# Upper mantle mush zones beneath low melt flux ocean island volcanoes: insights from Isla Floreana, Galápagos

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## ABSTRACT

The physicochemical characteristics of sub-volcanic magma storage regions have important implications for magma system dynamics and pre-eruptive behaviour. The architecture of magma storage regions located directly above high buoyancy flux mantle plumes (such as Kīlauea, Hawai'i and Fernandina, Galápagos) are relatively well understood. However, far fewer constraints exist on the nature of magma storage beneath ocean island volcanoes that are distal to the main zone of mantle upwelling or above low buoyancy flux plumes, despite these systems representing a substantial proportion of ocean island volcanism globally. To address this, we present a detailed petrological study of Isla Floreana in the Galápagos Archipelago, which lies at the periphery of the upwelling mantle plume and is thus characterised by an extremely low flux of magma into the lithosphere. Detailed *in situ* major and trace element analyses of crystal phases within exhumed cumulate xenoliths, lavas and scoria deposits, indicate that the erupted crystal cargo is dominated by disaggregated crystal-rich material (i.e., mush or wall rock). Trace element disequilibria between cumulus phases and erupted melts, as well as trace element zoning within the xenolithic clinopyroxenes, reveals that reactive porous flow (previously identified beneath mid-ocean ridges) is an important process of melt transport within crystal-rich magma storage regions. In addition, application of three petrological barometers reveal that the Floreana mush zones are located in the upper mantle, at a depth of 23.7±5.1 km. Our barometric results are compared to recent studies of high melt flux volcanoes in the western Galápagos, and other ocean island volcanoes worldwide, and

28   demonstrate that the flux of magma from the underlying mantle source represents a first-order control

29   on the depth and physical characteristics of magma storage.

## KEY WORDS

31   Galápagos; magma storage; reactive porous flow; barometry.

# 1  INTRODUCTION

33   The physicochemical characteristics (such as size, pressure, volatile content and geochemical

34   heterogeneity) of magma storage at volcanic centres located directly above high buoyancy flux mantle

35   plumes (e.g. Kīlauea, Hawai'i and Isabela, Galápagos) have been subject to intense study over the

36   past few decades (Bagnardi et al., 2013; Bernard et al., 2019; Clague and Denlinger, 1994; Geist et

37   al., 1998; Naumann and Geist, 1999; Neal et al., 2019; Park et al., 2007; Pietruszka et al., 2015;

38   Poland et al., 2015; Sides et al., 2014; Stock et al., 2018; Wieser et al., 2020, 2019). Systems such as

39   Kīlauea are characterised by frequent volcanic activity, and geophysical (seismicity, ground

40   deformation) and geochemical (gas emissions) monitoring is prevalent. Monitoring data, combined

41   with petrological and geochemical analysis of erupted products (mineral textures, deformation

42   characteristics and chemistry) provide important insights into the architecture and dynamics of their

43   sub-volcanic plumbing systems (Amelung et al., 2000; Davidge et al., 2017; Geist et al., 2014;

44   Hartley et al., 2018; McCormick Kilbride et al., 2016). However, these systems (which we term 'high

45   melt flux') represent only one endmember of global plume-derived volcanism. Low melt flux

46   systems, either above low buoyancy flux plumes (e.g. Canary Islands; Longpre et al., 2014) or at

47   volcanic systems distal to the centre of mantle melting at high buoyancy flux mantle plumes (e.g.

48   eastern and south-eastern Galápagos; Harpp and Geist, 2018), are the other endmember.

49   While a substantial number of hotspot-related volcanic systems that have been active during the

50   Holocene are located in regions characterised by a relatively low flux of magma into the lithosphere

51   (i.e., regions distal to the main zone of plume upwelling or above low buoyancy flux plumes; Samoa,

52   Canary Islands, Cape Verde; Global Volcanism Program, 2013), only a small number of eruptions

53   have been observed (and recorded) at these systems since the advent of modern volcano monitoring

54    techniques. As a result, few constraints exist on the conditions of magma storage in regions

55    characterised by a low flux of magma into the lithosphere, relative to volcanic centres located above

56    the centre of mantle plumes with a large buoyancy flux (and thus generating a large flux of magma

57    and more frequent eruptions).

58    The flux of mantle-derived magma into the lithosphere is thought to impart a first-order control on the

59    evolution of ocean island volcanoes and the homogeneity of erupted liquids (Geist et al., 2014).

60    Therefore, placing constraints on the physicochemical characteristics of magma storage at low melt

61    flux ocean island volcanoes is essential for determining the influence of mantle dynamics and melt

62    generation processes on the structure and physical characteristics of sub-volcanic magma plumbing

63    systems. In the absence of detailed monitoring data, petrological and geochemical analyses of

64    volcanic products from past eruptions represent the only available tools for determining the structure

65    and processes operating within these systems, as well as possible eruption precursors.

66    Isla Floreana in the south-eastern Galápagos is currently located ~100 km downstream from where the

67    centre of the Galápagos plume impacts on the base of the lithosphere beneath the island of Isabela in

68    the western archipelago (Fig. 1; Villagómez et al., 2014). Hence, although the Galápagos plume has a

69    relatively high buoyancy flux compared to regions such as the Canary Islands (Jackson et al., 2017),

70    Floreana's location relative to the main zone of mantle plume upwelling results in an extremely low

71    flux of magma entering the lithosphere and, consequently, very infrequent volcanic activity (Harpp et

72    al., 2014a; Harpp and Geist, 2018). Floreana is considered to be an infrequently active volcanic

73    centre, rather than extinct, owing to the persistent volcanic activity since ~1 Ma and the long-lived

74    nature of volcanism on the eastern Galapagos islands that lie >100 km 'downstream' of the Galapagos

75    mantle plume (e.g. the youngest lavas on San Cristobal are ~9 ka; Mahr et al., 2016).

76    In this paper, we present a thorough petrological study of scoria, lava and xenolith samples from

77    Floreana and place constraints of the structure, depth and crystallinity of magmatic systems beneath

78    this low melt flux ocean island volcano. We compare our results with more frequently active volcanic

79    centres in the western Galápagos (near the centre of plume upwelling; Geist et al., 1998; Naumann

80    and Geist, 1999; Stock et al., 2018), as well as other ocean island volcanoes worldwide (Hammer et

81   al., 2016; Hartley et al., 2018; Poland et al., 2015), to investigate how the flux of magma into the

82   lithosphere influences the depth and crystallinity of sub-volcanic magma storage regions.

## 2   GEOLOGICAL BACKGROUND

84   The Galápagos Archipelago in the eastern equatorial Pacific is one of the most volcanically active

85   regions on Earth, with eruptions typically occurring every 2–3 years (Global Volcanism Program,

86   2013). Although most historic Galápagos eruptions have taken place on the two westernmost islands

87   of Isabela and Fernandina (Fig. 1), infrequent volcanic activity has also occurred on several islands in

88   the eastern and south-eastern Galápagos (for example, Santiago in 1906 and Marchena in 1991;

89   Global Volcanism Program, 2013). In fact, volcanic activity in the eastern Galapagos, >100 km

90   'downstream' of the postulated position of the plume stem, has been shown to be long-lived, with

91   volcanic activity on San Cristobal extending over 2 Myrs with the most recent lavas erupted at ~9 ka

92   (Mahr et al., 2016).

93   Volcanoes in the western Galápagos likely emerged within the last 500 kyr (Naumann and Geist,

94   2000), whereas those in the eastern and south-eastern Galápagos are considerably older (eruption ages

95   up to 2.3 Ma and 3.2 Ma have been measured on San Cristobal and Espanola, respectively; Bailey,

96   1976; Geist et al., 1986). In addition, substantial differences in geomorphology and the style of

97   volcanic activity are observed across the archipelago (Geist et al., 1995; Harpp et al., 2014a; Harpp

98   and Geist, 2018). For example, volcanoes in the western archipelago are typified by large summit

99   calderas (<700m deep), which are not present on the eastern islands (Chadwick and Howard, 1991;

100  Cleary et al., 2020; Harpp and Geist, 2018).

101  Geochemical distinctions between the western and eastern/south-eastern Galápagos islands are also

102  observed, which are primarily related to variations in the composition of the underlying mantle source

103  (Geist et al., 1988; Gibson and Geist, 2010; Gleeson et al., 2020; Harpp and White, 2001; White et al.,

104  1993) or the volume flux of mantle-derived magma that ascends into the lithosphere (Geist et al.,

105  1995, 2014; Gibson et al., 2016; Harpp and Geist, 2018). For example, variations in the flux of mantle

106  derived magma are hypothesised to influence the geochemical heterogeneity of erupted basalts at each

107    island: volcanoes in the western archipelago typically erupt a very narrow range of basaltic

108    compositions over hundreds of millennia during their main shield building phase, whereas basalts

109    erupted from a single island in the eastern and/or south-eastern archipelago, such as Floreana, tend to

110    display far greater compositional heterogeneity (Geist et al., 2014; Harpp and Geist, 2018).

111    Floreana is characterised by numerous scoria cones and blocky, heavily vegetated lava flows that can

112    typically be traced to the cone from which they originated (Bow and Geist, 1992; Harpp et al., 2014a).

113    The crustal thickness beneath Floreana is ~16 km, similar to that observed in the western Galápagos

114    10–18 km (Feighner and Richards, 1994), and the lithospheric thickness beneath the western and

115    south-eastern Galápagos is very similar (~50–60 km; Gibson and Geist, 2010). Recent work has

116    shown that the average volumetric eruption rate on Floreana over the past 1–1.5 Myrs is 1–10 $m^3 \cdot yr^{-1}$,

117    6 orders of magnitude lower than the current volcanic productivity at volcanoes in the western

118    Galápagos (cf. ~$4.4 \cdot 10^6$ $m^3 \cdot yr^{-1}$ at Fernandina; Harpp et al., 2014a; Kurz et al., 2014). This difference

119    in volumetric eruption rate likely reflects a substantially lower flux of magma into the lithosphere

120    beneath Floreana than beneath each volcanic centre in the western Galápagos.

121    Despite the relatively low volcanic productivity of Floreana over the last ~1–1.5 Myrs, the erupted

122    products have several important characteristics that provide insights into the nature of the underlying

123    magmatic system. For example, Floreana has a high proportion of pyroclastic deposits compared to

124    the other Galápagos islands (Harpp et al., 2014a) and eruption deposits typically contain a large

125    number of cumulate xenoliths (Bow and Geist, 1992; Lyons et al., 2007), which have been interpreted

126    as evidence for very high magma ascent rates (Harpp et al., 2014a).

127    Floreana is the only Galápagos island that displays evidence for multiple stages in its volcanic

128    evolution. Submarine parts of the island have isotopic and trace element characteristics that are

129    similar to those measured in recent basalts erupted on southern Isabela, near the centre of plume

130    upwelling (e.g. Sierra Negra and Cerro Azul, Fig. 1A), whereas the subaerial material is isotopically

131    distinct (high $^{206}Pb/^{204}Pb$ and $^{87}Sr/^{86}Sr$ ratios; Harpp et al., 2014). The trace element and isotopic

132    differences between the erupted basalts is mirrored in xenoliths found in the Floreana lava and scoria

133    deposits: gabbroic xenoliths have radiogenic isotope ratios that are similar to modern Isabela basalts,

134    whereas wehrlitic xenoliths have trace element and isotopic compositions that resemble recent

135    Floreana subaerial basalts (Lyons et al., 2007). Differences in the isotopic characteristics of the

136    Floreana lavas (submarine vs subaerial) and xenoliths (gabbros vs wehrlites) are thought to indicate a

137    change in the mean composition of magma produced by mantle melting beneath the island at ~1–1.5

138    Ma (Harpp et al., 2014a). In this study, we focus on constraining the depth and physical

139    characteristics of magma storage during the most recent period of volcanic activity on Isla Floreana

140    (<1–1.5 Ma) using chemical and textural analysis of crystal phases in lava flows and xenolithic

141    nodules.

# 3   SAMPLES AND PETROGRAPHY

143    The Floreana samples analysed in this study consist of lavas (27 samples), scoria (2 samples) and

144    xenoliths (4 wehrlite, 3 dunite and 2 gabbro samples; Fig. 1B). Most lava samples were collected from

145    the unaltered, low vesicularity cores of blocky flows or glassy flow fronts. All samples form part of

146    the Main Series of Floreana lavas identified by Bow and Geist (1992). The scoria samples were

147    collected from two separate deposits and comprise rapidly cooled scoria lapilli (~0.5–2 cm across;

148    17MMSG16) and bombs (~10 cm across; 17MMSG20). Xenolithic fragments (3–15cm across) were

149    sampled from two different scoria cones on the north-east coast of Floreana; similar xenoliths are also

150    found within most lava flows across the entire island (Supplementary Information).

## 3.1   Lavas and scoria

152    The lava and scoria samples analysed in this study are typically olivine phyric with minor anhedral

153    clinopyroxene and very rare orthopyroxene. Except for small plagioclase laths in the microcrystalline

154    groundmass, plagioclase crystals are extremely rare in the Floreana lavas. Plagioclase macrocrysts are

155    only present in one of our lava samples (17MMSG29) where they occur as isolated phenocrysts and in

156    plagioclase-olivine crystal clots (Table S.1). The abundance of olivine and absence of plagioclase in

157    the Floreana lavas and scoria contrasts with basalts in the central, northern, and western parts of the

158    Galápagos Archipelago, where plagioclase-phyric and ultraphyric basalts are common (Geist et al.,

159    2002; Gibson et al., 2012; Harpp et al., 2014b).

160 Despite their relatively simple mineralogy, Floreana lava and scoria samples contain texturally diverse

161 olivine crystals which can be divided into five distinct groups (Fig. 2):

162 - Group 1 olivines are present in all lava and scoria samples and are the most abundant type of

163 olivine (~60-70% of all crystals). They are characterised by homogeneous cores, with respect

164 to major elements, and narrow normally zoned rims (Fig. 2A). Group 1 olivines are generally

165 subhedral to euhedral.

166 - Group 2 olivines are the second most abundant group (~20-30%) and display reverse zoning.

167 They are typically euhedral, with occasional small embayments (Fig. 2B).

168 - Group 3 olivines are also reversely zoned, but are distinguished by skeletal overgrowths,

169 indicating significant undercooling of the host magma and rapid crystal growth (Fig. 2C;

170 Donaldson, 1976; Welsch et al., 2014). Group 3 olivines are less abundant than Groups 1 and

171 2 olivines (<10%).

172 - Group 4 olivines are present in low abundance (<10%). They have homogeneous cores and

173 reverse-zoned rims (up to ~300 μm thick). The rims have sieve textures, potentially

174 suggesting resorption and chemical disequilibrium with their carrier melts (Fig. 2D).

175 - Group 5 olivines are only found in a minority of samples and are characterised by the

176 presence of 4 compositional zones with alternating high and low forsterite contents (visible in

177 back-scattered electron images; Fig. 2E and F).

178 ### 3.2 Xenoliths

179 *3.2.1 Gabbroic xenoliths*
180 Floreana gabbroic xenoliths predominantly comprise plagioclase (33–66 vol.%), clinopyroxene (28–

181 46 vol.%) and orthopyroxene (5–15 vol.%), with little or no olivine (Table S.2). Plagioclase and

182 pyroxene crystals are typically >500 μm and grain sizes are relatively uniform within a single xenolith

183 sample. Where three plagioclase grains meet at a triple junction, 120° grain boundaries indicate a high

184 degree of textural equilibrium (Fig. 3A; Holness et al., 2005). Some of the gabbros have

185 clinopyroxene-rich and plagioclase-rich layers of 2–5 mm thickness.

186 *3.2.2    Dunitic xenoliths*
187 Olivine dominates the dunitic Floreana xenoliths (>90 vol.%). The olivine crystals are subhedral to

188 euhedral and may have undergone partial textural re-equilibration, with some olivine triple junctions

189 approaching 120° grain boundaries. Minor intercumulus clinopyroxene is present along grain

190 boundaries and between pre-existing olivine crystals (Fig. 3B).

191 *3.2.3    Wehrlitic xenoliths*
192 Floreana wehrlitic xenoliths contain olivine (>50 vol.%), clinopyroxene (20–40 vol.%),

193 orthopyroxene (~0–7 vol.%) and minor spinel (<1 vol.%; Table S.2). Clinopyroxene typically occurs

194 as large (<5 mm) oikocrysts, which enclose rounded olivine chadacrysts <500 μm in diameter (Fig.

195 3C and D). Fine-scale orthopyroxene exsolution lamellae (<2 μm) is observed in the clinopyroxenes

196 of a single wehrlite (17MMSG03a; Fig. 3E and F). Olivine grains that are not enclosed by

197 clinopyroxene are typically larger (>1 mm) and more euhedral than the chadacrysts. In some samples,

198 the boundary between clinopyroxene and olivine crystals is characterised by a thin (<20-30 μm) layer

199 of glass and very fine-grained microcrysts. Excluding the rare exsolution lamellae in sample

200 17MMSG03a, orthopyroxene crystals are typically anhedral, infilling the space between earlier

201 formed clinopyroxene and olivine grains. Our observations of dunitic and wehrlitic xenoliths (which

202 have the isotopic signatures of modern day Floreana basalts; Lyons et al., 2007) indicate that the

203 typical order of crystallisation beneath Floreana is olivine, followed by clinopyroxene, with little to no

204 crystallisation of plagioclase.

# 4   ANALYTICAL METHODOLOGY

## 4.1   Electron microprobe analysis

207 Glass chips, olivine and clinopyroxene crystals were hand-picked from crushed scoria and lava

208 samples, mounted in epoxy or indium, and then ground and polished prior to analysis (crystals

209 mounted in indium were polished individually prior to mounting). Xenolithic crystals were analysed

210 as individual crystals mounted in indium or *in situ* in petrographic thin sections. The major and minor

211 element concentrations of olivine, clinopyroxene and glass were measured using a Cameca SX100

212  electron microprobe in the Department of Earth Sciences, University of Cambridge. Calibrations were

213  made using mineral and metal standards prior to each analytical session (see Gleeson and Gibson,

214  2019 for details). Glasses were analysed using a 6 nA, 15 kV, defocused (5 μm) beam for most

215  elements. Na and K were analysed first (10 s peak count time) to avoid alkali migration. Other

216  elements were analysed with peak count times of 10 s (Si), 20 s (Fe), 30 s (Al, P, Ca, Mg), 40 s (Mn),

217  or 60 s (Ti). Sulphur was analysed last using a 20 nA beam current and a 60 s peak count time.

218  Pyroxene compositions were determined by spot analyses using a 20 nA, 15 kV, focused (~1 μm)

219  beam, with Na, K and Si analysed first (10 s).  Element maps of Cr, Ti, and Al in key clinopyroxene

220  crystals from the Floreana xenoliths were created using a 60 nA, 15 kV, focused (~1 μm) beam, with

221  a dwell time of 150 ms. Cr counts were collected on a PET and a LIF crystal, Al counts were

222  collected on two TAP crystals, and Ti counts were collected on a PET crystal. Olivine electron

223  microprobe analysis was carried out using the method outlined in Gleeson and Gibson (2019).

224  Analytical uncertainties were tracked through analysis of appropriate Smithsonian Microbeam

225  Standards (Jarosewich et al., 1980). Accuracy was typically between 98 and 102% for all phases. $2\sigma$

226  analytical precision of clinopyroxene and olivine analyses are typically better than 2–3% for major

227  elements (>1 wt%) and typically ~5-10% for minor elements (<1 wt%). Similarly, the $2\sigma$ precision for

228  glass analysis was typically <3% for major elements, ~5% for Na, and ~10% for K (see

229  Supplementary Information).

230  ## 4.2  Laser ablation Inductively Coupled Plasma Mass Spectrometry

231  Trace element concentrations were measured in the apparent cores (i.e. as exposed in the 2D plane) of

232  clinopyroxene crystals from scoria and xenolith samples using an ESI193 laser coupled to a Nexion

233  350D inductively coupled plasma mass spectrometer in the Department of Earth Sciences, University

234  of Cambridge. Analyses were collected in spot mode using a 20 Hz laser repetition rate, 4 J/cm$^2$

235  fluence and 80 μm spot size, or in transect mode using a 10 Hz repetition rate, 3.5 J/cm$^2$ fluence and

236  30 μm spot size. For transects, individual spots were offset into two (alternating) lines to increase the

237  spatial resolution. Laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) data

238 reduction was carried out in Iolite©, with NIST 612SRM as the standard reference material (Hinton,

239 1999) and $^{43}$Ca (from electron microprobe analysis) as the internal reference standard. Analytical

240 accuracy was tracked using a USGS glass standard (Jochum et al., 2016) and was between 95% and

241 105% for most elements (See Supplementary File). 2σ analytical precision of spot analyses were

242 monitored through analysis of an in-house clinopyroxene standard and was 5–10% for the light rare-

243 earth elements (LREE), Y, Sr, and Zr and 10–20% for the heavy rare earth element (HREE). 2σ

244 analytical precision was ~10% for all elements of interest in transect analyses (Ce, Y).

# 5 GLASS AND MINERAL CHEMISTRY

## 5.1 Matrix glass compositions

247 The matrix glass compositions measured in one scoria (17MMSG16) and two glassy lava samples

248 (17MMSG12 and 17MMSG27) from Floreana have very similar MgO concentrations (mean

249 concentrations of 6.06–6.67 wt%) but exhibit differences in the concentrations of other elements (Fig.

250 4). For example, sample 17MMSG12 has consistently lower $TiO_2$ and $Al_2O_3$ concentrations than

251 17MMSG16, which must either reflect heterogeneity in the composition of primary mantle melts or

252 variations in crustal processing (e.g. the extent of plagioclase or clinopyroxene crystallisation; Fig.

253 4A,D). The largest variation in the glass major element composition, however, is seen in sample

254 17MMSG27 where, at a near constant MgO contents, the CaO, $Na_2O$, and $K_2O$ contents vary by ~6

255 wt%, 4.5 wt%, and 1.5 wt%, respectively (Fig. 4B,C). Differences between our matrix glass major

256 element analyses and previously-published whole-rock data from Floreana (Harpp et al., 2014a) are

257 primarily due to olivine accumulation in the whole-rock samples (additional accumulation of

258 clinopyroxene may explain the high CaO content of some whole-rock samples; Fig. 4C).

259 Our Floreana matrix glass analyses have higher $Al_2O_3$ concentrations, at a given MgO content, than

260 basaltic glass and whole-rock measurements from the western Galápagos Archipelago (excluding

261 whole-rock samples with accumulated plagioclase, Fig. 4D; Geist et al., 2002). This indicates

262 substantially lower extents of plagioclase fractionation in the Floreana magmatic system and is

263 consistent with the scarcity of plagioclase phenocrysts in erupted Floreana lavas. Reduced plagioclase

264   crystallisation could be due to the major element composition or $H_2O$ content of primary mantle melts

265   and/or increased pressure of magma storage (Asimow and Langmuir, 2003; Neave et al., 2019;

266   Thompson, 1987; Winpenny and Maclennan, 2011).

## 5.2   Olivine compositions

267

268   Olivine crystals in our Floreana lava and scoria samples show large variations in their forsterite

269   contents (Fo = 70–92, where Fo=(Mg/(Mg+Fe$^{2+}$) molar) with histograms showing a primary density

270   peak at Fo~85 (Fig. 5), more primitive than the olivine compositions in equilibrium with basaltic

271   glasses from Floreana ($K_D$=0.27-0.34, assuming a $Fe^{3+}/Fe_{tot}$ ratio of 0.15; Matzen et al., 2011;

272   Peterson et al., 2015; Roeder and Emslie, 1970).  Although there is no clear correlation between Fo

273   and Ca concentration in these crystals, the most forsteritic olivines (Fo>83) have extremely diverse Ca

274   contents (~250 to ~2600 ppm; Fig. 5), whereas crystals with lower Fo contents (<83) have

275   ubiquitously low Ca concentrations (<1500 ppm). All crystals with >1500 ppm Ca are classified as

276   Group 1 olivines. Crystals with <1500 ppm Ca, however, may be classified in any of the 5 olivine

277   groups, with the most evolved crystals (Fo ~70) the only to display the sieve-like rim texture of Group

278   4 olivines.

279   Floreana olivines separated from the lava and scoria samples also have a large range of Ni

280   concentrations (~700 to ~3200 ppm), consistent with crystallisation from a peridotite-derived melt

281   (Fig. 5; Gleeson and Gibson, 2019; Herzberg, 2011; Matzen et al., 2017b, 2017a). All olivine crystals

282   analysed in the wehrlite and dunite xenoliths have a narrow range of Fo contents (83-87) and,

283   crucially, have uniformly low Ca concentrations (<1000 ppm) and moderately high Ni contents

284   (~2000 ppm). Core-rim profiles were performed on a small number of the wehrlitic olivines, revealing

285   typically flat profiles in Fo, Ni, and Ca, with one profile displaying evidence for an increase in Ca in

286   the outer 10 – 20 μm (see Supplementary Information). The range in Ca contents of the Floreana

287   olivines contrasts with isolated olivine crystals from other Galápagos islands where Ca is typically

288   >1000 ppm (Gleeson and Gibson, 2019; Vidito et al., 2013). The Ca and Ni contents of our Floreana

289   olivines are inversely related at a set Fo content (Fig. 5B).

## 5.3 Clinopyroxene compositions

290

291 *5.3.1 Major elements*

292 The Floreana clinopyroxenes separated from the scoria and xenolith samples are augitic, and have a

293 relatively high Mg# (0.85–0.90; Mg# = Mg/(Mg+Fe$_t$) molar). The clinopyroxenes reach high Na

294 concentrations up to 1.58 wt% Na$_2$O (0.11 Na atoms per formula unit; Fig. 6A) and, correspondingly,

295 up to 10% of the jadeite component. In general, clinopyroxene separates from scoria samples display

296 a wide range of Na concentrations, although some xenolithic clinopyroxenes extend to higher Na

297 contents (Fig. 6A). The Floreana clinopyroxenes display a large range of Cr contents, ranging from

298 <0.05 wt% in the most evolved crystals to ~1.72 wt% in the more primitive crystals (Fig. 6B).

299 Clinopyroxene analyses are typically taken from the cores of crystals, but zoning was characterised by

300 a series of transects (and maps) on the xenolithic clinopyroxenes (see Supplementary Information).

301 Results indicate that the clinopyroxenes are unzoned with respect to most element (including MgO

302 and FeO), but some zoning is present in their Al$_2$O$_3$, TiO$_2$ and Cr$_2$O$_3$ contents. Specifically,

303 clinopyroxene crystal rims are typically characterised by elevated TiO$_2$, but lower Cr$_2$O$_3$ contents.

304 *5.3.2 Trace elements*

305 Our Floreana clinopyroxenes display a wide range of geochemical enrichment, with LREE to MREE

306 ratios varying from [La/Sm]$_n$ ~0.2 to [La/Sm]$_n$ ~3.1 (where n represents normalisation to the

307 primitive mantle composition of Sun and McDonough, 1989). Wehrlitic clinopyroxenes typically

308 have more enriched trace element ratios (such as [La/Sm]$_n$ or [Ce/Y]$_n$) than clinopyroxenes from the

309 scoria samples (Fig. 7). Furthermore, melt [La/Sm]$_n$ ratios calculated to be in equilibrium with

310 clinopyroxenes from the scoria and xenolith samples range from ~1 to ~15 (calculated using the major

311 element composition of the clinopyroxene at the location of LA-ICP-MS analysis and the elastic

312 strain model of Wood and Blundy (1997) at 1225°C and 700 MPa; Fig. 7B), significantly greater than

313 the range measured in Floreana whole-rock samples (~2–5, with a small number of outliers up to

314 ~7.5; Harpp et al. 2014a). Almost all of the xenolithic crystals, and a large proportion of the

315 clinopyroxenes separated from scoria deposits, are too enriched to be in equilibrium with the typical

316 composition of melts erupted on Floreana (Harpp et al.,2014a). In addition, there is a strong

317    correlation between the Na concentrations and highly/moderately incompatible trace element ratios of

318    the Floreana clinopyroxenes ($p<10^{-3}$), such that crystals with enriched trace element signatures

319    typically contain a high jadeite component (Fig. 7A). Finally, the Floreana clinopyroxenes all contain

320    Eu anomalies ($Eu^* = Eu_n/\sqrt{Sm_n \times Gd_n}$) within analytical uncertainty of 1, and Sr anomalies ($Sr^* =$

321    $Sr_n/\sqrt{Pr_n \times Nd_n}$) that have a similar range to that observed in the erupted basalts (Harpp et al.,

322    2014a).

# 6   MAGMA SYSTEM ARCHITECTURE BENEATH ISLA FLOREANA

## 6.1   Mush crystallisation and textural equilibration

326    Based on pyroxene trace element and radiogenic isotope ratios, Lyons et al. (2007) hypothesised that

327    gabbroic xenoliths in the Floreana lava and scoria deposits formed in an ancient (>1-1.5 Ma)

328    magmatic system, compositionally similar to those currently beneath Cerro Azul and Sierra Negra

329    volcanoes in the western Galápagos. In contrast, wehrlite xenoliths preserve isotopic ratios similar to

330    more recent subaerial lavas on Floreana, suggesting that they are fragments of the present-day

331    magmatic system (Lyons et al., 2007).

332    Our wehrlitic xenoliths preserve an original poikilitic igneous texture (clinopyroxene oikocrysts

333    surrounding olivine chadacrysts) and display no evidence for the metamorphic breakdown of

334    plagioclase (e.g. pseudomorphs or relict cores) as hypothesised by Lyons et al. (2007). We suggest

335    that the clinopyroxene crystals in our wehrlitic xenoliths grew within an olivine-dominated cumulate

336    mush (i.e. interstitial growth of clinopyroxene oikocrysts; Wager et al., 1960). If clinopyroxene

337    growth is principally within a crystal-rich (i.e. relatively viscous and immobile) mush zone, this could

338    explain its relatively low abundance in Floreana lava and scoria deposits (<5% of separated crystals).

339    In contrast, plagioclase triple junctions in the gabbroic xenoliths have ~120° dihedral angles (Fig. 3A),

340    indicating a high degree of textural equilibration (Holness et al., 2019, 2005). Plagioclase textural

341    equilibrium, along with the two-pyroxene phase assemblage, suggests that the gabbroic xenoliths

342  represent magmatic cumulates which were stored at high temperatures (>900°C) on long timescales

343  (Holness et al., 2006). These petrographic observations are consistent with the gabbroic nodules

344  sampling an ancient magmatic system beneath Floreana (>1 Ma; Lyons et al., 2007).

## 6.2   Mush disaggregation prior to eruption

### 6.2.1   Insights from olivine compositional heterogeneity

347  Olivine crystals separated from the Floreana lava and scoria samples have a wide range of zoning

348  patterns, morphologies, and compositions (Fig. 2). The five olivine groups identified in the Floreana

349  samples have distinct morphologies and zoning patterns (see Section 2 above), suggesting chemically

350  heterogeneous magma storage (Holness et al., 2019). In particular, the most evolved crystals (Group

351  4; Fo~70–75) are in equilibrium with melts that are more evolved than the Floreana erupted basalts

352  (likely basaltic andesites). This is consistent with a recent study which identified highly evolved

353  (andesitic - dacitic) magmas beneath basaltic volcanoes in the western Galápagos Archipelago (Stock

354  et al., 2020).

355  As Fe-Mg interdiffusion in olivine is geologically fast (Chakraborty, 2010; Costa et al., 2020),

356  preservation of forsterite zoning in the Floreana olivine crystals suggests that multiple magma batches

357  interacted on relatively short pre-eruptive timescales. In Group 5 olivines, for example, four

358  compositional zones are preserved over ~100-200 μm (Fig. 2E and F). Whilst we do not have enough

359  Group 5 olivine crystals to calculate statistically robust timescales of pre-eruptive magma interactions

360  using diffusion chronometry, complex forsterite zoning over ~100 μm is estimated to last <3 yrs at the

361  approximate temperature of basaltic magma storage (~1225°C; using diffusion coefficients from

362  Chakraborty, 2010). Therefore, we suggest that the range of crystal morphologies and major element

363  compositions displayed by the Floreana olivines in lava and scoria deposits indicates mixing of

364  chemically heterogeneous magma storage regions over relatively short timescales prior to eruption.

365  The minor element chemistry of the olivine crystals allows us to investigate the crystallinity of these

366  chemically diverse magma storage regions. Olivine crystals in our Floreana lava and scoria deposits

367  have an unusually low, and large range of Ca concentrations (~250–2600 ppm compared with ~1500-

368  3000 ppm in the eastern Galápagos; Gleeson and Gibson, 2019; Fig. 5A). The lower end of the range

369   in Ca concentrations measured in the Floreana lava and scoria deposits overlaps with those observed

370   in cumulate xenoliths (wehrlites) and thus are unlikely to represent mantle olivines (Thompson and

371   Gibson, 2000). Previous studies have shown that the Ca concentration of magmatic olivine is sensitive

372   to several parameters, including: (i) the major element composition of the co-existing melt phase

373   (Herzberg, 2011); (ii) the $H_2O$ content of the co-existing melt phase (Gavrilenko et al., 2016); and (iii)

374   the temperature of the system (Adams and Bishop, 1982; Köhler and Brey, 1990; Shejwalkar and

375   Coogan, 2013).

376   Variations in the Ca content of primary mantle melts are commonly hypothesised to result from the

377   presence of a lithologically heterogeneous mantle source (Herzberg, 2011; Sobolev et al., 2007;

378   Vidito et al., 2013). Specifically, melting of a pyroxene-rich mantle lithology is expected to produce

379   Ca-poor and Ni-rich melts which would go on to form Ca-poor, but Ni-rich, olivines. While the low

380   Ca contents of many of the Floreana olivines would therefore appear to indicate derivation from melts

381   of a pyroxenitic source lithology, the olivine Ni contents are relatively low (<3000 ppm), inconsistent

382   with this hypothesis (Fig. 5B; Gleeson and Gibson, 2019). As a result, if the low Ca contents of the

383   Floreana olivines represent equilibrium with low Ca melts, we require a different process to generate

384   these compositions. This process must reduce the Ca concentration of the melt phase (and co-existing

385   olivines), without simultaneously reducing the melt Mg# (as low Ca concentrations are observed

386   across the entire range of forsterite contents in the Floreana olivines; Fig. 5A).

387   Evidence for the origin of the low-Ca contents in the Floreana olivines from lava and scoria deposits

388   is present in the texture and composition of the wehrlitic xenoliths, which contain uniformly low-Ca

389   olivine crystals (<1000 ppm; Fig. 5A). The petrography of the wehrlitic xenoliths attests to

390   clinopyroxene growth within olivine-dominated mush regions. Clinopyroxene crystallisation within

391   this mush would extract CaO and MgO from the residual melt. However, in an olivine-rich mush, the

392   large reservoir of MgO contained within the cumulus olivine grains would buffer the residual melt at

393   a near-constant Mg# during clinopyroxene crystallisation (Meyer et al., 1989). In contrast, the CaO

394   concentration of the melt is not buffered and decreasing melt CaO contents, due to clinopyroxene

395   crystallisation, will cause the CaO concentration of cumulus olivine grains to decrease (as a result of

396     diffusive re-equilibration). Support for this interpretation comes from the anomalously low CaO

397     concentrations in the matrix glass of sample 17MMSG27.

398     Intercumulus clinopyroxene growth would also increase the $H_2O$ concentration of the residual melt

399     phase, decreasing the partition coefficient of Ca into olivine (Gavrilenko et al., 2016). Therefore,

400     variable amounts of clinopyroxene crystallisation within an olivine-dominated mush, and subsequent

401     disaggregation of this mush by an ascending melt, could explain the range of Ca contents measured in

402     the Floreana olivines derived from lava and scoria deposits. Specifically, we suggest that: (i)

403     forsteritic olivine crystals (Fo>83) with Ca contents >1500 ppm are consistent with those expected

404     from fractional crystallisation of mantle-derived melts in a liquid-rich magma storage region (Fig. 5A;

405     Gleeson and Gibson, 2019); (ii) olivines with Ca contents <1000 ppm, overlapping with the wehrlitic

406     olivines, reflect equilibrium with Ca-poor interstitial melts; and (iii) olivine crystals with intermediate

407     Ca concentrations (1000 – 1500 ppm) are sourced from regions where clinopyroxene growth is less

408     extensive, or ongoing at the time of eruption. Thus, the olivine crystal cargo of the Floreana magmas

409     is predominantly derived from crystal-rich domains with only a small number of olivine crystals

410     displaying compositions that are consistent with fractional crystallisation in liquid-rich storage regions

411     (Ca >1500 ppm; Gleeson and Gibson, 2019).

412     It is an important to note, however, that the partitioning of Ca between co-existing olivine and

413     clinopyroxene has been hypothesised to be sensitive to temperature (such that less Ca enters the

414     olivine structure at lower temperature; Shejwalkar and Coogan, 2013). As a result, the heterogeneity

415     in the Ca content of the Floreana olivines could instead represent disaggregation of xenolithic material

416     that has undergone variable amounts of cooling. Both hypotheses presented here can recreate the

417     range of Ca contents observed in the Floreana olivines and, as the majority of olivine analyses from

418     the Floreana lava and scoria deposits have low Ca concentrations (<1000 ppm) that overlap with those

419     in xenolithic nodules, indicate that a large proportion of the erupted crystal cargo derives from

420     disaggregated, highly crystalline magma storage regions.

421 *6.2.2    Insights from clinopyroxene major element compositions*

422 The compositions of clinopyroxene crystals from the Floreana scoria also overlap with those in our

423 xenolith samples, supporting the hypothesis that some of the erupted crystals are derived from

424 disaggregated sub-volcanic mush (Fig. 6). We used hierarchical cluster analysis to subdivide our 567

425 clinopyroxene major element analyses from the Floreana scoria and xenolith samples and determine

426 the proportion of material that is derived from each xenolith lithology in the erupted crystal cargo

427 (cluster analysis was performed in Python 3.8 using the scikit-learn package of Pedregosa et al. 2011).

428 We find that our clinopyroxene analyses form three distinct clusters (Fig. 8):

429 - Cluster 1 clinopyroxenes are predominantly from the wehrlite and dunite xenoliths and

430 include 90% of our analyses from these samples. 39% of clinopyroxenes analysed from the

431 scoria samples also fall into this cluster.

432 - Cluster 2 clinopyroxenes include all analyses from the gabbroic xenoliths, and ~10% of

433 analyses from crystals separated from the scoria samples.

434 - Cluster 3 clinopyroxenes are dominated by analyses of the scoria derived clinopyroxenes

435 (~50% of analyses from the scoria separates). However, 10% of analyses from the wehrlite

436 and dunite xenoliths also fall into this cluster.

437 Of our 248 clinopyroxenes analyses from the Floreana scoria, approximately half are classified as

438 Cluster 3 and thus have major element compositions that do not show a clear chemical affinity to

439 either the wehrlite/dunite or gabbroic cumulates. Therefore, these crystals may represent autocrysts

440 (defined here as crystals that are genetically related to primary mantle melts beneath Floreana, but are

441 not influenced by secondary cumulate processes) that grew in liquid-rich magma storage regions. The

442 remainder of clinopyroxene analyses from the scoria are either compositionally analogous to those in

443 the wehrlite and dunite xenoliths (Cluster 1; 39%) or the gabbroic xenoliths (Cluster 2; 11%); we

444 interpret these as representing disaggregated sub-volcanic mush or wall rock. The high proportion of

445 the clinopyroxene crystal cargo that is derived from highly crystalline storage regions beneath

446 Floreana is consistent with our interpretation of olivine minor element concentrations.

## 6.3 Reactive Porous Flow within a cumulate mush

447

448 Whilst the olivine and clinopyroxene major and minor element concentrations show that a large

449 proportion of the erupted crystal cargo is derived from highly crystalline magma storage regions,

450 clinopyroxene trace element concentrations (and zoning) reveal the magmatic processes that operate

451 within these crystal-rich domains. The trace element composition of melts in equilibrium with our

452 clinopyroxene crystals are calculated using the model of Wood and Blundy (1997). The results

453 indicate that many of our clinopyroxene analyses have incompatible trace element ratios (e.g.

454 $[Ce/Y]_n$) which are more enriched than any erupted basalt from Floreana (Harpp et al., 2014a). In fact,

455 nearly all clinopyroxene analyses from our xenolith samples, and ~50% of clinopyroxene analyses

456 from the scoria samples, record trace element disequilibrium with the typical composition of the

457 Floreana basalts (Fig. 7 and 9). Over-enriched trace element signatures are characteristic of Cluster 1

458 clinopyroxenes (i.e. chemical affinity to the wehrlitic or dunitic xenoliths), whereas crystals that are

459 near trace element equilibrium with Floreana basalts typically have Cluster 3 major element

460 compositions (i.e. the autocryst cluster).

461 Petrographic observations and olivine minor element data indicate that the Floreana sub-volcanic

462 system is characterised by clinopyroxene crystallisation within an olivine-dominated mush. If the

463 clinopyroxene grew from trapped melt within an olivine-dominated mush, progressive crystallisation

464 would increase the concentration of highly incompatible trace elements (e.g. Ba, La, Ce) relative to

465 less incompatible trace-elements (e.g. Sm, Y) in the residual melt. A simple fractional crystallisation

466 model indicates that ~70% crystallisation is required to generate melt $[Ce/Y]_n$ ratios that are in

467 equilibrium with enriched clinopyroxenes from the scoria samples and even greater extents of

468 crystallisation (~80%) would be required to generate the extremely high $[Ce/Y]_n$ ratios in some of the

469 xenolithic clinopyroxenes (Fig. 9).

470 Such extensive fractional crystallisation would be expected to result in the saturation and

471 crystallisation of plagioclase and other accessory phases (e.g. apatite, magnetite/ilmenite, quartz),

472 which are observed in more evolved xenoliths from Rabida island in the central Galápagos (Holness

473 et al., 2019). However, these phases are absent in the Floreana xenoliths, indicating that either

474    infiltration of melts from a highly enriched mantle source component or a different magmatic process

475    within the cumulate mush is responsible for generating the anomalous trace element signatures of the

476    Floreana clinopyroxenes. Infiltration of mantle-derived melts that are more enriched than anything

477    observed in erupted basalts on Floreana is considered unlikely owing to the overlapping isotopic

478    composition of xenoliths and erupted lavas  (Lyons et al., 2007), and the absence of a significant

479    correlation between trace element enrichment and radiogenic isotope compositions in the erupted

480    Floreana basalts (see Supplementary Information; Harpp et al., 2014a). As a result, a process other

481    than source heterogeneity must be responsible for generating the trace element variation in both the

482    Floreana xenoliths and erupted basalts.

483    One alternative mechanism that might be able to generate the observed trace element over-enrichment

484    is reactive porous flow. In this scenario, clinopyroxene is precipitated from ascending clinopyroxene-

485    saturated melts that continuously react with the existing, olivine-dominated crystal framework as the

486    system approaches equilibrium, and thus deviates from a simple fractional crystallisation trajectory

487    (Lissenberg and MacLeod, 2016). As a result, reactive porous flow in the olivine-dominated mush

488    beneath Floreana will likely result in substantial crystallisation of clinopyroxene, possibly at the

489    expense of pre-existing crystal phases, with little to no formation of olivine or saturation of minor

490    phases. Consequently, reactive porous flow can lead to enrichment of highly- to moderately-

491    incompatible trace elements in the resulting melt (Coogan et al., 2000; Gao et al., 2007; Lissenberg et

492    al., 2013; Lissenberg and MacLeod, 2016), and is consistent with the petrography of the Floreana

493    xenoliths. For example, major element maps of clinopyroxene crystals in the Floreana wehrlites show

494    that they are zoned, with Ti-rich rims (Fig. 10); equivalent zoning patterns have been attributed to

495    reactive porous flow in plutonic clinopyroxenes from the oceanic crust (e.g. Hess Deep; Lissenberg

496    and MacLeod, 2016). In addition, if pre-existing Cr-spinel was dissolved by the reacting melt, then

497    reactive porous flow could also explain the high Cr contents of clinopyroxene in our wehrlitic

498    xenoliths (Fig. 6; Lissenberg and MacLeod, 2016).

499    To test whether reactive porous flow of clinopyroxene saturated melts through an olivine-dominated

500    mush is consistent with the trace element compositions of melts calculated to be in equilibrium with

501    our Floreana clinopyroxenes, we use the zone refining model of Harris (1957; Fig. 9):

502
$$\frac{C_l}{C_l^o} = \frac{1}{D} - (\frac{1}{D} - 1)e^{-DI}$$

503                                                 (eq. 1)

504    where $D$ is the bulk partition coefficient; $C_l^o$ and $C_l$ are the initial and final concentration of that

505    element in the melt phase, respectively; and $I$ is the 'equivalent volumes of solid processed by the

506    liquid' (Lissenberg and MacLeod, 2016). The model assumes continuous reaction of the percolating

507    melt front with the existing crystal framework, and has previously been employed to investigate

508    geochemical signatures in oceanic gabbros (Lissenberg and MacLeod, 2016). Results indicate that

509    reactive porous flow can produce melts with trace element compositions that are comparable with

510    those in equilibrium with our Floreana clinopyroxenes (i.e. [Ce/Y]$_n$ ~8-13) using $I$ values that are

511    similar to those invoked in other magmatic settings worldwide (~2-5 compared to ~4-8 for the Hess

512    Deep; Lissenberg and MacLeod, 2016). Hence, reactive porous flow represents a realistic mechanism

513    for generating the geochemical diversity of Floreana clinopyroxenes, including the trace element

514    enriched crystals analysed in the wehrlitic nodules (Fig. 9).

515    In addition, detailed LA-ICP-MS transects of two clinopyroxene grains from the most enriched

516    wehrlitic xenolith analysed in this study (17MMSG02c) show clear trace element zoning (Fig. 11).

517    The core of the larger clinopyroxene crystal has low [Ce] and [Ce/Y]$_n$ contents that are approximately

518    in equilibrium with Floreana basalts (Harpp et al. 2014a; Fig. 11a), whereas the mantle and rim of the

519    crystal is characterised by increasing [Ce] and [Ce/Y]$_n$ contents. We interpret this as core

520    crystallisation from a melt with a trace element signature similar to that of erupted Floreana basalts

521    (Harpp et al., 2014a), followed by growth from a melt which became progressively enriched during

522    reactive porous flow (Fig. 11).

523    The mantle of the second, smaller xenolithic clinopyroxene shows a similar rim-ward increase in [Ce]

524    (interpreted as progressive melt enrichment during reactive porous flow). However, the [Ce] and

525    $[Ce/Y]_n$ values of the crystal core are too high to be in equilibrium with erupted Floreana basalts (Fig.

526    11C). This is consistent with our spot analyses of clinopyroxene cores in other crystals and samples.

527    The high [Ce] and $[Ce/Y]_n$ values measured in crystal cores cannot be explained by inward diffusion

528    of Ce, owing to significant differences in the diffusivities of Ce and Y and similar [Ce] and [Y]

529    zoning patterns in our two crystal transects (Fig. 11; Van Orman, 2001). Instead, we suggest that the

530    high apparent core [Ce] and $[Ce/Y]_n$ contents in many of the Floreana clinopyroxenes record

531    crystallisation from melts that had already undergone geochemical enrichment via reactive porous

532    flow. However, we cannot discount that our apparent clinopyroxene cores are fragments of larger

533    oikocrysts that have been broken during mush disaggregation or sample crushing and, as a result, do

534    not represent the true core compositions of each crystal.

535    Nevertheless, our clinopyroxene major and trace element data, as well as petrographic observations of

536    the wehrlitic xenoliths, provide substantial evidence that reactive porous flow is an important

537    mechanism of melt migration and melt differentiation in highly crystalline magma storage regions

538    beneath Floreana. Although reactive porous flow has been identified as an important process in MOR

539    gabbros, this is the first study to identify reactive porous flow in an ocean island setting.

540    ## 6.4   Petrographic estimates of magma storage pressures

541    Petrological and geophysical constraints on magma storage depths exist for several recently active

542    volcanoes in the western Galápagos Archipelago (Bagnardi et al., 2013; Case et al., 1973; Geist et al.,

543    1998; Stock et al., 2018; Vigouroux et al., 2008). However, in the absence of geophysical data (owing

544    to a paucity of recent eruptions), there are far fewer constraints on the structure of magma storage

545    regions in the eastern and south-eastern archipelago. To date, the only investigation of magma storage

546    depths beneath these volcanoes is by Geist et al. (1998), who undertook a visual comparison between

547    whole-rock lava compositions and the MORB olivine + plagioclase + augite + melt pseudoinvariant

548    point, parameterised by Grove et al. (1992). This approach is subject to substantial uncertainty, but the

549   authors suggest that the Floreana magmas consistently equilibrate at >5 kbar (typically >7 kbar) at a

550   depth >16 km, within the upper mantle.

551   We used three petrological barometers to provide improved constraints on magma storage depths

552   beneath Floreana. First, we applied the clinopyroxene-only barometer and thermometer of Putirka

553   (2008; equations 32b and 32d, respectively), in which pressure and temperature are solved iteratively

554   based solely on the clinopyroxene major element composition (primarily the jadeite component;

555   standard error of estimate [SEE] ±310 MPa). Second, we applied the clinopyroxene-melt barometer of

556   Neave and Putirka (2017; equation 1), which uses the composition of a co-existing melt phase and the

557   proportion of the Jadeite component in clinopyroxene to calculate the pressure of crystallisation (SEE

558   = ±140 MPa; pressure is solved iteratively with temperature using the clinopyroxene-melt

559   thermometer of Putirka, 2008; equation 33). Third, for the xenolithic nodules, we estimate the final

560   pressure and temperature of storage using the two-pyroxene thermobarometer of Putirka (2008;

561   equations 36 and 39; SEE = ±280 MPa).

562   Taken at face value, initial application of the clinopyroxene-only barometer to all clinopyroxene

563   analyses from the scoria and xenolith samples gives a range of pressure estimates between ~450 MPa

564   and ~1800 MPa. However, reactive porous flow has a substantial influence on the compositions of the

565   Cluster 1 (and Cluster 2) clinopyroxenes, which may influence the barometric results. Specifically,

566   crystals that show evidence for reactive porous flow also have elevated Na concentrations, leading to

567   an anomalously high jadeite component and thus calculated pressure. This likely originates from the

568   presence of unusual melt compositions that fall outside the calibration range of the clinopyroxene-

569   only barometer owing to reactive melt migration, which is supported by the unusual glass

570   compositions in sample 17MMSG27 (Fig. 4). Therefore, we filter our dataset to remove crystals that

571   show a chemical signature indicative of reactive porous flow and only use Cluster 3 clinopyroxenes

572   that have trace element compositions in equilibrium with the Floreana basalts (using the whole-rock

573   data from Harpp et al. 2014) in our barometric calculations (n=78). Barometric results from this

574   filtered dataset indicate that crystallisation beneath Floreana occurs at a pressure of 766 ±322 MPa

575    (2σ of calculated pressures), which equates to a depth of 25.2 ±9.9 km (using the crustal density

576    estimate of Putirka (1997) and a mantle density estimate of 3300 kg/m$^3$; Fig. 12).

577    Application of the Neave and Putirka (2017) clinopyroxene-melt barometer requires identification of

578    equilibrium clinopyroxene-liquid pairs. We achieve this using an automated melt-matching algorithm

579    (as in Winpenny and Maclennan, 2011, Neave and Putirka, 2017, Stock et al. 2018), with $K_D$(Fe-Mg),

580    diopside-hedenbergite, enstatite-ferrosillite and calcium Tschermak's equilibrium tests ($K_D$(Fe-Mg)

581    within ±0.03 other components within 2 SEE; Putirka, 1999, Putirka, 2008, Mollo et al., 2013). We

582    used the whole-rock data of Harpp et al. (2014a) and basaltic glass analyses from this study as

583    potential equilibrium liquids. Input crystal compositions were again filtered to remove analyses that

584    showed evidence of reactive porous flow (i.e. only Cluster 3 clinopyroxenes in trace element

585    equilibrium with the Floreana whole-rock were used). In total, 70 of the 78 input clinopyroxene

586    analyses returned at least one equilibrium match to either the basaltic glass or whole-rock

587    compositions. Where clinopyroxene compositions produced an equilibrium match with more than one

588    melt composition, an average melt composition was used in the barometric model. Results from this

589    barometer, coupled to the thermometer of Putirka (2008), indicate that magma crystallisation occurred

590    at 717 ±165 MPa (23.7 ±5.1 km) and 1224 ±33°C (Fig. 12).

591    Clinopyroxene-orthopyroxene thermobarometry records the final storage conditions of the cumulate

592    xenoliths, rather than the crystallisation conditions of clinopyroxene autocrysts (orthopyroxene is only

593    found as an intercumulus phase). Temperature and pressure estimates were only calculated from

594    orthopyroxene-clinopyroxene pairs in wehrlite and dunite xenoliths that passed the $K_D$(Fe-Mg)

595    equilibrium test of Putirka (2008; 1.09 ±0.14). Results suggest that the cumulates were stored at

596    ~975–1100°C and 600–900 MPa, with a mean storage pressure of 712 ±200 MPa (23.7 ±6.4 km; Fig.

597    12).

598    The depths of magma storage calculated from our three petrological barometers show an excellent

599    agreement within the model uncertainties. These new data provide robust evidence that magma

600    storage beneath Floreana occurs below the Moho (~16 km; Feighner and Richards, 1994), in the upper

601    mantle.

# 7   IMPLICATIONS FOR MAGMATIC PLUMBING SYSTEMS BENEATH LOW MELT FLUX OCEAN ISLAND VOLCANOES

Our new petrological and geochemical data show that magma storage beneath Floreana occurs in mush-dominated regions of the upper mantle (Fig. 13). Mineral chemistry (such as low olivine Ca concentrations and clinopyroxene major elements) reveal that a substantial portion of the erupted crystal cargo is derived from disaggregated mush and wall rock material which has been entrained into the ascending magmas. During ascent, magmas may entrain coherent nodules (xenoliths) as well as disaggregated mush (Fig. 13). Coherent nodules represent areas of the magmatic system beneath Floreana that have undergone cooling to temperatures <1100°C (compared to the clinopyroxene crystallisation temperatures of ~1225°C) and may represent material from the border of the active mush zone or older, almost completely solidified magma storage regions that are intersected during magma ascent (Fig. 13).

Petrographic observations and clinopyroxene trace element chemistry from both the xenolith and scoria samples reveal that clinopyroxene growth occurs via reactive porous flow in the mush-dominated areas beneath Floreana. Reactive porous flow causes distinct trace element enrichment in the percolating melt phase and crystallising clinopyroxene, which can explain the trace element disequilibrium between the erupted Floreana basalts and their clinopyroxene cargo. Nevertheless, the presence of some clinopyroxene crystals with major and trace element compositions in equilibrium with erupted Floreana basalts indicates that at least some crystallisation occurs in liquid-rich sub-volcanic storage regions, likely situated as localised melt pockets within the larger mush zone (Fig. 13).

Transport of melts modified by reactive porous flow into these melt pockets could impact the LREE enriched signature of the resultant hybridised melts. This could help to explain the unique, concave up REE signature of the Floreana basalts, which is not seen in other regions of the Galápagos Archipelago (Harpp et al., 2014a). However, it is important to note that similar trace element signatures are not observed in other, low melt-flux regions of the eastern Galapagos (such as San

629    Cristobal; Geist et al., 1986). As a result, we hypothesise that the unique REE pattern of the Floreana

630    basalts is primarily a source signature, likely associated with the highly radiogenic Pb and Sr isotope

631    signatures that characterise the Floreana basalts (Harpp et al., 2014a; Harpp and White, 2001).

632    Nevertheless, a few basalts on Floreana have trace element signatures that are far more enriched than

633    the majority of erupted basalts ($[La/Sm]_n$ up to 7.5), but are isotopically indistinguishable (Harpp et

634    al., 2014a); we therefore suggest that these basalts contain an anomalously large contribution of melts

635    that have undergone geochemical modification due to reactive porous flow.

636    Our results indicate substantial differences in the architecture of the magmatic systems beneath

637    Floreana and the frequently active shield volcanoes in the western Galápagos Archipelago. For

638    example, previous petrological and geophysical studies have identified that western Galápagos

639    magmatic systems are characterised by crustal magma storage, often with a large storage region in the

640    mid-to-lower crust (~7 km depth) and a smaller storage region at shallow levels, within the volcanic

641    edifice (~1 km depth; Geist et al. 1998; Bagnardi et al. 2013; Bagnardi and Hooper, 2018; Stock et al.,

642    2018; Fig 12). In contrast, our barometric data indicate that magmas beneath Floreana ascend directly

643    from the upper mantle and undergo no detectable crustal storage. In addition, although mush-rich

644    regions have been inferred beneath the western Galápagos shield volcanoes (based on whole-rock data

645    and the presence of gabbroic glomerocrysts; Chadwick et al., 2011; Geist et al., 1995, 2014; Stock et

646    al., 2018), magmatic differentiation appears to be driven by simple fractional crystallisation and

647    mixing of chemically diverse magmas (Geist et al., 1995; Naumann and Geist, 1999; Stock et al.,

648    2020).

649    One major factor that differentiates Floreana from shield volcanoes in the western archipelago is the

650    flux of magma into the lithosphere, which is evidenced by the large variations in the volumetric

651    eruption rate on Floreana and the western shields (Harpp et al., 2014a; Harpp and Geist, 2018; Kurz et

652    al., 2014). Hence, we suggest that the greater pressure of magma storage and prevalence of reactive

653    porous flow beneath Floreana, relative to volcanoes in the western archipelago, are related to the

654    substantially lower flux of magma into the lithosphere from the underlying mantle source (and thus

655    the thermal structure of the lithosphere). For example, the magma flux entering the lithosphere

656    beneath Wolf volcano (northern Isabela) has been substantially greater than that beneath Floreana for

657    several 100,000s of years (Geist et al., 2005). The high magma flux beneath Wolf maintains the

658    average temperature of the mid-to-lower crust at ~1125°C ($\Delta$T ~22 °C), with only small-scale thermal

659    and compositional heterogeneities present in the sub-volcanic plumbing system (Geist et al., 2014,

660    2005; Stock et al., 2020, 2018). In contrast, the flux of magma entering the magmatic system beneath

661    Floreana is much lower and the temperature of the mid-crust is likely to be significantly cooler than

662    the lowest temperature recorded by the Floreana xenoliths (that is, <<900°C; Fig. 12). As the flux of

663    magma (and heat) from the mantle is insufficient to maintain an elevated crustal geotherm beneath

664    Floreana, magmas that stall in the crust are likely to rapidly crystallise, increase their viscosity, and

665    become uneruptable. Therefore, eruptions must be fed by melts ascending from much deeper storage

666    regions (~700-750 MPa) where melts can persist over long time periods.

667    Our results have global implications for the architecture and dynamics of magma storage regions

668    beneath ocean island volcanoes worldwide. The observed difference in magma storage depths beneath

669    high and low melt flux volcanic systems in the Galapagos Archipelago is mirrored in a global

670    compilation of barometric data from ocean island volcanoes (Famin et al., 2009; Geist et al., 1998;

671    Hammer et al., 2016; Hartley et al., 2018; Klügel et al., 2015; Poland et al., 2015; Stock et al., 2018;

672    Zanon et al., 2020; Zanon and Pimentel, 2015). Using the average repose period between eruptions at

673    a particular basaltic volcanic centre as a proxy for the flux of magma entering the lithosphere from the

674    underlying mantle (Global Volcanism Program, 2013), Figure 14 shows that the most frequently

675    active volcanic centres (such as, Kīlauea, Hawai'i, and Piton de la Fournaise, Réunion) are

676    characterised by persistent magma storage in the mid to upper crust (Famin et al., 2009; Poland et al.,

677    2015). In contrast, less active centres located above low buoyancy flux plumes (e.g. El Hierro, Canary

678    Islands) and/or peripheral to the main zone of plume upwelling (e.g. Haleakalā, Hawai'i) are

679    characterised by longer repose periods and correspondingly greater magma storage pressures

680    (Hammer et al., 2016; Klügel et al., 2015; Zanon and Pimentel, 2015). In fact, although secondary

681    crustal magma staging can occur (Klügel et al., 2015), the main zone of magma storage beneath ocean

682    island volcanoes with repose periods >50 years is typically in the lithospheric mantle, below the base

683 of the crust (Longpre et al., 2014; Taracsák et al., 2019; Zanon et al., 2020). Hence, we speculate that

684 the flux of magma from the underlying mantle source has a first-order control on the depth of magma

685 storage beneath ocean island volcanoes and, correspondingly, high-pressure magma storage – as

686 observed beneath Floreana – is characteristic of low melt flux ocean island volcanoes globally.

687 # 8  CONCLUSIONS

688 Petrographic and geochemical analyses of lava, scoria and xenolith samples from Floreana in the

689 south-eastern Galápagos Archipelago provide new insights into the architecture and dynamics of

690 magma storage beneath low melt flux ocean island volcanoes. Comparison of olivine and

691 clinopyroxene major, minor and trace element contents between our different sample types reveal that

692 a substantial portion of the erupted crystal cargo is derived from mush-dominated magma storage

693 regions beneath Floreana. Mineral textures, highly enriched clinopyroxene trace element signatures

694 and trace element zoning in the xenoliths reveals that reactive porous flow is an important process of

695 chemical differentiation and melt transport within these mush-dominated regions. Mixing between

696 melts that have been geochemically enriched by reactive porous flow and those in overlying liquid-

697 rich storage regions could contribute to the anomalous, concave-up REE signature of the Floreana

698 basalts, which is absent in other parts of the Galápagos Archipelago where reactive porous flow has

699 not been identified.

700 Application of independent petrological barometers to crystals in Floreana scoria and xenolith

701 samples indicates that magmas are stored in the upper mantle ($\sim$23.7 $\pm$5.1 km). Floreana is in a distal

702 location to the Galápagos plume where the melt flux entering the lithosphere is low; the depth of

703 magma storage beneath Floreana contrasts with more proximal, higher melt flux volcanoes in the

704 western archipelago where magmas are stored in the crust (Geist et al., 1998; Stock et al., 2018).

705 Comparing our new data with ocean island volcanoes globally (e.g. Hawai'i, Iceland and the Canary

706 Islands) reveals that the Galápagos is not unique and that magma storage is ubiquitously shallower in

707 proximal magmatic systems above high buoyancy flux plumes than in off-axis systems, or above low

708 buoyancy flux plumes. We therefore suggest that the flux of mantle-derived magma entering the

709    lithosphere imparts a first-order control on the depth of magma storage beneath ocean island

710    volcanoes.

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1029  **FIGURES**
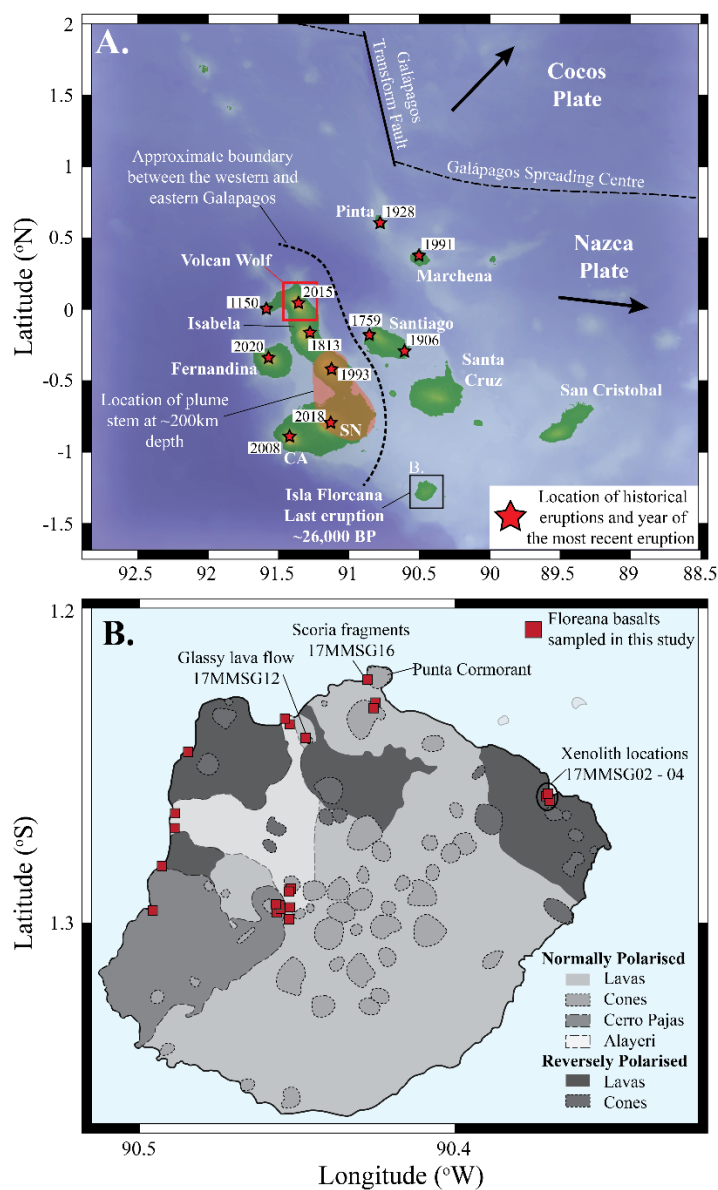


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1031  **Figure 1 - A.** Regional map of the Galápagos Archipelago highlighting the location of Isla Floreana,

1032  Cerro Azul (CA), Sierra Negra (SN) and Wolf volcanoes. Dates show the most recent eruptions at

1033  historically active volcanic centres. Black arrows show the direction of plate motion for the Nazca and

1034  Cocos tectonic plates. **B.** Geological map of Floreana adapted from Harpp et al. (2014a). Dashed lines

1035  delineate monogenetic scoria cones. Normally and reversely polarised lava flows are shown along

1036  with the largest (Cerro Pajas) and most recent (Alayeri; ~26,000 years) eruptions on the island.
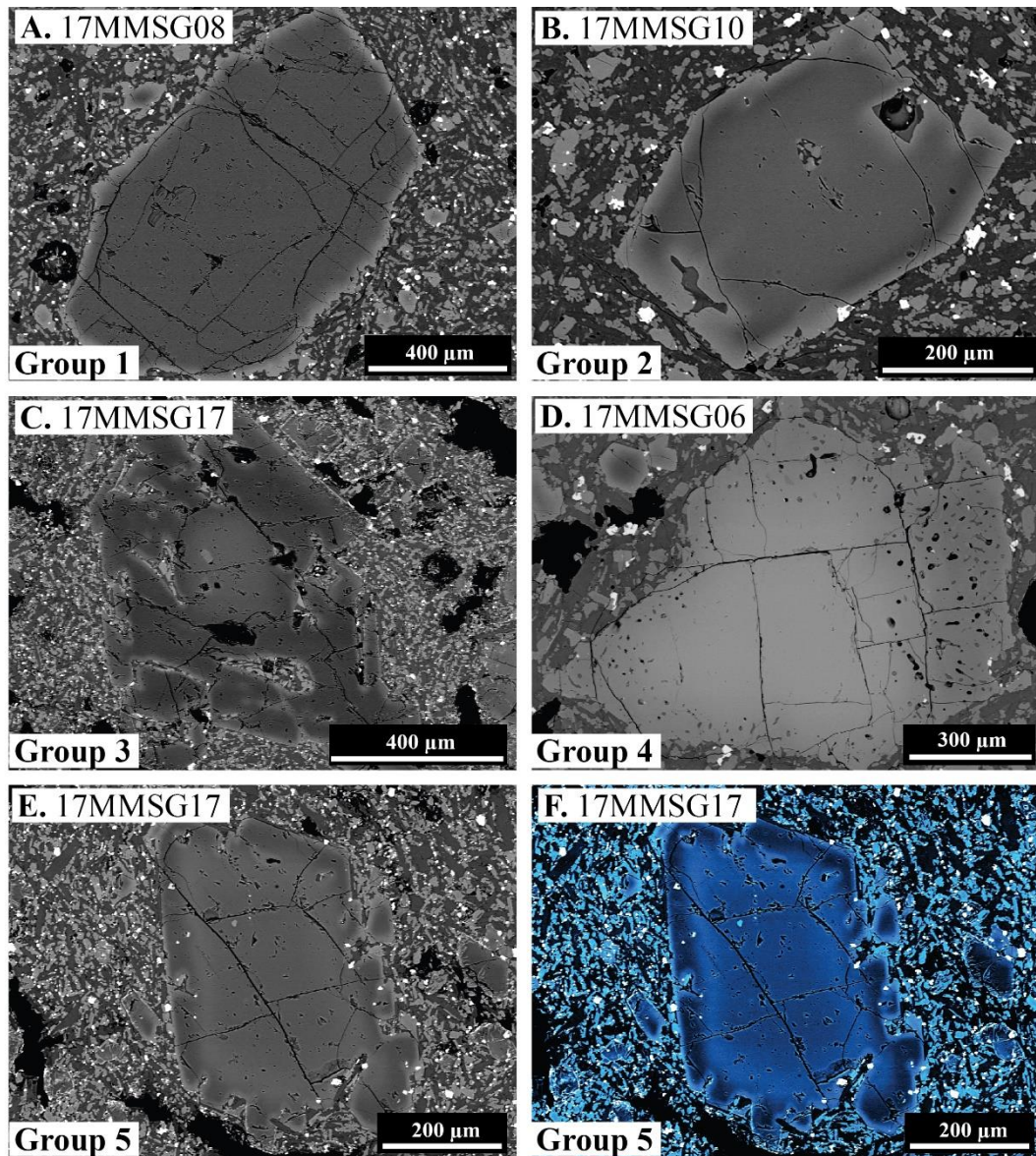
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**Figure 2 –** Backscatter Electron images. **A.** Group 1 olivines – euhedral to subhedral crystal

morphologies with large, unzoned, crystal cores and narrow, normally-zoned rims. **B.** Group 2

olivines – subhedral to euhedral crystals with clear, reverse-zoning profiles. **C.** Group 3 olivines –

skeletal crystals with high forsterite overgrowths on low forsterite cores. **D.** Group 4 olivines –

anhedral crystals with sieved textured, reverse zoned rims. **E.** (greyscale) and **F.** (false colour) Group

5 olivines – crystals preserve at least 4 composition zones over ~100-200 µm. False colour image (**F.**)

is used to highlight the compositional zoning of the Group 5 olivine, with the intensity of the blue

colour associated with the Fo composition of the crystal (darker = higher Fo).
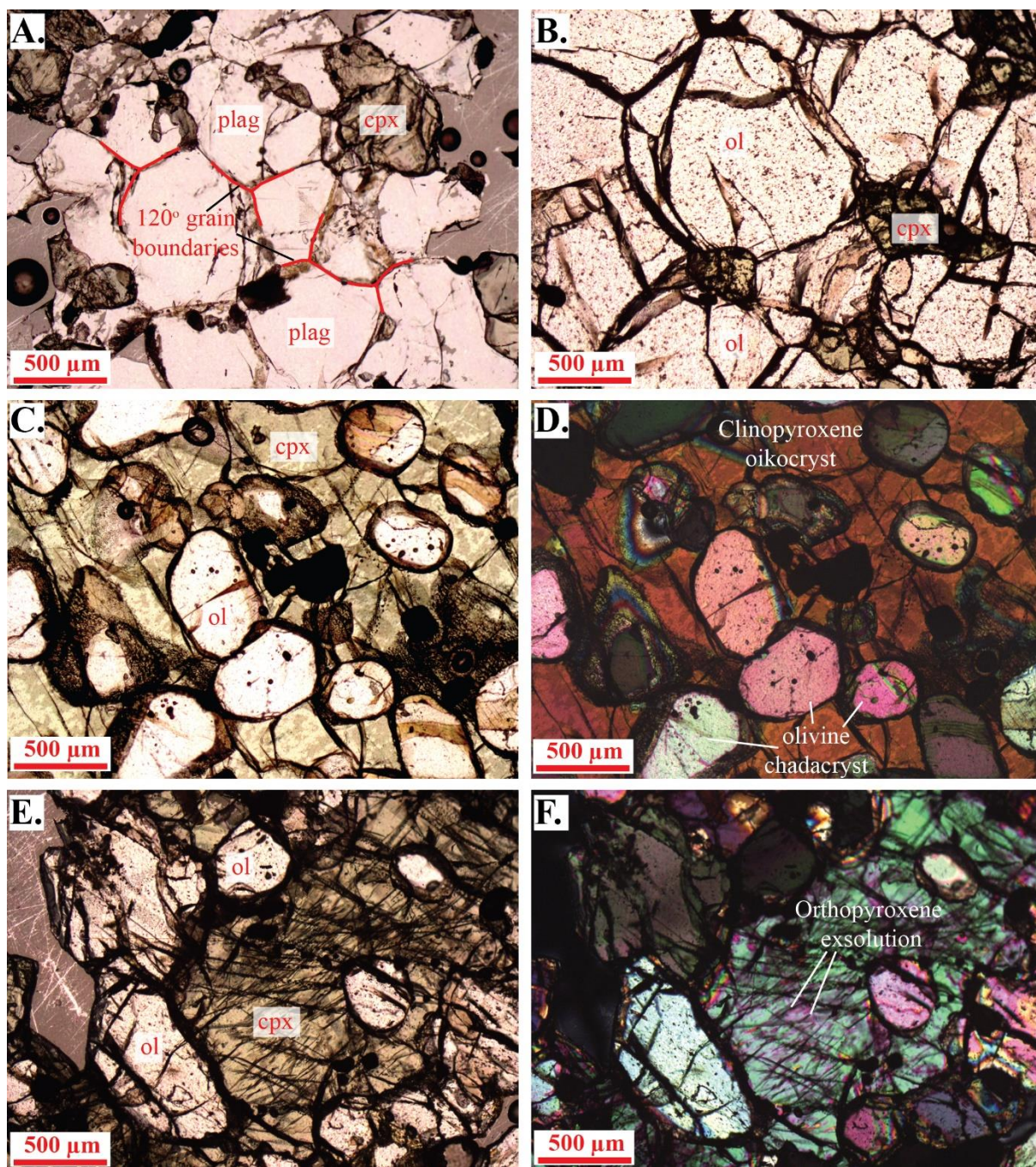
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**Figure 3 -** Plane Polarised Light (**A. – C.** and **E.**) and Crossed Polarised Light (**D., F.**) images of

Floreana xenoliths. **A.** – gabbroic xenolith (17MMSG04b), highlighting near 120° grain boundaries at

monomineralic plagioclase (plag) triple junctions. **B.** – dunitic xenolith (17MMSG04c) with

intercumulus clinopyroxene (cpx). **C.** and **D.** – wehrlitic xenolith (17MMSG02c) showing a large

clinopyroxene oikocryst surrounding olivine (ol) chadacrysts. **E.** and **F.** – wehrlitic xenolith (sample

17MMSG03a) showing olivine chadacrysts within a clinopyroxene oikocryst. Orthopyroxene

exsolution lamallae are visible within the clinopyroxene.
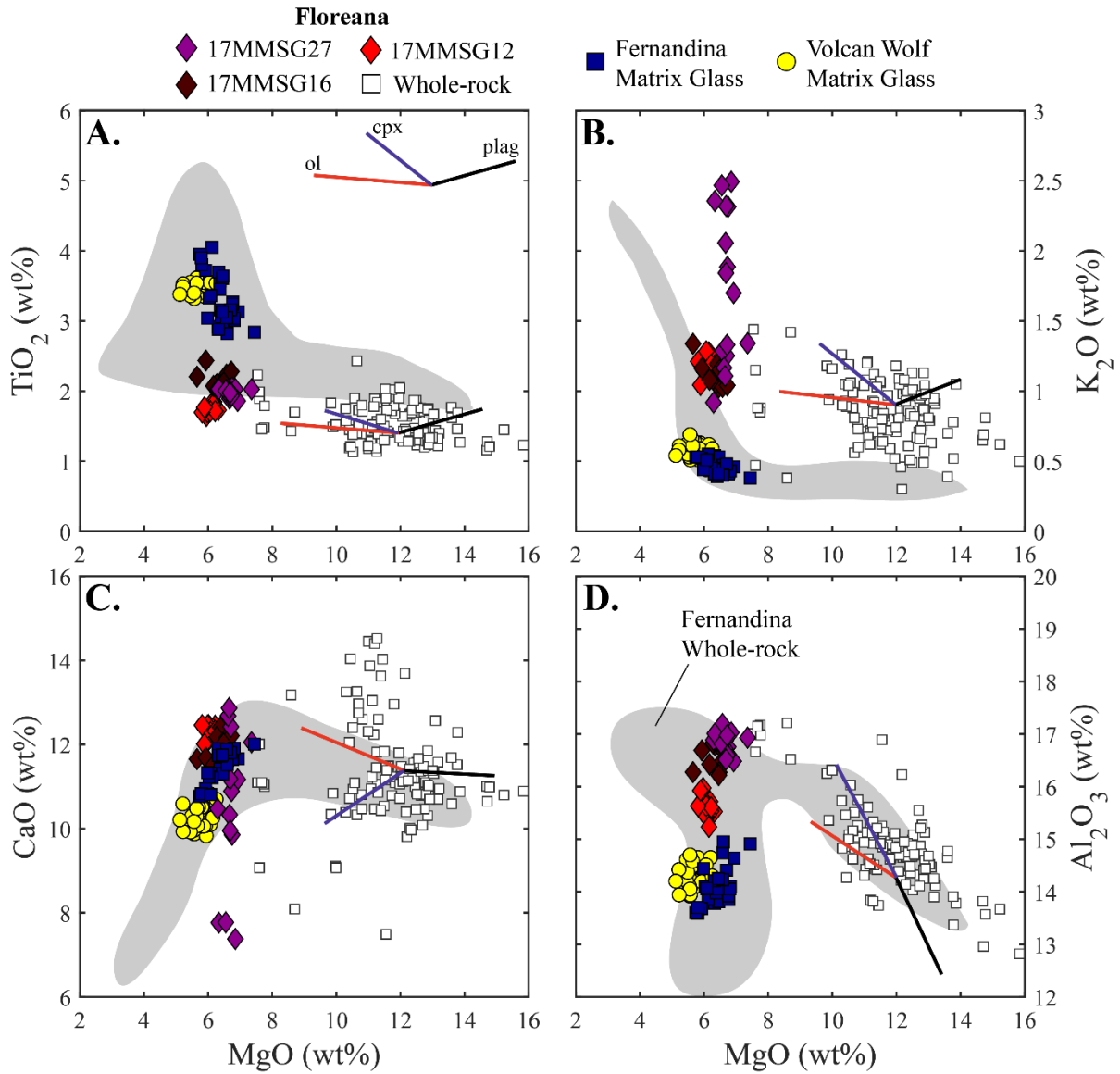
1054

**Figure 4** – Major element compositions of matrix glasses (this study) and whole-rocks (Harpp et al.,

2014a) from Floreana, as well as glasses from Fernandina (Peterson et al., 2017) and Wolf volcano

(Stock et al., 2018) in the western Galápagos Archipelago. Lines show trajectories of liquid

compositional evolution for olivine (ol; red), clinopyroxene (cpx; blue) and plagioclase (plag; black)

crystallisation. The grey field shows whole-rock data from Isla Fernandina in the western Galápagos

(Allan and Simkin, 2000; Geist et al., 2006). The $2\sigma$ precision of our matrix glass analyses is smaller

than the symbol size.
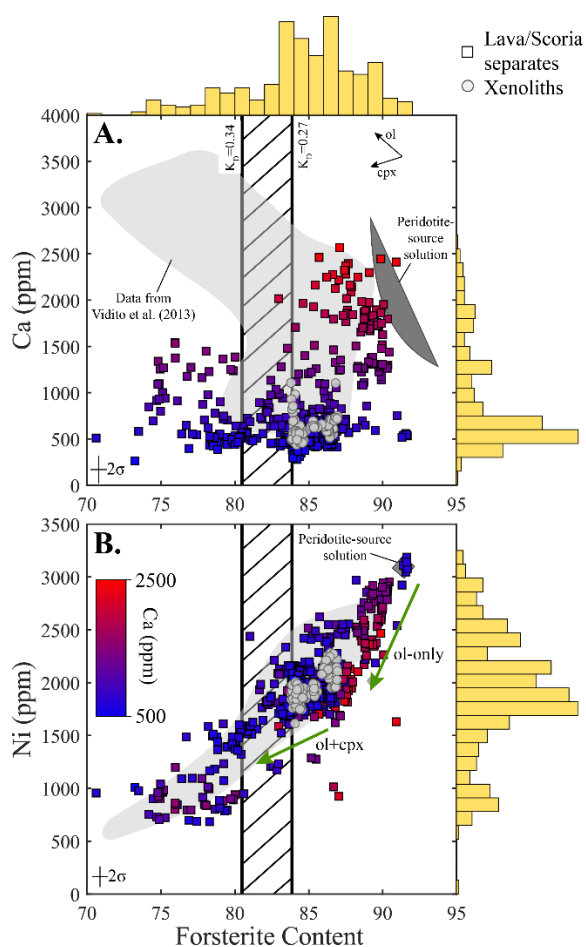
1062

**Figure 5 –** Major and minor element compositions of olivine crystals from the Isla Floreana basalts.

**A**. Fo vs. Ca and **B** Fo vs. Ni in Galápagos olivine crystals displaying analyses from our lava/scoria

separates and xenolith samples, as well as a compilation of available olivine data from Floreana

(Vidito et al. 2013). Our lava/scoria analyses are coloured according to their Ca concentration. The

histograms above and to the right of the plots show the data distributions (excluding *in situ* analyses

of xenolithic olivines). Peridotite source solutions are taken from Herzberg (2011) and Matzen et al.

(2017a). Black arrows in **A.** show the trajectory of crystal compositional evolution during olivine (ol)

and clinopyroxene (cpx) crystallisation (taken from Gleeson and Gibson, 2019). The green lines in **B.**

show the trajectories of crystal compositional evolution during olivine only and  olivine +

clinopyroxene fractional crystallisation (from Gleeson and Gibson, 2019). The vertical black lines

show the forsterite compositions of crystals calculated to be in equilibrium with the matrix glass

composition of tephra sample 17MMSG16 ($K_d$ = 0.27 – 0.34 after Matzen et al. 2011, and Roeder and

Emslie 1970).

**Floreana Clinopyroxenes**
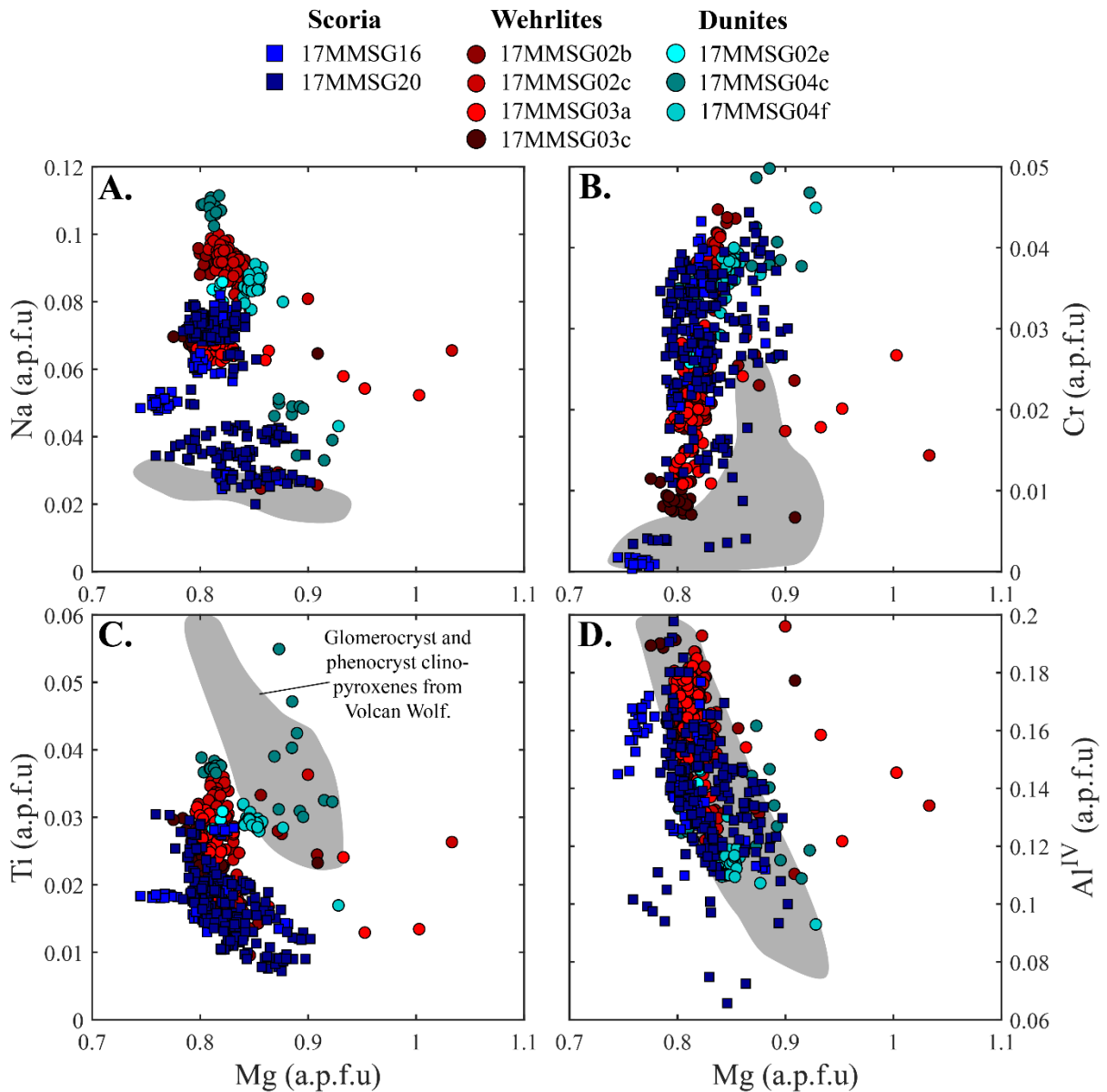
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1077 **Figure 6** – Major element composition of Floreana clinopyroxenes from our scoria, wehrlite and

1078 dunite samples. Data is shown as atoms per formula unit (a.p.f.u) on the basis of 6 oxygens. The grey

1079 field shows the compositions of clinopyroxenes from Wolf volcano in the western Galápagos

1080 Archipelago (from Stock et al. 2018). The 2σ precision of our clinopyroxene analyses is smaller than

1081 the symbol size.
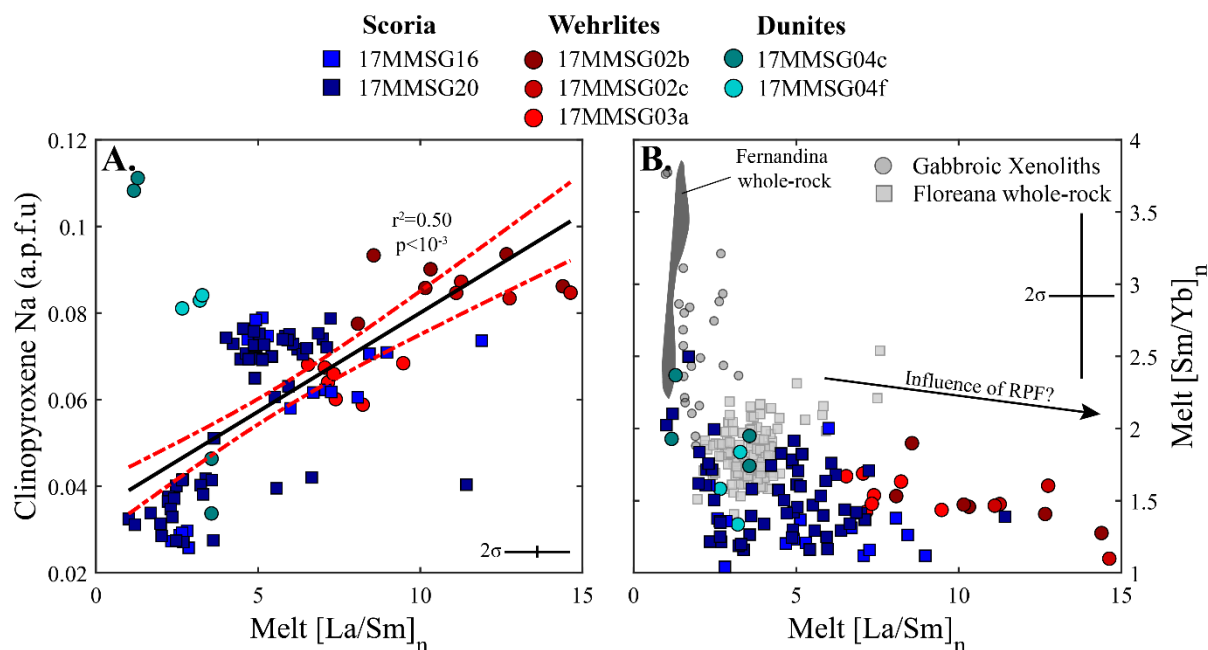
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**Figure 7** – Trace element composition of the Floreana clinopyroxenes and their relation to major

element systematics. **A.** $[La/Sm]_n$ vs. Na in clinopyroxenes from our scoria samples and wehrlite and

dunite xenoliths ($[La/Sm]_n$ is shown as the composition of melt in equilibrium with each

clinopyroxene). The black line shows a regression through the data ($r^2 = 0.50$) and the red dashed

lines show the 95% confidence limits on the regression. The correlation indicates that trace element

enrichment in the Floreana clinopyroxenes is associated with anomalously high Na contents. **B.**

$[La/Sm]_n$ vs. $[Sm/Yb]_n$ of melts calculated to be in equilibrium with our Floreana clinopyroxenes

using the model of Wood and Blundy (1997). The black arrow shows the approximate trend of crystal

compositional evolution hypothesised to occur as a result of clinopyroxene crystallisation during

reactive porous flow within the cumulate mush (RPF). The grey field shows whole-rock compositions

from Fernandina (Geist et al., 2006; White et al., 1993). **B.** additionally shows the whole-rock

compositions of erupted Floreana lavas (Harpp et al., 2014a) and analyses of the gabbroic xenoliths

from Floreana (this study). Error bars show the fully propagated $2\sigma$ precision of our analyses.
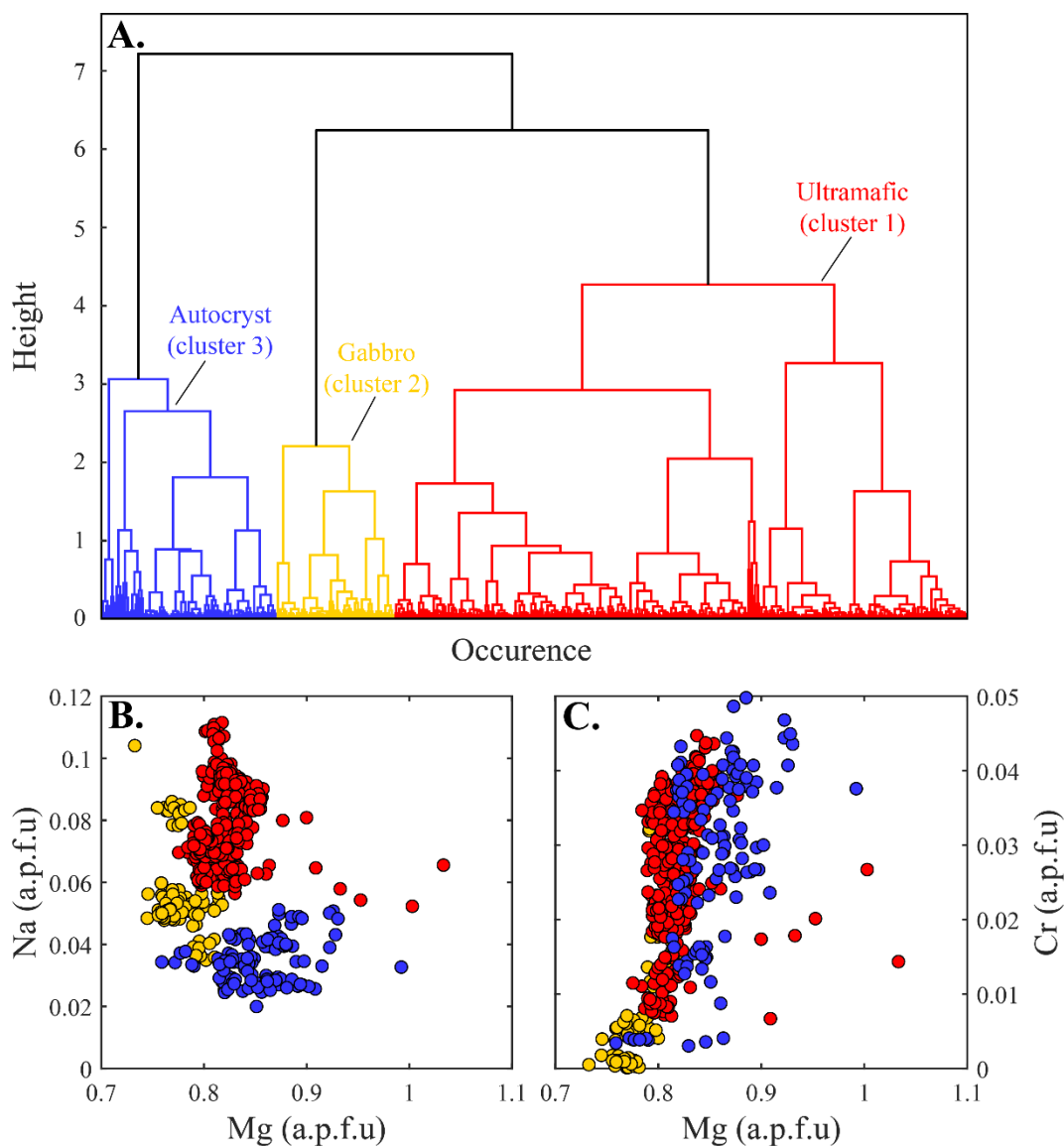
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Figure 8 – A. Hierarchical cluster analysis of our clinopyroxene major element analyses. Cluster

analysis was performed using Ward's method, which is built into the scikit-learn package in Python

(Pedregosa et al., 2011; Ward, 1963). Height above the x-axis is a measure of the distance of

separation of two clusters (i.e., the higher the join the more chemically distinct two clusters are).

Colours show the high-level division of crystal compositions into three clusters: Cluster 1 is

predominantly comprised of crystals from wehrlite and dunite xenoliths (red), Cluster 2 is composed

of crystals from gabbroic xenoliths (yellow) and Cluster 3 (blue) is dominated by crystals separated

from scoria samples. B. Na vs Mg and C. Cr vs Mg in our clinopyroxene analyses from the scoria and

xenoliths, coloured by their cluster.
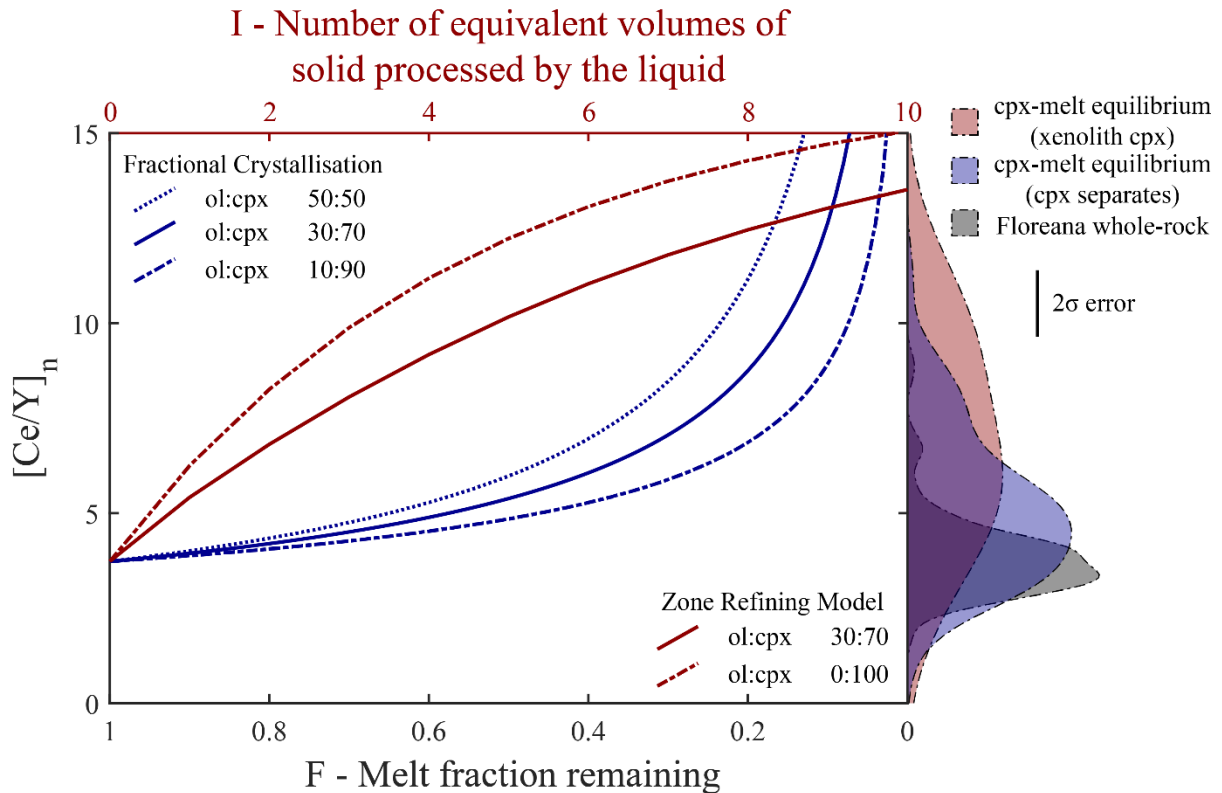
**Figure 9** – Models showing the evolution of melt $[Ce/Y]_n$ ratios as a function of the melt fraction remaining (F) during fractional crystallisation and as a function of the number of equivalent volumes of solid processed by the liquid (I) during reactive porous flow (red; using the zone-refining model of Harris, 1957). Line types show how models vary with different ratios of crystallising olivine (ol) and clinopyroxene (cpx). The kernel density plots show the $[Ce/Y]_n$ density distributions of liquids calculated to be in equilibrium with clinopyroxene crystals in our wehrlite and dunite xenoliths (red), scoria samples (blue; $2\sigma$ error for the clinopyroxene analyses is shown by the black bar), and whole-rock analyses of erupted Floreana basalts (grey; Harpp et al., 2014a). Clinopyroxene partition coefficients were calculated using the method of Wood and Blundy (1997), the mean major element composition of the Cluster 3 clinopyroxenes from this study, a temperature of 1225°C, and a pressure of 700 MPa. Initial Ce and Y concentrations for both models were taken as the mean values from the whole-rock dataset of Harpp et al. (2014a). Calculation increments are 0.01 in F and 0.1 in I.
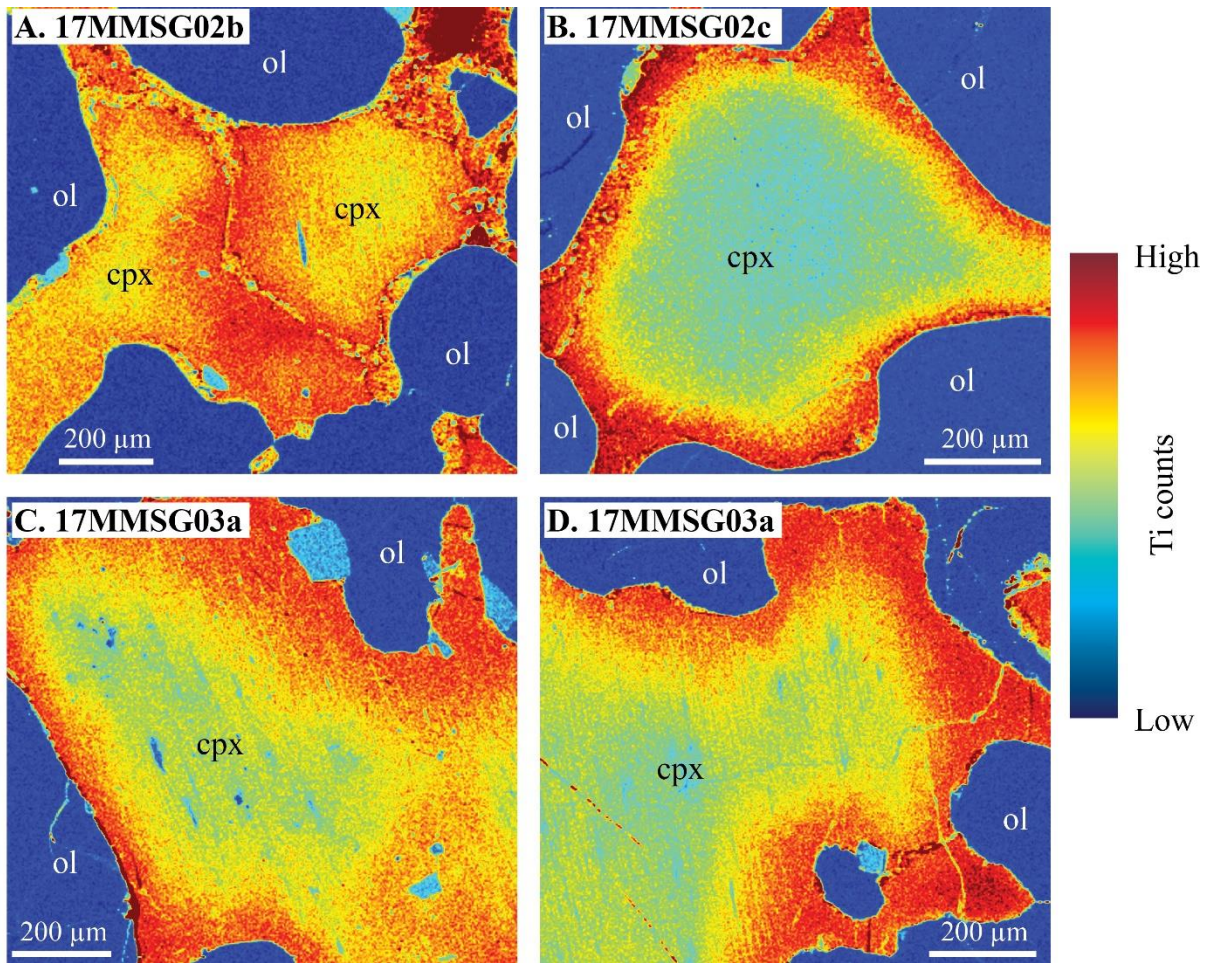
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1120 **Figure 10** – WDS maps of Ti counts in key clinopyroxene crystals from the Floreana wehrlite

1121 xenoliths. These maps display clear zoning in the xenolithic clinopyroxenes with Ti-poor cores and

1122 Ti-rich rims. EPMA transects across some of the wehrlitic clinopyroxenes indicates that the Ti

1123 concentrations in these crystals may vary from ~0.6 wt% to ~1.3 wt% $TiO_2$ (see Supplementary

1124 Information). The dark blue regions surrounding clinopyroxene grains are olivine crystals.
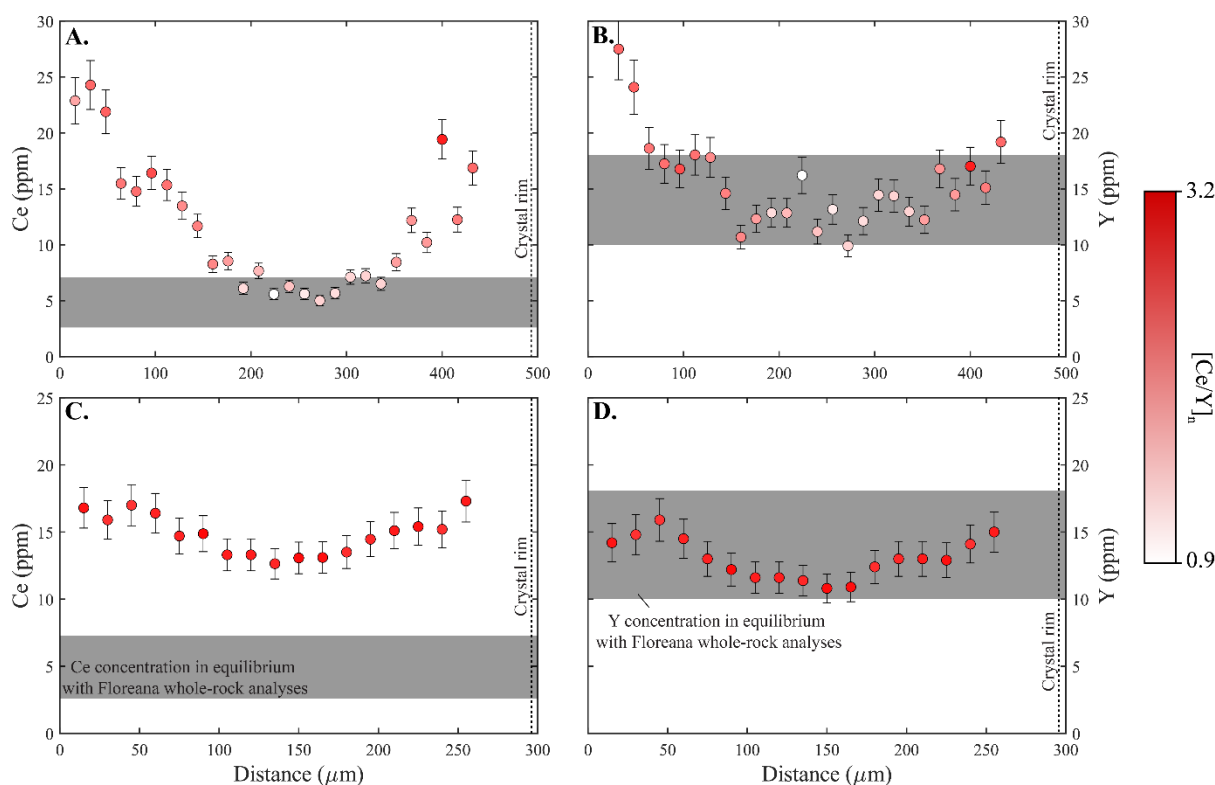
**Figure 11** – LA-ICP-MS transects showing Ce (**A, C**) and Y (**B, D**) zoning across two clinopyroxene grains in a wehrlitic xenolith (17MMSG02c). **A.** and **B.** show a transect across the core of a large clinopyroxene oikocryst. **C.** and **D.** show a transect across the apparent core of a smaller clinopyroxene oikocryst. Points are coloured according to their $[Ce/Y]_n$ ratio (see colour scale). The grey bars show the crystal compositions calculated to be in equilibrium with whole-rock analyses of erupted Floreana basalts (Harpp et al., 2014a).
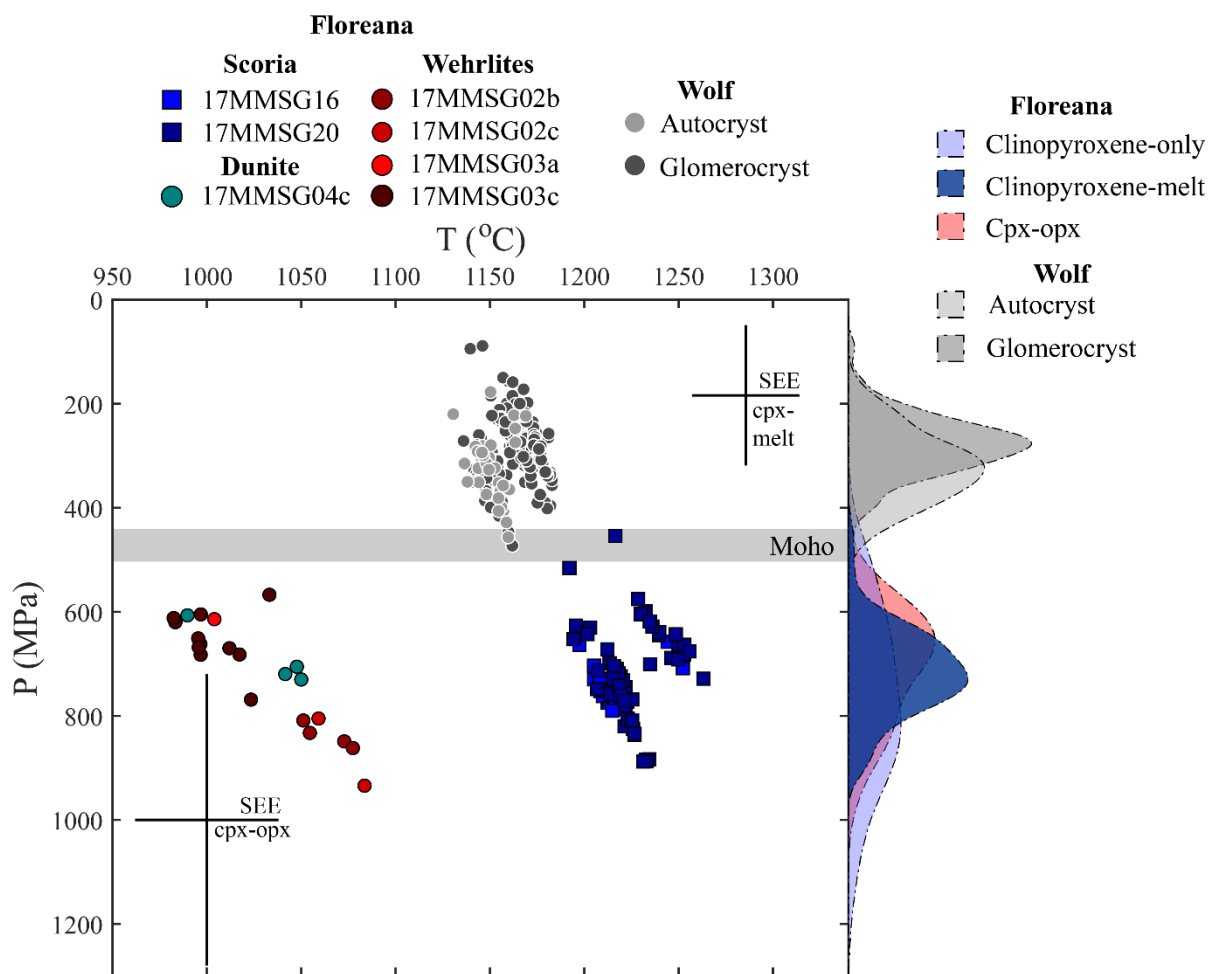
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**Figure 12** – Petrological thermobarometry results. The kernel density plots to the right show the density distributions of barometric results from different models (light blue – clinopyroxene-only, Putirka (2008); dark blue – clinopyroxene-melt, Neave and Putirka (2017); red – clinopyroxene-orthopyroxene Putirka (2008)). The grey bar shows the Moho depth beneath Fernandina (from Feighner and Richards, 1994) and the grey points and kernel density estimates show clinopyroxene-melt thermobarometric results for autocrysts and glomerocrysts from Wolf volcano (from Stock et al., 2018). The Standard Estimated Error (SEE) of the clinopyroxene-melt and orthopyroxene-clinopyroxene thermobarometers are given.
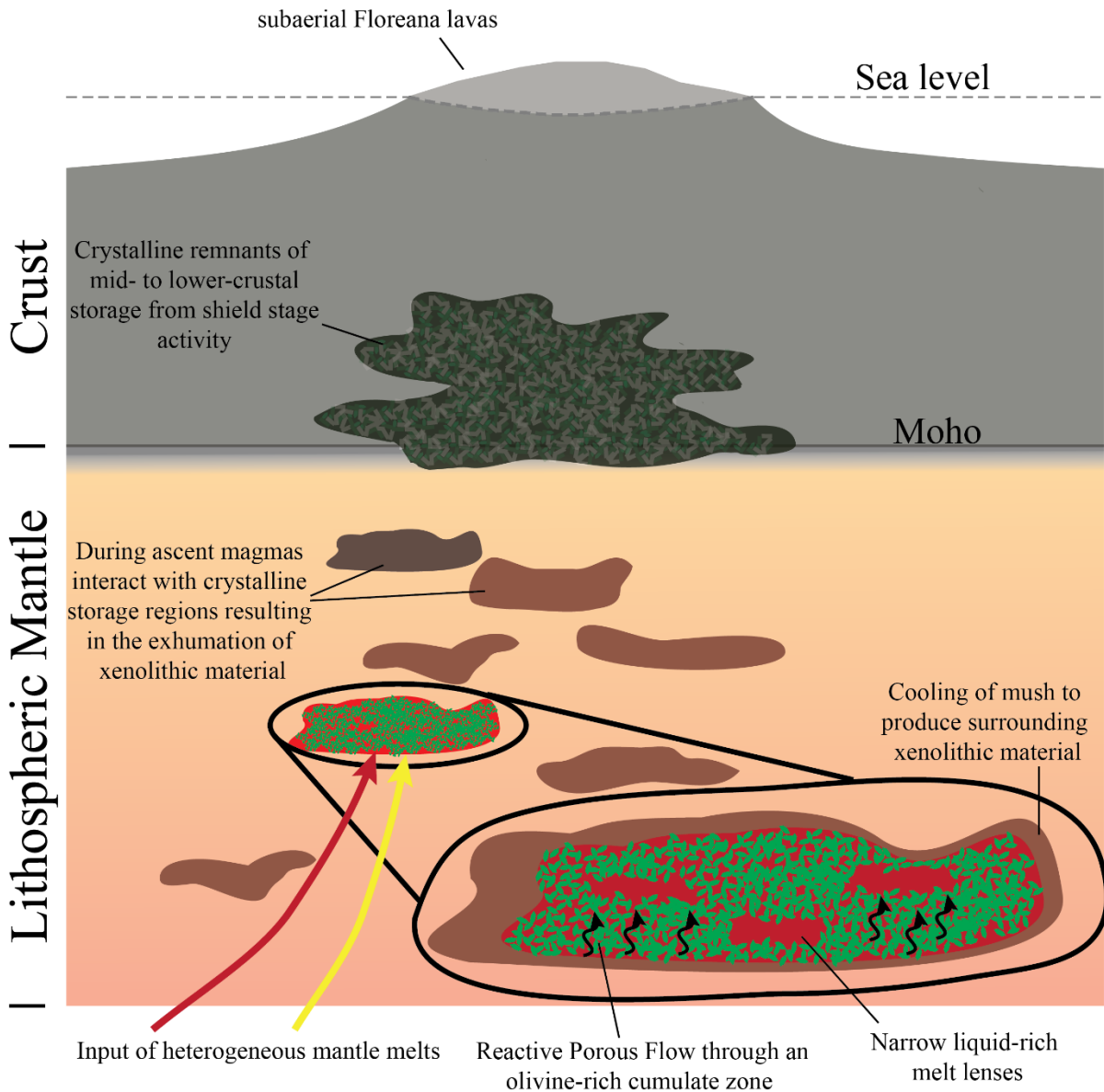
**Figure 13** – Schematic illustration of the magma plumbing system beneath Floreana. No magma storage is identified within the crust. Instead, our barometric results indicate that Floreana magmas ascend directly from the upper mantle, where they are stored at a depth of ~23.7 ±5.1 km. Floreana magma storage regions are dominated by crystal-rich domains (i.e. mush). Reactive porous flow is identified as an important mechanism of melt migration and magma differentiation in the crystal-rich storage regions, although our results shows that some crystallisation occurs within liquid-rich domains.
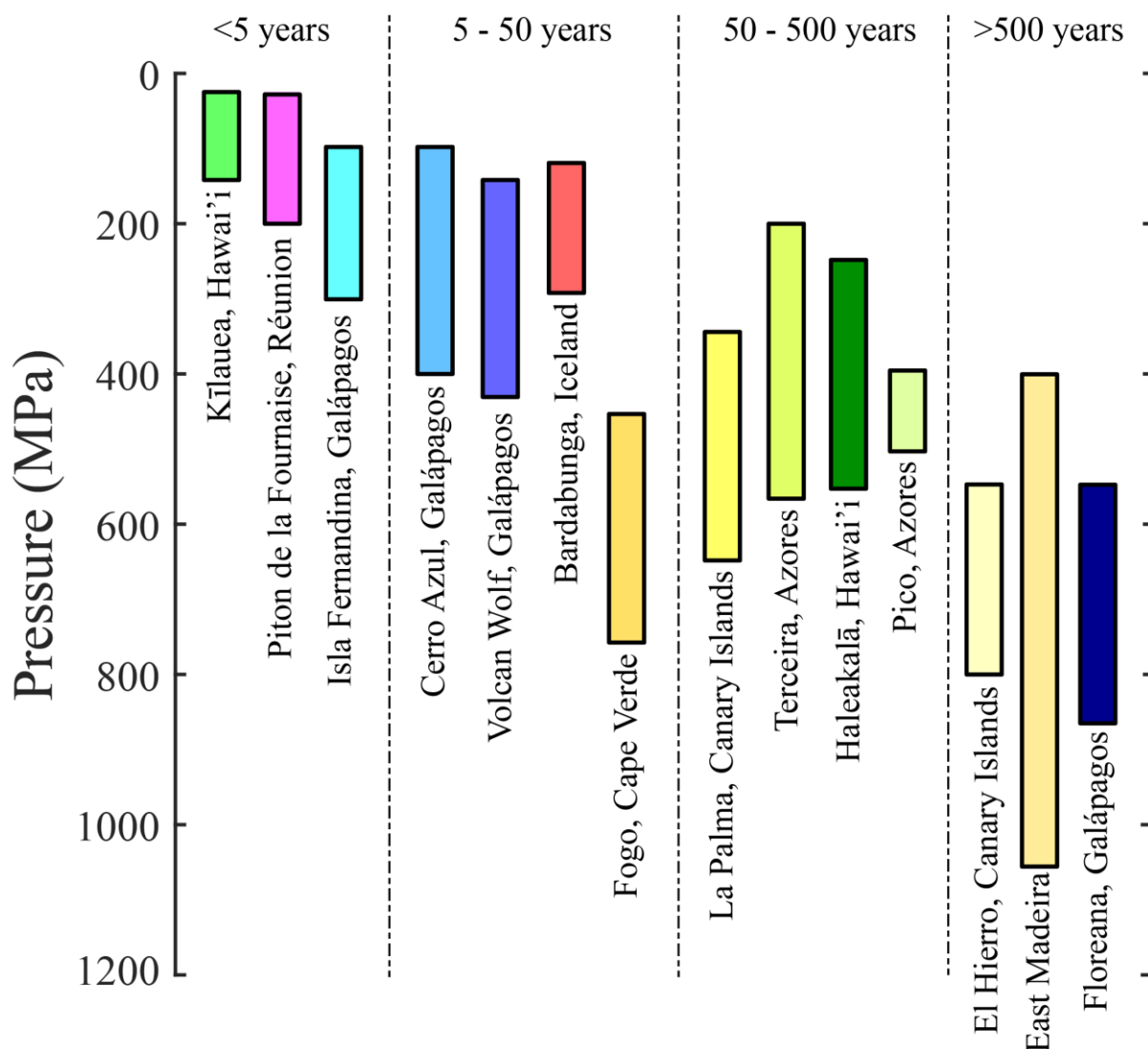
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**Figure 14 -** Compilation of barometric estimates for the primary magma storage region beneath various ocean island volcanoes worldwide. Data is divided according to the approximate repose period between eruptions at each volcano (estimated using data from Global Volcanism Program, 2013), which is used as a proxy for the flux of magma entering the lithosphere. A general trend to greater magma storage pressures is observed with increasing repose period, indicating that the flux of magma from the mantle has a first order control on the depth of magma storage. Data from (Famin et al., 2009; Geist et al., 1998; Hammer et al., 2016; Hartley et al., 2018; Klügel et al., 2015; Poland et al., 2015; Stock et al., 2018; Zanon et al., 2020; Zanon and Pimentel, 2015).