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1	Helium isotopes and olivine geochemistry of basalts and mantle xenoliths in
2	Jeju Island, South Korea: Evaluation of role of SCLM on the Cenozoic
3	intraplate volcanism in East Asia
4	Donghwan Kim ^a , Hyunwoo Lee ^{a,*} , Wonhee Lee ^a , Jonguk Kim ^b , Jihye Oh ^{a,b} , Jung-Hun Song ^a
5	Haemyeong Jung ^a , Finlay M. Stuart ^c
6	^a School of Earth and Environmental Sciences, Seoul National University, Seoul 08826,
7	Republic of Korea
8	^b Deep–Sea and Seabed Mineral Resources Research Center, Korea Institute of Ocean Science
9	and Technology, Busan 49111, Republic of Korea
10	°Scottish Universities Environmental Research Centre, East Kilbride G75 0QF, United
11	Kingdom
12	*Corresponding author.
13	E-mail address: lhw615@snu.ac.kr (H. Lee).
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20	SCLM

21 Abstract

22 Jeju Island, South Korea, is a Cenozoic intraplate volcano located in East Asia. The Jeju basalts display ocean island basalt (OIB)-like trace element patterns and enriched mantle type 23 2 (EM2) radiogenic isotope compositions, but the source that enriched the mantle remains 24 25 unclear. Here we report new geochemical compositions of basalts and xenoliths, including the first helium isotope and element analysis of olivine phenocrysts to constrain the source of basalt. 26 Olivines in the Jeju basalts have ${}^{3}\text{He}/{}^{4}\text{He}$ ratios of 3.5 to 7.3 R_a while olivines in the mantle 27 xenoliths are 2.9 to 6.3 R_a. They provide no evidence of a lower mantle plume but overlap the 28 range of the regional subcontinental lithospheric mantle (SCLM) and OIBs with low ³He/⁴He 29 30 values. Olivine phenocrysts included in the basalts show variations in Mn, Ni, and Ca concentrations with forsterite (Fo) contents, similar to the trend for olivines crystallized from 31 the pyroxenite-derived melts. Elemental ratios of Ca-Fe-Ni-Mg-Mn for the olivines also 32 33 indicate the pyroxenite contribution. Considering our results, it is suggested that the main 34 source of the Jeju basalts is the SCLM containing pyroxenite. In addition, the tomography image shows the low-velocity zone extending to the asthenosphere mantle beneath the central 35 36 Jeju Island. This implies that the localized asthenospheric upwelling is caused by edge-driven convection, and the high-velocity zones exist around Jeju island indicating a thick cratonic 37 lithosphere. Therefore, we propose that the interaction between the SCLM component 38 containing pyroxenite and the rising asthenosphere is the main mechanism to generate enriched 39 basaltic magmas for the Cenozoic intraplate volcanism in East Asia, including Jeju Island. 40

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1. Introduction

East Asia, including the Korean Peninsula, mainland China, and adjacent seas, is located 45 on the Eurasian Plate and records extensive Cenozoic intraplate volcanism (Fig. 1a). Although 46 47 trace element and radiogenic isotope compositions (e.g., Sr, Nd, Pb, and Hf) of the lavas are typical ocean island basalt (OIB) compositions (Chen et al., 2007; Choi et al., 2006; Kimura et 48 al., 2018), there is little consensus on the ultimate origin of this magmatism. Most of the basaltic 49 volcanism in this area appear to display mixing between the depleted mantle (DMM) and the 50 EM1 and EM2 enriched mantle components (Choi et al., 2006). There is a broad geographic 51 distinction in the enriched mantle contribution; the melting beneath northeastern China 52 53 contains EM1-type enriched mantle while the intraplate volcanism of Southeast Asia is sourced in mantle contaminated by the EM2 component (Choi et al., 2006). However, processes such 54 as the role of lower mantle plume, contribution from the stagnant slab beneath East Asia, and 55 small local convection in the asthenosphere have been extensively discussed, but there is no 56 clear agreement yet (Brenna et al., 2015; Choi et al., 2006; Kimura et al., 2018; Kuritani et al., 57 2019; Song et al., 2018; Tatsumi et al., 2005; Xu et al., 2017; Zhao et al., 2009). Among the 58 59 arguments, the metasomatized subcontinental lithospheric mantle (SCLM) has been proposed as the source of continental basalts in East Asia (Guo et al., 2020; Xu et al., 2017) as well as 60 61 Australia, East Africa, and West Antarctica (Lu et al., 2020; Nardini et al., 2009; Niu, 2008; Pilet et al., 2008; Rooney, 2020). Regarding the East Asian SCLM, it is suggested that it has 62 experienced thinning/delamination of the ancient cratonic lithosphere during the Late Jurassic-63 64 Early Cretaceous, subduction-related metasomatism during the Late Cretaceous-Early Cenozoic, and the lithospheric thinning since the Early Cretaceous (Correale et al., 2016; Guo 65 et al., 2014; Liu et al., 2019; Ma et al., 2019; Woo et al., 2014; Xu, 2002; Yamamoto et al., 66 2004). 67

In order to test the prevailing hypotheses, we report a study of Jeju Island, a large Cenozoic 68 intraplate volcanic center in the Korean Strait (Brenna et al., 2012a; Koh et al., 2013). 69 70 According to previous studies (Choi et al., 2006; Kim et al., 2019; Tatsumi et al., 2005), trace 71 element patterns are similar to the OIB trend, and radiogenic isotope compositions (Sr, Nd, Hf, 72 and Pb) show the DMM-EM2 mixing relationship, like basalts reported in southeast Asia. In a 73 recent study (Kim et al., 2019), it has been suggested that the Jeju magma source is likely garnet peridotite but contains recycled components such as rutile bearing eclogite, carbonate, and 74 sedimentary components. Lower mantle, asthenosphere, and SCLM components have been 75 proposed for the origin of the Cenozoic magmatism in East Asia (Brenna et al., 2015; Choi et 76 77 al., 2006; Kimura et al., 2018; Tatsumi et al., 2005). Also, it has been presented that the stagnant Pacific plate slab beneath this region played an important role in the genesis of enriched 78 79 magmas (Choi et al., 2006; Kim et al., 2019). The Jeju basalts were reported to have different 80 radiogenic isotope (Sr, Nd, and Pb) compositions compared to peridotite xenoliths, suggesting that magma has been enriched regardless of the contribution of the lithosphere (Baek et al., 81 2014; Choi et al., 2006). However, pyroxenite segregates in the metasomatized SCLM are 82 typically enriched, and subducted crustal components were stored in the lithosphere in the form 83 of pyroxenite, which contributed to the source of magma through tectonic reactivation (Lu et 84 85 al., 2020).

Helium isotope ratios (${}^{3}\text{He}/{}^{4}\text{He}$) are powerful for tracking the role of deep mantle and SCLM in intraplate volcanism. The lower mantle likely has ${}^{3}\text{He}/{}^{4}\text{He} > 50 R_{a}$ (e.g., Stuart et al., 2003) while the SCLM is typically lower (6 ± 1 R_a; e.g., Gautheron and Moreira, 2002) than depleted upper mantle (DMM) that melts beneath mid-ocean ridges (8 ± 1 R_a; Graham, 2002). Here we report the first analysis of the helium isotope compositions of young (< 100 ka) basalts and mantle xenoliths from Jeju Island. These are combined with major and trace element compositions of whole rocks and olivine phenocrysts to understand the characteristics of the 93 magma source.

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96 **2. Geological setting**

Jeju Island, South Korea, is a Cenozoic intraplate volcano located in the Korea Strait, 650 97 98 km from the Nankai Trough, the nearest convergent margin (Fig. 1). The subducting slab of the 99 Philippine Sea Plate is not present beneath Jeju (Wei et al., 2015). A stagnant slab of the 100 detached Pacific Plate has been detected in the mantle transition zone (MTZ) beneath the Korea 101 Strait (Huang and Zhao, 2006; Wei et al., 2015) although a more recent full-waveform seismic 102 tomography and P-wave anisotropic tomography study did not detect it (Ma et al., 2019; Tao et al., 2018). The East Asian SCLM has experienced lithospheric thinning since the Early 103 Cretaceous likely caused by the subducting Pacific plate (Correale et al., 2016; Guo et al., 2014; 104 105 Liu et al., 2019). Jeju Island sits on the continental crust (24.8 to 35 km) which is significantly 106 thinner than the Korean Peninsula and East China (Kim et al., 2015; Yoo et al., 2007). The 107 lithospheric thickness is estimated to be about 60 km beneath the central part of Jeju Island but thickens to the north, west, and east (Song et al., 2018). 108

Mt. Halla is the central volcanic edifice of Jeju. Volcanism in the past 0.1 Ma has been generated from about 300 scoria cones which are widely distributed across the island (Fig. 1b). The volcanic activity of Jeju is mainly divided into submarine (1.88 Ma to 0.5 Ma) and subaerial periods (0.5 Ma to Holocene), separated by the hydrovolcanic activity of the Seoguipo Formation (Koh et al., 2013). During the subaerial period (Stage 1 of Brenna et al., 2015), the melt flux was relatively low, and both high Al alkaline basalts and trachytic lava flows erupted (Brenna et al., 2015, 2012b). During the subaerial period, the eruption rate increased, forming the current morphology of the island (Brenna et al., 2015). The subaerial
period is divided into two stages; stage 2 (500-250 ka) and stage 3 (< 250 ka) (Brenna et al.,
2015). Stage 2 is characterized by the transitional alkaline basalts. During stage 3, low-Al alkali
basalts erupted dominantly as tholeiites in the western and eastern areas, and continued into
historical times (Lee and Yang, 2006).

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3. Samples and analytical methods

In this study, we have analyzed fresh stage 3 basalts from the Daepo-Dong (DB) region in the 123 south, Sinchang-Ri (SB) in the west, and Kwideok-Ri (KB) in the northwest of Jeju, 124 respectively (Fig. 1). We also collected mantle xenolith fragments hosted in basaltic rocks from 125 Sinsan-Ri (SX) in east Jeju (Fig. 1). The DB samples are low-Al alkaline basalts, while rocks 126 127 from SB and KB are tholeiitic basalts (Brenna et al. 2012b; Koh et al. 2013). The DB basalts contain olivine, clinopyroxene, and plagioclase phenocrysts while the SB and KB basalts 128 129 include only olivine phenocrysts. Mantle xenoliths discovered from Jeju Island can be divided into two groups. Group 1 xenoliths are spinel lherzolite and spinel harzburgite, containing Cr-130 131 rich clinopyroxene. Group 2 xenoliths are dunite, wehrlite to olivine clinopyroxenite, and 132 olivine websterite to websterite, having Al-Ti-rich clinopyroxene (Choi et al., 2005; Woo et al., 2014; Yang et al., 2012b). In this study, all xenolith samples are protogranular spinel lherzolites 133 composed of olivine, orthopyroxene (enstatite), and clinopyroxene (diopside). The Jeju 134 135 lherzolites imply that the SCLM was formed during the Paleoproterozoic (2.1 to 1.8 Ga) according to Os T_{RD} ages and experienced subduction-related metasomatism (Lee and Walker, 136 2006; Woo et al., 2014). Olivines in the xenoliths have Fo contents of 88.6 to 90.8 (Table 2) 137 typical of upper mantle xenoliths (Choi et al., 2005). 138

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Major element compositions were measured by X-ray fluorescence spectrometry (Shimadzu

XRF-1800) at the Cooperative Laboratory Center, Pukyong National University, Republic of 140 Korea. Mineral grains were epoxy-mounted and major elements were measured by a field 141 emission electron probe microanalyzer (FE-EPMA, JXA-8350F, JEOL) at the National Center 142 for Interuniversity Research Facilities (NCIRF) of Seoul National University, Republic of 143 144 Korea. Operating conditions were 15 kV accelerating voltage and 20 nA beam current with a 145 beam spot size of 3µm. The reproducibility of the data was verified using the Smithsonian standards (olivine; NMNH 111312-44, augite; NMNH 164905, and chromite; NMNH 117075), 146 with the reproducibility of major elements (RSD%) less than 1.2 % except for FeO (2.65 %) in 147 augite (Supplementary data; Table S3). 148

149 Trace element concentrations of whole-rock and mineral grains were measured by LA-ICP-MS (NWR 193 laser ablation system coupled to Agilent 7000x Inductively Coupled Plasma 150 Mass Spectrometer) at Korea Institute of Ocean Science and Technology (KIOST), Republic 151 of Korea. The analysis was performed with output energy of 3 J/cm² and a repetition rate of 5 152 Hz, with a spot size of 105 µm for each run for 40 seconds. The LA-ICP-MS analysis set the 153 same points as the EPMA targets for mineral grains to be used with the FE-EPMA results and 154 internal standards. NIST 612 was used for an external standard, and BCR-2G was used as a 155 secondary external standard to verify accuracy and precision. The Iolite 4 software was used 156 to reduce the amount of trace elements in samples from raw data. Internal standards included 157 Ca in clinopyroxene and the basalt beads, Si in orthopyroxene and olivine, and Cr in spinel, 158 measured by XRF and FE-EPMA. In the secondary external standard (BCR-2G), excluding Zn, 159 160 accuracy, and precision (RSD%) of most trace element concentrations were better than 17 and 8%, respectively, and the amount was measured between 33 and 56% higher than original 161 amounts (Supplementary data; Table S4). 162

163 The basalts and mantle xenoliths were crushed using titanium alloy mortar, sieved and 1-3

mm olivine and clinopyroxene were picked under a binocular microscope. The grains were 164 165 cleaned with distilled water using Ultrasonicator then ethyl alcohol. Helium isotope compositions (³He/⁴He) were measured at Noble Gas Isotope Laboratory, Scottish Universities 166 Environmental Research Centre (SUERC), from gases extracted from melt/fluid inclusions by 167 in vacuo crushing in a hydraulic press. Gas purification procedures are similar to those 168 described previously (Williams et al., 2005). Helium isotope composition was determined 169 170 using a ThermoFisher Helix-SFT mass spectrometer (Carracedo et al., 2019). The sensitivity and mass fractionation of the mass spectrometer were determined by repeated analyses of the 171 172 HESJ International standard (Matsuda et al., 2002).

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175 **4. Results**

Major and trace element compositions of whole rocks are summarized in Table 1 and plotted 176 in Fig. 2. As reported in previous studies on Jeju Island (Baek et al., 2014; Brenna et al., 2015; 177 Tatsumi et al., 2005), the alkaline and subalkaline basalts belong to the low-Al alkaline and 178 tholeiitic types based on SiO₂ and Al₂O₃ contents, respectively (Fig. 2b). The Ti-Mn-P diagram 179 shows that the Jeju basalts fall in the oceanic island basalt (OIB) and oceanic island tholeiite 180 (OIT) fields (Fig. S1). CI chondrite-normalized rare earth element (REE) abundances display 181 light rare earth element (LREE) enriched trends, similar to the typical OIB pattern (Fig. 3a), 182 183 while primitive mantle normalized trace element patterns are also similar to the OIB trend (Fig. 3b). The trace element patterns of the Jeju basalts are distinguished from the continental crust 184 185 trend, exhibiting enrichment of large ion lithophile elements (LILE) without depletion of high field strength elements (HFSE). This may indicate that there is no role for metasomatism 186 caused by subduction-related fluids in the mantle source (Fig. 3b). 187

The major and minor element contents of the Jeju xenoliths (Table 2) are homogenous and 188 similar to those previously reported for Jeju Island and East China (Choi et al., 2005; Correale 189 et al., 2016; Su et al., 2014; Woo et al., 2014). The REE patterns of clinopyroxenes in the Jeju 190 xenoliths are generally spoon-shaped (Fig. 4), with relatively flat trends of the middle (MREE) 191 192 and heavy rare earth elements (HREE). In addition, both LREE-depletion (SX-01 and SX-04) 193 and LREE-enrichment (SX-02) are observed (Fig. 4). These various LREE enrichments represent various degrees of metasomatism. Furthermore, most of the xenoliths exhibit Nb-Ta 194 depletion with U-Th enrichment (Fig. 4), and Woo et al. (2014) argued that subduction-related 195 fluids may have affected the lithosphere. 196

197 The ${}^{3}\text{He}/{}^{4}\text{He}$ ratios of the Jeju basalts range from 3.5 to 7.3 R_a (Table 5). There is no clear correlation between the ³He/⁴He ratios and He concentrations, nor any systematic differences 198 in ³He/⁴He of co-genetic olivine and clinopyroxene (Fig. 5). The ³He/⁴He ratios of olivines in 199 the xenoliths extend to slightly lower values but largely overlap the basalt values (2.9 to 6.5 200 R_a). The absence of any relationship between ${}^{3}He/{}^{4}He$ and He concentration rules out post-201 eruptive He ingrowth. The ³He/⁴He ratios have a distinctly narrower range than the East Asian 202 SCLM range (Korean Peninsula; 0.2 to 7.9 R_a, North China Craton; 2 to 10.5 R_a, Yangtze 203 Craton; 3.01 to 9.15 R_a, and Far East Russia; 0.3 to 8 R_a) (Correale et al., 2016; Kim et al., 204 2005; Tang et al., 2014; Yamamoto et al., 2004). This may reflect a lower degree of 205 heterogeneity in the mantle beneath Jeju. The crushing technique used in this study releases 206 less than 0.1% of any radiogenic and cosmogenic He in the mineral lattice (Carracedo et al., 207 2019). However, other approaches could release lattice-hosted radiogenic and cosmogenic He 208 (e.g., Sumino et al., 2000; Tang et al., 2014). Consequently, the narrower ³He/⁴He range that 209 we report might also reflect the absence of released radiogenic and cosmogenic He that seemed 210 to be present in other studies. 211

The olivine phenocrysts in the Jeju basalts have Fo contents ranging from 69.6 to 79.7 (Table 3). These are lower than typical primary magma compositions, likely reflecting fractional crystallization prior to eruption. The variations of Mn, Ni, and Ca with Fo values are similar to other olivine phenocrysts of basalts from North China and Hainan (Fig. 7). These element contents show small ranges in the same Fo values and good correlations with Fo contents, implying that fractional crystallization is the key process rather than source lithology, mantle temperature, and crystallization pressure (Rasmussen et al., 2020; Sobolev et al., 2007).

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221 **5. Discussion**

5.1. Crustal assimilation

The assimilation of the radiogenic He-bearing crustal basement by the basaltic magma is 223 224 likely to lower the initial magmatic ³He/⁴He ratio. However, the deep crystallization of olivines 225 prior to their long period of residence in the continental crust means that, despite evidence of contamination in basalt chemistry or isotopic composition, they can be almost immune to 226 crustal contamination (e.g., Stuart et al., 2003). The basement beneath Jeju Island consists of 227 Jurassic-Early Cenozoic granite and Paleo-Proterozoic anorthosite (Baek et al., 2014; Choi et 228 al., 2006; Kim et al., 2019; Tatsumi et al., 2005), both of which are expected to be concentrated 229 230 with radiogenic ⁴He. The trace element compositions of the Jeju basalts yield no strong evidence for crustal contamination, for example, enrichment of LILE (e.g., Rb, Ba, Th, and U) 231 or depletion of HFSE (e.g., Nb, Ta, and Ti) and Sr-Nd-Pb-Mg isotope compositions (Kim et al., 232 2019; Tatsumi et al., 2005). The Ce/Pb and Nb/U ratios for the basalts are slightly lower than 233 the OIB-MORB ranges (Hofmann et al., 1986) and are close to the upper continental crust 234

ratios (Rudnick and Gao, 2003) (Fig. 6a, b). However, the Th/Nb ratios are relatively uniform
compared to the La/Sm ratios (Fig. 6c), and there is no trend of crustal contamination. Previous
studies have shown that low-Al alkaline basalts have no meaningful correlation between
radiogenic isotope (Sr-Nd) compositions and SiO₂ contents, which indicate crustal assimilation
(Brenna et al., 2012b; Kim et al., 2019; Tatsumi et al., 2005). The slightly lower Ce/Pb ratios
of low-Al alkaline basalt samples could reflect source characteristics such as enriched sources
of the SCLM, which are not identical to MORB and OIB sources.

Baek et al. (2014) and Kim et al. (2019) suggested that some of the Jeju tholeiitic and high-242 Al alkaline basalts might be contaminated by anorthosite due to the presence of fragments in 243 the tholeiitic basalts (Baek et al., 2014; Yang et al., 2012a) showing positive anomalies of Sr 244 245 and Eu as well as positive ⁸⁷Sr/⁸⁶Sr ratio-Sr concentration trends (Tatsumi et al., 2005; Baek et al., 2014; Kim et al., 2019). Compared to the alkaline basalts, the tholeiitic basalts have 246 relatively high ²⁰⁷Pb/²⁰⁴Pb ratios with moderate ²⁰⁸Pb/²⁰⁴Pb ratios, which may indicate 247 248 assimilation with anorthosite (Kim et al., 2019; Baek et al., 2014). Since the elevated ⁸⁷Sr/⁸⁶Sr and ²⁰⁷Pb/²⁰⁴Pb ratios are more remarkable in the western region of Jeju Island compared to 249 other regions (Kim et al., 2019), the tholeiitic samples from western Jeju cannot ignore crustal 250 251 assimilation. Accordingly, the He isotope compositions from tholeiitic basalt samples can be modified by crustal assimilation. The low ${}^{3}\text{He}/{}^{4}\text{He}$ (3.5 R_a) ratios of basalt SB-01 may be due 252 253 to the addition of radiogenic ⁴He. Therefore, the ³He/⁴He ratios of the low-Al alkaline basalts 254 can represent the He isotope signatures of Jeju Island, and only the ³He/⁴He ratios of the low-255 Al alkali basalts will be discussed in the following sections in order to identify the source of 256 magma.

The ${}^{3}\text{He}/{}^{4}\text{He}$ ratios of the Jeju basalts (5.6 to 7.3 R_a) show no evidence for lower mantle 259 signatures represented by the value of >10 Ra (e.g., Stuart et al. 2003). This tends to refute 260 arguments in favor of a lower mantle plume origin for the hotspot volcanism (Kimura et al., 261 2018; Tatsumi et al., 2005) and is consistent with the absence of hotspot tracks, topographical 262 expansion, and deep thermal anomalies (Brenna et al., 2012b; Kim et al., 2019; Ma et al., 2019; 263 Song et al., 2018). The Sr-Nd-Pb-Hf isotope compositions of the Jeju basalts are consistent 264 with the incorporation of the EM2-type mantle component in the melt source region (Choi et 265 al., 2006; Kim et al., 2019). Given these backgrounds, there could be two possible causes of 266 267 low ³He/⁴He ratios with EM2 signatures in the Jeju volcanism, (i) enriched asthenosphere mantle or (ii) metasomatized SCLM. 268

(i) Many OIBs are characterized by low ³He/⁴He ratios and are typically ascribed to 269 heterogeneity in the asthenosphere mantle owing to the survival of the enriched components 270 (Day et al., 2015; Day and Hilton, 2011; Hanyu et al., 2014; Jackson et al., 2014; Parai et al., 271 272 2009). The northeastern Asian asthenosphere has been metasomatized by fluids released from the Pacific stagnant slab at the depth of the MTZ (Kim et al., 2019; Kuritani et al., 2019, 2011; 273 Zhao et al., 2009). However, it is unlikely that the subducted stagnant slab exists beneath Jeju 274 Island. Recent P-wave anisotropic tomography (Ma et al., 2019) and full-waveform seismic 275 tomography (Tao et al., 2018) show that the subducted Pacific plate is cut off beneath Jeju 276 Island. In addition, the subducted Philippine Sea plate extends to a depth of 300 km and, 277 according to the Vp and Vs tomography, does not reach beneath Jeju Island (Wei et al., 2015). 278 The trace elements and Sr-Nd-Pb isotope compositions, which reflect the DMM component 279

(BABB) of the Yamato basin (ODP 797) and the Japan basin (ODP 794) located in the East

with minor subducted sediment signatures, have been reported from the back-arc basin basalts

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282 Sea (Japan Sea) (Chen et al., 2015; Hirahara et al., 2015). It is known that helium in the

subducted sediments and the oceanic crust is degassed in the subduction zone, and the recycled 283 materials almost lost the He contents (Staudacher and Allègre, 1988). Most of the BABB 284 samples from the Mariana, Manus, Lau, and North Fiji basins exhibit MORB (8±1 R_a) like 285 helium isotope signatures, excluding deep mantle plume-related samples in the Lau and Manus 286 basins (Hilton et al., 2002). The BABBs from the Southern Okinawa trough have ³He/⁴He ratios 287 up to 7.9 R_a, despite their high ⁸⁷Sr/⁸⁶Sr ratios indicating subducted sediments (Yu et al., 2016). 288 Therefore, it is difficult to explain the low ³He/⁴He ratios of the basalts from Jeju Island only 289 290 with the heterogeneous asthenosphere mantle without the contribution of SCLM sources.

(ii) The Jeju basalts with ${}^{3}\text{He}/{}^{4}\text{He}$ ratios of 5.6 to 7.3 R_a overlap the range of the mantle 291 292 xenoliths (2.9 to 6.5 R_a). These are measurably lower than the depleted upper mantle value sampled by MORB (Graham, 2002), and more characteristics of SCLM (Chen et al., 2007; 293 294 Correale et al., 2016; Gautheron and Moreira, 2002; Tang et al., 2014; Yamamoto et al., 2004). 295 The trace elements and isotope compositions of the Jeju mantle xenoliths imply that the underlying SCLM has experienced metasomatism due to subduction-related fluids and melts 296 (Kim et al., 2005; Woo et al., 2014). Based on the Mg# values of xenolith olivines, Xia et al. 297 298 (2020) subdivided the lithosphere mantle underneath China into depleted (Mg# > 92) and fertile (Mg# < 90) regions, showing that the fertile lithosphere exists beneath East China, close to Jeju 299 Island. Olivine grains in the Jeju mantle xenoliths typically have the Mg# values of < 90 300 consistent with the metasomatized SCLM. 301

The Jeju xenoliths have three REE patterns: LREE depletion, LREE enrichment, and spoonshaped (LREE enrichment and MREE depletion) patterns (Fig. 4a). These indicate that the lherzolite xenoliths experienced different degrees of depletion or enrichment. Even other harzburgites and mylonitic lherzolite xenoliths from Jeju Island have more LREE enrichment than lherzolites (Woo et al., 2014). According to the Re-Os isotope compositions of the SCLM

beneath the Korean Peninsula, Paleoproterozoic harzburgites (1.9-1.8 Ga) are preserved, and 307 308 the isotope ratios represent the recent perturbation of Re concentrations in lherzolites (Lee and Walker, 2006). Woo et al. (2014) argued that metasomatism of the SCLM resulted from arc-309 related fluids because of the secondary orthopyroxene morphology and the presence of 310 phlogopite in the lherzolite xenoliths. Due to the differences in the radiogenic isotopes between 311 the peridotite xenoliths (DMM) with the host basalts (EM2), the SCLM has not been considered 312 313 the source of the magma, instead, the ancient recycled sediments wandering in the asthenosphere have been preferred as the source of EM2 signatures (Baek et al., 2014; Choi et 314 315 al., 2006). The Jeju basalt samples do not have any subduction-related signatures found in the Jeju lherzolite xenoliths. All of the xenoliths from Jeju Island are spinel lherzolites, and 316 pyroxenites is thought to be derived from the upper part of the SCLM. However, garnet has not 317 been observed in peridotite and pyroxenite xenoliths which are potentially considered to be the 318 source lithology of the Jeju basalts (Choi et al., 2005; Kim et al., 2019). 319

320 Recent mantle tomography studies (Song et al., 2020, 2018) have found that the thick cratonic lithosphere is located around Jeju Island, which could be associated with the East 321 Asian SCLM. In North China Craton, various pyroxenite xenoliths displaying EM1 and EM2 322 323 signatures, which are distinct from peridotite xenoliths (DMM), have been sampled and argued as being attributed to recycled crustal materials (Xu, 2002). The metasomatized SCLM could 324 contribute to the basalt source and affect the low ³He/⁴He ratios. However, the highest ³He/⁴He 325 ratio (7.3 R_a) of the Jeju basalts falls in the range of MORB (7 to 9 R_a), which represents the 326 asthenospheric mantle contribution. The radiogenic isotope compositions of the Jeju basalts 327 328 have a mixing trend between the DMM and EM2 components (Choi et al., 2005; Kim et al., 2019; Tatsumi et al., 2005). Hence, it is probable that the formation of the Jeju magma is mainly 329 due to the interaction between the enriched SCLM and depleted MORB mantle components. 330

The highest ${}^{3}\text{He}/{}^{4}\text{He}$ ratio (6.9 R_a) of the tholeiitic basalt sample (KB-01) falls within the range 331 of the alkali basalts (5.6 to 7.3 R_a) (Fig. 5). Compared to the alkaline basalts, the tholeiitic 332 basalts are less enriched in incompatible elements (Fig. 3) and have higher ⁸⁷Sr/⁸⁶Sr ratios, 333 lower ¹⁴⁴Nd/¹⁴³Nd ratios, and higher Δ 7/4 Pb values (Kim et al., 2019; Tatsumi et al., 2005). 334 335 These differences in the radiogenic isotopes between the tholeiitic and alkaline basalts have 336 been discussed in terms of the different depths of magma formation (Brenna et al., 2012b, 2012a; Tatsumi et al., 2005) or anorthosite assimilation (Kim et al., 2019). Nevertheless, the 337 338 tholeiitic and alkaline basalts have similar trends in the source lithologies based on the olivine compositions (Figs. 7 and 8) and whole-rock compositions (Kim et al., 2019). Therefore, the 339 340 tholeiite and alkaline basalts originated from the same source, but the divergence into two different basalt types can be attributed to different degrees of partial melting/fractional 341 crystallization. 342

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5.3. Olivine chemistry of the Jeju basalts

In general, basaltic magmas are formed from the melting of peridotite and pyroxenite 345 components. The pyroxenite component is considered to be related to recycled crustal materials 346 347 from the subducted slab (Sobolev et al., 2007). For this reason, the pyroxenite-derived melts can be distinguished from the peridotite-derived melts based on the minor and trace element 348 compositions (e.g., Mn, Ca, and Ni) in olivine phenocrysts of basaltic rocks (Foley et al., 2013; 349 350 Rasmussen et al., 2020). Olivines in the Jeju basalts have Fo contents of 68 to 80, which are lower than the primary mantle olivine values (88 to 92; Foley et al., 2013). Hence, it is 351 necessary to consider the chemical variation according to the Fo content of olivine. The 352 Petrolog program was adopted to examine some effects on magma compositions such as 353

fractional crystallization and crustal assimilation (Rasmussen et al., 2020). The melt components derived from peridotite and pyroxenite were referenced from Walter (1998) and Sobolev et al. (2007). According to Yang et al. (2012a), the depth of the Jeju magma chamber was considered to be the depth of the lower continental crust, and the crustal thickness beneath this region was reported to be 24.8~35 km (Kim et al., 2015; Yoo et al., 2007). Thus, we assumed a situation where the fractional crystallization of olivine, clinopyroxene, and plagioclase occurs at 7 kbar.

The Fo contents of olivines in the Jeju basalts show clear correlations with Mn, Ni, and Ca 361 concentrations (Fig. 7). Although olivines in the low-Al alkaline basalts have lower Fo contents 362 than the tholeiitic basalts, the minor and trace element compositions show similar trends (Fig. 363 7). In particular, most of the variations in the Mn and Ca contents are similar to the 364 crystallization trend of the pyroxenite melt at 7 kbar (Fig. 7), which can be explained by simple 365 crystallization of the pyroxenite-derived melt. As discussed in section 5.1, crustal assimilation 366 did not significantly contribute to the Jeju basalt composition. Thus, it is appropriate to consider 367 the contents of various elements in olivines mainly in terms of fractional crystallization. The 368 element (Mn, Ni, and Ca) contents and ratios (Mn/Fe, Ni/Mg, Ca/Fe and Ni/(Mg/Fe)/1000) in 369 370 olivine phenocrysts are used to distinguish melting of pyroxenite or peridotite (Rasmussen et al., 2020; Sobolev et al., 2007; Straub et al., 2008). The variation of Ni with Fo content is closer 371 to the pyroxenite trend rather than the peridotite trend (Fig. 7b), which is similar to the 372 characteristics of the Hainan and North China Craton regions (Gu et al., 2019; Li et al., 2016). 373 As noted by Straub et al. (2008), the high Ni contents in the olivines with low Fo contents may 374 375 be due to mixing between the more primitive magma and the evolved magma (Fig. 7b). The Mn and Ca contents also display similar trends to olivines crystallized in the pyroxenite-derived 376 melts (Fig. 7). 377

100*Mn/Fe ratios for all olivine phenocrysts are lower than 1.4, which is distinct from the 378 379 peridotitic values of 1.6 to 1.8 (Fig. 8) (Sobolev et al., 2007). The Mn/Fe ratio is not related to the variation in Fo content because it is known to be significantly controlled by fractional 380 crystallization (Rasmussen et al., 2020; Sobolev et al., 2007). 100Ca/Fe and 100Ni/Mg ratios 381 are lower than the pyroxenite values owing to fractional crystallization. On the other hand, the 382 corrected Ni/(Mg/Fe)/1000 values for fractional crystallization effects are similar to the 383 pyroxenite range. As a result, we believe that pyroxenite is an important source in the formation 384 of the Jeju magma, showing an agreement with Kim et al. (2019). 385

386 5.4. Implications for the Cenozoic intraplate volcanism in East Asia

According to the results of our olivine compositions and previous studies, it appears that 387 the source lithologies of the Cenozoic intraplate volcanism in East Asia, including Jeju Island, 388 are mixtures of garnet peridotite and pyroxenite (Gu et al., 2019; Kim et al., 2019; Li et al., 389 390 2016; Yang et al., 2016). The enriched pyroxenite xenoliths have been widely reported in East 391 Asia (Xu, 2002; Xu et al., 2002; Yu et al., 2010). Pyroxenite is a part that stores recycled crustal 392 components and is distributed in SCLM, showing various characteristics of trace elements and radiogenic isotopes, providing information on diverse sources of the East Asian Cenozoic 393 basalts (Xu et al., 2017). In addition, the low ³He/⁴He ratios (5.6 to 7.3 R_a) of the basalts support 394 395 that the pyroxenite melts in Jeju Island might be derived from the SCLM, unlike the Hawaiian shield basalts showing enriched mantle compositions through eclogite/pyroxenite recycled in 396 397 the lower mantle (Sobolev et al., 2005, 2007).

The reported noble gas isotope compositions (e.g., He and Ar) of the East Asian xenoliths show a wide range (Fig. 5), indicating the contribution of the crustal components to the SCLM (Correale et al., 2016; Tang et al., 2014; Yamamoto et al., 2004). The ancient East Asian SCLM, including North China Craton and Yangtze Craton, was eroded and delaminated from the Triassic to the Early Cretaceous and was replaced by the juvenile SCLM since the Late Cretaceous (Gan et al., 2018; Lee and Walker, 2006; Liu et al., 2019). Moreover, it is suggested that the SCLM has been metasomatized by the asthenospheric upwelling possibly due to the lithospheric thinning and delamination of the ancient lithosphere that occurred during the Late Jurassic-Early Cretaceous, or by subduction-related fluids/melts until the Early Cenozoic (Correale et al., 2016; Gan et al., 2018; Liu et al., 2019; Woo et al., 2014; Xu, 2002; Yamamoto et al., 2004).

Pyroxenite stored in the lithosphere has been proposed as a major source of continental 409 410 basalts occurring not only in East Asia, but also in other continents such as the East Africa Rift, West Antarctica, and Southeastern Australia (LeMasurier et al., 2016; Lu et al., 2020; Rooney, 411 2020). The stored pyroxenite may be involved in the magma formation by the interaction 412 between the SCLM containing fertile pyroxenite and the asthenospheric melts, which can 413 414 explain the mixing relationship of the radiogenic isotope compositions between the depleted and enriched mantle components (Xu et al., 2017). Additionally, helium isotope ratios of basalts 415 from East-Central China (0.6 to 7.5 R_a) and Jeju Island (3.5 to 7.3 R_a) are lower and wider than 416 the MORB range (7 to 9 R_a). This can be explained by that the metasomatized SCLM that has 417 a wide range of ³He/⁴He ratios have influenced the East Asian magma formation during the 418 Cenozoic era (Xu et al., 2014). 419

In Fig. 9, seismic tomography shows a low-velocity zone (LVZ) beneath Jeju Island (Song et al., 2020), indicating localized asthenospheric upwelling. The width of the LVZ increases with depth (Fig. 9 and Fig. S2 for model resolution), and high-velocity zones exist at the depth of SCLM (~55km) in the east, west, north of the Jeju Island, which is attributed to the relatively thick lithosphere of Sino-Korean Craton (Song et al., 2018). Variations in lithospheric thickness could lead to focused asthenospheric upwelling via edge-driven convection (Song et al., 2018;

references therein). The focused LVZ beneath the central part of Jeju Island at a depth of about 426 427 60 km represents decompressional melting and intensive interaction between the SCLM and the ascending asthenosphere (Song et al., 2018), resulting in the lithosphere weakening and 428 melting due to thermal perturbation (Rooney, 2020). Therefore, melting of pyroxenite 429 430 segregates contained in the SCLM by the localized asthenospheric upwelling could produce 431 the Jeju magma (Fig. 10). In Guo et al. (2016) and Song et al. (2020), the Cenozoic volcanic areas in East Asia are thought to be mostly related to the LVZs of the upper mantle, and the 432 heterogeneous lithosphere is observed beneath the Korean peninsula and North China Craton. 433 Thus, taking geophysical observations together with our geochemical results provides a robust 434 435 model of the interaction between the continental lithosphere and the convective upper mantle.

436

437 **6.** Conclusions

The ${}^{3}\text{He}/{}^{4}\text{He}$ ratios of the Jeju basalts and xenoliths are 3.5 to 7.3 R_a and 2.9 to 6.5 R_a, 438 439 respectively, falling within the range of the East Asian SCLM (0.2 to 10.5 R_a). Most Jeju basalt samples are unlikely affected by the crustal assimilation based on the trace element 440 compositions. Except for SB-01 showing a ³He/⁴He ratio of 3.5 Ra (SB-01) possibly due to the 441 addition of radiogenic ⁴He from crustal contamination, the ³He/⁴He ratios of the Jeju basalts 442 (5.6 to 7.3 R_a) imply that the SCLM played a major role in the formation of magma. Although 443 444 the HIMU-like plume or the subduction-related mantle wedge can be low ³He/⁴He reservoirs, they are excluded by trace element and radiogenic isotope compositions. Also, the wide range 445 of ³He/⁴He ratios of the Jeju xenoliths is attributed to the SCLM that was metasomatized by 446 lithospheric thinning/delamination of the ancient lithosphere occurred during the Late Jurassic-447 Early Cretaceous or subduction-related fluids/melts in the Late Cretaceous to the Early 448 449 Cenozoic. Our results of olivine geochemistry indicate that the source lithology is a hybrid of 450 pyroxenite and peridotite. In the tomography model (Fig. 9), the LVZ in the asthenosphere 451 beneath Jeju Island can reflect the localized asthenospheric upwelling as edge-driven 452 convection (Song et al., 2018, 2020). The asthenospheric upwelling could enhance the thermal 453 perturbation of the lithosphere, resulting in melting. Considering geochemical data and mantle 454 tomography together, we propose that the enriched Cenozoic East Asian magmas were created 455 by the interaction between the upwelling asthenospheric mantle and the metasomatized SCLM 456 that contained pyroxenite storing crustal components.

457

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Sample	DB-01	DB-02	DB-03	SB-01	KB-01
Major elem	ents (wt%) by X	RF			
SiO ₂	49.75	49.88	50.01	51.96	51.99
Al ₂ O ₃	16.00	15.68	15.72	14.65	14.86
TiO ₂	2.96	2.90	2.94	2.41	2.09
Fe ₂ O ₃	12.74	12.83	12.87	12.39	12.27
MnO	0.15	0.15	0.15	0.15	0.15
MgO	5.07	5.30	5.20	5.58	5.59
CaO	8.53	8.38	8.43	9.09	9.44
Na ₂ O	2.82	2.96	2.92	2.36	2.36
K ₂ O	1.37	1.45	1.45	0.67	0.61
P2O5	0.63	0.59	0.60	0.37	0.30
LOI	-0.18	-0.26	-0.41	0.20	0.20
Total	99.85	99.86	99.88	99.84	99.86
Trace eleme	ents (ppm) by LA	A-ICP-MS			
Sc	16.94	16.99	17.20	21.49	21.96
V	165.82	165.05	166.77	164.27	162.67
Cr	112.17	121.85	118.17	190.80	190.57
Mn	891.15	912.37	905.18	957.92	946.47
Со	140.53	120.09	144.42	182.12	162.94
Ni	78.18	74.21	81.22	84.09	76.20
Zn	105.61	112.83	106.72	110.74	109.08
Ga	18.81	18.63	18.21	18.37	18.36
Rb	20.94	25.38	24.93	12.68	13.44
Sr	499.41	487.00	491.00	318.13	298.49
Y	18.55	17.65	18.02	19.90	19.41
Zr	182.73	176.72	176.49	139.24	116.01
Nb	34.11	32.35	32.48	16.81	14.59
Мо	2.03	2.04	1.90	1.30	1.29
Cs	0.27	0.29	0.29	0.23	0.22
Ba	334.89	319.66	323.29	162.88	161.62
La	24.00	23.05	22.91	13.37	12.40
Ce	47.77	45.97	45.96	28.83	25.58
Pr	5.78	5.44	5.52	3.85	3.35
Nd	24.60	22.86	23.27	17.54	15.41
Sm	5.47	5.44	5.31	4.51	4.30
Eu	1.82	1.83	1.85	1.67	1.56
Gd	5.18	5.11	4.98	5.12	4.85
Tb	0.72	0.67	0.68	0.76	0.66

724 Table 1. Major and trace element compositions of the Jeju basalts

Dy	4.16	3.99	3.92	4.26	4.13	
Ho	0.70	0.69	0.68	0.79	0.79	
Er	1.77	1.80	1.65	1.99	1.90	
Tm	0.22	0.23	0.23	0.25	0.26	
Yb	1.48	1.35	1.35	1.56	1.49	
Lu	0.19	0.20	0.21	0.22	0.21	
Hf	4.53	4.09	4.31	3.74	3.15	
Та	2.17	2.06	2.07	1.15	1.00	
Pb	2.65	2.61	2.78	2.14	2.33	
Th	4.01	3.67	3.89	2.06	2.13	
U	0.87	0.78	0.83	0.46	0.54	

sample	SX-01								SX-02							SX-03							
mineral	Olivine		Clinopyroxene (n=3)		Orthopyroxene (n=3)		Spinel (n=3)		Olivine Clinopyroxene			Orthopy	roxene	Olivine (n=4)		Clinopyroxene (n=3)		Orthopyroxene (n=3)					
	(n=3)								(n=3)		(n=3)		(n=3)										
(wt%)	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%			
SiO2	41.27	0.3	51.86	0.5	55.09	0.2	0.04	12.5	40.32	0.1	51.72	0.2	54.84	0.1	40.25	0.2	51.93	0.2	54.93	0.5			
TiO2	0.01	94.5	0.47	3.6	0.12	5.5	0.11	3.9	0.01	42.0	0.52	0.9	0.13	15.1	0.01	110.4	0.35	0.8	0.09	19.1			
Al2O3	0.02	9.1	6.62	2.4	4.53	3.9	58.30	0.1	0.02	68.7	6.17	1.0	3.98	2.5	0.01	37.8	6.59	2.3	4.35	7.6			
FeO	9.60	1.4	2.79	5.9	6.40	13.2	11.16	1.2	11.03	0.9	3.12	3.9	6.55	3.2	10.95	5.4	2.81	1.7	6.31	0.4			
MnO	0.12	1.8	0.09	19.0	0.11	16.9	0.12	1.4	0.14	7.4	0.06	50.4	0.14	21.6	0.15	2.6	0.08	11.6	0.12	15.0			
MgO	49.31	0.1	15.63	0.8	33.23	1.2	20.50	0.2	47.98	0.1	15.44	0.8	33.44	0.3	48.01	0.8	15.51	0.5	33.56	0.4			
CaO	0.05	6.2	20.94	0.2	0.66	3.7	0.00		0.06	9.7	20.15	0.4	0.65	7.2	0.06	11.3	20.70	0.2	0.66	6.2			
K2O	0.00	77.6	0.01	76.6	0.01	95.4	0.00	141.4	0.00	141.4	0.00	141.4	0.00	141.4	0.00	100.0	0.00		0.01	88.2			
Na2O	0.01	141.4	1.32	2.8	0.07	9.5	0.01	75.0	0.02	58.7	1.61	2.8	0.09	9.2	0.02	68.8	1.47	1.2	0.08	11.3			
Cr2O3	0.00	141.4	0.74	3.9	0.35	9.6	9.08	0.9	0.01	100.9	1.26	1.4	0.47	13.6	0.01	149.6	0.96	4.1	0.40	13.1			
Total	100.40	0.1	100.47	0.3	100.55	0.3	99.32	0.1	99.59	0.1	100.06	0.3	100.30	0.2	99.47	0.1	100.40	0.1	100.51	0.0			
Mg#	90.15	0.1	90.88	0.6	90.26	1.4	76.60	0.3	88.57	0.1	89.81	0.5	90.09	0.3	88.66	0.7	90.79	0.1	90.46	0.1			
Cr#							9.46	0.7															

Table 2. Representative major element compositions of the Jeju xenoliths

Table 2. Continued

sample mineral	SX-04	X-04 SX-05 SX-06																		
	Olivine		Clinopyroxene Orthopyroxene		roxene	Olivine		Clinopyroxene		Orthopyroxene		Olivine		Clinopyroxene		Orthopyroxene		Spinel		
	(n=4)		(n=3)		(n=4)		(n=3)		(n=4)		(n=3)		(n=3)	(n=3)			(n=3)		(n=4)	
(wt%)	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%	Mean	RSD%
SiO2	40.69	0.1	52.15	0.2	54.94	0.4	40.96	0.1	52.25	0.4	54.84	0.3	40.60	0.4	51.95	0.1	54.92	0.7	0.04	13.1
TiO2	0.01	105.1	0.42	8.2	0.11	14.8	0.00	0.0	0.45	3.3	0.11	9.9	0.00	114.7	0.51	5.5	0.13	12.7	0.13	7.2
Al2O3	0.01	32.2	5.97	1.1	4.15	6.1	0.01	41.6	6.69	1.4	4.57	3.4	0.02	36.2	6.74	0.5	4.70	7.8	58.65	0.5
FeO	10.38	1.8	2.66	2.5	6.53	1.5	10.31	1.8	2.84	3.5	6.40	1.9	10.47	0.9	3.01	3.8	6.51	2.0	11.32	3.1
MnO	0.11	17.6	0.09	13.4	0.12	17.5	0.14	12.8	0.07	11.0	0.12	5.6	0.12	15.2	0.08	4.7	0.12	24.3	0.11	14.1
MgO	48.26	0.1	15.40	0.7	33.53	0.5	48.62	0.5	15.74	0.5	33.49	0.2	48.55	0.4	15.55	1.0	33.41	0.4	20.65	0.3
CaO	0.06	6.9	20.94	0.8	0.60	9.5	0.06	3.6	20.52	0.5	0.70	3.8	0.06	4.9	20.41	0.4	0.67	6.5	0.00	173.2
K2O	0.00	102.5	0.01	72.8	0.00	153.8	0.00	141.4	0.00		0.00	76.0	0.00	59.3	0.01	75.9	0.01	20.9	0.00	100.9
Na2O	0.00	173.2	1.47	2.7	0.08	17.4	0.00	141.4	1.44	2.6	0.10	11.7	0.01	97.8	1.53	1.9	0.08	5.9	0.00	173.2
Cr2O3	0.01	65.7	1.07	2.7	0.51	10.0	0.01	101.8	0.81	7.7	0.39	6.3	0.01	141.4	0.73	3.4	0.34	14.3	8.89	2.5
Total	99.52	0.2	100.18	0.2	100.57	0.2	100.12	0.0	100.80	0.2	100.72	0.1	99.83	0.2	100.52	0.2	100.89	0.3	99.79	0.3
Mg#	89.23	0.2	91.16	0.2	90.16	0.2	89.36	0.2	90.81	0.3	90.31	0.2	89.21	0.1	90.21	0.4	90.15	0.2	76.48	0.8
Cr#																			9.23	2.6

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sample	DB-01				DB-02					DB-03					
Туре	Olivine p	henocryst in	1 basalts							_					
Grain	ol-1	ol-2	ol-3	ol-4	ol-5	ol-1	ol-2	ol-3	ol-4	ol-5	ol-1	ol-2c	ol-2r	ol-3	ol-4
Major el	ement (wt%	%) by EPM	A												
SiO ₂	37.80	38.50	38.25	38.66	38.65	38.37	38.56	38.40	38.06	38.50	38.22	37.88	38.72	38.07	38.28
TiO ₂	0.04	0.02	0.00	0.02	0.02	0.02	0.03	0.01	0.01	0.02	0.03	0.04	0.03	0.01	0.04
Al ₂ O ₃	0.03	0.03	0.06	0.05	0.05	0.02	0.04	0.05	0.04	0.05	0.03	0.04	0.04	0.02	0.02
FeO	27.12	22.17	23.07	22.22	21.96	22.98	21.71	22.06	23.00	22.34	21.96	23.81	20.75	23.98	22.60
MnO	0.34	0.26	0.25	0.26	0.25	0.21	0.20	0.28	0.21	0.21	0.19	0.23	0.18	0.26	0.28
MgO	34.85	39.13	38.57	39.07	39.42	38.58	39.64	39.46	38.19	39.07	38.60	37.84	40.50	37.89	38.84
CaO	0.16	0.17	0.17	0.17	0.15	0.15	0.16	0.17	0.16	0.16	0.17	0.16	0.18	0.14	0.16
K ₂ O	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.01	0.02	0.02	0.00	0.02
Na ₂ O	0.00	0.01	0.00	0.02	0.00	0.02	0.00	0.02	0.00	0.01	0.02	0.05	0.02	0.02	0.04
Cr ₂ O ₃	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.01	0.02	0.03	0.00	0.00
Total	100.33	100.29	100.36	100.47	100.51	100.36	100.37	100.46	99.68	100.36	99.23	100.08	100.47	100.39	100.28
Mg#	69.61	75.88	74.87	75.81	76.19	74.95	76.49	76.13	74.74	75.71	75.81	73.91	77.67	73.79	75.39
Trace ele	ement (ppn	ı) by LA-IC	CP-MS												
Li	2.31	1.84	2.04	1.96		1.56	2.28	1.66	1.92	2.14	2.09	2.08		2.01	
Sc	5.38	5.04	4.91	4.83		6.96	7.39	6.90	6.25	6.82	5.40	4.95		5.69	
V	7.07	6.63	6.80	6.66		7.29	7.59	7.29	6.58	8.75	6.84	7.24		7.15	
Cr	LOD	33	30	29		25	53	36	22	22	25	31		14	
Со	207	204	206	217		228	211	215	228	201	222	228		214	
Ni	345	1308	1195	1315		1329	1677	1346	1210	1234	1292	1374		923	
Ga	0.18	0.18	0.17	0.19		0.20	0.23	0.24	0.18	0.25	0.20	0.15		0.22	

742 Table 3. Major and trace element compositions of olivine phenocrysts in the Jeju basalts

743 Table 3: Continued

sample	SB-01					KB-01					
Туре	Olivine pho	enocryst in basal	lts								
Grain	ol-1	ol-2	ol-3	ol-4	ol-5	ol-1	ol-2	ol-3c	ol-3r	ol-4	ol-5
Major elem	ent (wt%) by E	EPMA									
SiO ₂	39.10	39.13	39.03	39.13	39.06	39.21	38.98	38.65	38.52	39.07	38.91
TiO ₂	0.01	0.02	0.01	0.02	0.00	0.02	0.00	0.03	0.00	0.01	0.00
Al ₂ O ₃	0.03	0.03	0.02	0.03	0.05	0.01	0.05	0.03	0.02	0.02	0.04
FeO	19.90	18.87	18.93	18.93	18.85	18.79	19.79	20.32	21.61	19.08	19.45
MnO	0.18	0.21	0.23	0.23	0.21	0.21	0.22	0.24	0.27	0.19	0.19
MgO	40.96	41.72	41.67	41.69	41.71	41.29	41.10	40.51	39.72	41.39	41.20
CaO	0.20	0.21	0.20	0.20	0.19	0.21	0.20	0.22	0.20	0.21	0.23
K ₂ O	0.01	0.00	0.00	0.01	0.00	0.00	0.01	0.00	0.00	0.00	0.00
Na ₂ O	0.00	0.00	0.02	0.00	0.00	0.00	0.02	0.01	0.00	0.00	0.04
Cr ₂ O ₃	0.03	0.04	0.05	0.05	0.04	0.05	0.03	0.05	0.03	0.02	0.04
Total	100.42	100.23	100.17	100.29	100.11	99.80	100.39	100.05	100.38	99.99	100.09
Mg#	78.58	79.76	79.69	79.69	79.77	79.66	78.73	78.04	76.61	79.45	79.06
Trace elem	ent (ppm) by L	A-ICP-MS									
Li	1.99	1.71	1.73	1.91		1.95	1.86	1.91		1.60	
Sc	5.84	5.66	5.58	5.83		5.43	5.68	6.02		5.46	
V	7.33	5.95	5.95	6.77		6.41	7.04	7.21		6.54	
Cr	238	236	226	237		176	188	216		179	
Co	183	185	187	188		187	194	190		190	
Ni	1972	2008	1966	1977		1967	1980	1628		2029	
Ga	0.19	0.21	0.15	0.20		0.12	0.19	0.18		0.20	

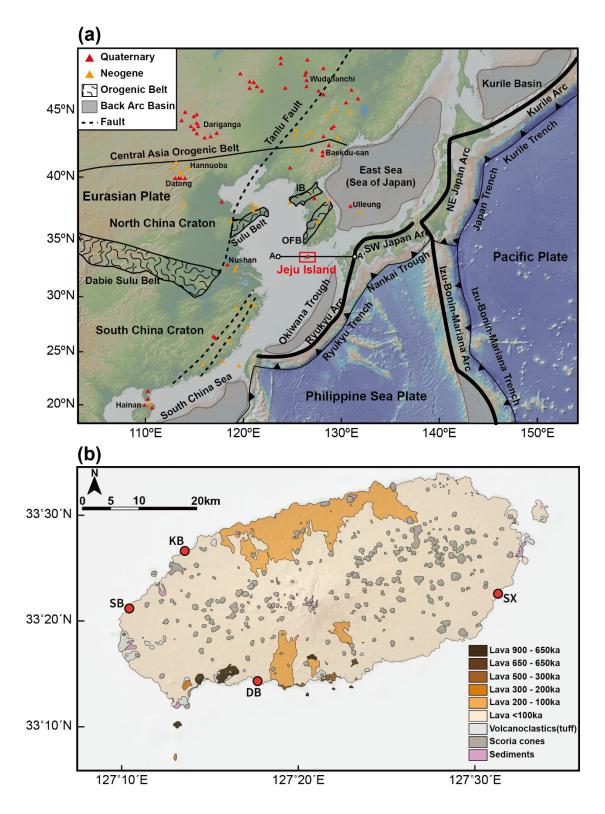
Sample	SX-01			SX-02			SX-03			SX-04			SX-05				SX-06		
Grain	cpx-4	cpx-5	cpx-6	cpx-1	cpx-2	cpx-3	cpx-1	cpx-2	cpx-3	cpx-1	cpx-2	cpx-3	cpx-1	cpx-2	cpx-3	cpx-4	cpx-1	cpx-2	cpx-3
Major ele	ement (wi	t%) by E	EPMA																
SiO ₂	51.7	52.2	51.7	51.6	51.8	51.8	52.0	51.8	52.0	52.0	52.3	52.2	52.5	52.1	52.4	52.0	52.0	52.0	51.9
TiO ₂	0.5	0.5	0.4	0.5	0.5	0.5	0.4	0.4	0.4	0.4	0.4	0.5	0.5	0.5	0.4	0.4	0.5	0.6	0.5
Al ₂ O ₃	6.4	6.6	6.8	6.2	6.1	6.2	6.4	6.8	6.6	6.1	5.9	5.9	6.8	6.6	6.6	6.7	6.8	6.7	6.8
FeO	2.6	2.8	3.0	3.1	3.0	3.3	2.8	2.8	2.7	2.6	2.8	2.7	2.9	3.0	2.7	2.8	3.1	3.1	2.9
MnO	0.1	0.1	0.1	0.1	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
MgO	15.8	15.7	15.5	15.4	15.6	15.3	15.6	15.5	15.5	15.3	15.5	15.4	15.8	15.8	15.7	15.6	15.8	15.4	15.5
CaO	20.9	20.9	21.0	20.0	20.2	20.2	20.6	20.7	20.7	20.9	20.8	21.2	20.6	20.3	20.6	20.6	20.4	20.5	20.3
K ₂ O	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Na ₂ O	1.34	1.35	1.27	1.55	1.66	1.61	1.45	1.49	1.48	1.49	1.50	1.41	1.50	1.42	1.40	1.43	1.49	1.53	1.56
Cr ₂ O ₃	0.7	0.8	0.8	1.3	1.2	1.3	0.9	1.0	1.0	1.1	1.0	1.1	0.7	0.8	0.9	0.9	0.7	0.8	0.7
Total	100.0	100.8	100.6	99.7	100.1	100.4	100.3	100.5	100.4	99.9	100.2	100.4	101.2	100.7	100.8	100.6	100.8	100.6	100.2
Mg#	91.5	91.0	90.1	89.8	90.3	89.3	90.7	90.7	91.0	91.3	91.0	91.2	90.8	90.5	91.2	90.7	90.1	89.9	90.7
Trace ele	ment (pp	m) by L	A-ICP-N	1 S															
Li	1.61	1.68	1.66	1.58	1.34	2.26	1.51	1.67	1.65	1.75	1.55	1.99	1.44	1.43	1.52	1.71	1.41	1.10	1.38
Sc	55.9	59.5	60.9	74.3	72.1	71.0	57.5	55.3	57.1	58.5	56.7	65.4	63.8	66.1	63.6	62.8	64.4	69.6	63.3
Ti	3023	3239	3159	3439	3076	3464	2210	2166	2325	2746	2641	2933	2892	2849	2903	2900	3341	3465	3348
V	272	287	284	279	263	271	261	256	264	253	248	259	272	275	270	271	279	289	279
Cr	4774	5093	5050	8520	6949	8838	6455	6754	6827	7951	8294	7389	6095	6086	5881	5953	4954	5069	5394
Mn	633	636	684	687	684	727	687	651	664	634	646	580	650	674	660	629	657	667	661
Co	20.8	21.1	21.6	22.0	22.2	22.8	21.4	20.7	20.7	20.0	20.2	20.1	21.6	22.3	21.9	21.2	22.0	22.0	21.0
Ni	339	348	349	367	362	363	335	332	334	321	329	341	339	341	330	326	340	340	326
Zn	13.2	10.5	18.1	13.3	12.4	14.9	10.5	10.6	10.9	9.8	12.5	10.3	11.2	15.8	10.7	10.2	10.4	12.6	10.8
Ga	3.52	3.60	3.64	4.24	3.85	4.64	4.01	3.46	3.63	3.68	4.14	3.52	3.88	3.73	3.62	3.68	4.19	4.20	4.26

744 Table 2. Major and trace element compositions of clinopyroxenes in the Jeju xenoliths

Rb	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD
Sr	6.6	6.3	13.2	99.7	75.0	118.6	40.0	19.4	18.3	32.1	31.8	31.3	15.4	18.2	31.1	15.6	25.2	51.9	40.1
Y	17.5	18.1	17.8	16.6	15.2	16.4	15.5	15.2	15.7	12.2	12.0	12.1	17.7	17.7	18.0	18.2	18.9	18.4	18.5
Zr	7.3	7.7	7.7	84.2	67.8	83.4	10.7	10.3	10.1	12.1	11.5	11.6	13.8	14.1	19.5	14.0	9.8	11.2	9.7
Nb	0.17	0.03	0.46	0.31	0.15	0.32	0.52	0.19	0.16	0.02	0.01	0.01	0.09	0.18	0.28	0.12	0.18	0.38	0.26
Мо	LOD	LOD	LOD	0.02	0.01	0.05	LOD												
Cs	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	LOD	0.01	LOD								
Ba	LOD	LOD	LOD	0.03	0.01	LOD	LOD	LOD	0.07	0.07	0.08	LOD	LOD	LOD	LOD	0.00	0.06	LOD	LOD
La	0.20	0.03	1.52	6.25	3.89	7.94	3.57	0.75	0.46	0.20	0.19	0.18	0.18	0.67	2.38	0.31	0.54	2.90	1.88
Ce	0.51	0.32	2.60	14.41	6.38	18.02	4.73	1.17	0.99	1.04	1.02	0.96	0.84	1.53	4.20	0.86	1.67	5.62	3.86
Pr	0.15	0.19	0.33	1.78	0.75	2.11	0.49	0.26	0.23	0.23	0.23	0.23	0.29	0.28	0.52	0.29	0.37	0.72	0.51
Nd	1.71	1.77	2.17	7.55	4.19	8.73	2.30	1.99	1.95	1.98	1.88	2.04	2.23	2.29	2.91	2.26	2.73	3.68	3.08
Sm	1.19	1.22	1.14	2.21	1.45	2.31	1.00	1.06	1.05	1.16	1.00	1.12	1.15	1.31	1.34	1.34	1.19	1.60	1.32
Eu	0.47	0.52	0.48	0.80	0.63	0.87	0.52	0.45	0.45	0.44	0.46	0.47	0.57	0.53	0.51	0.54	0.62	0.64	0.59
Gd	2.23	1.96	2.07	2.42	1.96	2.75	1.80	1.81	1.82	1.71	1.79	1.88	2.27	2.12	2.24	2.18	2.18	2.43	2.17
Tb	0.42	0.42	0.40	0.43	0.40	0.46	0.36	0.33	0.35	0.31	0.32	0.33	0.44	0.41	0.45	0.43	0.44	0.43	0.48
Dy	2.93	3.27	3.03	2.96	2.50	3.05	2.65	2.56	2.67	2.24	2.07	2.02	3.17	3.09	3.19	3.20	3.27	3.49	3.49
Но	0.67	0.71	0.66	0.67	0.56	0.65	0.59	0.63	0.58	0.47	0.43	0.47	0.76	0.70	0.68	0.70	0.70	0.73	0.69
Er	1.93	1.95	2.01	1.82	1.71	1.81	1.79	1.76	1.77	1.42	1.40	1.32	2.02	1.97	2.18	2.23	2.20	2.17	2.15
Tm	0.28	0.28	0.27	0.24	0.24	0.24	0.25	0.23	0.26	0.19	0.20	0.20	0.30	0.27	0.30	0.28	0.29	0.30	0.31
Yb	1.83	2.03	1.92	1.57	1.59	1.62	1.56	1.64	1.72	1.24	1.16	1.22	1.91	1.87	1.84	1.94	2.18	2.06	2.09
Lu	0.24	0.27	0.27	0.23	0.22	0.21	0.23	0.24	0.21	0.21	0.20	0.19	0.28	0.25	0.28	0.30	0.28	0.29	0.28
Hf	0.52	0.53	0.55	2.52	2.21	2.52	0.53	0.40	0.44	0.51	0.53	0.55	0.65	0.65	0.64	0.65	0.56	0.55	0.56
Та	LOD	LOD	0.01	0.02	LOD	0.01	0.02	LOD	LOD	0.00	LOD	LOD	LOD	LOD	0.04	LOD	0.02	0.02	0.01
Pb	0.06	0.03	0.06	0.31	0.35	0.32	0.31	0.23	0.25	LOD	LOD	LOD	0.05	0.05	0.11	0.07	0.09	0.13	0.17
Th	0.11	0.03	0.28	0.62	0.63	0.66	1.07	0.70	0.62	LOD	LOD	LOD	0.18	0.23	0.59	0.29	0.12	0.40	0.29
U	0.03	0.01	0.06	0.13	0.15	0.15	0.22	0.17	0.14	0.00	LOD	LOD	0.05	0.08	0.14	0.09	0.04	0.09	0.07

Sample	Location	Rock Type	Mineral	Mass (g)	Age (Ma)	R/Ra	d	d%	4He ccSTP/g	Error	3He ccSTP/g	Error
DB-01	Daepodong	Basalt	olivine	0.384	< 0.1	7.3	0.4	6%	1.2E-08	6.3E-12	1.2E-13	7.4E-15
DB-02	Daepodong	Basalt	olivine	0.327	< 0.1	6.8	0.3	5%	1.9E-08	1.0E-11	1.8E-13	1.1E-14
DB-03	Daepodong	Basalt	olivine	0.407	< 0.1	5.9	0.6	9%	1.2E-08	1.4E-11	4.0E-13	2.5E-14
SB-01	Sinchang-ri	Basalt	olivine	0.47	< 0.1	3.5	0.1	4%	8.2E-08	4.4E-11	4.0E-13	2.5E-14
KB-01	Kwideok-ri	Basalt	olivine	0.442	< 0.1	6.9	0.3	4%	2.1E-08	1.1E-11	2.0E-13	1.3E-14
DB-02	Daepodong	Basalt	clinopyroxene	0.655	< 0.1	5.6	0.2	4%	4.7E-08	2.5E-11	3.7E-13	2.3E-14
DB-03	Daepodong	Basalt	clinopyroxene	0.617	< 0.1	6.4	0.2	3%	4.1E-08	2.2E-11	3.7E-13	2.3E-14
SX-01	Sinsan-ri	xenolith	olivine	0.552	< 0.1	5.8	0.7	12%	1.5E-09	8.2E-12	1.3E-14	7.8E-16
SX-02	Sinsan-ri	xenolith	olivine	0.45	< 0.1	6.5	0.3	5%	1.3E-08	7.2E-12	1.2E-13	7.5E-15
SX-03	Sinsan-ri	xenolith	olivine	0.479	< 0.1	6.5	0.1	2%	1.8E-07	9.6E-11	1.6E-12	1.0E-13
SX-04	Sinsan-ri	xenolith	olivine	0.395	< 0.1	6.2	0.7	11%	6.1E-09	3.3E-12	5.3E-14	1.3E-15
SX-05	Sinsan-ri	xenolith	olivine	0.444	< 0.1	4.5	0.5	10%	5.3E-09	2.8E-12	3.4E-14	2.1E-15
SX-06	Sinsan-ri	xenolith	olivine	0.55	< 0.1	2.9	0.2	7%	1.5E-08	8.0E-12	5.6E-14	3.5E-15

745 Table 3. Helium isotope compositions of the basalt and xenolith samples in Jeju Island



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Fig. 1. (a) Geological map of East Asia with volcanic fields indicated by Choi et al. (2006), Chen et al. (2007), and Guo et al. (2014). Abbreviations in the diagram are IB (Imjingang Belt) and OFB (Okcheon Fold Belt). A-A' is the location of the vertical cross-section of mantle tomography (Fig. 9). (b) Geological map of Jeju Island (modified after Koh et al., 2013), and the sampling sites are shown. Abbreviations in the diagram are KB (Kwideok-Ri Basalt), SB (Sinchang-Ri Basalt), DB (Daepodong Basalt), and SX (Sinsan-Ri xenolith)



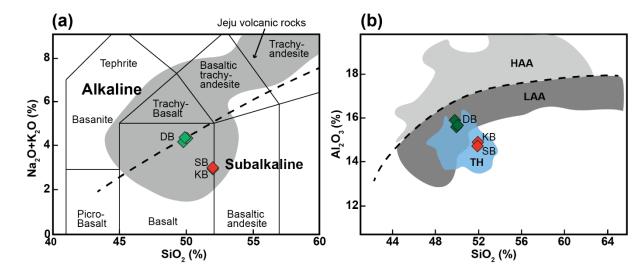




Fig. 2. Classification of the Jeju island basalt samples. (a) Total alkali (Na₂O + K_2 O) versus SiO₂ diagram, and (b) Al₂O₃ vs. SiO₂ diagram. The data sources indicating the areas in (a) and

(b) are from Baek et al. (2014) and references therein.

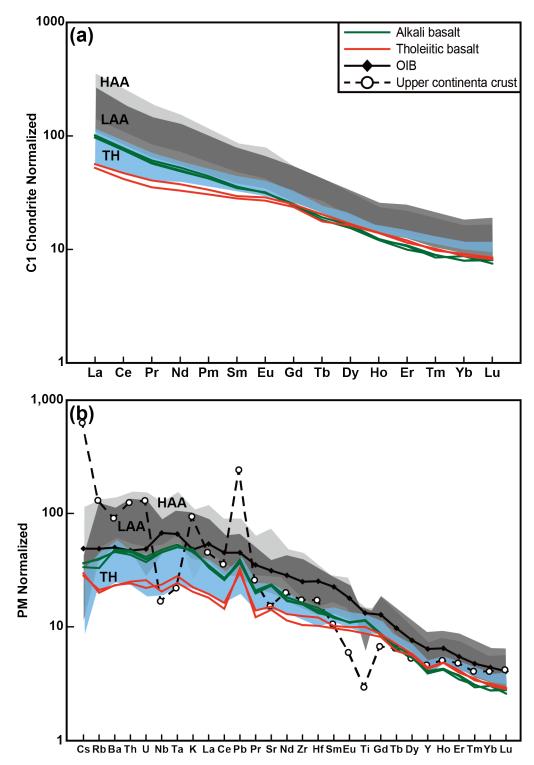




Fig. 3. Rare earth element (REE) and trace element patterns of the Jeju basalt samples (a) REE patterns normalized to the C1 chondrite values (Sun and McDonough, 1989). (b) Trace element patterns normalized to the primitive mantle (Sun and McDonough, 1989). The shaded areas indicate high-Al alkali (HAA), low-Al alkali (LAA), and tholeiite (TH) which are previously reported basalt samples (SiO₂ < 52%, Kim et al., 2019). Typical ocean island basalt (OIB) and upper continental crust compositions are from Sun and McDonough (1989) and Rudnick and Gao (2003).

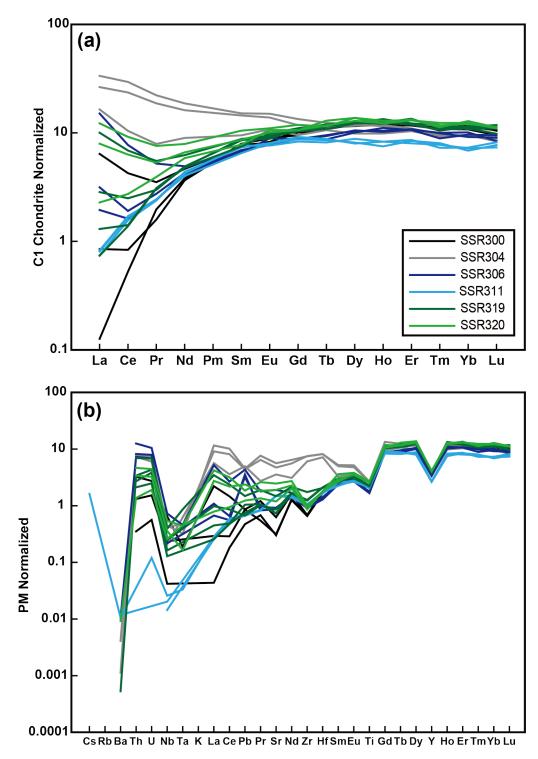




Fig. 4. Rare-earth-element (REE) and trace element patterns of clinopyroxenes in the Jeju
xenolith samples (a) REE patterns normalized to the C1 chondrite values (Sun and McDonough,
1989). Various LREE enrichments with relatively flat MREE and HREE patterns are shown.
(b) Trace element patterns normalized to the primitive mantle (Sun and McDonough, 1989).
Nb-Ta depletion and U-Th enrichment are observed.

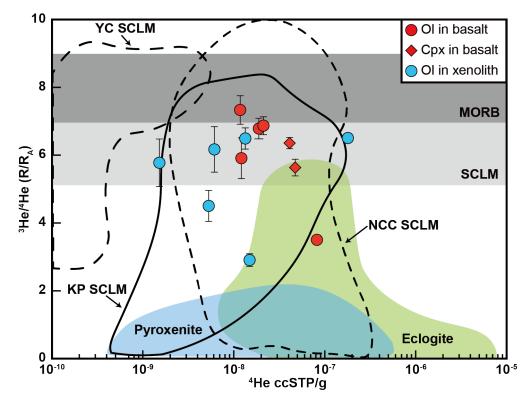




Fig. 5. ³He/⁴He ratios (R/Ra) vs. ⁴He contents of the Jeju Island basalts and xenoliths.
Reference data are from Graham (2002) for mid-ocean ridge basalt (MORB); Gautheron and
Moreira (2002) for Subcontinental lithospheric mantle (SCLM); Kim et al. (2005) for Korean
Peninsula (KP) SCLM; Correale et al. (2016) for Yangtze Craton (YC) SCLM; Tang et al.
(2014) for North China Craton (NCC) SCLM; Day et al. (2015) for pyroxenite and eclogite,
respectively.

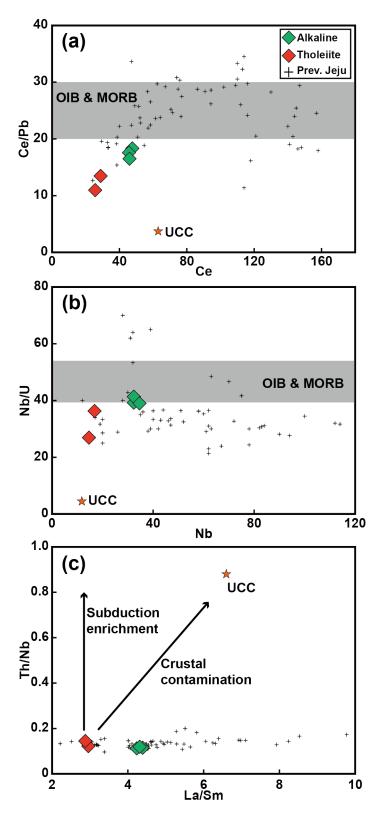


Fig. 6. Crustal assimilation indicators with (a) Ce/Pb vs. Ce (ppm), (b) Nb/U vs. Nb (ppm),
and (c) Th/Nb vs. La/Sm. The cross dots indicate the previously reported Jeju basalt data (Kim
et al., 2019) The orange star represents the upper continental crust (UCC) value (Rudnick and
Gao, 2003). The shaded area means the OIB and MORB ranges (Ce/Pb: 20-30; Nb/U: 37-57;
Hofmann et al., 1986)

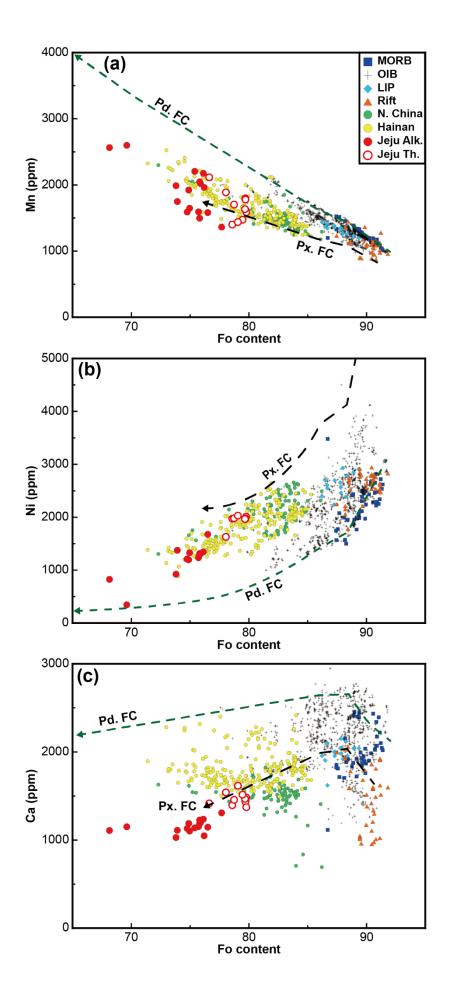


Fig. 7. Mn, Ni, and Ca vs. Fo contents in olivines. The black and green dotted lines represent
the modeled fractional crystallization (FC) path from peridotitic (Pd.) and pyroxenitic (Px.)
melts at 7kbar. Other various dots indicate mid-ocean ridge basalts (MORB; Sobolev et al.,
2007), ocean island basalts (OIB; Rasmussen et al., 2020; Reinhard et al., 2016; Sobolev et al.,
2007), large igneous province (LIP; Sobolev et al., 2007), continental rift (Foley et al., 2011;
Sobolev et al., 2007), and Cenozoic basalts in East Asia (North China; Li et al., 2016, Hainan;
Gu et al., 2019), respectively.

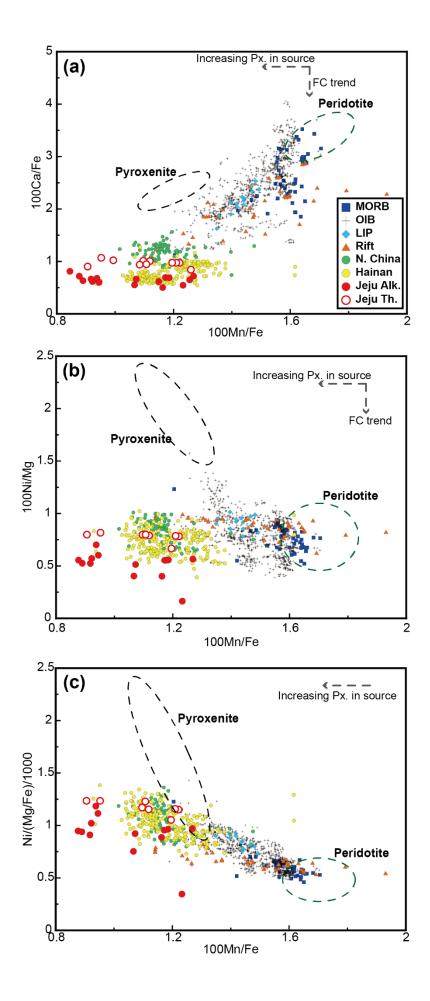
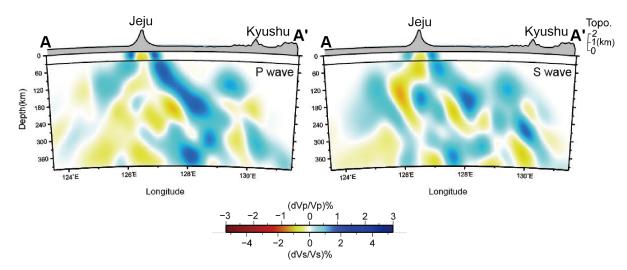


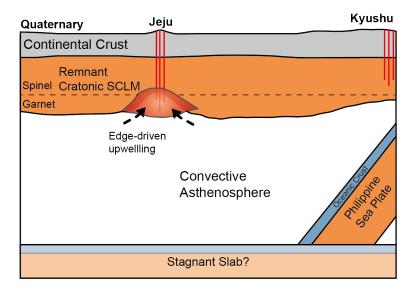
Fig. 8. 100Ca/Fe, 100Ni/Mg, and Ni/(Mg/Fe)/1000 vs. 100*Mn/Fe in olivine. The 100*Mn/Fe ratios of the Jeju olivines are similar to the olivines derived from the pyroxenitic melt. The black and green dotted circles mean the equilibrated olivine compositions from the peridotite and pyroxenite derived melts (Sobolev et al., 2007). Other various dots are from references in Fig. 7. (a) The 100Ca/Fe ratios of olivined from Jeju Island are relatively low, reflecting fractional crystallization (FC). (b) The 100Ni/Mg ratios of the Jeju Island olivines also show lower ratios (<1), caused by fractional crystallization (c) the Ni/(Mg/Fe)/1000 and 100*Mn/Fe ratios corrected for the fractional crystallization effect, and the area of the olivines from Jeju Island is close to the pyroxenite range.



829 Fig. 9. Vertical cross-sections of the P wave and S wave tomography along the A-A' transect in Fig 1a. Focused low-velocity zone is observed beneath Jeju Island, indicating the 830 asthenospheric upwelling interacting with the SCLM. The relatively thick and cold cratonic 831 832 lithosphere is located around Jeju Island. Detailed information of the mantle tomography is described in Song et al. (2020). The velocity perturbation scale is shown at the bottom. 833

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- Fig. 10. Schematic illustration for the magma formation of Jeju Island. The remnant cratonic 838
- SCLM is located in the east, west, and north of Jeju Island, a possible reservoir of the enriched 839
- components. Localized asthenosphere upwelling (e.g., edge-driven convection) could melt the 840
- pyroxenitic segregates in the lowermost SCLM. Interaction between the pyroxenite segregates 841 contained in the SCLM and the depleted asthenosphere might generate the magma of Jeju
- 842
- Island. 843