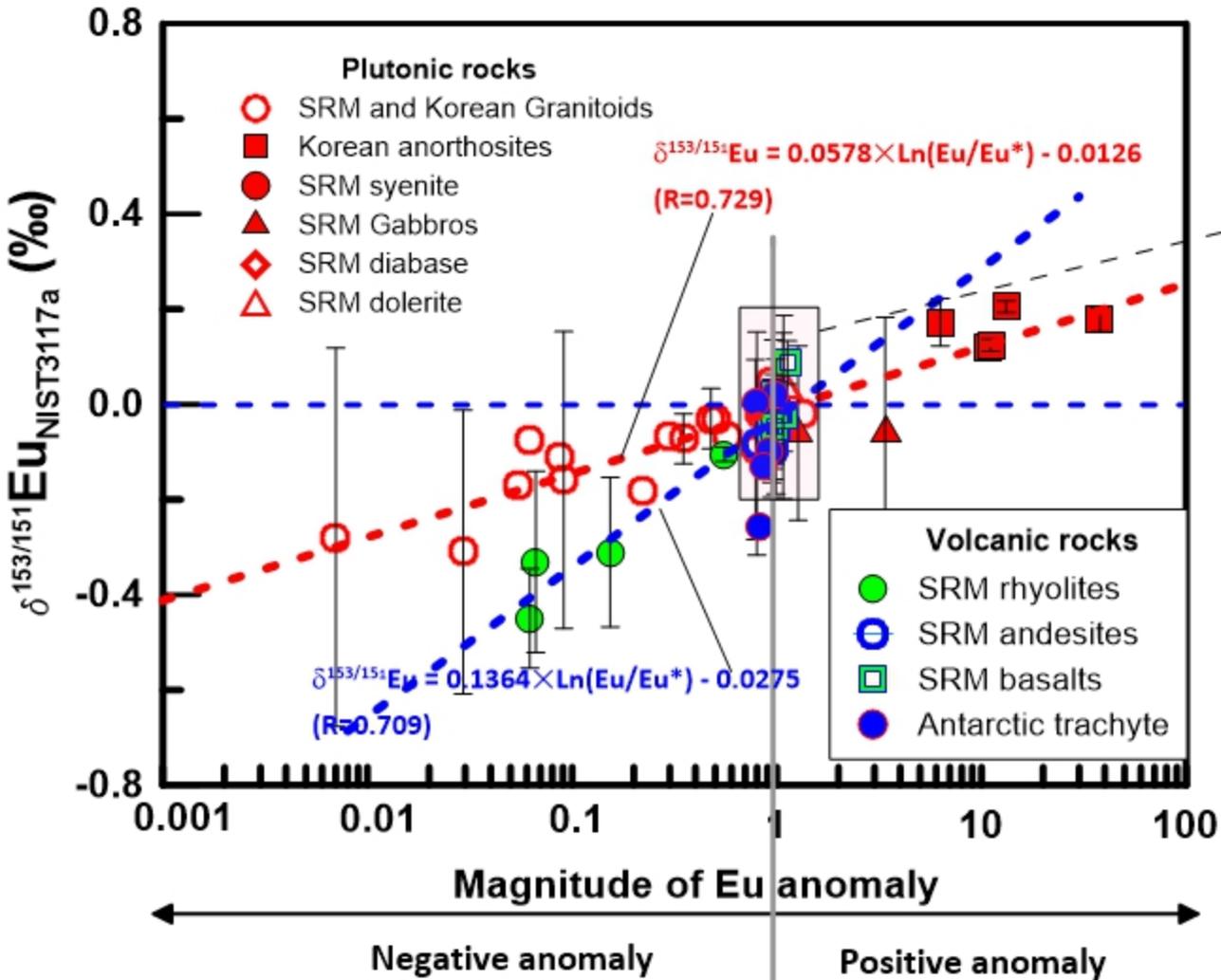


Table S1. Concentrations of Rare earth element of standard reference materials (SRMs) measured in this study

Type	Sample	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Eu/Eu* <sup>1)</sup>	Ref
basalt	<b>BCR2<sup>2)</sup></b>	<b>25.0</b>	<b>53.0</b>	<b>6.80</b>	<b>28.0</b>	<b>6.70</b>	<b>2.00</b>	<b>6.80</b>	<b>1.07</b>	<b>6.41</b>	<b>1.33</b>	<b>3.66</b>	<b>0.54</b>	<b>3.50</b>	<b>0.51</b>	<b>0.90</b>	<b>USGS</b>
		25.0 ± 0.9 (n=4, 1σ <sub>m</sub> )	53.7 ± 1.1 (n=4, 1σ <sub>m</sub> )	6.73 ± 0.16 (n=4, 1σ <sub>m</sub> )	28.5 ± 0.6 (n=4, 1σ <sub>m</sub> )	6.58 ± 0.15 (n=4, 1σ <sub>m</sub> )	2.05 ± 0.04 (n=4, 1σ <sub>m</sub> )	6.76 ± 0.17 (n=4, 1σ <sub>m</sub> )	1.03 ± 0.03 (n=4, 1σ <sub>m</sub> )	6.48 ± 0.13 (n=4, 1σ <sub>m</sub> )	1.32 ± 0.03 (n=4, 1σ <sub>m</sub> )	3.77 ± 0.09 (n=4, 1σ <sub>m</sub> )	0.52 ± 0.01 (n=4, 1σ <sub>m</sub> )	3.41 ± 0.05 (n=4, 1σ <sub>m</sub> )	0.51 ± 0.01 (n=4, 1σ <sub>m</sub> )	0.94	this study
	<b>BHVO2</b>	<b>15.00</b>	<b>38.00</b>	-	<b>25.00</b>	<b>6.20</b>	<b>2.05</b>	<b>6.30</b>	<b>0.90</b>	-	<b>1.04</b>	-	-	<b>2.00</b>	<b>0.28</b>	<b>1.00</b>	<b>USGS</b>
		14.9 ± 0.44 (n=3, 1σ <sub>m</sub> )	36.2 ± 1.80 (n=3, 1σ <sub>m</sub> )	5.17 ± 0.18 (n=3, 1σ <sub>m</sub> )	23.2 ± 1.22 (n=3, 1σ <sub>m</sub> )	5.76 ± 0.16 (n=3, 1σ <sub>m</sub> )	2.05 ± 0.10 (n=3, 1σ <sub>m</sub> )	5.90 ± 0.34 (n=3, 1σ <sub>m</sub> )	0.97 ± 0.07 (n=3, 1σ <sub>m</sub> )	4.89 ± 0.36 (n=3, 1σ <sub>m</sub> )	0.97 ± 0.03 (n=3, 1σ <sub>m</sub> )	2.39 ± 0.17 (n=3, 1σ <sub>m</sub> )	0.26 ± 0.02 (n=3, 1σ <sub>m</sub> )	1.66 ± 0.12 (n=3, 1σ <sub>m</sub> )	0.25 ± 0.02 (n=3, 1σ <sub>m</sub> )	1.07	this study
	<b>BIR1a</b>	<b>0.63</b>	<b>1.90</b>	-	<b>2.50</b>	<b>1.10</b>	<b>0.55</b>	<b>1.80</b>	-	<b>4.00</b>	-	-	-	<b>1.70</b>	<b>0.26</b>	<b>1.13</b>	<b>USGS</b>
		0.64 ± 0.05 (n=3, 1σ <sub>m</sub> )	1.99 ± 0.10 (n=3, 1σ <sub>m</sub> )	0.40 ± 0.05 (n=3, 1σ <sub>m</sub> )	2.49 ± 0.08 (n=3, 1σ <sub>m</sub> )	1.12 ± 0.05 (n=3, 1σ <sub>m</sub> )	0.53 ± 0.02 (n=3, 1σ <sub>m</sub> )	1.80 ± 0.23 (n=3, 1σ <sub>m</sub> )	0.36 ± 0.03 (n=3, 1σ <sub>m</sub> )	2.61 ± 0.20 (n=3, 1σ <sub>m</sub> )	0.57 ± 0.05 (n=3, 1σ <sub>m</sub> )	1.72 ± 0.17 (n=3, 1σ <sub>m</sub> )	0.34 ± 0.03 (n=3, 1σ <sub>m</sub> )	1.94 ± 0.05 (n=3, 1σ <sub>m</sub> )	0.26 ± 0.01 (n=3, 1σ <sub>m</sub> )	1.13	this study
	<b>JB1a</b>	<b>37.60</b>	<b>65.90</b>	<b>7.30</b>	<b>26.00</b>	<b>5.07</b>	<b>1.46</b>	<b>4.67</b>	<b>0.69</b>	<b>3.99</b>	<b>0.71</b>	<b>2.18</b>	<b>0.33</b>	<b>2.10</b>	<b>0.33</b>	<b>0.91</b>	<b>GSJ</b>
		33.7 ± 0.8 (n=6, 1σ <sub>m</sub> )	74.6 ± 9.7 (n=6, 1σ <sub>m</sub> )	6.47 ± 0.18 (n=6, 1σ <sub>m</sub> )	24.7 ± 0.6 (n=6, 1σ <sub>m</sub> )	4.72 ± 0.11 (n=6, 1σ <sub>m</sub> )	1.49 ± 0.03 (n=6, 1σ <sub>m</sub> )	4.43 ± 0.10 (n=6, 1σ <sub>m</sub> )	0.66 ± 0.01 (n=6, 1σ <sub>m</sub> )	3.76 ± 0.16 (n=6, 1σ <sub>m</sub> )	0.70 ± 0.02 (n=6, 1σ <sub>m</sub> )	2.12 ± 0.05 (n=6, 1σ <sub>m</sub> )	0.29 ± 0.01 (n=6, 1σ <sub>m</sub> )	1.91 ± 0.05 (n=6, 1σ <sub>m</sub> )	0.28 ± 0.01 (n=6, 1σ <sub>m</sub> )	0.99	this study
	<b>JB1b</b>	<b>41.20</b>	<b>71.80</b>	<b>7.73</b>	<b>27.10</b>	<b>5.17</b>	<b>1.59</b>	<b>4.38</b>	<b>0.69</b>	<b>3.73</b>	<b>0.67</b>	<b>1.97</b>	<b>0.31</b>	<b>2.10</b>	<b>0.31</b>	<b>1.02</b>	<b>GSJ</b>
		40.20	70.10	7.46	26.90	4.93	1.59	5.35	0.77	4.14	0.79	2.22	0.33	2.01	0.31	0.94	this study
<b>JB2</b>	<b>2.35</b>	<b>6.76</b>	<b>1.01</b>	<b>6.63</b>	<b>2.31</b>	<b>0.86</b>	<b>3.28</b>	<b>0.60</b>	<b>3.73</b>	<b>0.75</b>	<b>2.60</b>	<b>0.41</b>	<b>2.62</b>	<b>0.40</b>	<b>0.95</b>	<b>GSJ</b>	
	2.18 ± 0.8 (n=8, 1σ <sub>m</sub> )	6.39 ± 0.14 (n=8, 1σ <sub>m</sub> )	1.11 ± 0.05 (n=8, 1σ <sub>m</sub> )	6.26 ± 0.17 (n=8, 1σ <sub>m</sub> )	2.22 ± 0.04 (n=8, 1σ <sub>m</sub> )	0.83 ± 0.03 (n=8, 1σ <sub>m</sub> )	3.09 ± 0.07 (n=8, 1σ <sub>m</sub> )	0.56 ± 0.01 (n=8, 1σ <sub>m</sub> )	3.77 ± 0.09 (n=8, 1σ <sub>m</sub> )	0.81 ± 0.04 (n=8, 1σ <sub>m</sub> )	2.51 ± 0.07 (n=8, 1σ <sub>m</sub> )	0.37 ± 0.01 (n=8, 1σ <sub>m</sub> )	2.42 ± 0.06 (n=8, 1σ <sub>m</sub> )	0.37 ± 0.01 (n=8, 1σ <sub>m</sub> )	0.96	this study	
<b>JB3</b>	<b>8.81</b>	<b>21.50</b>	<b>3.11</b>	<b>15.60</b>	<b>4.27</b>	<b>1.32</b>	<b>4.67</b>	<b>0.73</b>	<b>4.54</b>	<b>0.80</b>	<b>2.49</b>	<b>0.42</b>	<b>2.55</b>	<b>0.39</b>	<b>0.90</b>	<b>GSJ</b>	
	8.82 ± 0.70 (n=3, 1σ <sub>m</sub> )	22.0 ± 2.5 (n=3, 1σ <sub>m</sub> )	3.45 ± 0.35 (n=3, 1σ <sub>m</sub> )	16.0 ± 1.78 (n=3, 1σ <sub>m</sub> )	4.31 ± 0.35 (n=3, 1σ <sub>m</sub> )	1.41 ± 0.16 (n=3, 1σ <sub>m</sub> )	4.55 ± 0.56 (n=3, 1σ <sub>m</sub> )	0.78 ± 0.07 (n=3, 1σ <sub>m</sub> )	4.64 ± 0.73 (n=3, 1σ <sub>m</sub> )	1.01 ± 0.12 (n=3, 1σ <sub>m</sub> )	2.71 ± 0.44 (n=3, 1σ <sub>m</sub> )	0.42 ± 0.05 (n=3, 1σ <sub>m</sub> )	2.57 ± 0.22 (n=3, 1σ <sub>m</sub> )	0.39 ± 0.05 (n=3, 1σ <sub>m</sub> )	0.97	this study	
Rhyolite	<b>JR1</b>	<b>19.70</b>	<b>47.20</b>	<b>5.58</b>	<b>23.30</b>	<b>6.03</b>	<b>0.30</b>	<b>5.06</b>	<b>1.01</b>	<b>5.69</b>	<b>1.11</b>	<b>3.61</b>	<b>0.67</b>	<b>4.55</b>	<b>0.71</b>	<b>0.17</b>	<b>GSJ</b>
		17.35	43.83	5.58	22.52	5.37	0.27	5.48	0.94	6.09	1.25	4.25	0.70	4.94	0.76	0.15	this study
	<b>JR2</b>	<b>16.30</b>	<b>38.80</b>	<b>4.75</b>	<b>20.40</b>	<b>5.63</b>	<b>0.14</b>	<b>5.83</b>	<b>1.10</b>	<b>6.63</b>	<b>1.39</b>	<b>4.36</b>	<b>0.74</b>	<b>5.33</b>	<b>0.88</b>	<b>0.07</b>	<b>GSJ</b>
		13.72	37.58	4.53	17.77	4.95	0.09	5.29	0.98	6.39	1.41	4.42	0.74	5.08	0.81	0.05	this study
	<b>JR3</b>	<b>179</b>	<b>327</b>	<b>33.1</b>	<b>107</b>	<b>21.3</b>	<b>0.53</b>	<b>19.7</b>	<b>4.29</b>	<b>21.5</b>	<b>4.70</b>	<b>14.0</b>		<b>20.3</b>	<b>2.80</b>	<b>0.06</b>	<b>GSJ</b>
		167 ± 5 (n=4, 1σ <sub>m</sub> )	309 ± 9 (n=4, 1σ <sub>m</sub> )	30.9 ± 1.5 (n=4, 1σ <sub>m</sub> )	101 ± 3 (n=4, 1σ <sub>m</sub> )	20.1 ± 0.7 (n=4, 1σ <sub>m</sub> )	0.41 ± 0.02 (n=4, 1σ <sub>m</sub> )	20.2 ± 0.72 (n=4, 1σ <sub>m</sub> )	3.99 ± 0.25 (n=4, 1σ <sub>m</sub> )	27.1 ± 1.3 (n=4, 1σ <sub>m</sub> )	6.00 ± 0.33 (n=4, 1σ <sub>m</sub> )	18.3 ± 0.7 (n=4, 1σ <sub>m</sub> )	2.96 ± 0.18 (n=4, 1σ <sub>m</sub> )	19.0 ± 0.8 (n=4, 1σ <sub>m</sub> )	2.71 ± 0.07 (n=4, 1σ <sub>m</sub> )	0.06	this study
<b>RGM2</b>	<b>25.00</b>	<b>48.00</b>	<b>5.00</b>	<b>20.00</b>	<b>4.00</b>	<b>0.70</b>	<b>3.60</b>	<b>0.60</b>	<b>3.30</b>	<b>0.80</b>	<b>2.20</b>			<b>0.40</b>	<b>0.56</b>	<b>USGS</b>	
	22.7 ± 1.8 (n=2, 1σ <sub>m</sub> )	47.2 ± 1.0 (n=2, 1σ <sub>m</sub> )	5.27 ± 0.31 (n=2, 1σ <sub>m</sub> )	19.4 ± 0.90 (n=2, 1σ <sub>m</sub> )	3.91 ± 0.48 (n=2, 1σ <sub>m</sub> )	0.70 ± 0.03 (n=2, 1σ <sub>m</sub> )	3.81 ± 0.31 (n=2, 1σ <sub>m</sub> )	0.57 ± 0.02 (n=2, 1σ <sub>m</sub> )	3.60 ± 0.19 (n=2, 1σ <sub>m</sub> )	0.73 ± 0.06 (n=2, 1σ <sub>m</sub> )	2.33 ± 0.09 (n=2, 1σ <sub>m</sub> )	0.36 ± 0.00 (n=2, 1σ <sub>m</sub> )	0.52 ± 0.09 (n=2, 1σ <sub>m</sub> )	0.38 ± 0.01 (n=2, 1σ <sub>m</sub> )	0.56	this study	
Andesite	<b>JA1</b>	<b>5.24</b>	<b>13.30</b>	<b>1.71</b>	<b>10.90</b>	<b>3.52</b>	<b>1.20</b>	<b>4.36</b>	<b>0.75</b>	<b>4.55</b>	<b>0.95</b>	<b>3.04</b>	<b>0.47</b>	<b>3.03</b>	<b>0.47</b>	<b>0.93</b>	<b>GSJ</b>
		4.69 ± 0.06 (n=6, 1σ <sub>m</sub> )	11.8 ± 1.52 (n=6, 1σ <sub>m</sub> )	2.01 ± 0.04 (n=6, 1σ <sub>m</sub> )	10.7 ± 0.2 (n=6, 1σ <sub>m</sub> )	3.30 ± 0.03 (n=6, 1σ <sub>m</sub> )	1.16 ± 0.02 (n=6, 1σ <sub>m</sub> )	4.31 ± 0.05 (n=6, 1σ <sub>m</sub> )	0.72 ± 0.01 (n=6, 1σ <sub>m</sub> )	4.65 ± 0.12 (n=6, 1σ <sub>m</sub> )	0.94 ± 0.01 (n=6, 1σ <sub>m</sub> )	3.04 ± 0.02 (n=6, 1σ <sub>m</sub> )	0.44 ± 0.00 (n=6, 1σ <sub>m</sub> )	2.91 ± 0.02 (n=6, 1σ <sub>m</sub> )	0.44 ± 0.00 (n=6, 1σ <sub>m</sub> )	0.93	this study
	<b>JA2</b>	<b>15.80</b>	<b>32.70</b>	<b>3.84</b>	<b>13.90</b>	<b>3.11</b>	<b>0.93</b>	<b>3.06</b>	<b>0.44</b>	<b>2.80</b>	<b>0.50</b>	<b>1.48</b>	<b>0.28</b>	<b>1.62</b>	<b>0.27</b>	<b>0.92</b>	<b>GSJ</b>
		14.5 ± 0.25 (n=6, 1σ <sub>m</sub> )	28.8 ± 3.9 (n=6, 1σ <sub>m</sub> )	3.48 ± 0.05 (n=6, 1σ <sub>m</sub> )	13.8 ± 0.2 (n=6, 1σ <sub>m</sub> )	2.93 ± 0.04 (n=6, 1σ <sub>m</sub> )	0.92 ± 0.02 (n=6, 1σ <sub>m</sub> )	2.99 ± 0.03 (n=6, 1σ <sub>m</sub> )	0.46 ± 0.00 (n=6, 1σ <sub>m</sub> )	2.78 ± 0.04 (n=6, 1σ <sub>m</sub> )	0.54 ± 0.01 (n=6, 1σ <sub>m</sub> )	1.69 ± 0.04 (n=6, 1σ <sub>m</sub> )	0.25 ± 0.00 (n=6, 1σ <sub>m</sub> )	1.59 ± 0.03 (n=6, 1σ <sub>m</sub> )	0.24 ± 0.01 (n=6, 1σ <sub>m</sub> )	0.95	this study
	<b>JA3</b>	<b>9.33</b>	<b>22.80</b>	<b>2.40</b>	<b>12.30</b>	<b>3.05</b>	<b>0.82</b>	<b>2.96</b>	<b>0.52</b>	<b>3.01</b>	<b>0.51</b>	<b>1.57</b>	<b>0.28</b>	<b>2.16</b>	<b>0.27</b>	<b>0.83</b>	<b>GSJ</b>
	8.91 ± 0.25 (n=6, 1σ <sub>m</sub> )	21.0 ± 3.9 (n=6, 1σ <sub>m</sub> )	2.74 ± 0.05 (n=6, 1σ <sub>m</sub> )	12.2 ± 0.2 (n=6, 1σ <sub>m</sub> )	3.02 ± 0.04 (n=6, 1σ <sub>m</sub> )	0.82 ± 0.02 (n=6, 1σ <sub>m</sub> )	3.23 ± 0.03 (n=6, 1σ <sub>m</sub> )	0.51 ± 0.00 (n=6, 1σ <sub>m</sub> )	3.21 ± 0.04 (n=6, 1σ <sub>m</sub> )	0.65 ± 0.01 (n=6, 1σ <sub>m</sub> )	1.99 ± 0.04 (n=6, 1σ <sub>m</sub> )	0.29 ± 0.00 (n=6, 1σ <sub>m</sub> )	1.94 ± 0.03 (n=6, 1σ <sub>m</sub> )	0.29 ± 0.01 (n=6, 1σ <sub>m</sub> )	0.80	this study	
<b>AGV2</b>	<b>38.00</b>	<b>68.00</b>	<b>8.30</b>	<b>30.00</b>	<b>5.70</b>	<b>1.54</b>	<b>4.69</b>	<b>0.64</b>	<b>3.60</b>	<b>0.71</b>	<b>1.79</b>	<b>0.26</b>	<b>1.60</b>	<b>0.25</b>	<b>0.91</b>	<b>USGS</b>	
	29.3 ± 0.4 (n=6, 1σ <sub>m</sub> )	74.6 ± 10.5 (n=6, 1σ <sub>m</sub> )	6.26 ± 0.08 (n=6, 1σ <sub>m</sub> )	24.2 ± 0.25 (n=6, 1σ <sub>m</sub> )	4.39 ± 0.05 (n=6, 1σ <sub>m</sub> )	1.41 ± 0.16 (n=6, 1σ <sub>m</sub> )	4.55 ± 0.56 (n=6, 1σ <sub>m</sub> )	0.78 ± 0.07 (n=6, 1σ <sub>m</sub> )	4.64 ± 0.73 (n=6, 1σ <sub>m</sub> )	1.01 ± 0.12 (n=6, 1σ <sub>m</sub> )	2.71 ± 0.44 (n=6, 1σ <sub>m</sub> )	0.42 ± 0.05 (n=6, 1σ <sub>m</sub> )	2.57 ± 0.22 (n=6, 1σ <sub>m</sub> )	0.39 ± 0.05 (n=6, 1σ <sub>m</sub> )	0.96	this study	
<b>G2</b>	<b>89.0</b>	<b>160</b>	<b>18.0</b>	<b>55.0</b>	<b>7.20</b>	<b>1.40</b>	<b>4.30</b>	<b>0.48</b>	<b>2.40</b>	<b>0.40</b>	<b>0.92</b>	<b>0.18</b>	<b>0.48</b>	<b>0.11</b>	<b>0.78</b>	<b>USGS</b>	
	96.1 ± 12.1 (n=2, 1σ <sub>m</sub> )	160 ± 2 (n=2, 1σ <sub>m</sub> )	16.1 ± 1.1 (n=2, 1σ <sub>m</sub> )	53.0 ± 1.3 (n=2, 1σ <sub>m</sub> )	7.18 ± 0.06 (n=2, 1σ <sub>m</sub> )	1.55 ± 0.09 (n=2, 1σ <sub>m</sub> )	4.07 ± 0.17 (n=2, 1σ <sub>m</sub> )	0.47 ± 0.01 (n=2, 1σ <sub>m</sub> )	2.13 ± 0.16 (n=2, 1σ <sub>m</sub> )	0.32 ± 0.04 (n=2, 1σ <sub>m</sub> )	0.89 ± 0.02 (n=2, 1σ <sub>m</sub> )	0.10 ± 0.05 (n=2, 1σ <sub>m</sub> )	0.66 ± 0.11 (n=2, 1σ <sub>m</sub> )	0.07 ± 0.02 (n=2, 1σ <sub>m</sub> )	0.86	this study	

	<b>GSP2</b>	<b>180</b>	<b>410</b>	<b>51.0</b>	<b>200</b>	<b>27.0</b>	<b>2.30</b>	<b>12.0</b>	<b>1.40</b>	<b>6.10</b>	<b>1.00</b>	<b>2.20</b>	<b>0.30</b>	<b>1.60</b>	<b>0.23</b>	<b>0.39</b>	<b>USGS</b>
		176 ± 19 (n=4, 1σ <sub>m</sub> )	453 ± 30 (n=4, 1σ <sub>m</sub> )	53.1 ± 4.4 (n=4, 1σ <sub>m</sub> )	197 ± 15 (n=4, 1σ <sub>m</sub> )	25.6 ± 1.5 (n=4, 1σ <sub>m</sub> )	2.30 ± 0.11 (n=4, 1σ <sub>m</sub> )	12.1 ± 0.6 (n=4, 1σ <sub>m</sub> )	1.29 ± 0.06 (n=4, 1σ <sub>m</sub> )	5.82 ± 0.27 (n=4, 1σ <sub>m</sub> )	0.94 ± 0.03 (n=4, 1σ <sub>m</sub> )	2.34 ± 0.09 (n=4, 1σ <sub>m</sub> )	0.27 ± 0.01 (n=4, 1σ <sub>m</sub> )	1.61 ± 0.03 (n=4, 1σ <sub>m</sub> )	0.22 ± 0.01 (n=4, 1σ <sub>m</sub> )	0.40	this study
Granite	<b>JG1a</b>	<b>21.30</b>	<b>45.00</b>	<b>5.63</b>	<b>20.40</b>	<b>4.53</b>	<b>0.70</b>	<b>4.10</b>	<b>0.81</b>	<b>4.44</b>	<b>0.82</b>	<b>2.57</b>	<b>0.38</b>	<b>2.70</b>	<b>0.44</b>	<b>0.50</b>	<b>GSJ</b>
		20.9 ± 0.69 (n=5, 1σ <sub>m</sub> )	45.0 ± 1.9 (n=5, 1σ <sub>m</sub> )	5.02 ± 0.11 (n=5, 1σ <sub>m</sub> )	19.2 ± 0.5 (n=5, 1σ <sub>m</sub> )	4.32 ± 0.11 (n=5, 1σ <sub>m</sub> )	0.68 ± 0.02 (n=5, 1σ <sub>m</sub> )	4.21 ± 0.16 (n=5, 1σ <sub>m</sub> )	0.72 ± 0.03 (n=5, 1σ <sub>m</sub> )	4.70 ± 0.24 (n=5, 1σ <sub>m</sub> )	0.96 ± 0.06 (n=5, 1σ <sub>m</sub> )	2.90 ± 0.17 (n=5, 1σ <sub>m</sub> )	0.44 ± 0.03 (n=5, 1σ <sub>m</sub> )	2.98 ± 0.22 (n=5, 1σ <sub>m</sub> )	0.43 ± 0.03 (n=5, 1σ <sub>m</sub> )	0.48	this study
	<b>JG2</b>	<b>19.90</b>	<b>48.30</b>	<b>6.20</b>	<b>26.40</b>	<b>7.78</b>	<b>0.10</b>	<b>8.01</b>	<b>1.62</b>	<b>10.50</b>	<b>1.67</b>	<b>6.04</b>	<b>1.16</b>	<b>6.85</b>	<b>1.22</b>	<b>0.04</b>	<b>GSJ</b>
		19.0 ± 0.72 (n=4, 1σ <sub>m</sub> )	49.6 ± 1.3 (n=4, 1σ <sub>m</sub> )	6.00 ± 0.26 (n=4, 1σ <sub>m</sub> )	24.4 ± 0.75 (n=4, 1σ <sub>m</sub> )	7.97 ± 0.22 (n=4, 1σ <sub>m</sub> )	0.08 ± 0.02 (n=4, 1σ <sub>m</sub> )	9.63 ± 0.47 (n=4, 1σ <sub>m</sub> )	1.87 ± 0.13 (n=4, 1σ <sub>m</sub> )	12.4 ± 0.8 (n=4, 1σ <sub>m</sub> )	2.69 ± 0.17 (n=4, 1σ <sub>m</sub> )	8.01 ± 0.38 (n=4, 1σ <sub>m</sub> )	1.26 ± 0.07 (n=4, 1σ <sub>m</sub> )	8.11 ± 0.39 (n=4, 1σ <sub>m</sub> )	1.21 ± 0.07 (n=4, 1σ <sub>m</sub> )	0.03	this study
	<b>JG3</b>	<b>20.60</b>	<b>40.30</b>	<b>4.70</b>	<b>17.20</b>	<b>3.39</b>	<b>0.90</b>	<b>2.92</b>	<b>0.46</b>	<b>2.59</b>	<b>0.38</b>	<b>1.52</b>	<b>0.24</b>	<b>1.77</b>	<b>0.26</b>	<b>0.87</b>	<b>GSJ</b>
		19.7 ± 0.23 (n=6, 1σ <sub>m</sub> )	37.2 ± 4.8 (n=6, 1σ <sub>m</sub> )	4.47 ± 0.07 (n=6, 1σ <sub>m</sub> )	16.9 ± 0.03 (n=6, 1σ <sub>m</sub> )	3.20 ± 0.03 (n=6, 1σ <sub>m</sub> )	0.91 ± 0.02 (n=6, 1σ <sub>m</sub> )	2.95 ± 0.02 (n=6, 1σ <sub>m</sub> )	0.44 ± 0.01 (n=6, 1σ <sub>m</sub> )	2.64 ± 0.03 (n=6, 1σ <sub>m</sub> )	0.50 ± 0.00 (n=6, 1σ <sub>m</sub> )	1.58 ± 0.01 (n=6, 1σ <sub>m</sub> )	0.23 ± 0.00 (n=6, 1σ <sub>m</sub> )	1.60 ± 0.02 (n=6, 1σ <sub>m</sub> )	0.24 ± 0.00 (n=6, 1σ <sub>m</sub> )	0.90	this study
diabase	<b>W2a</b>	<b>10.00</b>	<b>23.00</b>	<b>-</b>	<b>13.00</b>	<b>3.30</b>	<b>1.00</b>	<b>-</b>	<b>0.63</b>	<b>3.60</b>	<b>0.76</b>	<b>2.50</b>	<b>0.38</b>	<b>2.10</b>	<b>0.33</b>	<b>-</b>	<b>USGS</b>
		10.0 ± 0.07 (n=6, 1σ <sub>m</sub> )	20.6 ± 2.8 (n=6, 1σ <sub>m</sub> )	2.86 ± 0.02 (n=6, 1σ <sub>m</sub> )	12.7 ± 0.06 (n=6, 1σ <sub>m</sub> )	3.19 ± 0.04 (n=6, 1σ <sub>m</sub> )	1.12 ± 0.02 (n=6, 1σ <sub>m</sub> )	3.76 ± 0.05 (n=6, 1σ <sub>m</sub> )	0.60 ± 0.01 (n=6, 1σ <sub>m</sub> )	3.74 ± 0.04 (n=6, 1σ <sub>m</sub> )	0.72 ± 0.01 (n=6, 1σ <sub>m</sub> )	2.23 ± 0.01 (n=6, 1σ <sub>m</sub> )	0.32 ± 0.00 (n=6, 1σ <sub>m</sub> )	2.02 ± 0.02 (n=6, 1σ <sub>m</sub> )	0.30 ± 0.01 (n=6, 1σ <sub>m</sub> )	0.98	this study
dolerite	<b>DNC1a</b>	<b>3.60</b>		<b>1.20</b>	<b>5.20</b>	<b>1.41</b>	<b>0.59</b>	<b>2.00</b>	<b>0.42</b>	<b>3.00</b>	<b>0.62</b>	<b>1.70</b>	<b>0.33</b>	<b>2.00</b>	<b>0.27</b>	<b>1.07</b>	<b>USGS</b>
		4.20 ± 0.39 (n=4, 1σ <sub>m</sub> )	10.3 ± 0.9 (n=4, 1σ <sub>m</sub> )	1.37 ± 0.12 (n=4, 1σ <sub>m</sub> )	6.31 ± 0.57 (n=4, 1σ <sub>m</sub> )	1.80 ± 0.17 (n=4, 1σ <sub>m</sub> )	0.77 ± 0.08 (n=4, 1σ <sub>m</sub> )	2.61 ± 0.25 (n=4, 1σ <sub>m</sub> )	0.48 ± 0.04 (n=4, 1σ <sub>m</sub> )	3.40 ± 0.25 (n=4, 1σ <sub>m</sub> )	0.74 ± 0.03 (n=4, 1σ <sub>m</sub> )	2.47 ± 0.22 (n=4, 1σ <sub>m</sub> )	0.37 ± 0.03 (n=4, 1σ <sub>m</sub> )	2.44 ± 0.20 (n=4, 1σ <sub>m</sub> )	0.38 ± 0.03 (n=4, 1σ <sub>m</sub> )	1.08	this study
syenite	<b>STM2</b>	<b>154</b>	<b>256</b>	<b>25.0</b>	<b>81.0</b>	<b>12.0</b>	<b>3.45</b>	<b>8.00</b>	<b>1.38</b>	<b>8.01</b>	<b>1.55</b>	<b>4.40</b>	<b>0.55</b>	<b>4.20</b>	<b>0.60</b>	<b>1.07</b>	<b>USGS</b>
		153 ± 5 (n=2, 1σ <sub>m</sub> )	258 ± 17 (n=2, 1σ <sub>m</sub> )	26.8 ± 0.9 (n=2, 1σ <sub>m</sub> )	79.5 ± 7 (n=2, 1σ <sub>m</sub> )	12.5 ± 0.3 (n=2, 1σ <sub>m</sub> )	3.72 ± 0.08 (n=2, 1σ <sub>m</sub> )	12.5 ± 0.1 (n=2, 1σ <sub>m</sub> )	1.30 ± 0.28 (n=2, 1σ <sub>m</sub> )	7.90 ± 0.11 (n=2, 1σ <sub>m</sub> )	1.54 ± 0.07 (n=2, 1σ <sub>m</sub> )	4.25 ± 0.05 (n=2, 1σ <sub>m</sub> )	0.66 ± 0.04 (n=2, 1σ <sub>m</sub> )	0.69 ± 0.13 (n=2, 1σ <sub>m</sub> )	0.06 ± 0.00 (n=2, 1σ <sub>m</sub> )	1.01	this study
gabbro	<b>JGb1</b>	<b>3.60</b>	<b>8.17</b>	<b>1.13</b>	<b>5.47</b>	<b>1.49</b>	<b>0.62</b>	<b>1.61</b>	<b>0.29</b>	<b>1.56</b>	<b>0.33</b>	<b>1.04</b>	<b>0.16</b>	<b>1.06</b>	<b>0.15</b>	<b>1.22</b>	<b>GSJ</b>
		3.40 ± 0.09 (n=5, 1σ <sub>m</sub> )	7.55 ± 1.2 (n=5, 1σ <sub>m</sub> )	1.12 ± 0.03 (n=5, 1σ <sub>m</sub> )	5.29 ± 0.14 (n=5, 1σ <sub>m</sub> )	1.40 ± 0.04 (n=5, 1σ <sub>m</sub> )	0.61 ± 0.01 (n=5, 1σ <sub>m</sub> )	1.52 ± 0.19 (n=5, 1σ <sub>m</sub> )	0.27 ± 0.01 (n=5, 1σ <sub>m</sub> )	1.70 ± 0.12 (n=5, 1σ <sub>m</sub> )	0.33 ± 0.03 (n=5, 1σ <sub>m</sub> )	1.00 ± 0.07 (n=5, 1σ <sub>m</sub> )	0.14 ± 0.01 (n=5, 1σ <sub>m</sub> )	0.92 ± 0.04 (n=5, 1σ <sub>m</sub> )	0.13 ± 0.01 (n=5, 1σ <sub>m</sub> )	1.27	this study
	<b>JGb2</b>	<b>1.50</b>	<b>3.00</b>	<b>0.39</b>	<b>1.80</b>	<b>0.51</b>	<b>0.59</b>	<b>0.48</b>	<b>0.15</b>	<b>0.60</b>	<b>0.15</b>	<b>0.36</b>	<b>0.06</b>	<b>0.39</b>	<b>0.06</b>	<b>3.63</b>	<b>GSJ</b>
		1.28 ± 0.13 (n=2, 1σ <sub>m</sub> )	2.27 ± 0.42 (n=2, 1σ <sub>m</sub> )	0.37 ± 0.01 (n=2, 1σ <sub>m</sub> )	1.74 ± 0.04 (n=2, 1σ <sub>m</sub> )	0.49 ± 0.01 (n=2, 1σ <sub>m</sub> )	0.59 ± 0.00 (n=2, 1σ <sub>m</sub> )	0.58 ± 0.06 (n=2, 1σ <sub>m</sub> )	0.10 ± 0.03 (n=2, 1σ <sub>m</sub> )	0.63 ± 0.02 (n=2, 1σ <sub>m</sub> )	0.12 ± 0.01 (n=2, 1σ <sub>m</sub> )	0.40 ± 0.02 (n=2, 1σ <sub>m</sub> )	0.06 ± 0.00 (n=2, 1σ <sub>m</sub> )	0.39 ± 0.00 (n=2, 1σ <sub>m</sub> )	0.06 ± 0.00 (n=2, 1σ <sub>m</sub> )	3.38	this study

<sup>1)</sup> The magnitude of Eu anomaly is defined as the ratio  $Eu_N/Eu^*$  where  $Eu^*$  is  $SQRT(Sm_N \times Gd_N)$ . The magnitude was calculated based on the values of Sm, Eu and Gd from the reference.

<sup>2)</sup> The bold number of this row are recommended value by United States of Geological Survey (USGS) or Geological Survey of Japan (GSJ)