

1 Geochemical constraints on the structure of the Earth's deep 2 mantle and the origin of the LLSVPs

3
4 Matthew Gleeson¹, Caroline Soderman², Simon Matthews³, Sanne Cottaar², Sally Gibson²

5 ¹School of Earth and Environmental Sciences, Cardiff University, Main Building, Park Place, CF10 3AT,
6 UK.

7 ²Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge, CB2 3EQ, UK.

8 ³Institute of Earth Sciences, University of Iceland, Sturlugata 7, 102 Reykjavik, Iceland.

9 KEY POINTS

- 10 1. Compositional variability in erupted basalts and olivine crystals reveals the distribution of
11 recycled material in the Galápagos plume.
- 12 2. The eastern Pacific LLSVP does not represent piles of subducted oceanic crust.
- 13 3. Geochemical and geophysical data indicate the presence of recycled crustal material near
14 the eastern margin of the Pacific LLSVP.

15 ABSTRACT

16 Geophysical analysis of the Earth's lower mantle has revealed the presence of two superstructures
17 characterized by low shear wave velocities on the core-mantle boundary. These Large Low Shear
18 Velocity Provinces (LLSVPs) play a crucial role in the dynamics of the lower mantle and act as the
19 source region for deep-seated mantle plumes. However, their origin, and the characteristics of the
20 surrounding deep mantle, remain enigmatic. Mantle plumes located above the margins of the
21 LLSVPs display evidence for the presence of this deep-seated, thermally and/or chemically
22 heterogeneous mantle material ascending into the melting region. As a result, analysis of the spatial
23 geochemical heterogeneity in OIBs provides constraints on the structure of the Earth's lower mantle
24 and the origin of the LLSVPs. In this study, we focus on the Galápagos Archipelago in the eastern
25 Pacific, where bilateral asymmetry in the radiogenic isotopic composition of erupted basalts has

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26 been linked to the presence of LLSVP material in the underlying plume. We show, using spatial
27 variations in the major element contents of high-MgO basalts, that the isotopically enriched south-
28 western region of the Galápagos mantle – assigned to melting of LLSVP material – displays no
29 evidence for lithological heterogeneity in the mantle source. As such, it is unlikely that the Pacific
30 LLSVP represents a pile of subducted oceanic crust. Clear evidence for a lithologically heterogeneous
31 mantle source is, however, found in the north-central Galápagos, indicating that a recycled crustal
32 component is present near the eastern margin of the Pacific LLSVP, consistent with seismic
33 observations.

34 **1 INTRODUCTION**

35 Volcanic archipelagos such as the Galápagos, Hawai'i and Samoa represent the surface expression of
36 deep-seated mantle plumes that likely originate near the core-mantle boundary (Morgan, 1971;
37 Wilson, 1973). Such regions of ocean island volcanism provide an important window into the
38 composition and structure of the Earth's lower mantle, which plays a critical role in the
39 geodynamical and thermochemical evolution of the planet. For example, unradiogenic He (and Ne)
40 isotope systematics (that is, $^3\text{He}/^4\text{He} > 8 R/R_A$; where R/R_A indicates the measured ratio of $^3\text{He}/^4\text{He}$
41 relative to the $^3\text{He}/^4\text{He}$ ratio of air) in ocean island basalts (OIBs) indicates that an undegassed,
42 primordial component is likely preserved in the lower mantle over >4 billion years despite vigorous
43 mantle convection (Farley et al., 1992; Jackson et al., 2010; Kurz and Geist, 1999; Stuart et al., 2003).
44 In addition, the radiogenic isotope variability of OIBs provides evidence for recycling of lithospheric
45 material into the deep mantle, resulting in the presence of several isotopically distinct mantle
46 reservoirs (e.g. EM-1, EM-2, HIMU; Chauvel et al., 1992; Hofmann, 1997; Stracke et al., 2005; White
47 and Hofmann, 1982; Willbold and Stracke, 2006). However, to fully understand the long-term
48 evolution of the Earth's lower mantle, it is necessary to relate the chemical heterogeneities
49 observed in OIBs to the detailed picture of the Earth's lower mantle that has been developed
50 through various geophysical techniques.

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51 Seismic tomography images of the Earth's lower mantle reveal that it is far from homogeneous.
52 Most notably, seismic models highlight the presence of two 'superstructures' on the core mantle
53 boundary that are characterized by lower shear wave velocities than the surrounding mantle
54 (Cottaar and Lekic, 2016; Dziewonski and Woodhouse, 1987; Garnero et al., 2016; Ritsema et al.,
55 2011) and are argued to have higher densities than surroundings (Lau et al., 2017; Moulik and
56 Ekström, 2016) at least at their base (Davaille and Romanowicz, 2020; Richards et al., 2021). These
57 superstructures, known as Large Low Shear Velocity Provinces (LLSVPs), are located beneath Africa
58 and the Pacific and play a critical role in mantle dynamics, the rise of mantle plumes (Heyn et al.,
59 2020), and the configuration of True Polar Wander events (Steinberger et al., 2017). Despite their
60 importance, however, the origin of the LLSVPs remain enigmatic, with their presence having
61 previously been assigned to piles of subducted oceanic crust (Brandenburg and van Keken, 2007;
62 Niu, 2018) or primordial material that has undergone differentiation early in Earth's history (such as
63 magma ocean cumulates; Deschamps et al., 2012; Labrosse et al., 2007; Peters et al., 2018).

64 Global seismic tomography models reveal that many mantle plumes are rooted within, or at the
65 margins of the LLSVPs, indicating that these regions represent 'plume nurseries' (Fig. 1; Doubrovine
66 et al., 2016; French and Romanowicz, 2015; Jackson et al., 2018). Furthermore, several mantle
67 plumes worldwide display bilateral asymmetry; where isotopically enriched signals are assigned to
68 melting of upwelling LLSVP material, and isotopically depleted compositions are related to melting
69 of the surrounding peridotitic mantle (Harpp et al., 2014b; Harpp and Weis, 2020; Hoernle et al.,
70 2015; Huang et al., 2011; Weis et al., 2011; Zhou et al., 2020). Therefore, a critical analysis of the
71 compositional variability displayed by one such mantle plume, considering all available isotopic,
72 trace element and major element data, has the potential to reveal new insights into the origin of the
73 LLSVPs, with implications for the evolution of the solid Earth over billion-year timescales. Specifically,
74 correlation of the isotopic and lithological heterogeneity (the latter inferred by major elements in
75 basalts and minor elements in olivine) observed in LLSVP-rooted plumes will allow the distribution of

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76 recycled components in the lower mantle to be investigated and thus determine whether LLSVPs
77 truly represent piles of subducted oceanic crust.

78 Here, we choose to focus on the Galápagos Archipelago, which is located above the eastern margin
79 of the Pacific LLSVP and displays bilateral asymmetry in the trace element and radiogenic isotope
80 composition of the erupted basalts (Harpp et al., 2014b; Harpp and Weis, 2020). We present new
81 olivine data from the central Galápagos that, alongside published olivine and whole-rock data from
82 across the archipelago, allows us to determine the spatial variability in the lithological structure of
83 the underlying mantle plume. In doing so, we identify where recycled crustal components are
84 directly involved in the genesis of Galápagos magmas (Herzberg, 2011; Rosenthal et al., 2015;
85 Sobolev et al., 2005; Yaxley and Green, 1998). Finally, we discuss the implications of our findings for
86 the structure of the lower mantle beneath the Pacific and the origin of the Pacific LLSVP.

87 2 GEOLOGICAL BACKGROUND

88 The Galápagos Archipelago is located ~1000 km off the western coast of Ecuador in the eastern
89 equatorial Pacific and represents one of the most volcanically active regions in the world. Volcanism
90 in the Galápagos is driven by melting in an upwelling mantle plume that has a mantle potential
91 temperature (T_p) >30 – 150 °C above that of the ambient mantle (Gibson et al., 2015). Seismic
92 tomography provides evidence that the Galápagos mantle plume originates below the mantle
93 transition zone, likely at the core-mantle boundary near the eastern margin of the Pacific LLSVP
94 (Hooft et al., 2003; Nolet et al., 2019).

95 The Galápagos Archipelago lies on the eastward-moving Nazca tectonic plate (plate velocity of ~50
96 km/Myr; Argus et al., 2011), about 150 – 200 km south of the Galápagos Spreading Centre, a plume-
97 influenced segment of the global mid-ocean ridge system. Seismic tomography indicates that the
98 Galápagos mantle plume is centered beneath the south-western Archipelago at ~200 km depth, but
99 is deflected to the north-east at ~100 – 80 km depth owing to the presence of the nearby spreading

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100 centre (Villagómez et al., 2014). The Galápagos Spreading Centre itself clearly shows the influence of
101 the nearby mantle plume in the geochemical composition of the erupted basalts and the crustal
102 thickness of the ridge (Canales et al., 2002; Christie et al., 2005; Cushman et al., 2004; Detrick et al.,
103 2002; Gibson and Richards, 2018; Gleeson et al., 2020; Gleeson and Gibson (2021); Ingle et al., 2010;
104 Schilling et al., 1982; Sinton et al., 2003).

105 Owing to the west-to-east motion of the Nazca tectonic plate, Galápagos volcanoes follow a general
106 west-to-east age progression, with the youngest volcanic activity in the west and the oldest lavas
107 observed on the eastern islands of San Cristobal and Espanola (Bailey, 1976; Geist et al., 1986;
108 Naumann and Geist, 2000). However, volcanic activity in the central and eastern Galápagos persists
109 into the Holocene, with historical eruptions observed on the central island of Santiago (Global
110 Volcanism Program, 2013) and lavas as young as ~9 ka found on the eastern-most island of San
111 Cristobal (Mahr et al., 2016). Nevertheless, some of the basalts from the eastern-most Galápagos
112 may have erupted when the islands were located several 10s of km west of their current location.
113 Yet, owing to the size of the Galápagos Archipelago (>400 km from west to east), and the dominance
114 of more recent volcanic activity on the central and western Galápagos volcanoes, the composition of
115 basalts erupted across the Galápagos Archipelago can be used to evaluate the spatial heterogeneity
116 of the underlying mantle plume.

117 2.1 ISOTOPIC HETEROGENEITY OF THE GALÁPAGOS ARCHIPELAGO

118 Since the 1980s, analysis of radiogenic isotope ratios has dominated understanding of source
119 heterogeneity in the Galápagos mantle plume (Geist et al., 1988; Harpp and Weis, 2020; Harpp and
120 White, 2001; Hoernle et al., 2000; White et al., 1993). Specifically, spatial variability in the Pb, Nd, Hf
121 and Sr isotope composition of the Galápagos basalts has traditionally been interpreted to represent
122 the contribution of melts from at least 4 isotopically distinct end-members (Blichert-Toft and White,
123 2001; Harpp and White, 2001). These end-members, known as PLUME, FLO, WD (Wolf-Darwin), and

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124 DGM (Depleted Galápagos Mantle), are most strongly expressed in spatially defined regions of the
125 Galápagos Archipelago and associated ridges (Harpp and White, 2001; Hoernle et al., 2000).

126 The PLUME end-member is dominant in basalts from the westernmost island of Fernandina, and is
127 characterized by moderately radiogenic Sr and Pb isotope signatures, similar to the common plume
128 component referred to as 'FOZO' or 'C' (Hanan and Graham, 1996; Harpp and White, 2001). Notably,
129 the Fernandina basalts have unradiogenic He and Ne isotope signatures (e.g. $^3\text{He}/^4\text{He}$ up to 30 R/R_A;
130 Kurz and Geist, 1999), which indicates the presence of an undegassed primordial reservoir in the
131 Galápagos plume.

132 To the south of Fernandina, basalts trend towards more radiogenic Sr and Pb isotope signatures,
133 which reach a maximum on the southern island of Floreana (Harpp et al., 2014a; Harpp and White,
134 2001; Kurz and Geist, 1999). The extreme radiogenic isotope composition of Floreana (that is,
135 elevated $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{187}\text{Os}/^{188}\text{Os}$ ratios) is used to define the FLO mantle end-member, which
136 displays similar $^{206}\text{Pb}/^{204}\text{Pb}$ signatures to the global HIMU mantle (Gibson et al., 2016; Harpp et al.,
137 2014a). As a result, the FLO mantle is hypothesized to represent recycled Archean oceanic crust
138 (Gibson et al., 2016; Harpp et al., 2014a), although the absence of evidence for a pyroxene-rich
139 component in the mantle source of the Floreana lavas casts doubt on the recycled crustal origin
140 (Gleeson et al., 2021; Vidito et al., 2013).

141 Basalts in the eastern Galápagos (i.e. Genovesa, San Cristobal and eastern Santiago) are dominated
142 by melts of the DGM (Gibson et al., 2012; Harpp and White, 2001). However, whether the DGM
143 component is entrained upper mantle, or derives from the lower mantle, is an ongoing area of
144 debate (Blichert-Toft and White, 2001; Gibson et al., 2012; Harpp and Weis, 2020; Hoernle et al.,
145 2000). Finally, the WD isotopic end-member is restricted to a small number of seamounts and minor
146 islands in the northernmost Galápagos and appears to have little to no influence on the composition
147 of basalts elsewhere in the Galápagos (Harpp and White, 2001). The origin of this localized
148 component is unknown and is not addressed in this study.

149 The complex relationship between the different mantle end-members identified in the Galápagos
150 mantle plume has made correlation of these signatures to the structure of the underlying deep
151 mantle very challenging. It is possible, however, to simplify the spatial heterogeneity observed in the
152 radiogenic isotope composition of the Galápagos basalts and instead describe their variability in
153 terms of overall isotopic enrichment (Harpp and Weis, 2020). The most enriched isotopic signatures
154 (here used to describe radiogenic Pb and Sr and unradiogenic Nd isotope compositions) are
155 observed in the western and southern Galápagos, whereas isotopically depleted compositions
156 dominate in the north-eastern Galápagos (apart from Pinta, whose anomalously enriched
157 composition is likely related to plume-ridge interactions in the Galápagos; Fig. 2a).

158 Critically, this isotopic variability mirrors the structure of the deep mantle at the base of the
159 Galápagos mantle plume, with LLSVP material to the south-west and 'normal' lower mantle to the
160 north-east (Fig. 1 & 2; Cottaar and Lekic, 2016; Garnero et al., 2016; Ritsema et al., 2011). As a result,
161 the enriched isotopic signatures of the south-western Galápagos have been assigned to melting of
162 LLSVP material ascending as part of the Galápagos mantle plume, whereas the depleted nature of
163 the north-eastern Galápagos basalts is hypothesized to result from melting of the surrounding
164 peridotitic mantle (Harpp et al., 2014b; Harpp and Weis, 2020).

165 2.2 LITHOLOGICAL PROPERTIES OF THE GALÁPAGOS MANTLE PLUME

166 Our current understanding of the structure and composition of the Galápagos mantle plume is
167 complicated by the unclear relationship between lithological and radiogenic isotopic heterogeneity
168 (Gleeson et al., 2020; Gleeson and Gibson, 2019; Vidito et al., 2013). Lithological heterogeneity, that
169 is, the presence of fusible, pyroxene-rich components in the underlying mantle, is believed to result
170 from recycled crustal components in the mantle and their incorporation into upwelling mantle
171 plumes (Gibson, 2002; Hauri, 1996; Lambart, 2017; Lambart et al., 2013; Mallik and Dasgupta, 2012;
172 Rosenthal et al., 2015; Shorttle et al., 2014; Sobolev et al., 2007, 2005; Yaxley and Green, 1998). As
173 such, identification of lithological heterogeneity in the mantle source region of basaltic lavas

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174 provides evidence for the contribution of recycled crustal components to mantle plumes. Therefore,
175 linking signatures of lithological heterogeneity to the isotopic heterogeneity of the Galápagos mantle
176 plume might help to identify whether the LLSVPs truly represent piles of subducted oceanic crust.

177 Lithological heterogeneity in the mantle is commonly tracked through the minor element
178 composition of olivine phenocrysts (Gurenko et al., 2013, 2009; Herzberg, 2011; Sobolev et al., 2007,
179 2005). Specifically, high Ni but low Mn and Ca contents in primitive olivine phenocrysts are thought
180 to be characteristic of a contribution from pyroxenite-derived melts, owing to the large differences
181 in the bulk partition coefficient of these elements during melting of an olivine-rich (peridotite) and a
182 pyroxene-rich (pyroxenite) lithology.

183 The composition of olivine crystals from the Galápagos Archipelago has previously been used to
184 evaluate the lithological structure of the underlying plume, with initial interpretations suggesting
185 that lithologically distinct components are present in both isotopically enriched and isotopically
186 depleted regions of the Galápagos (Vidito et al., 2013). However, the presence of a pyroxenitic
187 component in the mantle source region of the eastern Galápagos basalts contradicts their
188 isotopically depleted nature and trace element systematics (e.g. Gibson et al., 2012). As a result, the
189 anomalously high Ni and low Ca contents of the eastern Galápagos olivines were recently revisited,
190 with numerical models of fractional crystallisation, magma recharge and diffusive re-equilibration
191 demonstrating that these 'pyroxenitic' olivine compositions could be generated through crustal
192 processing of basaltic lavas (Gleeson and Gibson, 2019).

193 Nevertheless, it remains possible that pyroxenitic source components contribute to basalts from
194 other regions of the Galápagos. In fact, analysis of Fe-isotope ratios in basaltic lavas from plume-
195 influenced regions of the Galápagos Spreading Centre (GSC) revealed that an enriched pyroxenitic
196 component is present in the Galápagos mantle plume (Gleeson et al., 2020). However, it remains
197 uncertain whether the enriched pyroxenite in the mantle source region of the GSC basalts is related
198 to the isotopically enriched signatures assigned to melting of the LLSVP material contained within

199 the Galápagos mantle plume. To address this, we collect new olivine data from the central
200 Galápagos, which is used alongside published olivine and whole-rock data from across the Galápagos
201 Archipelago to evaluate the spatial variability in the lithological properties of the underlying mantle.

202 **3 METHODS**

203 We present high-precision analyses of the major and minor element composition of olivine crystals
204 in geochemically enriched basalts from western Santiago in the central Galápagos. Previous work on
205 the major element, trace element and isotopic composition of the Santiago basalts has led to the
206 classification of four chemical groups: low-K tholeiites, high ϵNd transitional basalts, low ϵNd
207 transitional basalts, and mildly-alkaline basalts (Gibson et al., 2012). In general, the low-K tholeiites
208 are found on the eastern side of the island, with enriched isotopic signatures found in mildly-alkaline
209 basalts further west (Gibson et al., 2012). Our new data from well-characterised mildly alkaline and
210 low ϵNd transitional basalts fills a crucial gap in the olivine data from the Galápagos, as olivine
211 compositions from western and eastern Galápagos basalts (including eastern Santiago) have
212 previously been characterized (Gleeson and Gibson, 2019; Vidito et al., 2013).

213 All data were collected using a Cameca SX100 electron microprobe in the Department of Earth
214 Sciences, University of Cambridge. Analysis was carried out using a defocused (5 μm) spot and a 15
215 kV accelerating voltage. Analysis of Si, Fe, and Mg was carried out using a 20 nA beam current. To
216 increase the analytical precision on low concentration elements, a 100 nA beam current was used
217 for analysis of Ni, Mn, Ca and Al. Mineral and metal standards were used to calibrate at the start of
218 the analytical session and precision and accuracy were tracked through repeat analysis of a San
219 Carlos Olivine secondary standard. Recovery for all elements is between 99 and 103%. The 2-sigma
220 analytical precision of analysis is $\sim 3\%$ for Fe, better than 1.5% for Mg and Si, $\sim 4\%$ for Ni, $\sim 15\%$ for Ca,
221 and $\sim 8\%$ for Mn (Supplementary Data).

222 4 RESULTS

223 Our new analyses reveal that olivines in mildly alkaline basalts from Isla Santiago (such as sample
224 08DSG33) are relatively evolved, with forsterite (Fo) contents ranging from $\sim 70 - 84$, and contain
225 moderately high Ni contents ($\sim 800 - 2900$ ppm; Fig. 3). Olivines in mildly alkaline basalts also contain
226 relatively low Mn contents ($\sim 1500 - 2500$ ppm), and correspondingly high Fe/Mn ratios (72.8 ± 5.2 ;
227 Fig. 3). In contrast, the Fo contents of olivines from transitional basalt 07DSG61 are slightly more
228 primitive than those observed in the mildly alkaline basalts (Fo $\sim 81-85$). Furthermore, the Ni content
229 and Fe/Mn ratio of olivines in sample 07DSG61 are lower than those observed in the mildly alkaline
230 basalts ($\sim 1200 - 2300$ ppm and 71.1 ± 5.1 , respectively; Fig. 3).

231 5 DISCUSSION

232 5.1 OLIVINE MINOR ELEMENT SYSTEMATICS

233 Olivine minor elements provide a powerful method for investigating the lithological properties of the
234 mantle (Gurenko et al., 2009; Herzberg et al., 2014; Sobolev et al., 2007), as long as the influence of
235 crustal processes and the conditions of mantle melting are considered (Gleeson and Gibson, 2019;
236 Matzen et al., 2013, 2017b). Olivine data from the eastern Galápagos (that is, San Cristobal,
237 Genovesa and Espanola) indicate that the mantle source regions of these basalts are dominated by
238 peridotite (Vidito et al., 2013; Fig. 3a,b). However, interpretation of olivine data from elsewhere in
239 the Galápagos is not so simple (Gleeson and Gibson, 2019).

240 Taken at face value, the Ca content (and to a lesser extent the Ni content) of olivines in basalts from
241 Floreana in the southern Galápagos, which originates from melting of LLSVP material with highly
242 radiogenic Pb isotope signatures (Harpp and Weis, 2020), suggests that there is a notable
243 contribution from melts from a pyroxenitic source component (Gleeson et al., 2021; Harpp et al.,
244 2014a; Vidito et al., 2013). However, the low Ca contents and moderately high Ni contents observed
245 in some of the Floreana olivines can instead be explained by chemical modification in a cumulate

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246 mush (Gleeson et al., 2021). As such, there is no significant evidence in the olivine minor element
247 systematics of the Floreana basalts to indicate that there is a substantial contribution of melts from
248 a pyroxenitic source component (Fig. 3).

249 Olivine data from the western Galápagos (that is, the islands of Isabela, Roca Redonda, and
250 Fernandina) display a range of compositions. Notably, it is clear that the Ni, Fe/Mn and Ca contents
251 of olivines from Fernandina and Cerro Azul (on the southern margin of Isabela), which fall into the
252 isotopically enriched south-western region of the archipelago, are consistent with the presence of a
253 peridotite source (especially once the influence of crustal processes are taken into account; Gleeson
254 and Gibson, 2019; Vidito et al., 2013). However, evidence for the contribution of melts from a
255 pyroxenitic source component is found in the Fe/Mn ratio of olivines from Roca Redonda and
256 Volcans Ecuador, Wolf and Darwin on Northern Isabela (Fe/Mn >70; Fig. 3 & 4; Vidito et al., 2013).
257 Notably, the Ni contents of olivine from Roca Redonda and Volcan Ecuador are also higher than the
258 olivine compositions predicted by the magma mixing models of Gleeson and Gibson (2019),
259 supporting the interpretation that a pyroxenitic source contributes to the olivine composition of
260 these Northern Isabela and Roca Redonda basalts. High Fe/Mn ratios (>75) are also found in olivines
261 from Sierra Negra, but the evolved nature of the Sierra Negra basalts (often <5 wt% MgO) and
262 olivines mean that we cannot rule out these signatures originating through crustal processing (cf.
263 Trela et al., 2015).

264 In the central Galápagos, olivine data from mildly-alkaline basalts E-76 and 08GSD33 on western
265 Santiago, reveal Ni and Fe/Mn contents that are too high to be explained by melting of a peridotitic
266 source, even if the influence of crustal processes are considered (Gleeson and Gibson, 2019).

267 Although care must be used when comparing the composition of magmatic olivines to the
268 composition of their host basalt, as olivine crystals might not be directly related to their carrier melt
269 (Wieser et al., 2019), it is notable that these basalts display the most enriched trace element and
270 isotopic signatures of any basalt found on Santiago (Gibson et al., 2012). Conversely, olivine data

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271 from isotopically depleted basalts on eastern Santiago are consistent with a dominant contribution
272 of melts from a peridotitic source lithology (Gibson et al., 2016; Gleeson and Gibson, 2019).
273 Transitional basalts from Santiago (such as 07DSG61) display intermediate olivine compositions at
274 moderately high Ni and Fe/Mn contents, confirming that these basalts represent a mixture of
275 pyroxenite and peridotite derived melts.

276 Olivine compositions from Santa Cruz in the central Galápagos fall into two groups, although the
277 compositions measured within each sample are typically relatively uniform (Vidito et al., 2013). One
278 group displays Fe/Mn contents between 60 and 72 (that is, consistent with a peridotitic source;
279 Herzberg, 2011), whereas the other contains Fe/Mn ratios >70. The high Fe/Mn group also contains
280 high Ni contents that cannot be easily explained by crustal processing of peridotite-derived basaltic
281 magmas (Gleeson and Gibson, 2019). As such, the variability in the olivine composition of basalts
282 from Santa Cruz likely results from changes in the proportion of pyroxenite-derived melt. The olivine
283 compositional characteristics of Santa Cruz does not appear to define a geographic geochemical
284 trend, unlike on Santiago (Gibson et al., 2012), with shorter length-scale variability in the
285 composition of erupted basalts dominating.

286 Overall, the new and compiled olivine data indicates that a pyroxenitic, recycled component is
287 present in the Galápagos mantle plume. However, this pyroxenitic component is not dominant in
288 basalts of the isotopically enriched south-western region of the Galápagos. Instead, it is most
289 prevalent in basalts from the north-central Galápagos, that is, mildly-enriched basalts from northern
290 Isabela, Roca Redonda, western Santiago, and Santa Cruz (Fig. 4a). Importantly, our results indicate
291 that geochemically enriched peridotite and pyroxenite components exist in the Galápagos mantle
292 plume; as a result, there is no simple relationship between host-basalt enrichment and olivine Ni and
293 Mn contents across the archipelago. Nevertheless, the proposed contribution of pyroxenitic melts to
294 the north-central Galápagos basalts is supported by the isotopic similarity of these basalts (with

295 regards to Sr, Nd and Pb) to the pyroxenitic end-member previously identified in the mantle source
296 region of the GSC basalts (Gleeson et al., 2020; Gleeson and Gibson, 2021; Fig. 5).

297 5.2 MAJOR ELEMENT SYSTEMATICS OF THE GALÁPAGOS BASALTS

298 Alongside olivine minor element compositions and Fe-isotope ratios of basaltic lavas, information
299 about the lithological properties of the mantle source is contained in the major element systematics
300 of high-MgO basalts (i.e., those that have not undergone significant fractionation of clinopyroxene
301 or plagioclase; Dasgupta et al., 2010; Hauri, 1996; Lambart et al., 2016, 2013; Shorttle et al., 2014;
302 Shorttle and Maclennan, 2011). Specifically, melts of a pyroxenitic source lithology often have lower
303 CaO contents, and higher FeO_t contents (where FeO_t represents the total Fe content of the melt
304 expressed as FeO), than melts of a peridotite (Herzberg, 2011; Hirose and Kushiro, 1993; Lambart et
305 al., 2016, 2012).

306 To evaluate the spatial variability in the major element composition of primary mantle melts from
307 the Galápagos, we compiled whole-rock major element data from across the archipelago and filtered
308 the resulting dataset to exclude any samples with MgO contents <8 wt% (i.e., those that display
309 substantial evidence for clinopyroxene and plagioclase fractionation; see Supplementary
310 Information). Basalts in the filtered database contain a relatively narrow range of Mg# compositions
311 (from ~55 to ~75 where $\text{Mg\#} = \text{Mg}/(\text{Mg}+\text{Fe}_t)$ molar). However, variations in other major element
312 parameters are observed.

313 Notably, the FeO_t content of high-MgO basalts from the western, southern and eastern Galápagos
314 are consistently <11 wt%, and typically less than 10.5 wt%. Melting experiments at pressures
315 between 1.5 and 3 GPa on the KLB-1 peridotite (a commonly used experimental analogue for the
316 upper mantle) provide mean FeO_t contents of 8.88 wt%, and maximum FeO_t contents of 10.05 wt%,
317 broadly consistent with the compositions observed in the western, southern and eastern Galápagos
318 (Fig. 2b; Fig. 4; Hirose and Kushiro, 1993; Takahashi et al., 1993). Furthermore, melting calculations
319 performed in the KNCFMASCr system using THERMOCALC v3.4.7 reveal that melts produced by

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320 the KLB-1 peridotite between 1.5 and 3 GPa, and at melt fractions less than 20% (see Supplementary
321 Information), contain approximately $9.77^{+1.09}_{-1.99}$ wt% FeO_t (Holland et al., 2018; Holland and Powell,
322 2011; Powell et al., 1998), almost perfectly overlapping with the FeO_t content of the western,
323 southern and eastern Galápagos basalts ($9.74^{+0.94}_{-0.98}$ wt% FeO_t; Fig. 4). Melts produced in hydrous
324 melting regimes at significantly higher pressures (>3 GPa) are volumetrically minor, and therefore
325 unlikely to influence the major element systematics of the Galápagos basalts and not considered
326 here. In addition, the CaO contents of primitive basaltic magmas in the southern, eastern and
327 western Galápagos are also consistent with the CaO content predicted from melting of a peridotite
328 source (Hirose and Kushiro, 1993; Takahashi et al., 1993). As such, basalts in these regions of the
329 Galápagos are likely dominated by melts of a peridotite source, with only a minor-to-moderate
330 contribution from melts of a pyroxenitic component (<0 - 30 %), broadly consistent with the
331 compositions observed in olivine crystals from these basalts (see above).

332 Some basalts in the north-central Galápagos, that is, Santa Cruz and western Santiago, the northern
333 margin of Isabela (Volcan Ecuador) and Roca Redonda, display FeO_t contents >11 wt%. In fact, on
334 western Santiago and Roca Redonda, the whole-rock FeO_t contents extend to >12.5 wt%, well
335 outside the range of FeO_t contents that can result from melting of a peridotite source lithology (Fig.
336 4; Gibson et al., 2000; Hirose and Kushiro, 1993). Even if basalts from Roca Redonda with MgO
337 contents >15 wt%, which may have assimilated high-FeO_t olivine, are excluded the whole-rock FeO_t
338 contents of the remaining basalts are notably higher than those of the south-western and eastern
339 Galápagos (~11 – 12 wt%; Supplementary Information). In addition, it is unlikely that a substantial
340 increase in the depth of melting beneath the north-central Galápagos (relative to the western
341 Galápagos) could explain this shift to higher FeO_t contents, as the lithosphere is thickest and the
342 mantle potential temperature is greatest beneath the south-western region of the archipelago
343 where low FeO_t values are observed (Gibson and Geist, 2010).

344 The high FeO_t contents of the north-central Galápagos are consistent with the FeO_t concentrations
345 measured in experimental melts of pyroxenitic lithologies such as MIX-1g and M5-40 (Hirschmann et
346 al., 2003; Kogiso et al., 2003; Lambart et al., 2013). Melting simulations in THERMOCALC v3.4.7 again
347 support the experimental data, and demonstrate that melts of MIX-1g at pressures above 1.5 GPa,
348 and melt fractions below 60%, contain $11.54^{+2.22}_{-1.88}$ wt% FeO_t, indicating a strong contribution of
349 melts from a pyroxenitic source to the basalts of the north-central Galápagos ($10.95^{+1.44}_{-1.13}$ wt% FeO_t;
350 Fig. 4). Notably, the regions of the northern and central Galápagos that display basalt FeO_t contents
351 >11 wt% all plot very close to the region where olivine minor element chemistry indicates the
352 presence of a recycled pyroxenitic source component (Fig. 3 & 4).

353 In addition, a more detailed look at the central Galápagos (that is, Santiago, Santa Cruz, Santa Fe and
354 Rabida), demonstrates that the major element variability observed across the Galápagos Archipelago
355 is related to the degree of trace element and isotopic enrichment (Fig. 6 & 7). For example, high-
356 MgO basalts with high FeO_t and low CaO contents, which are inconsistent with the composition of
357 melts produced by a peridotite source, are typically characterized by moderately radiogenic Pb and
358 Sr isotope ratios and enriched trace element systematics (e.g. Nb/Y > 0.4; Gibson et al., 2012).
359 Furthermore, comparison of experimental melt compositions to the observed major element
360 systematics of the central Galápagos basalts has previously shown that the compositional variations
361 across Isla Santiago are consistent with the presence of both pyroxenite and peridotite components
362 in the mantle source, an observation that is consistent with the THERMOCALC v3.4.7 calculations
363 presented here (Supplementary Information; Gleeson et al., 2020).

364 The high FeO_t contents of the central Galápagos and Northern Isabela/Roca Redonda basalts are also
365 expressed in their anomalously high Fe/Mn ratios (>65; Fig. 5b). However, unlike the FeO_t and CaO
366 contents of basalts from across the Galápagos Archipelago, there is a slight difference in the Fe/Mn
367 ratio of basalts from the western Galápagos (Fernandina and Southern Isabela; ~60) and the eastern
368 Galápagos (Espanola and San Cristobal; ~55). Variations in the Fe/Mn ratio of basaltic magmas have

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369 traditionally been assigned to the presence of a fusible, pyroxenitic component (Herzberg, 2011) or a
370 core component (Humayun et al., 2004) in the mantle source. Yet, the slight variation in the Fe/Mn
371 ratio of the western and eastern Galápagos basalts could be explained by differences in the depth of
372 melting (Matzen et al., 2017a), consistent with the greater lithospheric thickness in the western
373 Galápagos compared to the eastern Galápagos (Gibson et al., 2012; Gibson and Geist, 2010).
374 Nevertheless, the higher Fe/Mn ratio of the north-central Galápagos basalts requires a substantial
375 contribution from melts of a pyroxenitic source, as these signatures cannot be generated by
376 variations in the melting processes alone (Fig. 5b).

377 Overall, variations in the major element systematics of primitive basaltic lavas from across the
378 Galápagos Archipelago indicate that a pyroxenitic component is present in the underlying mantle
379 plume and is most strongly expressed in the composition of basalts from the north-central
380 Galápagos basalts (northern Isabela, Roca Redonda, western Santiago and Santa Cruz). This
381 hypothesis is supported by the similarity between the radiogenic isotope composition of the most
382 enriched basalts from the north-central Galápagos (on Roca Redonda and western Santiago;
383 Standish et al., 1998; Gibson et al. 2012) and the proposed isotopic composition of the pyroxenitic
384 end-member in the mantle source region of the GSC basalts (Fig. 5; Gleeson et al., 2020; Gleeson
385 and Gibson, 2021). As a result, the major element systematics of the primitive Galápagos basalts
386 indicate that an isotopically enriched pyroxenitic source component contributes to both basalts from
387 the north-central Galápagos, consistent with our interpretations based on the available olivine data
388 shown above, and the GSC.

389 The spread of isotopic compositions observed in basalts from both the south-western and eastern
390 Galápagos indicates that a small contribution of Sr, Pb and Nd-rich melts from this pyroxenitic
391 component (i.e., <<25% pyroxenitic melt) might influence the radiogenic isotope ratios of basalts
392 erupted across the entire archipelago (Fig. 8). There is, however, no evidence in the major element
393 systematics of basalts from Fernandina, Southern Isabela and Floreana to indicate that there is a

394 large contribution of melts from this pyroxenitic component to the major element composition of
395 basalts in the isotopically enriched south-western region of the archipelago (Fig. 4). Additionally, the
396 major element systematics of the eastern Galápagos basalts provide no evidence to support
397 previous interpretations of a depleted pyroxenitic component is dominant in the eastern Galápagos
398 (Vidito et al., 2013).

399 5.3 VARIATIONS IN SOURCE PYROXENITE PROPORTIONS

400 The major element systematics of high-MgO basalts, and the minor element contents of their olivine
401 cargo, reveal clear variations in the contribution of pyroxenitic melts to basalts erupted across the
402 Galápagos Archipelago. However, it is important to consider whether the prevalence of pyroxenitic
403 melt signatures in the north-central region of the Galápagos represents true spatial heterogeneity in
404 the distribution of pyroxenitic components in the underlying Galápagos mantle plume, or if these
405 signatures can instead be caused by variations in mantle potential temperature, melt extents, and
406 melt extraction processes. Addressing this question is critical to understanding the distribution of
407 lithologically distinct, recycled components in the Earth's lower mantle.

408 As pyroxenitic source components are typically more fusible than 'normal' mantle peridotite, the
409 pyroxenite solidus will be crossed at higher pressures than the peridotite solidus during adiabatic
410 decompression melting (Gibson et al., 2000; Kogiso et al., 2003; Lambart et al., 2016, 2013; Sobolev
411 et al., 2007; Yaxley and Green, 1998). Therefore, melts of a pyroxenitic source dominate at low total
412 melt fractions during melting of a two- or three-component mantle, with peridotite-derived melts
413 becoming more dominant at shallower pressures (Lambart et al., 2016). As a result, the proportion
414 of pyroxenite-derived melt contributing to the composition of basaltic lavas is influenced by
415 variations in the mantle potential temperature and lithospheric thickness, as well as the proportion
416 of pyroxenite in the source.

417 To address whether variations in melting parameters could explain the spatial variability in the
418 contribution of pyroxenitic melts to the Galápagos Archipelago, we calculate the proportion of

419 pyroxenite-derived melt that results from melting of a two-component mantle under various
420 conditions. Calculations were performed using the pymelt Python module (Matthews et al., 2020),
421 and recent empirical parameterisations for the melting of a lherzolitic peridotite and silica-
422 undersaturated pyroxenite (KLB-1 and KG1, respectively; Matthews et al., 2021). We ran the
423 calculations over a range of mantle potential temperatures ($T_p = 1400 - 1460$ °C) and lithospheric
424 thicknesses (46 – 60 km; $\sim 1.5 - 1.85$ GPa) appropriate to the Galápagos Archipelago (Gibson et al.,
425 2015, 2012; Gibson and Geist, 2010; Herzberg and Asimow, 2008; Vidito et al., 2013), and consider
426 how these conditions may influence the relative contribution of melts from a mixed peridotite-
427 pyroxenite mantle source. For example, the pymelt models indicate that a mantle containing $\sim 10\%$
428 pyroxenite, melting at a T_p of 1400 °C under 60 km thick lithosphere produces magmas with a
429 pyroxenite melt proportion of $\sim 70\%$. Melting of the same mantle with a T_p of 1460°C and a
430 lithospheric thickness of ~ 46 km gives a pyroxenitic melt proportion of only $\sim 30\%$.

431 To determine whether variations in the conditions of mantle melting across the Galápagos
432 Archipelago can cause the observed differences in the relative contribution of pyroxenitic melts to
433 the Galápagos basalts, we compare the results of our melting calculations to first-order estimates of
434 the proportion of pyroxenitic melt that contributes to each region of the Galápagos. These estimates
435 are derived from the mean FeO_t content of the Galápagos basalts, an assumed peridotite melt FeO_t
436 content of 8.8 – 9.77 wt% and a pyroxenite melt FeO_t content of 12.8 wt% (representing the mean
437 FeO_t content of the experimental and thermodynamic KLB-1 melts and the highest FeO_t content
438 observed in any of the Galápagos basalts, respectively). Results indicate that basalts from the
439 western and eastern Galápagos contain, on average, a 0 – 24% contribution of melts from a
440 pyroxenitic source (mean FeO_t of 9.75 wt%), whereas basalts from western Santiago contain 57 –
441 80% pyroxenitic melts (FeO_t contents between 11.5 and 12 wt%). This variation is similar in
442 magnitude to the maximum difference in the proportion of pyroxenitic melt that can be caused by
443 variations in the melting conditions of a homogeneous mantle source beneath the Galápagos
444 Archipelago ($\sim 30 - 70\%$; Fig. 9).

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445 Regions of the Galápagos Archipelago that are dominated by melts of peridotitic source lithologies,
446 however, do not only occur in regions where the lithosphere is thinnest or where the mantle
447 potential temperature is highest. For example, Isla Fernandina and Volcan Cerro Azul, on southern
448 Isabela, display no evidence for the contribution of pyroxenitic melts, despite the fact that seismic
449 data indicates the lithosphere is thickest in this region of the archipelago (Fig. 2c; Gibson and Geist,
450 2010; Rychert et al., 2014). As such, it is unlikely that variations in the contribution of pyroxenitic
451 melts to basalts erupted across the Galápagos Archipelago results purely from variations in the
452 melting conditions. Instead, we suggest that the difference in the proportion of pyroxenitic melt
453 contributing to basalts from the south-western, north-central and north-eastern regions of the
454 Galápagos Archipelago must result from variations in the proportion of pyroxenite present in the
455 mantle source. We note that the first-order estimates for the proportion of pyroxenitic melt
456 contributing to each region of the Galápagos presented above can be recreated when the mantle
457 source region of the western, southern and eastern Galápagos contains <5 % pyroxenite, but the
458 mantle source region of the north-central Galápagos basalts contains >20% pyroxenite (Fig. 9).

459 In addition, the presence of at least three distinct components in the Galápagos mantle plume are
460 required by the radiogenic isotope variability of the Galápagos basalts. Specifically, we note that the
461 $^{87}\text{Sr}/^{86}\text{Sr}$ isotope signature of the north-central Galápagos basalts (pyroxenite source) are lower than
462 that observed in basalts from the south-western Galápagos (peridotite source; Fig. 5). This indicates
463 that basalts in the south-western archipelago cannot be a mixture of melts derived from an enriched
464 pyroxenite and a depleted peridotite, otherwise they would display a less enriched radiogenic
465 isotope composition than the basalts from the north-central Galápagos (where the pyroxenitic
466 source component is most strongly expressed). As a result, the presence of one or more isotopically
467 enriched south-western peridotite components are required (Fig. 8), alongside a pyroxenite source
468 component that is focused beneath the north-central Galápagos and an isotopically depleted north-
469 eastern peridotite (Fig. 10).

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470 As indicated above, we cannot exclude the possibility that a small fraction of pyroxenitic material
471 (i.e., <5%) exists in the mantle source of all Galápagos basalts and contributes to their isotopic
472 compositions (Fig. 8). However, the analysis presented here clearly shows that this pyroxenitic
473 component is present in much higher proportions in the mantle source region of the north-central
474 Galápagos basalts, separating the isotopically enriched domain of the south-western Galápagos from
475 the isotopically depleted eastern Galápagos.

476 6 IMPLICATIONS FOR THE STRUCTURE OF THE DEEP MANTLE

477 Owing to the location of the Galápagos Archipelago above the eastern margin of the Pacific LLSVP,
478 and the asymmetric structure of the Galápagos mantle plume (with regards to isotopic composition),
479 it is hypothesised that the plume stem is rooted at the eastern boundary of the Pacific LLSVP (Harpp
480 and Weis, 2020; Jackson et al., 2018; Ritsema et al., 2011; Fig. 1). Therefore, placing constraints on
481 the spatial distribution of lithologically distinct components in the Galápagos mantle plume, as
482 achieved above, can be used to identify the contribution of recycled material to the deep mantle.

483 Seismic tomography reveals that the structure and slope of the LLSVP boundaries are not uniform
484 (Cottaar and Lekic, 2016). For example, the boundary of the eastern Pacific LLSVP near the base of
485 the Galápagos mantle plume is relatively steep ($>60^\circ$), displaying a sharp transition between the
486 LLSVP and seismically faster material to the east (Fig. 1; Frost and Rost, 2014). Conversely, the
487 northern boundary of the Pacific LLSVP, which may represent the source region of the Hawaiian
488 mantle plume (Weis et al., 2011), is shallower ($\sim 25\text{-}35^\circ$; Frost and Rost, 2014). These variations in the
489 slope of the LLSVP margins have been hypothesized to result from changes in mantle dynamics and,
490 specifically, the presence of recycled slabs in the Earth's mantle (Frost and Rost, 2014). Steeper
491 margins, such as that observed at the eastern margin of the Pacific LLSVP, are attributed to the
492 presence of subducted slabs, which push into the LLSVP and cause an increased thermal and
493 compositional gradient. Additionally, a compilation of seismic tomography models indicate that
494 there is considerable evidence to suggest that recycled slabs are present in the Earth's lowermost

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495 mantle beneath the eastern margin of the Pacific Ocean (Cottaar and Lekic, 2016; Shephard et al.,
496 2017).

497 The distribution of lithologically distinct components in the Galápagos mantle plume allows us to
498 compare the geochemical signatures of plume-related lavas to these seismic interpretations.

499 Geochemical evidence for pyroxenitic source components is most strongly observed in the

500 composition of basalts from the volcanoes of northern Isabela (Ecuador and Wolf), Roca Redonda,

501 western Santiago and Santa Cruz. These locations lie along the border between the isotopically

502 enriched south-western domain and the isotopically depleted north-eastern domain of the

503 Galápagos mantle plume identified by Harpp and Weis (2020). As such, our observations suggest

504 that the Galápagos mantle plume contains a pyroxenitic, recycled component and that this

505 component is most prevalent within the boundary zone between the enriched LLSVP material to the

506 south-west and depleted peridotitic mantle to the north-east. It is unclear how the shallow level

507 (<100-200 km depth) deflection of the Galápagos mantle plume to the north-east influences the

508 projection of spatial variations in basalt chemistry to features in the deep mantle, but, if we assume

509 that the spatial distribution of lower mantle material is maintained during plume ascent (Dannberg

510 and Gassmöller, 2018; Farnetani et al., 2018), our observations suggest that subducted crustal

511 material is present near the margin of the Pacific LLSVP and is entrained into the core of the

512 upwelling Galápagos plume (Fig. 11). This distribution of recycled crustal material in the Pacific lower

513 mantle can explain the localized expression of lithological heterogeneity at the surface, and is

514 consistent with the presence of a seismically fast body near the eastern margin of the Pacific LLSVP

515 (Frost and Rost, 2014).

516 Critically, there is no evidence in either the major element systematics of the Galápagos basalts, or

517 the minor element contents of their olivine cargo, to indicate that the isotopically enriched LLSVP

518 material melting beneath the south-western portion of the Galápagos Archipelago is pyroxenitic

519 (Vidito et al., 2013). Consequently, there is little to no data in the Galápagos to support the popular

520 hypothesis that the LLSVPs represent piles of subducted oceanic crust (Niu, 2018). Notably, our
521 interpretation that the Pacific LLSVP cannot be dominated by piles of subducted oceanic crust is
522 consistent with recent ab initio calculations of the density and seismic velocities of subducted crustal
523 material, which indicate that such bodies should be visible as high velocity regions in the lower
524 mantle (as opposed to the low seismic velocities of the LLSVPs; Wang et al., 2020). Instead, the
525 eastern Pacific LLSVP likely contains a contribution from a primordial, or undegassed mantle
526 component, consistent with the elevated $^3\text{He}/^4\text{He}$ signature of the Fernandina basalts. In addition,
527 the isotopic data from the south-western Galápagos and the Loa trend of Hawaii clearly
528 demonstrate that the LLSVP material is heterogeneous at a range of different length scales, and it is
529 therefore unlikely that one single process is responsible for the formation of these deep mantle
530 superstructures (Harpp and Weis, 2020; Jackson et al., 2018).

531 Additionally, our interpretation that recycled crustal components are external to the LLSVPs is
532 consistent with dynamical models of mantle circulation, which demonstrate that only ~10% of
533 subducted oceanic crust can be stored in the deep mantle superstructures (Li et al., 2014).
534 Therefore, the distribution of pyroxenitic components in the Galápagos mantle plume demonstrates
535 that recycled crustal components are present along the eastern margin of the Pacific LLSVP, and
536 potentially contribute to the steep, sharp transition at the LLSVP margin (Frost and Rost, 2014).

537 7 CONCLUSIONS

538 The Galápagos Archipelago offers an opportunity to investigate the structure of the Earth's lower
539 mantle and the origin of the LLSVPs through the geochemical analysis of erupted basalts. In this
540 study we have used the major element composition of high-MgO basalts, and the minor element
541 contents of their olivine cargo, to map out the distribution of lithologically distinct components in
542 the Galápagos mantle plume. By comparing our results with the spatial heterogeneity in the
543 radiogenic isotope composition of basalts from across the Archipelago we have constrained the

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544 distribution of recycled crustal components in the upwelling mantle plume and, by extension, at the
545 core mantle boundary.

546 Our results indicate that the south-western and north-eastern regions of the Galápagos mantle
547 plume, corresponding to upwelling LLSVP material and depleted mantle respectively, are dominated
548 by peridotite, with little evidence for lithological heterogeneity. In the central and northern
549 Galápagos, however, high FeO_t contents in primitive basalts, and Fe/Mn ratios >70 in olivine crystals,
550 provides substantial evidence for the presence of a lithologically distinct, pyroxenitic component in
551 the mantle source. We interpret this signature to represent the presence of recycled oceanic crust in
552 the Galápagos mantle plume, likely dragged up from the margins of the Pacific LLSVP. We also note
553 that there is no evidence in the geochemical composition of the Galápagos basalts to suggest that
554 upwelling LLSVP material is lithologically distinct from the surrounding mantle. As a result, the Pacific
555 LLSVP is unlikely to be formed through accumulation of subducted oceanic crust.

556 DATA AVAILABILITY STATEMENT

557 The data used in this study, and the python scripts used for data plotting, are available via
558 <https://zenodo.org/badge/latestdoi/384184976>

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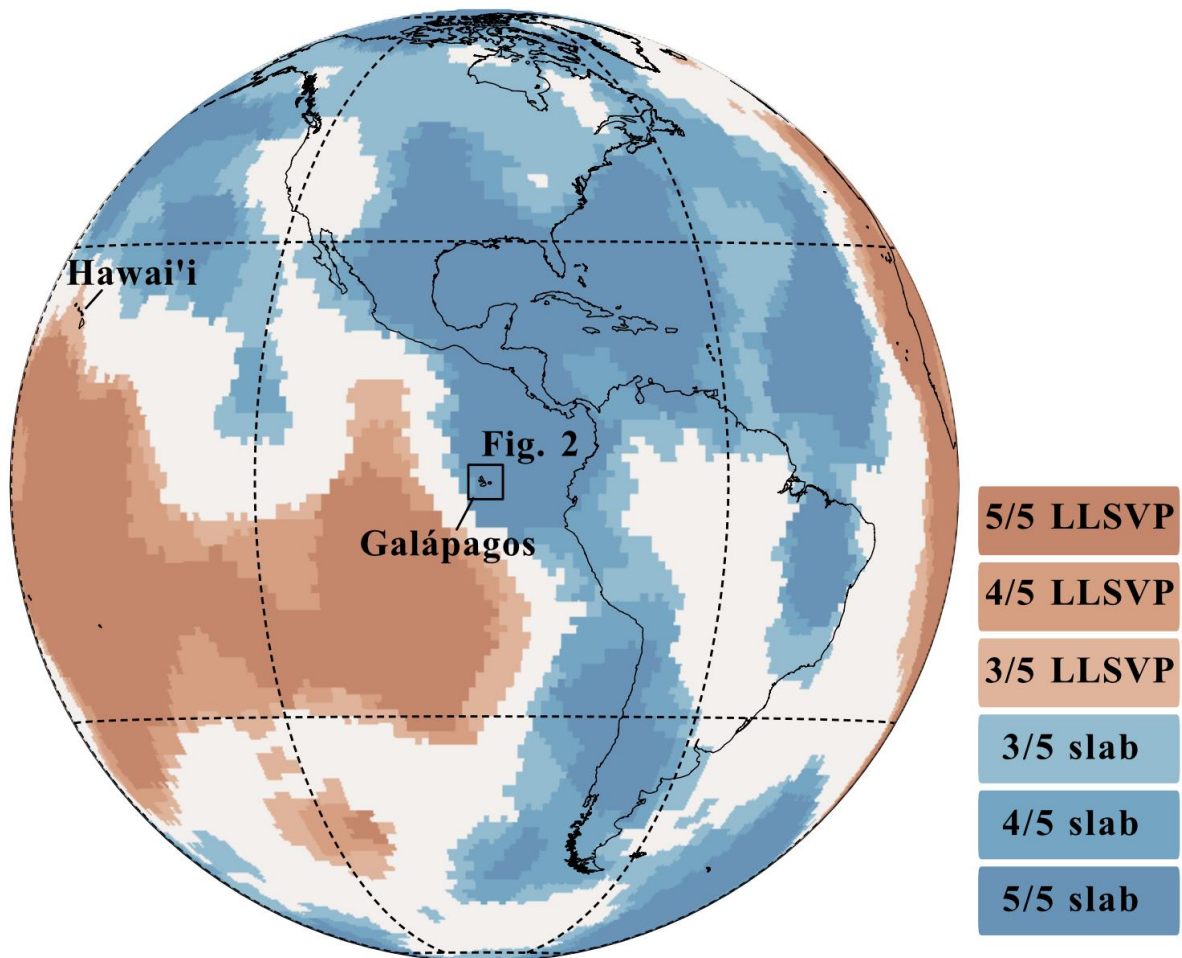
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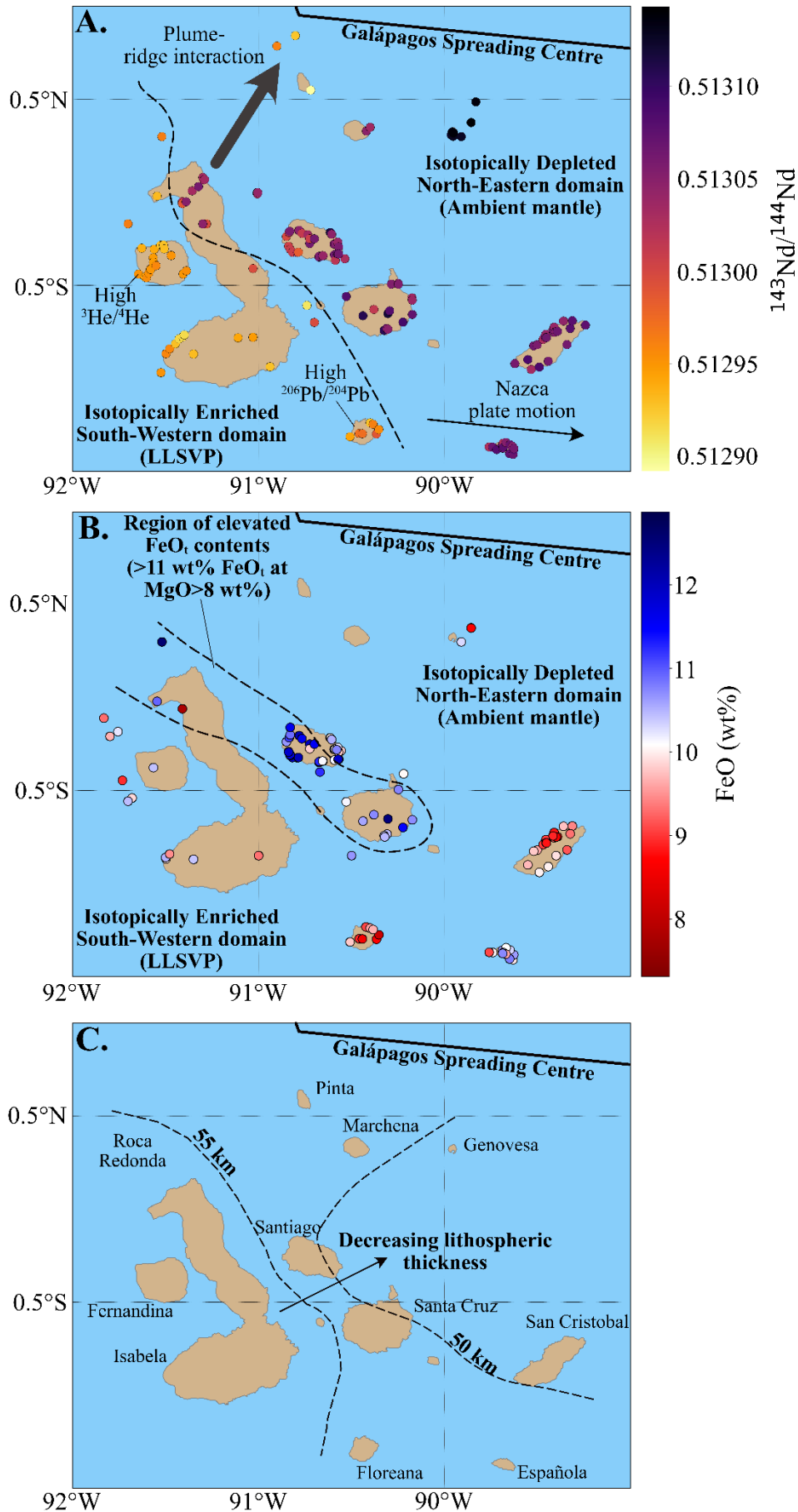
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923 **FIGURE CAPTIONS**



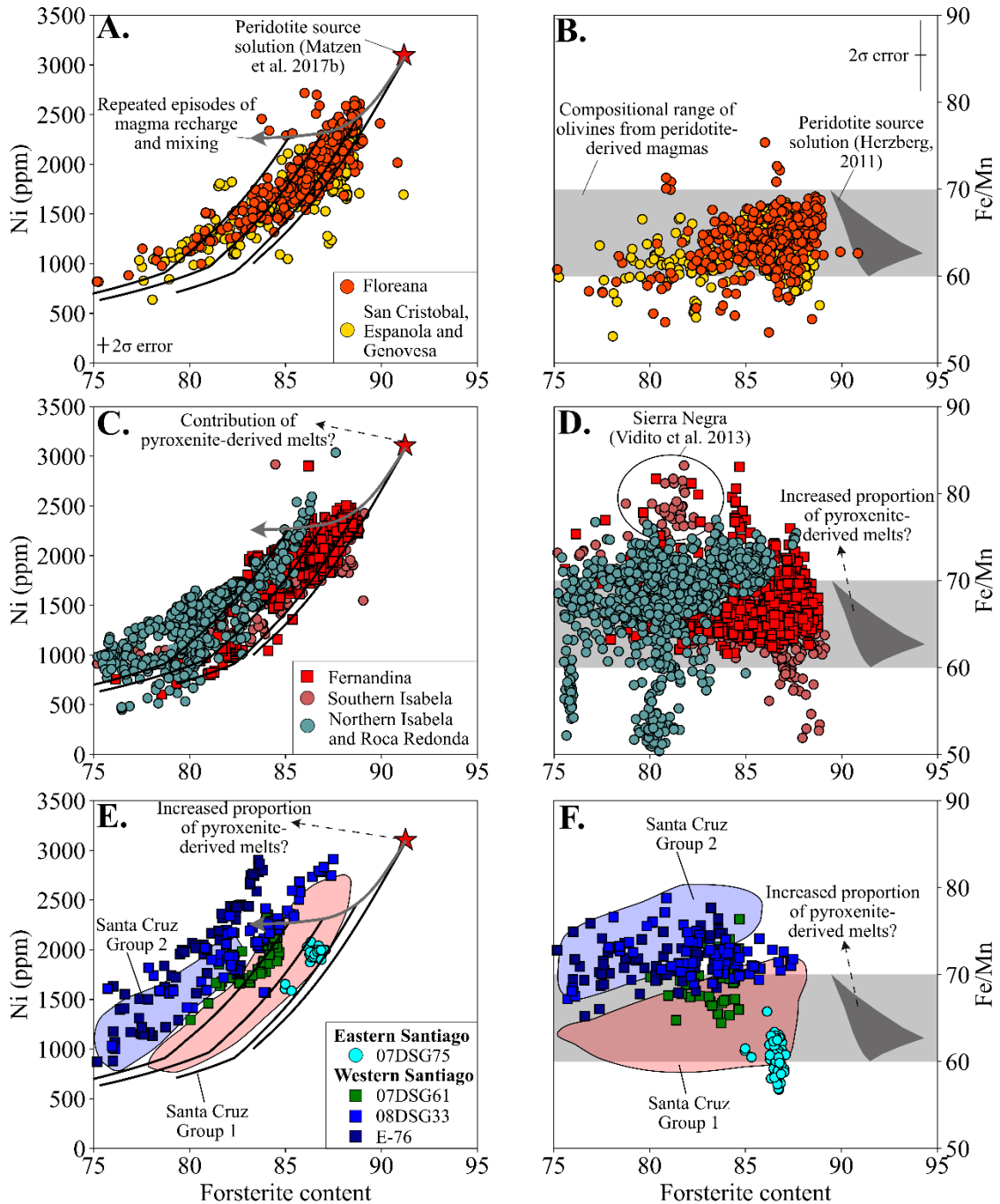
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925 **Figure 1** – Coloured regions on the map show where different tomographic models agree on the
926 lowermost mantle being slow (red colours) and fast (blue colours) (Cottaar and Lekic, 2016, see
927 reference therein for the five tomographic models included). Slow regions are defined as LLSVPs
928 while fast regions are generally interpreted as subducted slab material. The geographic location of
929 the Galápagos Archipelago is located near the NW-SE striking boundary between these two regions
930 at the core-mantle boundary.



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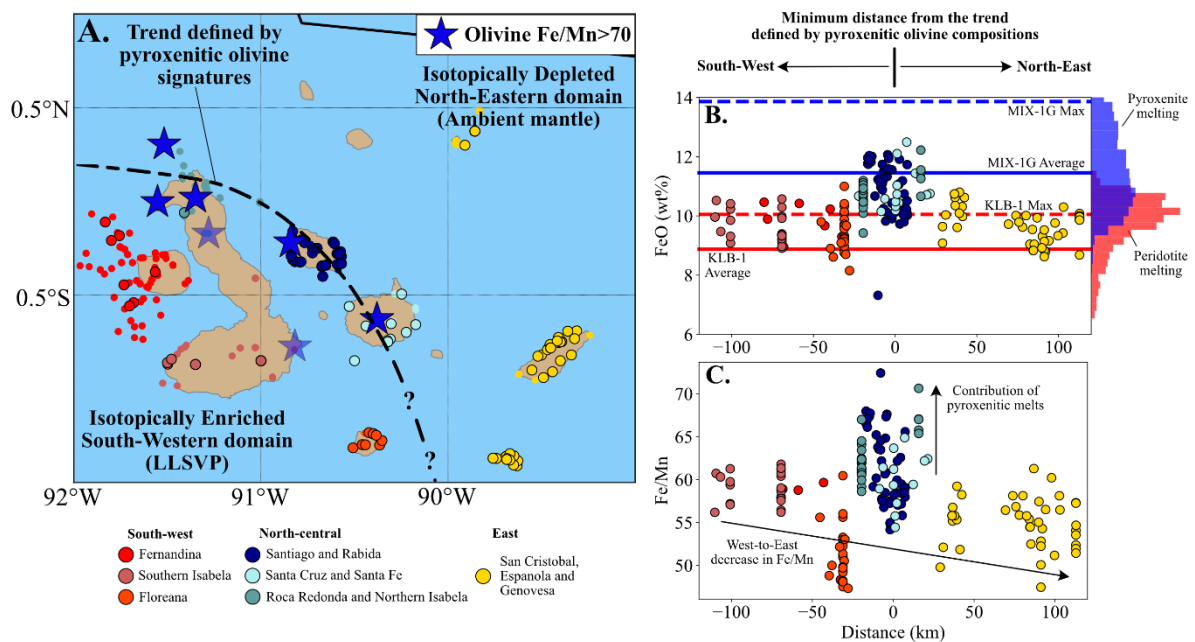
932 **Figure 2** – Variation in the composition of basalts erupted across the Galápagos Archipelago. **A.**
933 Spatial variations in the $^{143}\text{Nd}/^{144}\text{Nd}$ composition of the Galápagos basalts. Less radiogenic, and thus
934 more enriched, Nd isotope signatures are observed in the south-western Galápagos. The enriched
935 isotopic signature of Pinta is likely related to the transfer of compositionally enriched melts to the
936 nearby Galápagos Spreading Centre (Gleeson and Gibson, 2021). **B.** Variations in the FeO_t content of
937 high-MgO basalts erupted in the Galápagos (basalts with MgO contents above 8 wt% are shown).
938 Notably, areas with the highest FeO_t contents are found in the north and central Galápagos, on the
939 islands of Santiago, Santa Cruz, Roca Redonda, and on the northern margins of Isabela. **C.** Contours
940 of lithospheric thickness (taken from Gibson and Geist 2010) that reveal the thickness of the
941 lithosphere decreases eastwards in the Galápagos Archipelago. Data from Allan and Simkin, 2000;
942 Bow and Geist, 1992; Geist et al., 2002, 2006, 2005; Gibson et al., 2012; Gibson and Geist, 2010;
943 Harpp et al., 2003; Harpp and Weis, 2020; Kurz and Geist, 1999; McBirney and Williams, 1969;
944 Naumann et al., 2002; Saal et al., 2007; Standish et al., 1998; Swanson et al., 1974; Teasdale et al.,
945 2005; and White et al., 1993.



946

947 **Figure 3** – Composition of olivines from the eastern and southern Galápagos (**A.**, **B.**), western
 948 Galápagos (**C.**, **D.**), and central Galápagos (**E.**, **F.**). **A.** Ni contents of olivines from islands in the
 949 eastern Galápagos (Genovesa, Espanola, and San Cristobal) and Floreana in the southern Galápagos
 950 are consistent with the compositions predicted to form from melts of a peridotite source. Data from
 951 the western Galápagos (panels **C.** and **D.**) is typically consistent with the presence of a peridotitic

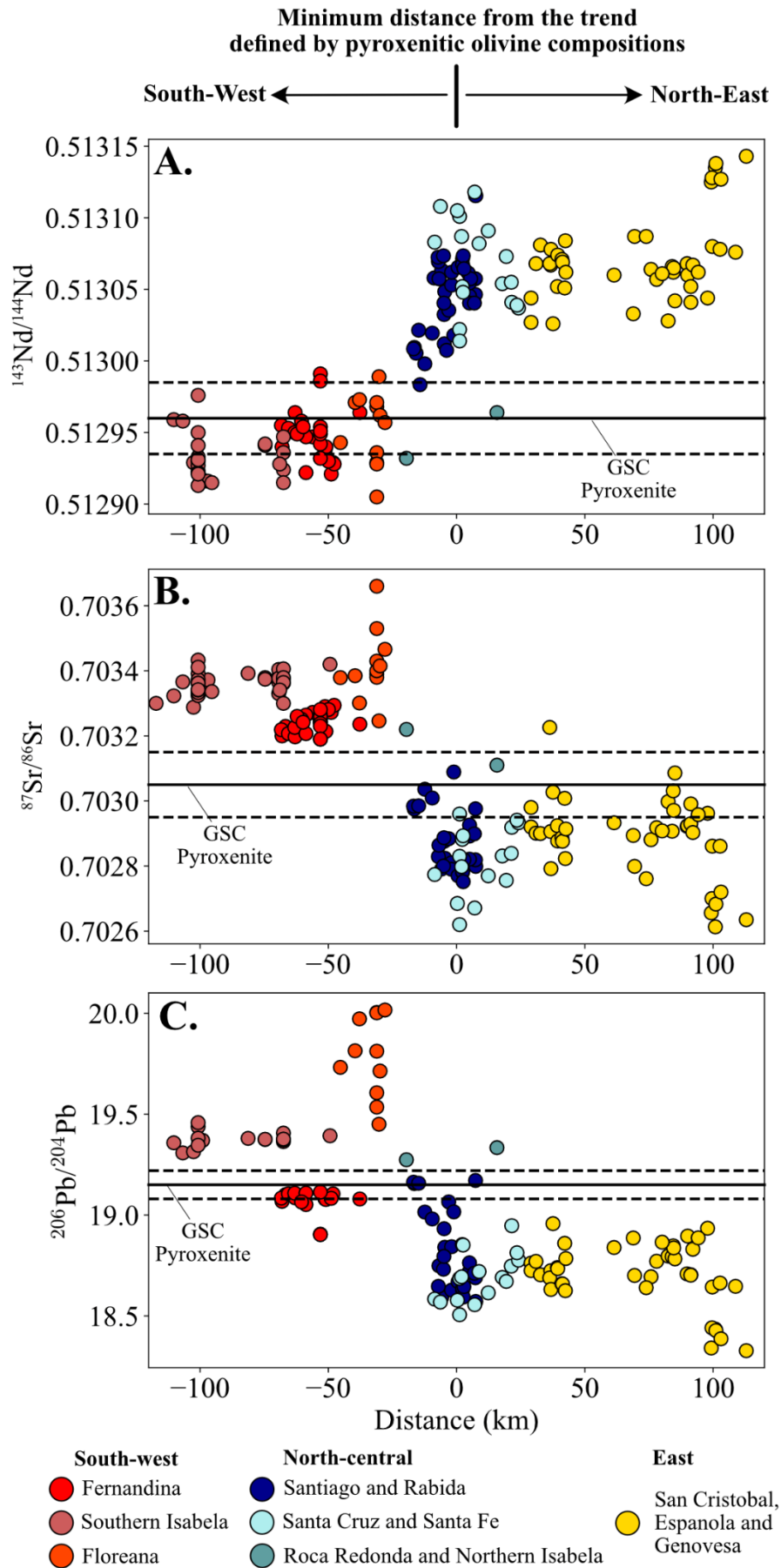
952 source. The Ni and Fe/Mn contents of olivines from northern Isabela and Roca Redonda, however,
 953 are difficult to explain without invoking the presence of a lithologically distinct source component.
 954 Olivine data from the central Galápagos (E. and F.) is more complex, the composition of olivines in
 955 tholeiitic basalts from Santiago and Group 1 olivines from Santa Cruz are consistent with a peridotitic
 956 source. Group 2 olivines from Santa Cruz and olivines in mildly alkaline basalts from Santiago,
 957 however, require the presence of a lithologically distinct component in their mantle source.
 958 Fractional crystallisation paths in A., C., and E. are taken from Gleeson and Gibson (2019). The range
 959 of olivine Fe/Mn contents that are consistent with derivation from a peridotite source is taken from
 960 (Herzberg, 2011). Peridotite source component taken from Matzen et al. (2017b) and Herzberg
 961 (2011). Data from this study, Vidito et al. (2013), and Gleeson and Gibson (2019).



962

963 **Figure 4** – Major element systematics of the Galápagos basalts. **A.** Location of basalts considered in
 964 this study, those with MgO contents above 8 wt% are displayed with a black outline. Samples with
 965 olivine Fe/Mn > 70 are highlighted by the blue stars. Samples from Sierra Negra and Darwin are
 966 partially transparent as the olivines measured from these volcanoes are very evolved. The black line
 967 represents the approximate trend through this region with ‘pyroxenitic’ olivine compositions. Basalts
 968 are broadly sub-divided into 3 categories: south-western basalts (reds); north-central basalts (blues);

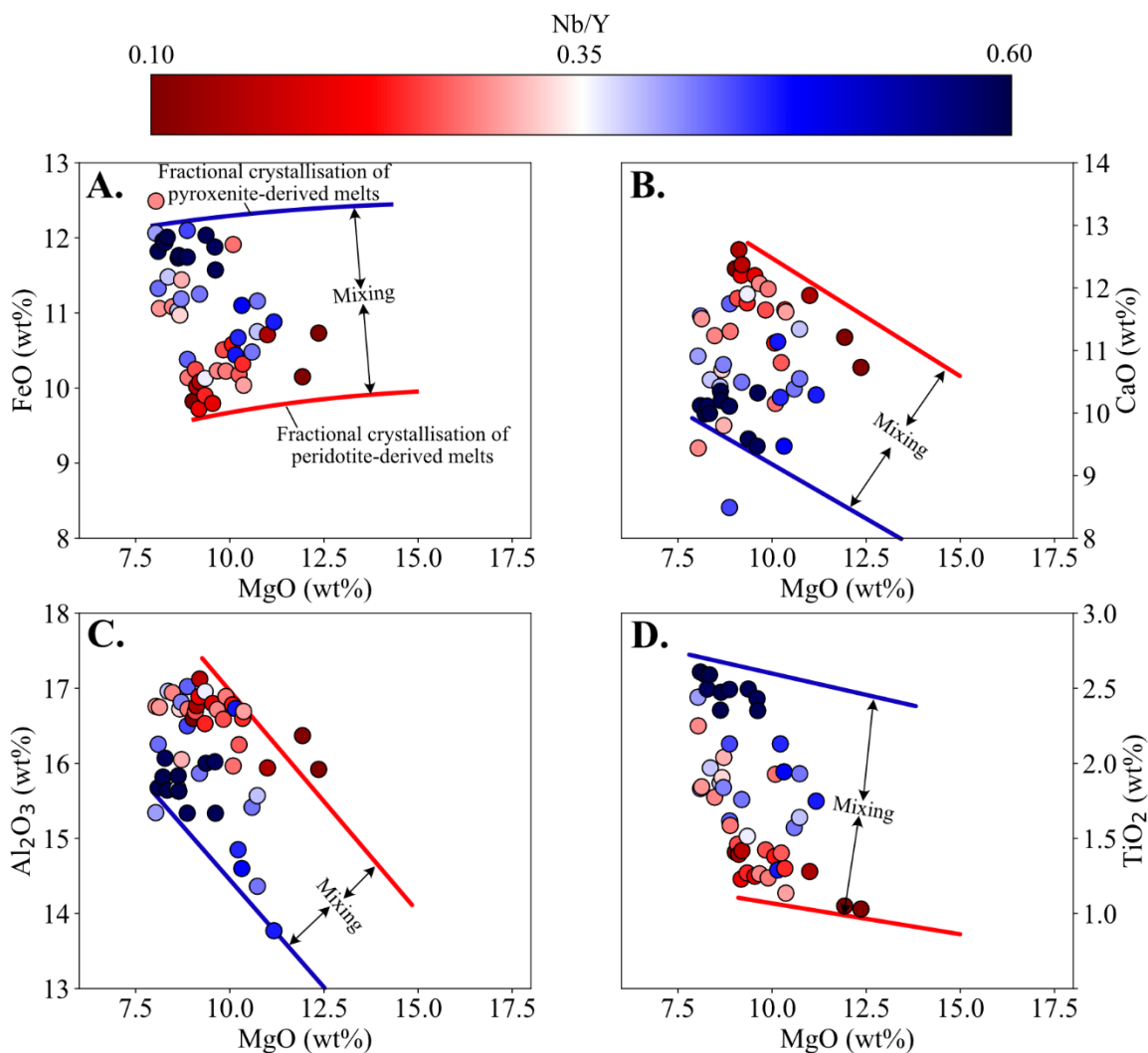
969 and eastern basalts (yellow). **B.** The FeO_t contents of high-MgO basalts are compared to their
970 minimum distance to the black line plotted in **A.** (i.e., the location of basalts with 'pyroxenitic' olivine
971 compositions). We find that the FeO_t content of basalts from the south-western Galápagos and the
972 north-eastern Galápagos are relatively constant, typically between 9 and 10.5 wt%. Notably, these
973 FeO_t contents are consistent with those measured in experimental melts of the KLB-1 peridotite
974 (Hirose and Kushiro, 1993; Takahashi et al., 1993) and THERMOCALC v3.4.7 calculations of melting
975 the KLB-1 peridotite (red histogram). Basalts from the north-central Galápagos, which plot within
976 ~25 km of the black line shown in **A.**, have higher FeO_t contents, up to 12.8 wt%. Such high FeO_t
977 contents require the presence of lithological heterogeneity in the mantle source. The average and
978 max FeO_t content of melting experiments on the pyroxenitic lithology MIX-1g is shown for reference
979 (Hirschmann et al., 2003; Kogiso et al., 2003), and the FeO_t contents predicted for melting of the
980 MIX-1g pyroxenite in THERMOCALC v3.4.7 is shown by the blue histogram. **C.** The Fe/Mn ratio of
981 high-MgO basalts from across the Galápagos shows a general decrease from west to east. Notable
982 exceptions to this trend are the basalts from the north-central Galápagos. Data from Allan and
983 Simkin, 2000; Bow and Geist, 1992; Geist et al., 2002, 2006, 2005; Gibson et al., 2012; Gibson and
984 Geist, 2010; Harpp et al., 2003; Harpp and Weis, 2020; Kurz and Geist, 1999; McBirney and Williams,
985 1969; Naumann et al., 2002; Saal et al., 2007; Standish et al., 1998; Swanson et al., 1974; Teasdale et
986 al., 2005; and White et al., 1993.



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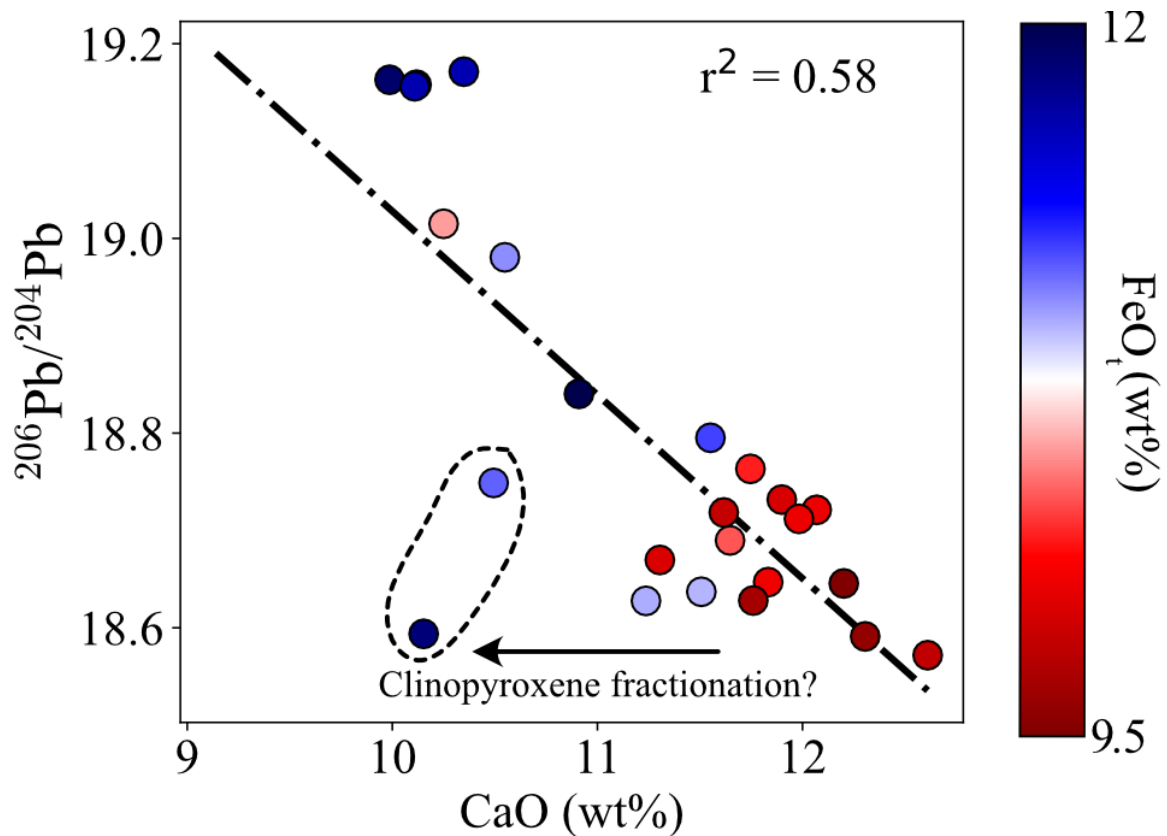
988 **Figure 5** – Radiogenic isotope composition of basalts from the Galápagos Archipelago. In all panels,
989 the proposed isotopic composition of the pyroxenite component in the source region of the
990 Galápagos Spreading Centre basalts is shown by the black horizontal lines. These values are taken
991 from the work of Gleeson et al. (2020) and Gleeson and Gibson (2021) and the uncertainties in these
992 isotopic compositions were constrained using the python code presented in Gleeson and Gibson
993 (2021). Basalts from Roca Redonda, northern Isabela, and the most enriched basalts from Santiago,
994 have very similar isotopic systematics to this proposed end-member. Data from Allan and Simkin,
995 2000; Bow and Geist, 1992; Geist et al., 2002, 2006, 2005; Gibson et al., 2012; Gibson and Geist,
996 2010; Harpp et al., 2003; Harpp and Weis, 2020; Kurz and Geist, 1999; McBirney and Williams, 1969;
997 Naumann et al., 2002; Saal et al., 2007; Standish et al., 1998; Swanson et al., 1974; Teasdale et al.,
998 2005; and White et al., 1993.

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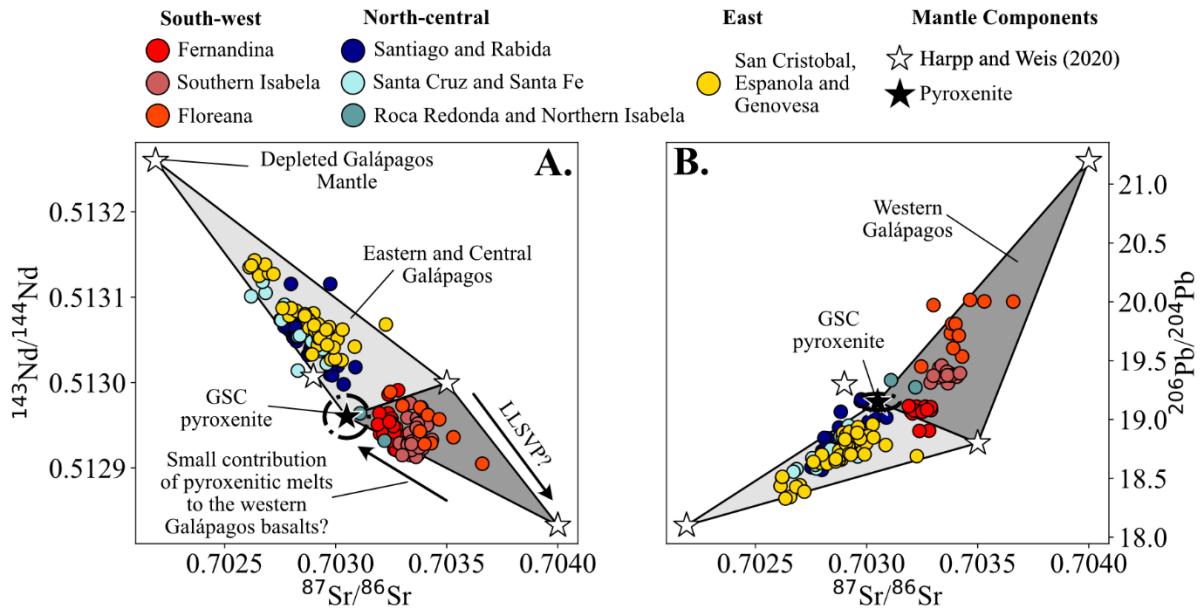
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1001 **Figure 6** – Major element systematics of basalts from Santiago, Santa Cruz, Rabida and Santa Fe with
1002 MgO contents >8 wt%. The major element systematics of the high-MgO basalts are related to their
1003 isotopic and trace element signatures (represented here by their Nb/Y ratio). Pyroxenite melts
1004 contain high FeO_t and TiO₂, but lower CaO and Al₂O₃ contents than the peridotitic melts, consistent
1005 with experimental data (Lambart et al., 2013). Blue and red lines display the olivine fractionation
1006 curves, calculated by removing olivine whose composition is calculated using the olivine K_d of
1007 Herzberg and O’Hara (2002), for hypothetical pyroxenite-derived and peridotite-derived melts,
1008 respectively. Data from Gibson et al., 2012; Gibson and Geist, 2010; Harpp and Weis, 2020; McBirney
1009 and Williams, 1969; Saal et al., 2007; and White et al., 1993.



1010

1011 **Figure 7** – Correlation between the major element systematics of the high-MgO Santiago basalts
1012 (MgO >8 wt%) and radiogenic isotopes. Strong correlations that are significant at the 99%
1013 confidence level are observed between CaO or FeO_t and the radiogenic isotope ratios considered in
1014 this study ($^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$, $^{206}\text{Pb}/^{204}\text{Pb}$) and confirm the relationship between isotopic
1015 enrichment and pyroxenitic contribution in the central Galápagos. Two samples with low CaO
1016 contents and unradiogenic $^{206}\text{Pb}/^{204}\text{Pb}$ signatures might result from unfiltered clinopyroxene
1017 fractionation, but we expect the influence of plagioclase and clinopyroxene fractionation to be
1018 minor in most Galápagos basalts considered here (see Supplementary Information). Data from
1019 Gibson et al. (2012).



1020

1021 **Figure 8** – Isotopic composition of the Galápagos basalts compared to the proposed mantle end-

1022 members from Harpp and Weis (2020) and the proposed isotopic composition of the Galápagos

1023 pyroxenite component (determined using the models presented by Gleason and Gibson (2021)).

1024 There is a clear divide between the isotopic composition of basalts from the south-western

1025 Galápagos and those from the central and eastern Galápagos. We suggest that the isotopic

1026 composition of basalts from the eastern and central Galápagos are controlled by mixing of melts

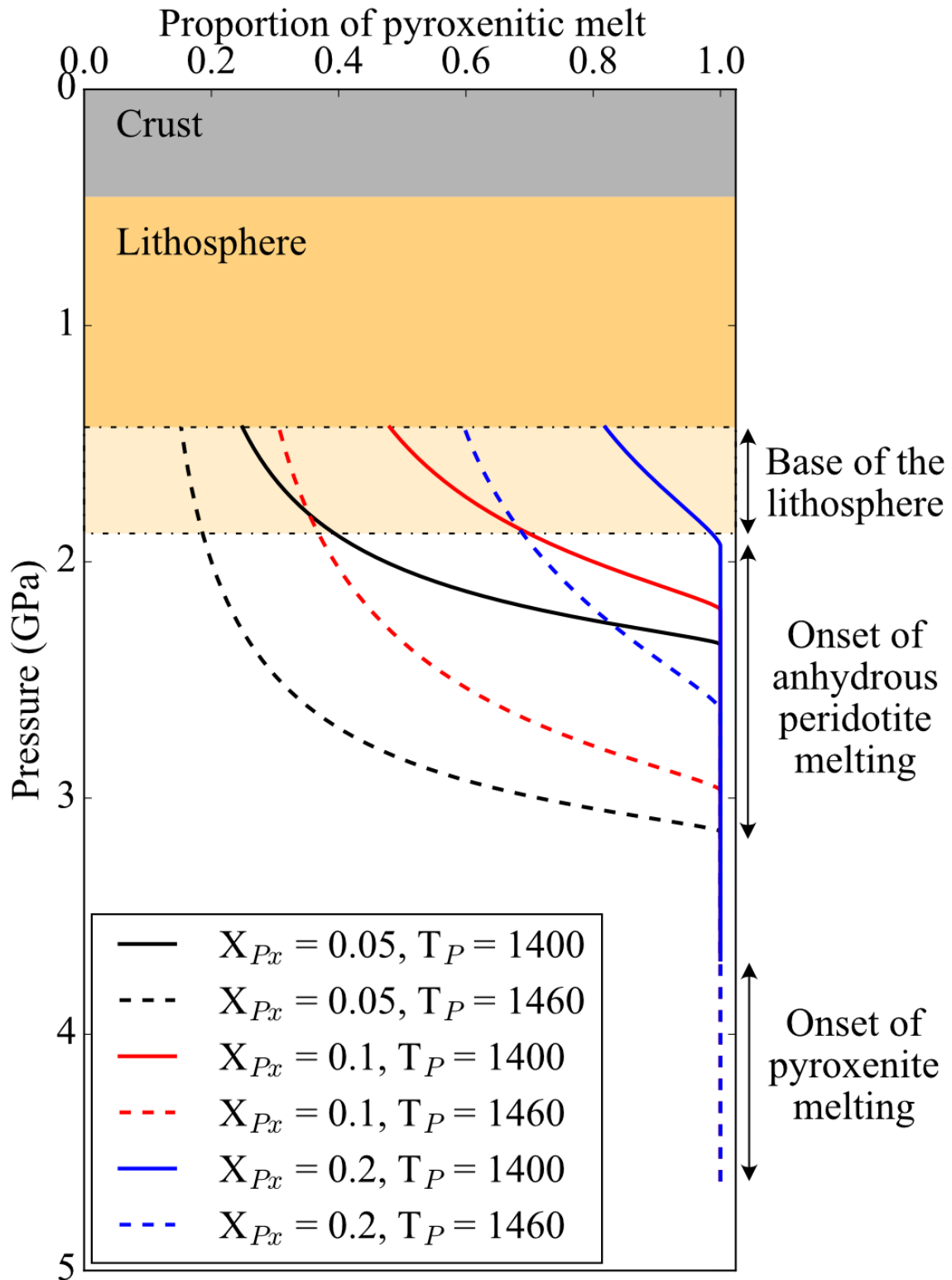
1027 from the DGM and the proposed pyroxenitic end-member (potentially with a minor contribution

1028 from LLSVP material). On the other hand, basalts from the south-western Galápagos are primarily

1029 sourced from LLSVP material, but small contributions of pyroxenitic material may influence their

1030 isotopic systematics.

1031



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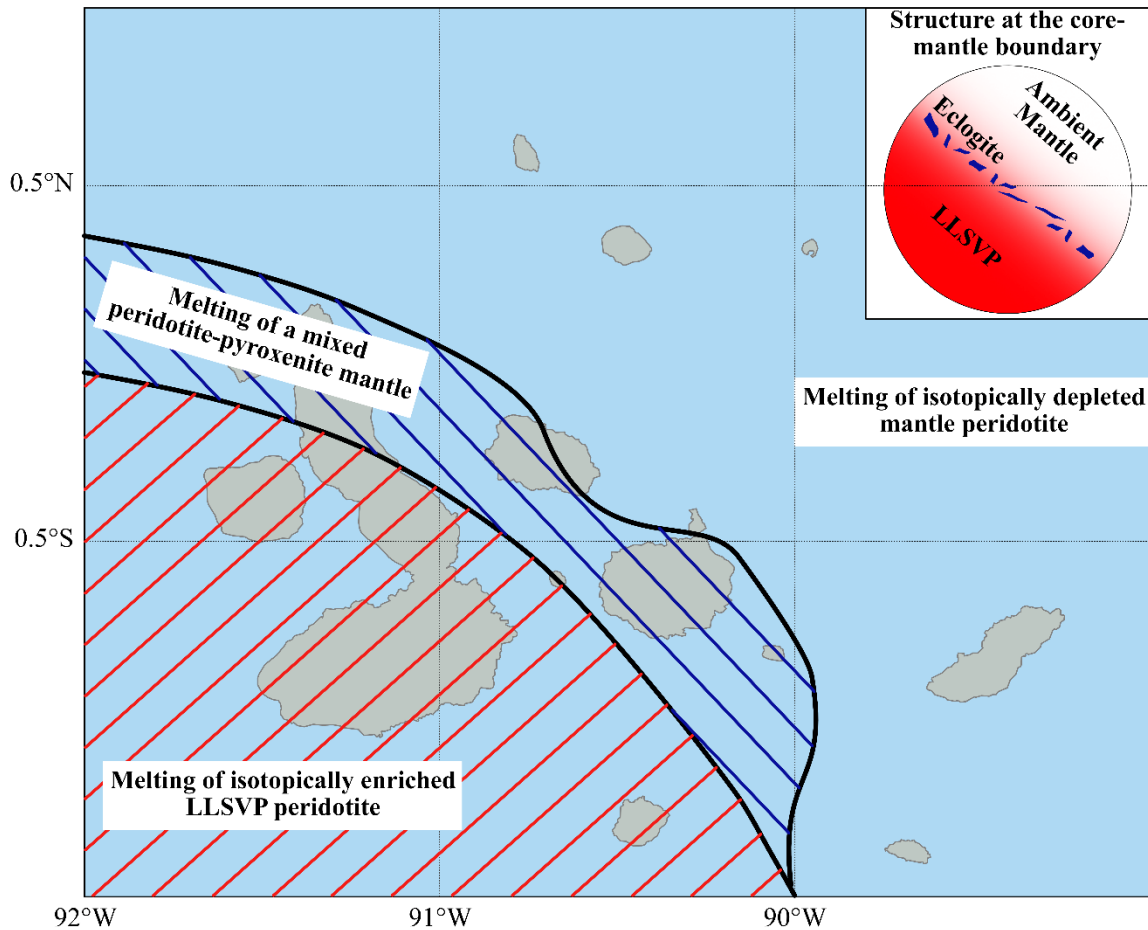
1033 **Figure 9** – Proportion of pyroxenitic melt predicted from melting of a two-component mantle.

1034 Calculations were performed in the pymelt module (Matthews et al., 2020) over a range of initial

1035 parameters, including the proportion of pyroxenite (formed as the reaction product of melts of

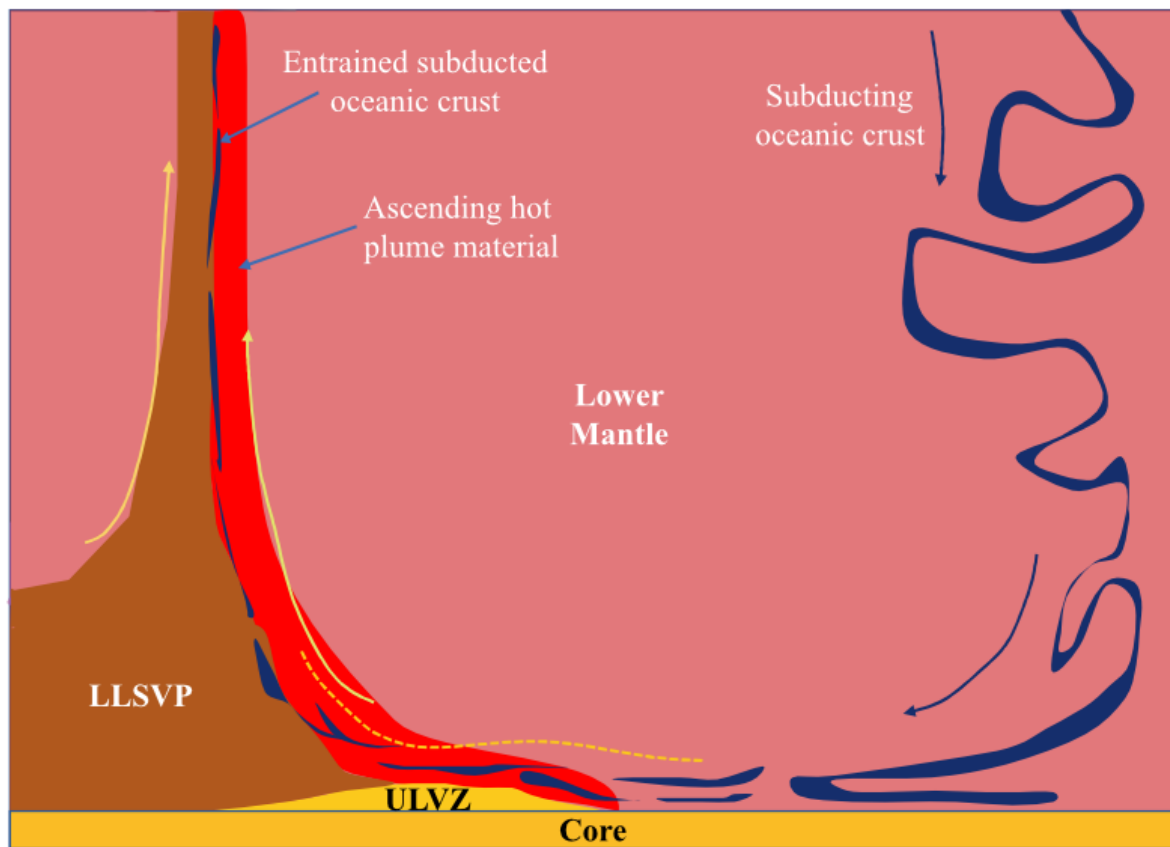
1036 subducted oceanic crust and peridotite) in the source (X_{Px}), the mantle potential temperature (T_p),
1037 and the pressure at the base of the lithosphere.

1038



1040 **Figure 10** – Distribution of peridotite and pyroxenite in the mantle source region of the Galápagos
1041 basalts. The isotopically enriched mantle beneath the south-western Galápagos displays no evidence
1042 for lithological heterogeneity and is thus interpreted to be peridotitic. As a result, there is no
1043 evidence in the Galápagos to suggest that the Pacific LLSVP represents a pile of subducted oceanic
1044 crust. In the north-central Galápagos the chemistry of the erupted basaltic lavas is controlled by
1045 mixing of melts from a pyroxenitic mantle source, formed through the reaction of melts from
1046 subducted oceanic crust (eclogite) with surrounding mantle peridotite, and upwelling mantle

1047 peridotite. In the eastern Galápagos, the depleted nature of the basalts indicates that the mantle
1048 source is dominated by isotopically depleted peridotitic mantle.



1049

1050 **Figure 11** – Schematic of the possible structure of the Pacific LLSVP margin at the base of the
1051 Galápagos mantle plume. Subducted oceanic lithosphere is present near the margin of the LLSVP
1052 leading to the steep LLSVP margin and the spatial distribution of lithological heterogeneity in the
1053 Galápagos mantle plume. The Galápagos plume has a complex asymmetric structure, with peridotitic
1054 mantle rising on the north-eastern side of the plume and enriched LLSVP material rising on the
1055 south-western side. The LLSVP is likely formed from primordial material. Figure adapted from
1056 Stevenson (2019).