¹ Upper mantle mush zones beneath low

² melt flux ocean island volcanoes: ³ insights from Isla Floreana, Galápagos

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9 ABSTRACT

10 The physicochemical characteristics of sub-volcanic magma storage regions have important 11 implications for magma system dynamics and pre-eruptive behaviour. The architecture of magma 12 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 13 14 the nature of magma storage beneath ocean island volcanoes that are distal to the main zone of mantle 15 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 16 17 study of Isla Floreana in the Galápagos Archipelago, which lies at the periphery of the upwelling 18 mantle plume and is thus characterised by an extremely low flux of magma into the lithosphere. 19 Detailed *in situ* major and trace element analyses of crystal phases within exhumed cumulate 20 xenoliths, lavas and scoria deposits, indicate that the erupted crystal cargo is dominated by 21 disaggregated crystal-rich material (i.e., mush or wall rock). Trace element disequilibria between 22 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 23 24 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 25 26 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 27

- 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.

30 KEY WORDS

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

32 **1 INTRODUCTION**

The physicochemical characteristics (such as size, pressure, volatile content and geochemical 33 heterogeneity) of magma storage at volcanic centres located directly above high buoyancy flux mantle 34 plumes (e.g. Kīlauea, Hawai'i and Isabela, Galápagos) have been subject to intense study over the 35 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; Poland et al., 2015; Sides et al., 2014; Stock et al., 2018; Wieser et al., 2020, 2019). Systems such as 38 Kīlauea are characterised by frequent volcanic activity, and geophysical (seismicity, ground 39 40 deformation) and geochemical (gas emissions) monitoring is prevalent. Monitoring data, combined with petrological and geochemical analysis of erupted products (mineral textures, deformation 41 characteristics and chemistry) provide important insights into the architecture and dynamics of their 42 sub-volcanic plumbing systems (Amelung et al., 2000; Davidge et al., 2017; Geist et al., 2014; 43 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 systems, either above low buoyancy flux plumes (e.g. Canary Islands; Longpre et al., 2014) or at 46 volcanic systems distal to the centre of mantle melting at high buoyancy flux mantle plumes (e.g. 47 48 eastern and south-eastern Galápagos; Harpp and Geist, 2018), are the other endmember. While a substantial number of hotspot-related volcanic systems that have been active during the 49 50 Holocene are located in regions characterised by a relatively low flux of magma into the lithosphere (i.e., regions distal to the main zone of plume upwelling or above low buoyancy flux plumes; Samoa, 51 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

techniques. As a result, few constraints exist on the conditions of magma storage in regions
characterised by a low flux of magma into the lithosphere, relative to volcanic centres located above
the centre of mantle plumes with a large buoyancy flux (and thus generating a large flux of magma
and more frequent eruptions).

The flux of mantle-derived magma into the lithosphere is thought to impart a first-order control on the evolution of ocean island volcanoes and the homogeneity of erupted liquids (Geist et al., 2014). Therefore, placing constraints on the physicochemical characteristics of magma storage at low melt flux ocean island volcanoes is essential for determining the influence of mantle dynamics and melt generation processes on the structure and physical characteristics of sub-volcanic magma plumbing systems. In the absence of detailed monitoring data, petrological and geochemical analyses of volcanic products from past eruptions represent the only available tools for determining the structure

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 the western archipelago (Fig. 1; Villagómez et al., 2014). Hence, although the Galápagos plume has a 68 relatively high buoyancy flux compared to regions such as the Canary Islands (Jackson et al., 2017), 69 Floreana's location relative to the main zone of mantle plume upwelling results in an extremely low 70 71 flux of magma entering the lithosphere and, consequently, very infrequent volcanic activity (Harpp et al., 2014a; Harpp and Geist, 2018). Floreana is considered to be an infrequently active volcanic 72 centre, rather than extinct, owing to the persistent volcanic activity since ~1 Ma and the long-lived 73 74 nature of volcanism on the eastern Galapagos islands that lie >100 km 'downstream' of the Galapagos mantle plume (e.g. the youngest lavas on San Cristobal are ~9 ka; Mahr et al., 2016). 75

In this paper, we present a thorough petrological study of scoria, lava and xenolith samples from
Floreana and place constraints of the structure, depth and crystallinity of magmatic systems beneath
this low melt flux ocean island volcano. We compare our results with more frequently active volcanic
centres in the western Galápagos (near the centre of plume upwelling; Geist et al., 1998; Naumann
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.

83 2 GEOLOGICAL BACKGROUND

84 The Galápagos Archipelago in the eastern equatorial Pacific is one of the most volcanically active regions on Earth, with eruptions typically occurring every 2–3 years (Global Volcanism Program, 85 2013). Although most historic Galápagos eruptions have taken place on the two westernmost islands 86 of Isabela and Fernandina (Fig. 1), infrequent volcanic activity has also occurred on several islands in 87 the eastern and south-eastern Galápagos (for example, Santiago in 1906 and Marchena in 1991; 88 Global Volcanism Program, 2013). In fact, volcanic activity in the eastern Galapagos, >100 km 89 'downstream' of the postulated position of the plume stem, has been shown to be long-lived, with 90 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

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 compositions over hundreds of millennia during their main shield building phase, whereas basalts 108 109 erupted from a single island in the eastern and/or south-eastern archipelago, such as Floreana, tend to display far greater compositional heterogeneity (Geist et al., 2014; Harpp and Geist, 2018). 110 Floreana is characterised by numerous scoria cones and blocky, heavily vegetated lava flows that can 111 typically be traced to the cone from which they originated (Bow and Geist, 1992; Harpp et al., 2014a). 112 The crustal thickness beneath Floreana is ~16 km, similar to that observed in the western Galápagos 113 10-18 km (Feighner and Richards, 1994), and the lithospheric thickness beneath the western and 114 115 south-eastern Galápagos is very similar (~50-60 km; Gibson and Geist, 2010). Recent work has shown that the average volumetric eruption rate on Floreana over the past 1-1.5 Myrs is 1-10 m³·yr⁻¹, 116 6 orders of magnitude lower than the current volcanic productivity at volcanoes in the western 117 Galápagos (cf. ~4.4·10⁶ m³·yr⁻¹ at Fernandina; Harpp et al., 2014a; Kurz et al., 2014). This difference 118

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.

Despite the relatively low volcanic productivity of Floreana over the last ~1–1.5 Myrs, the erupted products have several important characteristics that provide insights into the nature of the underlying magmatic system. For example, Floreana has a high proportion of pyroclastic deposits compared to the other Galápagos islands (Harpp et al., 2014a) and eruption deposits typically contain a large number of cumulate xenoliths (Bow and Geist, 1992; Lyons et al., 2007), which have been interpreted 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

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

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

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

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

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 Floreana subaerial basalts (Lyons et al., 2007). Differences in the isotopic characteristics of the 135 Floreana lavas (submarine vs subaerial) and xenoliths (gabbros vs wehrlites) are thought to indicate a 136 change in the mean composition of magma produced by mantle melting beneath the island at $\sim 1-1.5$ 137 138 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 (<1–1.5 Ma) using chemical and textural analysis of crystal phases in lava flows and xenolithic 140 141 nodules.

142 **3 SAMPLES AND PETROGRAPHY**

The Floreana samples analysed in this study consist of lavas (27 samples), scoria (2 samples) and 143 144 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 the Main Series of Floreana lavas identified by Bow and Geist (1992). The scoria samples were 146 collected from two separate deposits and comprise rapidly cooled scoria lapilli (~0.5-2 cm across; 147 148 17MMSG16) and bombs (~10 cm across; 17MMSG20). Xenolithic fragments (3-15cm across) were sampled from two different scoria cones on the north-east coast of Floreana; similar xenoliths are also 149 150 found within most lava flows across the entire island (Supplementary Information).

151 3.1 Lavas and scoria

The lava and scoria samples analysed in this study are typically olivine phyric with minor anhedral 152 clinopyroxene and very rare orthopyroxene. Except for small plagioclase laths in the microcrystalline 153 groundmass, plagioclase crystals are extremely rare in the Floreana lavas. Plagioclase macrocrysts are 154 155 only present in one of our lava samples (17MMSG29) where they occur as isolated phenocrysts and in plagioclase-olivine crystal clots (Table S.1). The abundance of olivine and absence of plagioclase in 156 the Floreana lavas and scoria contrasts with basalts in the central, northern, and western parts of the 157 Galápagos Archipelago, where plagioclase-phyric and ultraphyric basalts are common (Geist et al., 158 159 2002; Gibson et al., 2012; Harpp et al., 2014b).

160	Despit	e 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
- pyroxene crystals are typically >500 µm and grain sizes are relatively uniform within a single xenolith
- sample. Where three plagioclase grains meet at a triple junction, 120° grain boundaries indicate a high
- 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
- approaching 120° grain boundaries. Minor intercumulus clinopyroxene is present along grain
- 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
- as large (<5 mm) oikocrysts, which enclose rounded olivine chadacrysts <500 µm in diameter (Fig.
- 195 3C and D). Fine-scale orthopyroxene exsolution lamellae ($<2 \mu m$) 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 μ m) 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
- have the isotopic signatures of modern day Floreana basalts; Lyons et al., 2007) indicate that the
- typical order of crystallisation beneath Floreana is olivine, followed by clinopyroxene, with little to no
- 204 crystallisation of plagioclase.

205 4 ANALYTICAL METHODOLOGY

206 4.1 Electron microprobe analysis

Glass chips, olivine and clinopyroxene crystals were hand-picked from crushed scoria and lava
samples, mounted in epoxy or indium, and then ground and polished prior to analysis (crystals
mounted in indium were polished individually prior to mounting). Xenolithic crystals were analysed
as individual crystals mounted in indium or *in situ* in petrographic thin sections. The major and minor
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 μ m) 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 μ m)
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 μ m) 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. 2σ
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 2σ precision for
228	glass analysis was typically $<3\%$ for major elements, $\sim5\%$ for Na, and $\sim10\%$ for K (see
229	Supplementary Information).

230 4.2 Laser ablation Inductively Coupled Plasma Mass Spectrometry

Trace element concentrations were measured in the apparent cores (i.e. as exposed in the 2D plane) of clinopyroxene crystals from scoria and xenolith samples using an ESI193 laser coupled to a Nexion 350D inductively coupled plasma mass spectrometer in the Department of Earth Sciences, University of Cambridge. Analyses were collected in spot mode using a 20 Hz laser repetition rate, 4 J/cm² fluence and 80 µm spot size, or in transect mode using a 10 Hz repetition rate, 3.5 J/cm² fluence and 30 µm spot size. For transects, individual spots were offset into two (alternating) lines to increase the spatial resolution. Laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) data

reduction was carried out in Iolite[®], with NIST 612SRM as the standard reference material (Hinton,

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

- accuracy was tracked using a USGS glass standard (Jochum et al., 2016) and was between 95% and
- 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-
- earth elements (LREE), Y, Sr, and Zr and 10–20% for the heavy rare earth element (HREE). 2σ
- analytical precision was ~10% for all elements of interest in transect analyses (Ce, Y).

245 **5 GLASS AND MINERAL CHEMISTRY**

246 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

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

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

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

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

17MMSG27 where, at a near constant MgO contents, the CaO, Na₂O, and K₂O contents vary by ~6

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

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

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

259 Our Floreana matrix glass analyses have higher Al₂O₃ concentrations, at a given MgO content, than

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

- whole-rock samples with accumulated plagioclase, Fig. 4D; Geist et al., 2002). This indicates
- 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 H₂O content of primary mantle melts
- and/or increased pressure of magma storage (Asimow and Langmuir, 2003; Neave et al., 2019;
- 266 Thompson, 1987; Winpenny and Maclennan, 2011).

267 5.2 Olivine compositions

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

269 contents (Fo = 70–92, where Fo= $(Mg/(Mg+Fe^{2+}) \text{ molar})$ 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 Fe³⁺/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
- and Ca concentration in these crystals, the most forsteritic olivines (Fo>83) have extremely diverse Ca
- contents (~250 to ~2600 ppm; Fig. 5), whereas crystals with lower Fo contents (<83) have
- 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 \sim 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
- concentrations (~700 to ~3200 ppm), consistent with crystallisation from a peridotite-derived melt
- (Fig. 5; Gleeson and Gibson, 2019; Herzberg, 2011; Matzen et al., 2017b, 2017a). All olivine crystals
- analysed in the wehrlite and dunite xenoliths have a narrow range of Fo contents (83-87) and,
- 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
- typically flat profiles in Fo, Ni, and Ca, with one profile displaying evidence for an increase in Ca in
- the outer $10 20 \ \mu m$ (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).

290 **5.3 Clinopyroxene compositions**

291 5.3.1 Major elements

292 The Floreana clinopyroxenes separated from the scoria and xenolith samples are augitic, and have a relatively high Mg# (0.85–0.90; Mg# = Mg/(Mg+Fe_t) molar). The clinopyroxenes reach high Na 293 294 concentrations up to 1.58 wt% Na₂O (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). Results indicate that the clinopyroxenes are unzoned with respect to most element (including MgO 301 and FeO), but some zoning is present in their Al₂O₃, TiO₂ and Cr₂O₃ contents. Specifically, 302

303 clinopyroxene crystal rims are typically characterised by elevated TiO_2 , but lower Cr_2O_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 $[La/Sm]_n \sim 0.2$ to $[La/Sm]_n \sim 3.1$ (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 scoria samples (Fig. 7). Furthermore, melt $[La/Sm]_n$ ratios calculated to be in equilibrium with 309 310 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 strain model of Wood and Blundy (1997) at 1225°C and 700 MPa; Fig. 7B), significantly greater than 312 the range measured in Floreana whole-rock samples ($\sim 2-5$, with a small number of outliers up to 313 \sim 7.5; Harpp et al. 2014a). Almost all of the xenolithic crystals, and a large proportion of the 314 315 clinopyroxenes separated from scoria deposits, are too enriched to be in equilibrium with the typical composition of melts erupted on Floreana (Harpp et al., 2014a). In addition, there is a strong 316

- 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
- 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).

323 6 MAGMA SYSTEM ARCHITECTURE BENEATH 324 ISLA FLOREANA

325 6.1 Mush crystallisation and textural equilibration

Based on pyroxene trace element and radiogenic isotope ratios, Lyons et al. (2007) hypothesised that gabbroic xenoliths in the Floreana lava and scoria deposits formed in an ancient (>1-1.5 Ma) magmatic system, compositionally similar to those currently beneath Cerro Azul and Sierra Negra volcanoes in the western Galápagos. In contrast, wehrlite xenoliths preserve isotopic ratios similar to more recent subaerial lavas on Floreana, suggesting that they are fragments of the present-day magmatic system (Lyons et al., 2007).

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

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

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

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

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

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

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

345 **6.2** Mush disaggregation prior to eruption

- 346 6.2.1 Insights from olivine compositional heterogeneity
- Olivine crystals separated from the Floreana lava and scoria samples have a wide range of zoning
 patterns, morphologies, and compositions (Fig. 2). The five olivine groups identified in the Floreana
 samples have distinct morphologies and zoning patterns (see Section 2 above), suggesting chemically
 heterogeneous magma storage (Holness et al., 2019). In particular, the most evolved crystals (Group
 4; Fo~70–75) are in equilibrium with melts that are more evolved than the Floreana erupted basalts
 (likely basaltic andesites). This is consistent with a recent study which identified highly evolved
 (andesitic dacitic) magmas beneath basaltic volcanoes in the western Galápagos Archipelago (Stock
- et al., 2020).
- 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
- interacted on relatively short pre-eruptive timescales. In Group 5 olivines, for example, four
- 358 compositional zones are preserved over ~100-200 μ m (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
- using diffusion chronometry, complex forsterite zoning over $\sim 100 \ \mu m$ is estimated to last <3 yrs at the
- 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.
- The minor element chemistry of the olivine crystals allows us to investigate the crystallinity of these chemically diverse magma storage regions. Olivine crystals in our Floreana lava and scoria deposits 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

in Ca concentrations measured in the Floreana lava and scoria deposits overlaps with those observed
in cumulate xenoliths (wehrlites) and thus are unlikely to represent mantle olivines (Thompson and
Gibson, 2000). Previous studies have shown that the Ca concentration of magmatic olivine is sensitive
to several parameters, including: (i) the major element composition of the co-existing melt phase
(Herzberg, 2011); (ii) the H₂O content of the co-existing melt phase (Gavrilenko et al., 2016); and (iii)
the temperature of the system (Adams and Bishop, 1982; Köhler and Brey, 1990; Shejwalkar and
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

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

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

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

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

Evidence for the origin of the low-Ca contents in the Floreana olivines from lava and scoria deposits 387 is present in the texture and composition of the wehrlitic xenoliths, which contain uniformly low-Ca 388 olivine crystals (<1000 ppm; Fig. 5A). The petrography of the wehrlitic xenoliths attests to 389 390 clinopyroxene growth within olivine-dominated mush regions. Clinopyroxene crystallisation within this mush would extract CaO and MgO from the residual melt. However, in an olivine-rich mush, the 391 large reservoir of MgO contained within the cumulus olivine grains would buffer the residual melt at 392 a near-constant Mg# during clinopyroxene crystallisation (Meyer et al., 1989). In contrast, the CaO 393 394 concentration of the melt is not buffered and decreasing melt CaO contents, due to clinopyroxene crystallisation, will cause the CaO concentration of cumulus olivine grains to decrease (as a result of 395

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

397 concentrations in the matrix glass of sample 17MMSG27.

Intercumulus clinopyroxene growth would also increase the H₂O concentration of the residual melt 398 399 phase, decreasing the partition coefficient of Ca into olivine (Gavrilenko et al., 2016). Therefore, variable amounts of clinopyroxene crystallisation within an olivine-dominated mush, and subsequent 400 disaggregation of this mush by an ascending melt, could explain the range of Ca contents measured in 401 the Floreana olivines derived from lava and scoria deposits. Specifically, we suggest that: (i) 402 forsteritic olivine crystals (Fo>83) with Ca contents >1500 ppm are consistent with those expected 403 404 from fractional crystallisation of mantle-derived melts in a liquid-rich magma storage region (Fig. 5A; Gleeson and Gibson, 2019); (ii) olivines with Ca contents <1000 ppm, overlapping with the wehrlitic 405 olivines, reflect equilibrium with Ca-poor interstitial melts; and (iii) olivine crystals with intermediate 406 Ca concentrations (1000 - 1500 ppm) are sourced from regions where clinopyroxene growth is less 407 408 extensive, or ongoing at the time of eruption. Thus, the olivine crystal cargo of the Floreana magmas is predominantly derived from crystal-rich domains with only a small number of olivine crystals 409 410 displaying compositions that are consistent with fractional crystallisation in liquid-rich storage regions 411 (Ca >1500 ppm; Gleeson and Gibson, 2019).

It is an important to note, however, that the partitioning of Ca between co-existing olivine and 412 413 clinopyroxene has been hypothesised to be sensitive to temperature (such that less Ca enters the olivine structure at lower temperature; Shejwalkar and Coogan, 2013). As a result, the heterogeneity 414 in the Ca content of the Floreana olivines could instead represent disaggregation of xenolithic material 415 that has undergone variable amounts of cooling. Both hypotheses presented here can recreate the 416 417 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

- 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
- 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.

447 **6.3 Reactive Porous Flow within a cumulate mush**

Whilst the olivine and clinopyroxene major and minor element concentrations show that a large 448 449 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 within these crystal-rich domains. The trace element composition of melts in equilibrium with our 451 clinopyroxene crystals are calculated using the model of Wood and Blundy (1997). The results 452 indicate that many of our clinopyroxene analyses have incompatible trace element ratios (e.g. 453 454 [Ce/Y]_n) which are more enriched than any erupted basalt from Floreana (Harpp et al., 2014a). In fact, nearly all clinopyroxene analyses from our xenolith samples, and ~50% of clinopyroxene analyses 455 from the scoria samples, record trace element disequilibrium with the typical composition of the 456 Floreana basalts (Fig. 7 and 9). Over-enriched trace element signatures are characteristic of Cluster 1 457 clinopyroxenes (i.e. chemical affinity to the wehrlitic or dunitic xenoliths), whereas crystals that are 458 near trace element equilibrium with Floreana basalts typically have Cluster 3 major element 459 compositions (i.e. the autocryst cluster). 460

Petrographic observations and olivine minor element data indicate that the Floreana sub-volcanic 461 system is characterised by clinopyroxene crystallisation within an olivine-dominated mush. If the 462 clinopyroxene grew from trapped melt within an olivine-dominated mush, progressive crystallisation 463 would increase the concentration of highly incompatible trace elements (e.g. Ba, La, Ce) relative to 464 less incompatible trace-elements (e.g. Sm, Y) in the residual melt. A simple fractional crystallisation 465 model indicates that \sim 70% crystallisation is required to generate melt [Ce/Y]_n ratios that are in 466 467 equilibrium with enriched clinopyroxenes from the scoria samples and even greater extents of crystallisation (\sim 80%) would be required to generate the extremely high [Ce/Y]_n ratios in some of the 468 xenolithic clinopyroxenes (Fig. 9). 469

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 within the cumulate mush is responsible for generating the anomalous trace element signatures of the 475 Floreana clinopyroxenes. Infiltration of mantle-derived melts that are more enriched than anything 476 observed in erupted basalts on Floreana is considered unlikely owing to the overlapping isotopic 477 478 composition of xenoliths and erupted lavas (Lyons et al., 2007), and the absence of a significant correlation between trace element enrichment and radiogenic isotope compositions in the erupted 479 480 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.

One alternative mechanism that might be able to generate the observed trace element over-enrichment 483 is reactive porous flow. In this scenario, clinopyroxene is precipitated from ascending clinopyroxene-484 saturated melts that continuously react with the existing, olivine-dominated crystal framework as the 485 486 system approaches equilibrium, and thus deviates from a simple fractional crystallisation trajectory (Lissenberg and MacLeod, 2016). As a result, reactive porous flow in the olivine-dominated mush 487 beneath Floreana will likely result in substantial crystallisation of clinopyroxene, possibly at the 488 expense of pre-existing crystal phases, with little to no formation of olivine or saturation of minor 489 490 phases. Consequently, reactive porous flow can lead to enrichment of highly- to moderatelyincompatible trace elements in the resulting melt (Coogan et al., 2000; Gao et al., 2007; Lissenberg et 491 492 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).

To test whether reactive porous flow of clinopyroxene saturated melts through an olivine-dominated mush is consistent with the trace element compositions of melts calculated to be in equilibrium with our Floreana clinopyroxenes, we use the zone refining model of Harris (1957; Fig. 9):

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

5	0	3
_	_	_

(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 melt front with the existing crystal framework, and has previously been employed to investigate 507 geochemical signatures in oceanic gabbros (Lissenberg and MacLeod, 2016). Results indicate that 508 509 reactive porous flow can produce melts with trace element compositions that are comparable with those in equilibrium with our Floreana clinopyroxenes (i.e. $[Ce/Y]_n \sim 8-13$) using I values that are 510 similar to those invoked in other magmatic settings worldwide (~2-5 compared to ~4-8 for the Hess 511 Deep; Lissenberg and MacLeod, 2016). Hence, reactive porous flow represents a realistic mechanism 512 for generating the geochemical diversity of Floreana clinopyroxenes, including the trace element 513 enriched crystals analysed in the wehrlitic nodules (Fig. 9). 514 515 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 in equilibrium with Floreana basalts (Harpp et al. 2014a; Fig. 11a), whereas the mantle and rim of the 518 519 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 (Harpp et al., 2014a), followed by growth from a melt which became progressively enriched during 521

522 reactive porous flow (Fig. 11).

523 The mantle of the second, smaller xenolithic clinopyroxene shows a similar rim-ward increase in [Ce] (interpreted as progressive melt enrichment during reactive porous flow). However, the [Ce] and 524 [Ce/Y]_n values of the crystal core are too high to be in equilibrium with erupted Floreana basalts (Fig. 525 11C). This is consistent with our spot analyses of clinopyroxene cores in other crystals and samples. 526 527 The high [Ce] and $[Ce/Y]_n$ values measured in crystal cores cannot be explained by inward diffusion of Ce, owing to significant differences in the diffusivities of Ce and Y and similar [Ce] and [Y] 528 529 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 flow. However, we cannot discount that our apparent clinopyroxene cores are fragments of larger 532 oikocrysts that have been broken during mush disaggregation or sample crushing and, as a result, do 533 534 not represent the true core compositions of each crystal.

Nevertheless, our clinopyroxene major and trace element data, as well as petrographic observations of
the wehrlitic xenoliths, provide substantial evidence that reactive porous flow is an important
mechanism of melt migration and melt differentiation in highly crystalline magma storage regions
beneath Floreana. Although reactive porous flow has been identified as an important process in MOR
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

Petrological and geophysical constraints on magma storage depths exist for several recently active 541 542 volcanoes in the western Galápagos Archipelago (Bagnardi et al., 2013; Case et al., 1973; Geist et al., 1998; Stock et al., 2018; Vigouroux et al., 2008). However, in the absence of geophysical data (owing 543 to a paucity of recent eruptions), there are far fewer constraints on the structure of magma storage 544 regions in the eastern and south-eastern archipelago. To date, the only investigation of magma storage 545 546 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 point, parameterised by Grove et al. (1992). This approach is subject to substantial uncertainty, but the 548

authors suggest that the Floreana magmas consistently equilibrate at >5 kbar (typically >7 kbar) at a
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 (2008; equations 32b and 32d, respectively), in which pressure and temperature are solved iteratively 553 based solely on the clinopyroxene major element composition (primarily the jadeite component; 554 standard error of estimate [SEE] ± 310 MPa). Second, we applied the clinopyroxene-melt barometer of 555 Neave and Putirka (2017; equation 1), which uses the composition of a co-existing melt phase and the 556 557 proportion of the Jadeite component in clinopyroxene to calculate the pressure of crystallisation (SEE $= \pm 140$ MPa; pressure is solved iteratively with temperature using the clinopyroxene-melt 558 thermometer of Putirka, 2008; equation 33). Third, for the xenolithic nodules, we estimate the final 559 pressure and temperature of storage using the two-pyroxene thermobarometer of Putirka (2008; 560

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

562 Taken at face value, initial application of the clinopyroxene-only barometer to all clinopyroxene analyses from the scoria and xenolith samples gives a range of pressure estimates between ~450 MPa 563 and ~1800 MPa. However, reactive porous flow has a substantial influence on the compositions of the 564 Cluster 1 (and Cluster 2) clinopyroxenes, which may influence the barometric results. Specifically, 565 566 crystals that show evidence for reactive porous flow also have elevated Na concentrations, leading to an anomalously high jadeite component and thus calculated pressure. This likely originates from the 567 presence of unusual melt compositions that fall outside the calibration range of the clinopyroxene-568 only barometer owing to reactive melt migration, which is supported by the unusual glass 569 570 compositions in sample 17MMSG27 (Fig. 4). Therefore, we filter our dataset to remove crystals that show a chemical signature indicative of reactive porous flow and only use Cluster 3 clinopyroxenes 571 that have trace element compositions in equilibrium with the Floreana basalts (using the whole-rock 572 data from Harpp et al. 2014) in our barometric calculations (n=78). Barometric results from this 573 574 filtered dataset indicate that crystallisation beneath Floreana occurs at a pressure of 766 ±322 MPa

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

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/m³; Fig. 12).

577

equilibrium clinopyroxene-liquid pairs. We achieve this using an automated melt-matching algorithm (as in Winpenny and Maclennan, 2011, Neave and Putirka, 2017, Stock et al. 2018), with K_D (Fe-Mg), diopside-hedenbergite, enstatite-ferrosillite and calcium Tschermak's equilibrium tests (K_D (Fe-Mg) within ±0.03 other components within 2 SEE; Putirka, 1999, Putirka, 2008, Mollo et al., 2013). We

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

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

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

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

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

melt composition, an average melt composition was used in the barometric model. Results from this
barometer, coupled to the thermometer of Putirka (2008), indicate that magma crystallisation occurred

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

Clinopyroxene-orthopyroxene thermobarometry records the final storage conditions of the cumulate xenoliths, rather than the crystallisation conditions of clinopyroxene autocrysts (orthopyroxene is only found as an intercumulus phase). Temperature and pressure estimates were only calculated from orthopyroxene-clinopyroxene pairs in wehrlite and dunite xenoliths that passed the K_D (Fe-Mg) equilibrium test of Putirka (2008; 1.09 ±0.14). Results suggest that the cumulates were stored at ~975–1100°C and 600–900 MPa, with a mean storage pressure of 712 ±200 MPa (23.7 ±6.4 km; Fig. 12).

The depths of magma storage calculated from our three petrological barometers show an excellent
agreement within the model uncertainties. These new data provide robust evidence that magma
storage beneath Floreana occurs below the Moho (~16 km; Feighner and Richards, 1994), in the upper
mantle.

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 605 606 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 607 crystal cargo is derived from disaggregated mush and wall rock material which has been entrained 608 609 into the ascending magmas. During ascent, magmas may entrain coherent nodules (xenoliths) as well 610 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 611 crystallisation temperatures of ~1225°C) and may represent material from the border of the active 612 mush zone or older, almost completely solidified magma storage regions that are intersected during 613 614 magma ascent (Fig. 13).

615 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-616 dominated areas beneath Floreana. Reactive porous flow causes distinct trace element enrichment in 617 the percolating melt phase and crystallising clinopyroxene, which can explain the trace element 618 619 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 620 with erupted Floreana basalts indicates that at least some crystallisation occurs in liquid-rich sub-621 volcanic storage regions, likely situated as localised melt pockets within the larger mush zone (Fig. 622 623 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 basalts is primarily a source signature, likely associated with the highly radiogenic Pb and Sr isotope 630 signatures that characterise the Floreana basalts (Harpp et al., 2014a; Harpp and White, 2001). 631 632 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 that have undergone geochemical modification due to reactive porous flow. 635 Our results indicate substantial differences in the architecture of the magmatic systems beneath 636 637 Floreana and the frequently active shield volcanoes in the western Galápagos Archipelago. For example, previous petrological and geophysical studies have identified that western Galápagos 638 magmatic systems are characterised by crustal magma storage, often with a large storage region in the 639 mid-to-lower crust (~7 km depth) and a smaller storage region at shallow levels, within the volcanic 640 641 edifice (~1 km depth; Geist et al. 1998; Bagnardi et al. 2013; Bagnardi and Hooper, 2018; Stock et al., 2018; Fig 12). In contrast, our barometric data indicate that magmas beneath Floreana ascend directly 642

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

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

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

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).

One major factor that differentiates Floreana from shield volcanoes in the western archipelago is the flux of magma into the lithosphere, which is evidenced by the large variations in the volumetric eruption rate on Floreana and the western shields (Harpp et al., 2014a; Harpp and Geist, 2018; Kurz et al., 2014). Hence, we suggest that the greater pressure of magma storage and prevalence of reactive porous flow beneath Floreana, relative to volcanoes in the western archipelago, are related to the substantially lower flux of magma into the lithosphere from the underlying mantle source (and thus 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 average temperature of the mid-to-lower crust at ~1125°C ($\Delta T \sim 22$ °C), with only small-scale thermal 658 and compositional heterogeneities present in the sub-volcanic plumbing system (Geist et al., 2014, 659 660 2005; Stock et al., 2020, 2018). In contrast, the flux of magma entering the magmatic system beneath Floreana is much lower and the temperature of the mid-crust is likely to be significantly cooler than 661 the lowest temperature recorded by the Floreana xenoliths (that is, <<900°C; Fig. 12). As the flux of 662 magma (and heat) from the mantle is insufficient to maintain an elevated crustal geotherm beneath 663 664 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 regions (~700-750 MPa) where melts can persist over long time periods. 666 Our results have global implications for the architecture and dynamics of magma storage regions 667 668 beneath ocean island volcanoes worldwide. The observed difference in magma storage depths beneath high and low melt flux volcanic systems in the Galapagos Archipelago is mirrored in a global 669 compilation of barometric data from ocean island volcanoes (Famin et al., 2009; Geist et al., 1998; 670 Hammer et al., 2016; Hartley et al., 2018; Klügel et al., 2015; Poland et al., 2015; Stock et al., 2018; 671 672 Zanon et al., 2020; Zanon and Pimentel, 2015). Using the average repose period between eruptions at a particular basaltic volcanic centre as a proxy for the flux of magma entering the lithosphere from the 673 674 underlying mantle (Global Volcanism Program, 2013), Figure 14 shows that the most frequently active volcanic centres (such as, Kīlauea, Hawai'i, and Piton de la Fournaise, Réunion) are 675 676 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 Islands) and/or peripheral to the main zone of plume upwelling (e.g. Haleakalā, Hawai'i) are 678 679 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 crustal magma staging can occur (Klügel et al., 2015), the main zone of magma storage beneath ocean 681

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

of the crust (Longpre et al., 2014; Taracsák et al., 2019; Zanon et al., 2020). Hence, we speculate that
the flux of magma from the underlying mantle source has a first-order control on the depth of magma
storage beneath ocean island volcanoes and, correspondingly, high-pressure magma storage – as
observed beneath Floreana – is characteristic of low melt flux ocean island volcanoes globally.

687 8 CONCLUSIONS

Petrographic and geochemical analyses of lava, scoria and xenolith samples from Floreana in the 688 south-eastern Galápagos Archipelago provide new insights into the architecture and dynamics of 689 magma storage beneath low melt flux ocean island volcanoes. Comparison of olivine and 690 691 clinopyroxene major, minor and trace element contents between our different sample types reveal that a substantial portion of the erupted crystal cargo is derived from mush-dominated magma storage 692 693 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 chemical differentiation and melt transport within these mush-dominated regions. Mixing between 695 melts that have been geochemically enriched by reactive porous flow and those in overlying liquid-696 697 rich storage regions could contribute to the anomalous, concave-up REE signature of the Floreana basalts, which is absent in other parts of the Galápagos Archipelago where reactive porous flow has 698 not been identified. 699

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 western archipelago where magmas are stored in the crust (Geist et al., 1998; Stock et al., 2018). 704 705 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 proximal magmatic systems above high buoyancy flux plumes than in off-axis systems, or above low 707 buoyancy flux plumes. We therefore suggest that the flux of mantle-derived magma entering the 708

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

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1029 **FIGURES**



Figure 1 - A. Regional map of the Galápagos Archipelago highlighting the location of Isla Floreana,
Cerro Azul (CA), Sierra Negra (SN) and Wolf volcanoes. Dates show the most recent eruptions at
historically active volcanic centres. Black arrows show the direction of plate motion for the Nazca and
Cocos tectonic plates. B. Geological map of Floreana adapted from Harpp et al. (2014a). Dashed lines
delineate monogenetic scoria cones. Normally and reversely polarised lava flows are shown along
with the largest (Cerro Pajas) and most recent (Alayeri; ~26,000 years) eruptions on the island.



1038 Figure 2 – Backscatter Electron images. A. Group 1 olivines – euhedral to subhedral crystal 1039 morphologies with large, unzoned, crystal cores and narrow, normally-zoned rims. B. Group 2 1040 olivines – subhedral to euhedral crystals with clear, reverse-zoning profiles. C. Group 3 olivines – 1041 skeletal crystals with high forsterite overgrowths on low forsterite cores. **D.** Group 4 olivines – 1042 anhedral crystals with sieved textured, reverse zoned rims. E. (greyscale) and F. (false colour) Group 1043 5 olivines – crystals preserve at least 4 composition zones over ~100-200 μ m. False colour image (F.) is used to highlight the compositional zoning of the Group 5 olivine, with the intensity of the blue 1044 1045 colour associated with the Fo composition of the crystal (darker = higher Fo).



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.



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 2σ 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. 1063 1064 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 1065 1066 (Vidito et al. 2013). Our lava/scoria analyses are coloured according to their Ca concentration. The 1067 histograms above and to the right of the plots show the data distributions (excluding *in situ* analyses 1068 of xenolithic olivines). Peridotite source solutions are taken from Herzberg (2011) and Matzen et al. 1069 (2017a). Black arrows in A. show the trajectory of crystal compositional evolution during olivine (ol) 1070 and clinopyroxene (cpx) crystallisation (taken from Gleeson and Gibson, 2019). The green lines in **B.** 1071 show the trajectories of crystal compositional evolution during olivine only and olivine + 1072 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 1073 composition of tephra sample 17MMSG16 ($K_d = 0.27 - 0.34$ after Matzen et al. 2011, and Roeder and 1074 1075 Emslie 1970).



Figure 6 – Major element composition of Floreana clinopyroxenes from our scoria, wehrlite and
dunite samples. Data is shown as atoms per formula unit (a.p.f.u) on the basis of 6 oxygens. The grey
field shows the compositions of clinopyroxenes from Wolf volcano in the western Galápagos
Archipelago (from Stock et al. 2018). The 2σ precision of our clinopyroxene analyses is smaller than
the symbol size.



1083 Figure 7 – Trace element composition of the Floreana clinopyroxenes and their relation to major 1084 element systematics. A. $[La/Sm]_n$ vs. Na in clinopyroxenes from our scoria samples and wehrlite and 1085 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 1086 lines show the 95% confidence limits on the regression. The correlation indicates that trace element 1087 1088 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 1089 1090 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 1091 1092 reactive porous flow within the cumulate mush (RPF). The grey field shows whole-rock compositions 1093 from Fernandina (Geist et al., 2006; White et al., 1993). B. additionally shows the whole-rock 1094 compositions of erupted Floreana lavas (Harpp et al., 2014a) and analyses of the gabbroic xenoliths 1095 from Floreana (this study). Error bars show the fully propagated 2σ precision of our analyses.

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Figure 8 – A. Hierarchical cluster analysis of our clinopyroxene major element analyses. Cluster 1097 1098 analysis was performed using Ward's method, which is built into the scikit-learn package in Python 1099 (Pedregosa et al., 2011; Ward, 1963). Height above the x-axis is a measure of the distance of 1100 separation of two clusters (i.e., the higher the join the more chemically distinct two clusters are). 1101 Colours show the high-level division of crystal compositions into three clusters: Cluster 1 is 1102 predominantly comprised of crystals from wehrlite and dunite xenoliths (red), Cluster 2 is composed 1103 of crystals from gabbroic xenoliths (yellow) and Cluster 3 (blue) is dominated by crystals separated 1104 from scoria samples. B. Na vs Mg and C. Cr vs Mg in our clinopyroxene analyses from the scoria and 1105 xenoliths, coloured by their cluster.







Figure 10 – WDS maps of Ti counts in key clinopyroxene crystals from the Floreana wehrlite
xenoliths. These maps display clear zoning in the xenolithic clinopyroxenes with Ti-poor cores and
Ti-rich rims. EPMA transects across some of the wehrlitic clinopyroxenes indicates that the Ti
concentrations in these crystals may vary from ~0.6 wt% to ~1.3 wt% TiO₂ (see Supplementary
Information). The dark blue regions surrounding clinopyroxene grains are olivine crystals.

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1126Figure 11 – LA-ICP-MS transects showing Ce (A, C) and Y (B, D) zoning across two clinopyroxene1127grains in a wehrlitic xenolith (17MMSG02c). A. and B. show a transect across the core of a large1128clinopyroxene oikocryst. C. and D. show a transect across the apparent core of a smaller1129clinopyroxene oikocryst. Points are coloured according to their $[Ce/Y]_n$ ratio (see colour scale). The1130grey bars show the crystal compositions calculated to be in equilibrium with whole-rock analyses of1131erupted Floreana basalts (Harpp et al., 2014a).



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 – clinopyroxeneorthopyroxene 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 clinopyroxenemelt 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-

1140 clinopyroxene thermobarometers are given.



1141

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.



1150 Figure 14 - Compilation of barometric estimates for the primary magma storage region beneath 1151 various ocean island volcanoes worldwide. Data is divided according to the approximate repose 1152 period between eruptions at each volcano (estimated using data from Global Volcanism Program, 1153 2013), which is used as a proxy for the flux of magma entering the lithosphere. A general trend to 1154 greater magma storage pressures is observed with increasing repose period, indicating that the flux of 1155 magma from the mantle has a first order control on the depth of magma storage. Data from (Famin et 1156 al., 2009; Geist et al., 1998; Hammer et al., 2016; Hartley et al., 2018; Klügel et al., 2015; Poland et 1157 al., 2015; Stock et al., 2018; Zanon et al., 2020; Zanon and Pimentel, 2015).