

1 **Timescales of crystal mush mobilization in the Bárðarbunga-Veiðivötn**  
2 **volcanic system based on olivine diffusion chronometry**

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21 Abstract

22 The timescales of magmatic processes within a volcanic system may be variable over a volcano's  
23 geological history. Crystals reflect environmental perturbations under which they grew, and  
24 compositional gradients quenched inside crystals on eruption can be exploited to extract timescales of  
25 magmatic processes. Here, we use multi-element diffusion chronometry in olivine macrocrysts to  
26 recover their residence time in a melt which ultimately erupted at the surface. The macrocrysts were  
27 mobilized by the carrier melt from mushy layers in the magma reservoir, and diffusion timescales likely  
28 reflect the time interval between mush disaggregation, ascent and eruption. To unravel the evolution  
29 of mush disaggregation timescales with time, we target early-Holocene, middle-Holocene and historical  
30 magmatic units erupted in the Bárðarbunga-Veiðivötn volcanic system in Iceland's Eastern Volcanic  
31 Zone. Macrocryst contents vary between samples; early-Holocene samples are highly phyrlic (10-45  
32 vol% macrocrysts) and contain gabbroic nodules, whereas middle-Holocene (5-15 vol%) and historical  
33 units (5-10 vol%) tend to be generally less phyrlic. Early-Holocene olivine macrocrysts have core  
34 compositions in the range  $Fo_{84-87}$ , whilst middle-Holocene and historical samples record a wider range  
35 in core compositions from  $Fo_{80}$  to  $Fo_{86.5}$ . Olivine rims are in chemical equilibrium with their carrier liquid,  
36 and are slightly more evolved in early-Holocene units ( $Fo_{76-81}$ ) compared to middle-Holocene ( $Fo_{78-80}$ )  
37 and historical ( $Fo_{81-83}$ ) units. Diffusion chronometry reveals that the timescale between mush  
38 disaggregation and eruption has changed over time, with timescales getting shorter approaching recent  
39 times. Early-Holocene olivine macrocrysts dominantly record Fe-Mg diffusion timescales between 200-  
40 400 days, whilst middle-Holocene and historical units typically record timescales of about 70 and 60  
41 days, respectively. Barometric studies suggest that melts and crystals are likely stored and gradually  
42 transferred throughout an interconnected multi-tiered system that ultimately culminate in a mid-  
43 crustal reservoir(s) at about  $6.8-7.5 \pm 2.5$  km depth, where final disaggregation by the carrier liquid took  
44 place. We argue that, as a result of extensional processes enhanced by rifting events, well-mixed melts

45 got drawn into mid-crustal reservoir(s) causing crystal mush loosening and mobilization. In addition, we  
46 propose that more energy in the form of heat and/or melt supply was required in the early Holocene  
47 to break up the dense mush fabric and convert it into an eruptible magma. Conversely, as evidenced by  
48 the diverse macrocryst content of the historical units and by the lack of gabbroic nodules, the system  
49 has become characterized by a less compact mush fabric since at least the middle-Holocene, such that  
50 fresh injection of melt would easily loosen and mobilize the mush, resulting in an eruption within a  
51 couple of months. This study provides evidence that along axial rift settings, rifting-related processes  
52 can help to “pull the mush apart” with no requirement for primitive magma injection as an eruption  
53 trigger. Furthermore, we provide evidence that in the Bárðarbunga-Veiðivötn volcanic system  
54 specifically, the time between mush disaggregation and eruption has decreased considerably with time,  
55 indicating shorter warning times before imminent eruptions.

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## 68 **1. INTRODUCTION**

69 Understanding the time and duration of magmatic processes has been a central topic in volcanology,  
70 as time is a critical parameter to assess volcanic risk (Sparks and Sigurdsson 1977). Great strides have  
71 been made over the past decades in developing diffusion chronometry as a fundamental tool to retrieve  
72 time information of magmatic processes recorded in minerals (Martin et al. 2008; Costa and Morgan  
73 2010; Kahl et al. 2011, 2013, 2015, 2017; Ruprecht and Plank 2013; Cooper and Kent 2014; Viccaro et  
74 al. 2016; Hartley et al. 2016; Dohmen et al. 2017; Mutch et al. 2019a, 2019b).

75 The chemistry of igneous minerals reflects the environment(s) under which they grew, and any  
76 perturbation of pressure, temperature, melt composition or oxygen fugacity might be recorded by their  
77 composition (Ginibre et al. 2007; Blundy and Cashman 2008; Streck 2008). Changing growth conditions  
78 result in chemical zoning in minerals. At magmatic temperatures, compositional gradients tend to be  
79 smoothed out via diffusion as long as the system stays above the closure temperature. Diffusion  
80 chronometry is used to model the re-equilibration of different elements across compositionally distinct  
81 zones in crystals to derive the time that elapsed between the onset of the perturbation in growth  
82 conditions (e.g. magma injection) and the point at which diffusion effectively ceased to play a significant  
83 role (e.g. volcanic eruption) (Turner and Costa 2007; Costa et al. 2008; Costa and Morgan 2010; Dohmen  
84 et al. 2017). Diffusion chronometry in plagioclase, olivine and pyroxene crystals has been widely applied  
85 to estimate timescales of mixing processes and recharge events (Costa and Chakraborty 2004; Kahl et  
86 al. 2011, 2015; Ruprecht and Cooper 2012; Chamberlain et al. 2014; Rae et al. 2016; Viccaro et al. 2016;  
87 Rasmussen et al. 2018), magma ascent rates (Demouchy et al., 2006; Hartley et al., 2018; Mutch et al.,  
88 2019a; Ruprecht and Plank, 2013), cooling rates (Coogan et al. 2005; Faak et al. 2013; Sio et al. 2013;  
89 Newcombe et al. 2014) and crystal mush disaggregation timescales (Costa et al. 2010; Cooper and Kent  
90 2014; Moore et al. 2014; Hartley et al. 2016; Pankhurst et al. 2018; Nikkola et al. 2019; Mutch et al.  
91 2020).

92 In Iceland, magma storage bodies occur over a large range of depths (Neave et al. 2013; Haddadi  
93 et al. 2017; Neave and Putirka 2017; Hartley et al. 2018; Caracciolo et al. 2020) and understanding the  
94 rate of magma transfer between the different active storage zones in such active magmatic systems is  
95 vital for the interpretation of signals of volcanic unrest. The 2010 Eyjafjallajökull eruption provided a  
96 significant step forward in linking petrological and geophysical data: pre-eruptive geophysical signals  
97 (e.g. ground displacement and earthquake frequency) (Sigmundsson et al. 2010) registered between  
98 November 2009 and March 2010 are consistent with the onset of magma injection in the Eyjafjallajökull  
99 system obtained through diffusion chronometry on zoned olivine crystals (Pankhurst et al. 2018).

100 Diffusion chronometry applied to recent Icelandic eruptions indicates very short timescales of  
101 magma mixing (Viccaro et al. 2016) and crystal mush disaggregation (Hartley et al. 2016; Pankhurst et  
102 al. 2018; Nikkola et al. 2019) beneath Icelandic volcanoes. However, these studies mainly focus on  
103 recent and single eruptions in different volcanic systems and it is still unknown whether timescales of  
104 magmatic processes are uniform within the lifespan of a single volcanic system. Here we employ  
105 diffusion chronometry by modeling the diffusive relaxation of Fe-Mg, Mn and Ni in olivine macrocrysts  
106 to retrieve timescales of crystal mush disaggregation beneath the Bárðarbunga-Veiðivötn volcanic  
107 system in central Iceland. Secondly, we merge diffusion modeling results with prior geobarometric  
108 constraints in order to reconstruct magmatic processes in the Bárðarbunga-Veiðivötn volcanic system  
109 throughout the Holocene. To do this, we have targeted well-characterized magmatic units of different  
110 age (early-Holocene to historical) within the same volcanic system. We determine the time that has  
111 elapsed between magma injection into the crystal mush reservoirs and the subsequent eruption, and  
112 importantly, we discuss how these timescales have changed over the Holocene period.

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## 115 **2. GEOLOGICAL BACKGROUND, SAMPLE LOCALITIES AND PREVIOUS WORK**

116 Iceland is the only place in the world where a mid-ocean ridge intersects a mantle plume directly. The  
117 on-land manifestation of the Mid-Atlantic Ridge is split into four main neovolcanic rift zones (Fig. 1a),  
118 the Reykjanes Volcanic Belt (RVB), the Western Volcanic Zone (WVZ), the Eastern Volcanic Zone (EVZ)  
119 and the Northern Volcanic Zone (NVZ). The Bárðarbunga volcanic system is the largest volcanic system  
120 of the EVZ and it rises above the current presumed centre of the mantle plume (Bjarnason 2008) (Fig.  
121 1a). The southwesternmost segment of the Bárðarbunga volcanic system is commonly referred to as  
122 the Bárðarbunga-Veiðivötn volcanic system and it consists of a fissure swarm that fades out towards  
123 the south into the Torfajökull volcanic system (Thordarson and Larsen 2007; Larsen and Guðmundsson  
124 2014).

125 The volcanic units in the Bárðarbunga-Veiðivötn volcanic system considered in this study are  
126 represented by a suite of well-characterized samples, which have previously been studied by Caracciolo  
127 et al. (2020). Here we summarize key findings of that study and we guide the reader to this work for  
128 further details on these units and the geology of the area. The sampled units are: Brandur, Fontur and  
129 Saxi (early Holocene), Þjórsárdalshraun and Drekahraun (middle Holocene) and Veiðivötn 1477  
130 (historical) (Fig. 1b). Brandur, Fontur and Saxi are three early-Holocene tephra cones and the samples  
131 consist of fresh, macrocryst-rich, glassy tephra material and highly crystalline gabbroic nodules.  
132 Þjórsárdalshraun and Drekahraun are dated as middle-Holocene units (3-4 ka, Pinton et al. 2018) and  
133 the samples come from lava and fresh vesicular scoria, respectively. Veiðivötn 1477 was produced in a  
134 65 km long fissure eruption that occurred in 1477 AD (Fig. 1b). Caracciolo et al. (2020) found that all  
135 samples contain macrocrysts and polymineralic clots, indicative of a crystal mush origin. The  
136 compositions of the macrocrysts vary with time, with early-Holocene magmatic units containing more

137 primitive compositions compared to middle-Holocene and historical magmatic units. Application of  
138 different thermobarometers to crystals, groundmass glasses and melt inclusions (MIs) revealed  
139 temporally invariant crystallization conditions in the middle crust, at about 1.9-2.2 kbar (6.8-7.8 km),  
140 along with the occurrence of deep-seated storage reservoir(s) during the time covered by the oldest  
141 magmatic units. This deep crystallization record has been linked to a different crustal response  
142 associated with post-glacial isostasy rebound (Gee et al. 1998; Sinton et al. 2005; Le Breton et al. 2016).

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### 145 **3. SAMPLE PREPARATION AND ANALYTICAL METHODS**

146 Unaltered olivine macrocysts in the size range 0.5-2.4 mm were hand-picked from crushed tephra and  
147 lava samples and mounted in epoxy resin to expose a flat surface. High-contrast backscattered electron  
148 (BSE) images were taken of olivines using the JEOL JXA-8230 SuperProbe electron probe microanalyzer  
149 (EPMA) at the Institute of Earth Sciences, University of Iceland. ImageJ software was used to process  
150 the BSE images to identify crystals with zoning patterns and to select traverses suitable for diffusion  
151 modeling.

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#### 153 **3.1. Electron microprobe analyses (EMPA)**

154 Olivine concentration profiles of major and minor elements (Si, Ti, Al, Cr, Fe, Mn, Ni, Mg, Ca) were  
155 acquired using the EPMA at the University of Iceland. Chemical transects were measured using a  
156 focused beam with an accelerating voltage of 15 kV and a probe current of 20 nA. A counting time of  
157 30 s at the peak and background was used for Ti, Al, Mn, Cr and Ni. Si, Fe, Mg and Ca were analyzed  
158 using a counting time of 30 s at peak and 15 s at the background. Chemical profiles with point spacing  
159 between 3-6  $\mu\text{m}$  (25 to 100 points) were measured perpendicular to macrocryst margins along different

160 directions in a total of 121 olivine grains from all studied eruptive units. Multiple standard samples were  
161 analyzed prior to and after each session. Error bars reported in the chemical diagrams indicate  $2\sigma$  of  
162 multiple standard analyses ( $2\sigma_{\text{Fe}}=0.34$  mol%,  $2\sigma_{\text{Mn}}=0.038$  wt%,  $2\sigma_{\text{Ni}}=0.018$  wt%). In order to test for  
163 equilibrium conditions, groundmass glasses immediately around olivine macrocrysts were analyzed  
164 with the EPMA. In this study, we include some olivine-hosted MIs ( $n=8$ ) analyzed by Caracciolo et al.  
165 (2020), along with new MI data ( $n=13$ ) (supplementary material S1). See Caracciolo et al. (2020) for  
166 analytical details about MI analyses and post-entrapment process correction.

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### 168 **3.2. Electron backscatter diffraction (EBSD)**

169 Crystallographic orientations of olivine macrocrysts were obtained using electron backscatter  
170 diffraction (EBSD; Prior et al., 1999; Costa and Chakraborty, 2004) on the FEI Quanta 650 FEG-SEM at  
171 the University of Leeds Electron Microscopy and Spectroscopy Centre (LEMAS). Characterizing the  
172 crystallographic directions in olivine with respect to the micro-analytical traverses is essential for  
173 accurate diffusion modeling, as the diffusivity of different elements (e.g. Fe–Mg or Ni) in olivine is  
174 strongly anisotropic, with diffusion along the c-axis being six times faster than that along the a- and b-  
175 axes (e.g. Clark and Long 1971; Dohmen et al. 2007).

176 To minimize uncertainty in the determination of the orientation data, orientation maps consisting of  
177 hundreds of EBSD point determinations were conducted for each macrocryst (Kahl et al., 2017). Maps  
178 were acquired using an accelerating voltage of 20 kV and a spot size of 5.5  $\mu\text{m}$ . EBSD maps over crystal  
179 areas of interest were generated and processed using the HKL CHANNEL5 EBSD post-processing  
180 software, enabling the extraction of hundreds to thousands of orientation measurements. Measured  
181 Euler angles were converted into trends and plunges of the olivine crystallographic a-, b- and c-axes  
182 using Stereo32 software developed at the Ruhr-Universität Bochum. Using this software, it is possible



183 to calculate the angles between the measured electron microprobe traverses and the crystallographic  
184 a-, b- and c-axes in olivine (Supplementary material S2) (Costa and Chakraborty 2004; Kahl et al. 2011).

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## 187 **4. KINETIC MODELING**

188 At high temperatures, compositional heterogeneities in magmatic crystals will homogenize due to  
189 diffusive relaxation. Following the modeling approach outlined in Kahl et al. (2011, 2013) and Costa and  
190 Chakraborty (2004), we modeled the diffusive relaxation of Fe-Mg, Ni and Mn zoning in olivine  
191 macrocrysts from all studied localities. Details of the methods and the criteria for the choice of  
192 concentration profiles for modeling, including tests for robustness, have been reported in full detail by  
193 Costa et al. (2008) and Kahl et al. (2011, 2013).

194 Among 121 olivine grains representing all eruptive units (Table 1), 21 grains show dipping plateaux  
195 (Shea et al. 2015) or zoning patterns probably related to crystal growth (Costa et al. 2008). These grains  
196 are not suitable for olivine diffusion modeling and were excluded from further consideration. For the  
197 diffusion models, the spacing was set to 5 $\mu$ m, which is a good compromise between calculation speed  
198 and EMPA resolution of the analyzed diffusion profiles. For each crystal, the modelling curves were  
199 calculated by manually changing the number of time steps until a best-fit curve for a given Fe-Mg  
200 diffusion profile was observed. Therefore, depending on the zonation pattern, the number of time steps  
201 was different for each crystal. Subsequently, we used the same number of time steps and the identical  
202 initial conditions to model diffusion profiles of Mn and Ni (Fig. S1.4). In most of the cases, best-fit curves  
203 (and comparable timescales) were observed for Fe-Mg, Mn and Ni (Fig. S1.4), suggesting that zoning  
204 profiles are likely to be controlled by diffusion.

### 205 **4.1. Modeling approach**

206 We used the composition-dependent, one-dimensional expression of the diffusion equation (i.e. Fick's  
207 second law):

$$208 \quad \frac{\partial C_i(x, t)}{\partial t} = \frac{\partial}{\partial x} \left( D_i \frac{\partial C_i(x, t)}{\partial x} \right)$$

209 where  $C_i$  is the concentration of element  $i$ ,  $x$  denotes the distance,  $D_i$  is the diffusion coefficient of  
210 element  $i$ , and  $t$  is time. The evolution of the concentration with time ( $t$ ) at different spatial coordinates  
211 ( $x$ ),  $C_i(x, t)$ , is obtained numerically using a one-dimensional finite difference scheme (see Costa et al.  
212 2008). In this study, we mainly focused on Fe-Mg but also on Ni and Mn zoning when possible, to  
213 increase the robustness of the calculated timescales. Diffusion coefficients are functions of pressure,  
214 temperature, composition, oxygen fugacity and crystallographic orientation, and were taken from  
215 experimentally determined data (Fe–Mg and Mn: Dohmen et al., 2007; Ni: Petry et al., 2004 and  
216 Holzapfel et al., 2007).

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## 218 **4.2. Modeling parameters and error propagation**

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### 220 *4.2.1. Initial and boundary conditions*

221 The initial condition refers to the shape of the zoning pattern before diffusive modification. The  
222 strategies outlined in Kahl et al. (2011) were adopted to identify initial conditions for each profile. In  
223 most cases, we assumed a homogeneous initial profile (Fig. S1.1a), guided by the occurrence of  
224 extended compositional core plateaux. Only in a few cases, plateaux at the rims (Fig. S1.1b) were  
225 observed. These were interpreted as stranded diffusion profiles and a stepped initial profile shape was  
226 invoked.

227 In our modeling, we assume that macrocrysts re-equilibrated with their host liquid (i.e. open boundary  
228 conditions) and that the composition of the liquid is constant with time within a specific magmatic unit.

#### 229 *4.2.2. Input parameters*

230 Clinopyroxene-liquid, OPAM and liquid-only thermobarometry constraints suggest that the final  
231 equilibration depth of the carrier liquids before eruption and olivine rim crystallization occurred in the  
232 mid-crust at about 1.1-2.7 kbar pressure, at temperatures of 1160-1175 °C (Caracciolo et al. 2020).  
233 Therefore, we have modeled the chemical zoning with temperature and pressure set according to the  
234 magmatic unit under consideration (Caracciolo et al. 2020). A summary of input temperature and  
235 pressure conditions is reported in Table 1. Oxygen fugacity was set to the FMQ (Fayalite-Magnetite-  
236 Quartz) buffer based on spinel-olivine oxybarometry.

#### 237 *4.2.3. Error propagation*

238 Timescale uncertainties have been calculated following the method described by Kahl et al. (2015), with  
239 temperature showing the largest effect on the propagated error. For instance, a 1 $\sigma$  temperature  
240 uncertainty of  $\pm 5$  °C yields errors of  $\pm 134$  days for a calculated diffusion timescale of 1000 days,  $\pm 13.4$   
241 days for a timescale of 100 days and  $\pm 1.4$  days for a timescale of 10 days.

242

## 243 **5. RESULTS**

### 244 **5.1. Petrography of the samples**

245 Among all studied samples, plagioclase is the principal macrocryst (>0.5 mm) phase, followed by olivine  
246 and clinopyroxene. Early-Holocene units consist of tephra and high-crystallinity gabbroic nodules.  
247 Tephra are slightly vesicular (~15-20 %) and contain between ~15 and ~45 vol% macrocrysts (Fig. 2a).  
248 Nodules (Fig. 2b) consist of pseudo-spherical olivine gabbros (up to 10 cm in diameter) with a

249 crystallinity between ~80-95%, and they are made of a network of plagioclase macrocrysts (~80-85%),  
250 interspersed with olivine macrocrysts (~5-10%), clinopyroxene (~5%) and interstitial glass (~5-10%) (Fig.  
251 2 b-c). Olivine crystals found in the gabbroic nodules are morphologically and texturally similar to those  
252 found in the tephra samples. Lava and scoria samples from middle-Holocene units show variable  
253 vesicularity (50-80 %) and a low abundance of macro- and microcrysts (Fig. 2d). The macrocryst content  
254 ranges between ~5-15 %, with plagioclase macrocrysts being more abundant than olivine and  
255 clinopyroxene (Fig. 2d). Historical samples consist of fresh, highly vesicular (~70-90 %), crystal-poor  
256 scoria with the most crystalline samples having ~5-10 % macrocrysts. Detailed sample petrography is  
257 provided by Caracciolo et al. (2020).

258

259

## 260 **5.2. Olivine - chemistry and zoning**

261 We have measured a total of 173 chemical profiles in 121 olivine macrocrysts from all localities (Table  
262 1). The olivine core and rim compositions range between  $Fo_{76-87}$  and  $Fo_{74-84}$  [ $Fo=100 \cdot Mg / (Mg + Fe)$ , in  
263 mol%], respectively, although we find systematic compositional differences between eruptions (Fig. 3).

264 Brandur, Fontur and Saxi olivine macrocryst cores record the smallest compositional variation,  
265 whilst rims show the largest range among all magmatic units (Fig. 3a). Olivine cores from Brandur are  
266 in the range  $Fo_{84.5-86.5}$  and rims in the range  $Fo_{77-81}$ . Fontur olivines have core and rim compositions of  
267  $Fo_{83.5-87.5}$  and  $Fo_{76-80.5}$ , respectively. Saxi olivine macrocrysts preserve cores of  $Fo_{85-86.5}$  and rims of  $Fo_{77.5-}$   
268  $79$ . Olivine cores from Fontur and Saxi gabbroic nodule samples display a larger Fo range ( $Fo_{80-87.5}$ ) than  
269 olivine cores in tephra samples ( $Fo_{85-87}$ ) (Fig. S1.2). Brandur olivines record remarkably homogeneous  
270 core compositions within gabbroic nodule and tephra samples, with 1 mol% difference between the  
271 two. Most of the tephra olivines from Brandur are slightly more primitive ( $Fo_{\sim 86}$ ) than nodule olivines

272 (Fo<sup>~85</sup>) (Fig. S1.2), although this difference is not observed in Fontur and Saxi. Drekahraun and  
273 Þjórsárdalshraun record similar olivine core composition range (Fo<sub>80-85.5</sub>), while olivine rims are in the  
274 range Fo<sub>75-81</sub> and Fo<sub>81-83</sub>, respectively (Fig. 3a). Olivine macrocrysts from the Veiðivötn 1477 eruption  
275 display core compositions in the range Fo<sub>80-86.5</sub> and rims in the range Fo<sub>78-81</sub>. The majority of olivine  
276 macrocrysts display normal zoning (Fig. 3a and Fig. S1.1a), with forsterite-rich cores surrounded by less  
277 forsteritic rims. Two macrocrysts from Veiðivötn 1477 and one macrocryst from Drekahraun display  
278 reverse zonation (Fig. 3a and Fig. S1.1b), with the latter showing a strong compositional gradient  
279 between core (Fo<sub>76</sub>) and rim (Fo<sub>82</sub>). Some olivine macrocrysts from Drekahraun (n=4), Þjórsárdalshraun  
280 (n=3) and Veiðivötn 1477 (n=9) show complex zoning patterns with reversely zoned macrocryst  
281 interiors followed by normally zoned outermost rims (Fig. S1.1c). The difference in Fo content between  
282 the core and the intermediate zone is always lower than 1 mol% but still above the analytical  
283 uncertainty ( $2\sigma_{Fo} = 0.34$ ). This intermediate pattern is not recorded by other elements. Finally, we do  
284 not observe any unzoned olivine macrocrysts in the magmatic units discussed in this work (Fig. 3a).

285 At any given Fo content, olivine cores from early-Holocene, middle-Holocene and historical units  
286 do not show any difference in terms of Mn and Ni. Primitive olivine cores (Fo<sub>85-87</sub>) from early-Holocene  
287 samples have Mn contents in the range 1400-1700 ppm and Ni contents in the range 1200-1700 ppm.  
288 Less primitive (Fo<sub>78-82</sub>) olivine cores are found in the historical samples and they record Mn contents  
289 between 2000-2500 ppm and Ni contents between 800-1300 ppm. The measured Mn and Ni contents  
290 overlap with those of olivine macrocrysts from the 2014-15 Holuhraun eruption (Halldórsson et al.  
291 2018) (Fig. S1.3).

292

### 293 **5.3. Relation to the carrier liquid**

294 Each eruptive unit is characterized by a specific range in groundmass glass compositions (Fig.  
295 3b), which is assumed to represent the carrier liquid composition. Early-Holocene groundmass glasses  
296 have the largest chemical variability (MgO 5.5-7.5 wt%, TiO<sub>2</sub> 1.0-2.5 wt%), whereas recent units are  
297 more homogeneous (Caracciolo et al., 2020). Middle-Holocene carrier liquids have MgO and TiO<sub>2</sub>  
298 contents in the range 7.0-7.5 wt% and 1.6-1.8 wt% respectively, while historical carrier liquids are in  
299 the range 6.3-6.9 wt% for MgO and 1.6-2.0 wt% for TiO<sub>2</sub>. The melt Mg# ( $Mg\# = 100 * Mg / (Mg + Fe^{2+})$ ) of  
300 the carrier liquid was obtained by calculating Fe<sup>2+</sup> and Fe<sup>3+</sup> contents following the model of Kress and  
301 Carmichael (1991) at the FMQ buffer. Following this procedure, 91% of olivine macrocryst rims are in  
302 chemical equilibrium with the carrier liquid (Fig. 3b).

303

#### 304 **5.4. Diffusion modeling results**

305 We modeled 100 olivine crystals with zoning patterns related to diffusive relaxation of Fe and Mg and  
306 well-suited for diffusion modelling (Fig. 4). Independent Fe-Mg timescale results for each locality are  
307 reported in Fig. 5 and in S1, while a summary is listed in Table 1. Kernel density estimate (KDE)  
308 distribution curves are shown in Fig. 6.

309

##### 310 *5.4.1. Brandur, Fontur and Saxi (early-Holocene)*

311 Olivine macrocrysts from the three tephra cones, Brandur, Fontur and Saxi, record a large range of  
312 timescales, varying from 20 to 1330 days (Fig. 5a-c and Fig. 6a). The large range of timescales is observed  
313 both, in olivines found in gabbroic nodules and in tephra samples (Fig. 5a-b). KDE distribution curves  
314 largely overlap (Fig. 6a), with the majority of olivines (63%) recording timescales between 150 and 400  
315 days (0.4-1.1 years). Only 17% of the olivines give timescales shorter than 150 days and 20% longer than  
316 400 days. Brandur olivines (n=29) show a narrowly focused major peak at 340 days. Fontur olivines

317 (n=16) display the widest timescale distribution, with a major peak located at 444 days and a large  
318 shoulder with timescales between ~100 and ~230 days (n=5). The shortest timescale of 19 days is  
319 recorded by one macrocryst from Fontur (Fig. 5b). Finally, Saxi olivines (n=11) show most probable  
320 timescales of 210-350 days (Fig. 6a), with a main peak at 220 days, and a minor band located at a  
321 timescale >800 days (n=2).

322

#### 323 *5.4.2. Þjórsárdalshraun and Drekahraun (middle-Holocene)*

324 Diffusion timescales calculated for Þjórsárdalshraun olivines show a scattered KDE distribution (Fig. 6b).  
325 Timescales are all below 110 days (Fig. 4d), with a major peak at 24 days. However, the Þjórsárdalshraun  
326 samples come from a section of the lava that had flowed about 50-60 km before ultimately forming a  
327 rootless cone field. For this reason, olivines from this unit are likely affected by post-eruptive diffusion  
328 and/or crystallization processes and timescale results are treated with caution. Indeed, only a few  
329 macrocrysts (n=7) suitable for diffusion measurements were found within the Þjórsárdalshraun samples  
330 and sample bias might affect the distribution. In contrast, samples from the Drekahraun unit consist of  
331 fresh scoria. KDE timescale distribution for Drekahraun olivines show a prominent peak at 78 days and  
332 a minor peak at 30 days (Fig. 5e and Fig. 6b). Only one macrocryst records a timescale above 100 days  
333 (118 days). Intermediate zoning patterns observed in some middle-Holocene macrocrysts were  
334 modeled sequentially and return diffusion timescales between 2-29 days.

335

336

337

#### 338 *5.4.3. Veiðivötn 1477 (historical)*

339 83% of olivine macrocrysts from Veiðivötn 1477 register diffusion timescales shorter than 100 days (Fig.  
340 5f), with a main peak in the KDE distribution at 55 days (Fig. 6c). A minor peak is observed at 15 days,  
341 while only 4 macrocrysts out of 22 record timescales longer than 100 days (Fig. 5f). Finally, the  
342 intermediate chemical zoning found in some macrocrysts gives an average timescale of 25 days.

343

## 344 **6. DISCUSSION**

### 345 **6.1. Relationship between melt composition, magma depth and timescales**

346 Barometry constraints show that the carrier liquid last equilibrated with the crystal cargo in a  
347 reservoir(s) located in the middle-crust (7-11 km depth), before being erupted (Caracciolo et al. 2020).  
348 The cores of 21 olivine macrocrysts contain large (up to 120  $\mu\text{m}$  in diameter), naturally quenched melt  
349 inclusions (e.g. Fig. 4g and Fig. S1.4i), which are mostly hosted in early-Holocene units (Fig. 7a). These  
350 melt inclusions have MgO contents between 7.6 and 9.2 wt% and  $\text{TiO}_2$  in the range 0.8-1.8 wt% (Mg#55-  
351 65). We estimated MI equilibration pressures within the Bárðarbunga-Veiðivötn plumbing system by  
352 applying the Olivine-Plagioclase-Augite-Melt (OPAM) barometer (Yang et al. 1996; Hartley et al. 2018)  
353 with the goal to locate the macrocrysts within the crust. MI equilibration pressures (n=3) of middle-  
354 Holocene and historical samples range between 1.8 and 1.9 kbar (Fig. 7a) and they are hosted in olivine  
355 cores with compositions in the range  $\text{Fo}_{81.6-84.6}$ . However, MIs hosted in plagioclase macrocrysts record  
356 pressures down to 4 kbar (Caracciolo et al. 2020). Early-Holocene units register a much larger pressure  
357 variation, between 1.5 and 6 kbar, with a clear positive relationship between olivine composition, MI  
358 composition and MI equilibration pressure (Fig. 7a). The olivine crystals become more evolved as MI  
359 equilibration pressures decrease, with primitive olivines ( $\text{Fo}>86$ ) recording the highest pressures (4-6  
360 kbar) and the most primitive MI compositions (Mg# 64-65) (Fig. 7a). These findings, which are in good



361 agreement with the data of Caracciolo et al. (2020), support a stacked-sills model (Kelemen et al. 1997;  
362 Maclennan 2019) in which olivine macrocrysts are exposed to slightly more evolved environments as  
363 they move shallower in the crust (Fig. 8).

364 The shortest diffusion timescales (6-108 days) are recorded by macrocrysts from middle-  
365 Holocene and historical units, which likely crystallized at mid-crustal (7-11 km) levels (Fig. 7b and  
366 Caracciolo et al. 2020). In contrast, early-Holocene olivines cluster at timescales between 200-400 days  
367 (Fig. 7b) and they most likely crystallized throughout the crustal section. Although there is a positive  
368 correlation between early-Holocene olivine depths and their compositions (Fig. 7a), we do not observe  
369 any clear relationship between MI equilibration pressure and olivine diffusion timescales (Fig. 7b) nor  
370 between olivine Fo content and diffusion timescales (Fig. S1.5). This suggests that olivine macrocrysts  
371 are likely to have been transported up to mid-crustal (7-11 km) mush piles during earlier remobilization  
372 events throughout a multi-tiered system of interconnected stacked sills in which crystals are gradually  
373 transported to higher levels until they accumulate in the main mid-crustal reservoir at about 7-11 km.  
374 Indeed, the absence of intermediate zoning sectors may indicate that these macrocrysts resided in the  
375 mid-crust for a period sufficiently long to erase any chemical gradient previously formed. Eventually, at  
376 some point in the early-Holocene, injection of melts with variable composition (Caracciolo et al. 2020)  
377 into the mid-crustal reservoir(s) caused remobilization of these deep-origin olivines and their  
378 incorporation in the carrier melt.

379

## 380 **6.2. Timing of crystal mush disaggregation and triggering mechanism**

381 Gabbroic nodules found in early-Holocene magmatic units are unique samples whose  
382 mineralogical and textural characteristics indicate that they represent exhumed pieces of the crystal  
383 mush (Hansen and Grönvold 2000; Caracciolo et al. 2020). The lack of notable chemical differences

384 between olivines in the gabbroic nodules and in the tephra samples (Fig. S1.2) support their common  
385 origin. Except for Brandur samples (Fig. S1.2), no notable differences in composition are observed  
386 between olivines coming from the mush and olivines carried in the melt as a result of disaggregation  
387 processes (Fig. 5a-c). While the majority of olivine rim compositions are found to be in chemical  
388 equilibrium with the carrier liquid (Fig. 3b), olivine cores are not. They most likely crystallized from a  
389 set of compositionally diverse primitive melts, as supported by MI data (Caracciolo et al. 2020). In most  
390 cases, olivine cores are homogenous, forming long compositional plateaux, which can extend up to 200  
391  $\mu\text{m}$  within the largest grains (Fig. 4b, h). This observation suggests long residence time whereby olivine  
392 cores have either been re-equilibrated completely through diffusion or they have grown in a reservoir(s)  
393 under steady chemical and physical conditions before rim crystallization occurred. In contrast, some  
394 olivine cores in the more recent samples (middle-Holocene and historical) record more complex zoning  
395 patterns, with Fo variations of  $\sim 1$  mol% (Fig. S1.1c). These jumps likely relate to changes in the intensive  
396 thermodynamic variables (composition, pressure, temperature and/or oxygen fugacity) of the  
397 magmatic environment (e.g. Kahl et al. 2017), so that the next increment of olivine that grew had a  
398 slightly different composition.

399         The decrease in the Fo content at the rims is an evidence for changes in the chemical and or  
400 physical conditions of the reservoir. We infer that the outermost normal zoning is related to interaction  
401 of a relatively evolved melt (e.g. the final carrier liquid) within an undisturbed crystal mush reservoir(s);  
402 the evolved liquids got injected into the reservoir and permeated the mush, thereby starting the  
403 diffusion clock after efficient mixing was achieved (Fig. 8). This process most likely caused  
404 disaggregation of mush clots and addition of mush crystals (Sigmundsson et al. 2020) into the carrier  
405 melt. If this is the case, then diffusion timescales correspond to the time elapsed between the  
406 disaggregation of crystal mush following magma injection, when chemical re-equilibration with the

407 carrier liquid started, and the eruption, when diffusion ceased to play a role (Fig. 8). Specifically, the  
408 composition of the tholeiitic basalt magma being injected into the mush reservoir has not remained  
409 constant over the Holocene period (see 5.2 above and Fig. 1. in Caracciolo et al. 2020).

410 Olivine macrocrysts with complex and reverse zoning patterns are widespread in off-rift  
411 Icelandic settings and their zoning features have been associated with late mafic magma replenishment  
412 events (e.g. Mattsson and Oskarsson 2005; Pankhurst et al. 2014; Viccaro et al. 2016), which could have  
413 triggered the eruptions. In contrast, rift-related olivine macrocrysts commonly preserve normal zoning  
414 patterns (e.g. Neave et al. 2013, 2015; Thomson and Maclennan 2013; Hartley et al. 2016; Halldórsson  
415 et al. 2018), which could indicate a different triggering mechanism. In fact, about 95% of the olivine  
416 macrocrysts studied in this work do not preserve direct evidence for late mafic recharge events. It is  
417 possible that slightly deeper, well-mixed and relatively evolved melts pockets could get remobilized and  
418 intrude the mid-crustal reservoir as a result of rift-related events that would “pull the mush apart”,  
419 promoting mush disaggregation and the vertical rearrangement of crystal and melt layers within the  
420 crust. If so, rifting-related events can play a fundamental role on eruption triggering mechanisms along  
421 axial rift zones.

422

### 423 **6.3. Temporal variation of crystal mush disaggregation timescales in the Bárðarbunga-Veiðivötn** 424 **volcanic system**

425 The older Holocene macrocrysts appear to have resided longer in their carrier liquid than the  
426 younger macrocrysts (Fig. 6 and S1.5). One noteworthy observation is that early-Holocene units contain  
427 mush nodules and a larger macrocryst load than the younger units (Fig. 2 and Fig. 9). The gabbroic  
428 nodules contain up to 90 vol% macrocrysts and confirm the existence of a crystal-rich mush fabric. In  
429 Fig. 9 we show diffusion modeling timescales yielded by olivine macrocrysts in relation to the estimated

430 macrocryst content of the samples for each studied time period. We also report diffusion durations for  
431 the subglacial Skuggafjöll eruption (Mutch et al. 2020) that occurred in the Bárðarbunga-Veiðivötn  
432 volcanic system and which shares many features with our samples. The Skuggafjöll products consist of  
433 a highly phyrlic (6-45% by volume), plagioclase-rich basalts (Neave et al. 2014) that crystallized at about  
434  $11 \pm 4$  km (Neave and Putirka 2017). Subglacial and early-Holocene units record the largest timescale  
435 spread and the most abundant crystal content (up to 45 vol%). On the other hand, middle-Holocene  
436 and historical units preserve a smaller macrocryst content (5-15 vol %) than early-Holocene and  
437 subglacial units, which is coupled with much shorter timescales and narrower timescale ranges (Fig. 9).  
438 Therefore, we suggest that during subglacial and early-Holocene times the mid-crustal mush network  
439 required more time to be converted into an eruptible magma (Cashman et al. 2017) because of the  
440 occurrence of a more compact mush fabric. It is possible that multiple injections of melt fractions were  
441 needed to eventually mobilize the rigid network of the mush reservoir(s) (Fig. 8a-b), causing long  
442 residence times of macrocrysts in the reservoir(s). Furthermore, we find important to highlight that  
443 crystals found in the gabbroic nodules and crystals found in the tephra samples reveal the same  
444 timescales (Fig. 5). Those units were sourced from packed mush piles, which required more heat or a  
445 larger volume of intruding magma to be weakened and mobilized compared to the present. The larger  
446 volume of magma or energy (i.e. heat) supply is consistent with the increase of magma production rates  
447 observed in Iceland during the early Holocene that were likely caused by glacio-isostasy effects (Sinton  
448 et al. 2005; Le Breton et al. 2016; Caracciolo et al. 2020). In contrast, the present-day system is most  
449 likely characterized by a less dense mush fabric and a larger melt-mineral ratio (Fig. 8c-d). If this is true,  
450 then since the middle-Holocene, injections of smaller melt fractions would be enough to mobilize the  
451 mush network almost instantly (Fig. 8d), with magma flowing towards the surface within short  
452 timescales. The shortening of timescales approaching the present day is consistent with magma

453 transport durations of about 1-12 days (Fig. 10), estimated for the 2014-15 Holuhraun lava (Hartley et  
454 al. 2018) that was emplaced in the northern segment of the Bárðarbunga volcanic system.

455

#### 456 **6.4. Timescales of magmatic processes in Iceland and MOR settings**

457 Our study is the first to obtain diffusion timescales for a temporally diverse magmatic suite  
458 erupted within the same volcanic system (Fig. 10). Our results can be combined with diffusion  
459 timescales estimated for the subglacial Skuggafjöll eruption (Mutch et al. 2020) to evaluate the  
460 temporal evolution of crystal mush disaggregation from the subglacial period until historical time within  
461 the Bárðarbunga-Veiðivötn volcanic system. Diffusion modeling of plagioclase and olivine macrocrysts  
462 from Skuggafjöll yield most probable timescales in the range 50-400 days (Mutch et al. 2020),  
463 suggesting that mush disaggregation processes started about a year or less prior to the eruption. These  
464 estimates are in good agreement with the early-Holocene timescales calculated in this study and  
465 consistently longer than middle-Holocene and historical mush disaggregation times (Fig. 10).

466 To our knowledge, published diffusion chronometry data for Icelandic volcanoes ascribed to  
467 crystal mush disaggregation processes only concern single eruptions that occurred in the EVZ. Diffusion  
468 kinetic data are available for a subglacial unit erupted in the Bárðarbunga-Veiðivötn volcanic system  
469 (Mutch et al. 2020), for the 2014-15 Holuhraun lava flow emplaced in the northern segment of the  
470 Bárðarbunga volcanic system (Hartley et al. 2018), for the 1783-84 Laki eruption of the Grímsvötn  
471 volcanic system (Hartley et al. 2016) and for the 2010 (Pankhurst et al. 2018) and middle-Pleistocene  
472 (Nikkola et al. 2019) Eyjafjallajökull eruptions in the southernmost section of the EVZ. With the  
473 exception of Skuggafjöll (Mutch et al. 2020), these studies reveal short diffusion times between mush  
474 disaggregation and entrainment prior to eruption, on the order of days up to 1-2 months (Fig. 10). These  
475 estimates overlap and are consistent with timescales calculated for middle-Holocene and historical

476 units from the Bárðarbunga-Veiðivötn system (Fig. 10). Eyjafjallajökull is the only system studied so far  
477 for which there are timescale estimates available for eruptions of different ages. Timescales between  
478 mobilization of mush macrocrysts and eruption of about 10-30 days were calculated for the 2010  
479 Eyjafjallajökull eruption (Pankhurst et al. 2018), in good agreement with mush mobilization times of  
480 about 9-37 days estimated for the middle-Pleistocene Eyjafjallajökull units (Nikkola et al. 2019).

481 At the present, Iceland is the only place above a slow-spreading ridge where numerous  
482 timescales of mush crystal entrainment to eruption are available and only very few data exist at fast-,  
483 intermediate- and slow-spreading MOR settings (Fig. 10). Timescales between mush disruption and  
484 eruption are on the order of few days for the East Pacific Rise (fast-spreading ridge) (Moore et al. 2014)  
485 and for the Costa Rica Rift (intermediate-spreading ridge) (Costa et al. 2010), while mush disruption  
486 beneath Serocki Volcano (slow-spreading ridge) occurred about 1.5 years before an eruption on the  
487 seafloor (Costa et al. 2010). Mush disruption to eruption timescales estimated for slow-spreading MOR  
488 settings are consistent with timescales obtained from the oldest units from this work. Overall, most  
489 timescales calculated at slow-spreading ridges (Fig. 10) tend to be longer (months to years) than the  
490 ones obtained at intermediate and fast spreading ridges (days). The fact that the time between mush  
491 disintegration and eruption increases with decreasing spreading rates (Zellmer et al. 2012) is consistent  
492 with thickening of the lithosphere and increase in the depth of melt levels at slow-spreading ridges  
493 (Wanless and Shaw 2012). However, further geospeedometric work on intermediate- and fast-  
494 spreading ridges is required to elucidate whether or not there is spreading-rate control of the mush  
495 disaggregation-to-eruption timescales along MOR settings.

496

## 497 **7. Implications**

498 Samples studied in this work include crystalline gabbroic nodules that most likely preserve the  
499 physical and mineralogical features of the mush itself. We modeled Fe-Mg, Mn and Ni diffusion in  
500 olivine crystals from Holocene samples of the Bárðarbunga-Veiðivötn volcanic system in central Iceland  
501 to reconstruct crystal residence time in the carrier melt, which we link to the time elapsed between  
502 melt injection (i.e. mush disaggregation) and eruption.

503 One important implication highlighted by our study is that not every magma replenishment  
504 event results in an eruption. In the early Holocene, a year or more may have elapsed between initial  
505 perturbation of the shallow reservoir(s) and final eruption, but the timescale between perturbation and  
506 eruption appears to get shorter approaching the present day. This difference in crystal residence times  
507 in the carrier liquid appears to be associated with the occurrence of a much denser and more rigid mush  
508 fabric in the early Holocene, which required more energy to be loosened, disaggregated and mobilized.  
509 In terms of hazard assessment, this study highlights that the Bárðarbunga-Veiðivötn volcanic system  
510 has been responding faster to melt injection as time proceeds, providing less warning between magma  
511 injection and eruption. This could possibly indicate the recent establishment of dominant magma  
512 transfer routes along which magma can quickly migrate towards the surface. More studies on the  
513 temporal evolution of magma transport timescales underneath single volcanic systems are needed to  
514 better constrain the behavior of volcanic systems through time, with a direct implication for volcanic  
515 hazard assessment, especially in regions where the last known eruption occurred long before the  
516 installation of modern volcano monitoring networks.

517 The crystal cargos in our samples do not record any evidence of late-stage primitive magma  
518 replenishment that could have triggered the eruption. This feature is observed also in crystals from  
519 other rift settings in Iceland. Primitive melts with different compositions have been supplied to the  
520 Bárðarbunga-Veiðivötn system during earlier stages of its magmatic history (Caracciolo et al. 2020), but

521 these magmas are typically homogenized by concurrent mixing and crystallization (Maclennan 2008;  
522 Caracciolo et al. 2020) before being injected into the shallow crystal mush reservoir. These processes  
523 likely take place in a multi-tiered magmatic system made up of stacked sills in which macrocrysts are  
524 gradually transported upwards and exposed to melts that become more evolved as they move toward  
525 the mid-crustal reservoir(s). A plausible eruption trigger mechanism is that rifting-associated events  
526 can enable the vertical rearrangement of crystal and melt horizons and facilitate ascent of relatively  
527 evolved and mixed melt pockets stored at mid-crustal levels, with no requirement to supply hot and  
528 primitive magmas. Our findings have broad implications for the dynamics of magmatic processes  
529 operating in axial ridge settings, since we emphasize the possibility that rifting events can significantly  
530 control the vertical arrangement of crystal and melt layers.

531

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543 MATLAB® code for Fe-Mg, Ni and Mn diffusion can be requested from the authors.



544

545

546

547

548 **Figure captions**

549 **Fig. 1** a) Map of Iceland showing the neovolcanic rift zones outlined with dashed lines, individual  
550 volcanic systems in yellow (subglacial parts not shown) and the inferred location of the mantle plume  
551 is shown as a dotted circle. b) Hillshade map of the southernmost part of the Bárðarbunga volcanic  
552 system with sample localities and eruptive units indicated with different colors. SVB: Snæfellsnes  
553 Volcanic Belt; RVB: Reykjanes Volcanic Belt; WVZ: Western Volcanic Zone; MIB: Mid-Iceland Belt; EVZ:  
554 Eastern Volcanic Zone; NVZ: Northern Volcanic Zone.

555

556

557 **Fig. 2.** Macroscopic features of studied samples. a) Thin section image of a lava sample from Brandur  
558 tephra cone. Note the occurrence of a nodule-like aggregate made of a large plagioclase (~1 cm) and  
559 olivine grains. Macrocysts are found in a glassy, light brown groundmass. b-c) Macroscopic photo of a  
560 crystalline olivine gabbro nodule from Fontur tephra cone. This gabbroic nodule contains around ~90  
561 vol% macrocrysts, most of which are plagioclase (white greasy macrocrysts), followed by olivines  
562 (yellow-green macrocrysts) and clinopyroxene (dark green macrocrysts). The crystal framework is  
563 permeated by interstitial glass (dark areas). d) Thin section image of a gabbroic nodule from Saxi. The  
564 framework comprises plagioclase macrocrysts with minor olivine macrocrysts. Clinopyroxene crystals  
565 are mostly found in glomerophytic clots in interstitial positions. e) Thin section image from Drekahraun.  
566 Note the lower abundance of macrocrysts compared to Brandur samples. Thin section images were

567 acquired with the use of a high-resolution scanner and the thin sections placed between two polarizing  
568 sheets to display the interference colors of different phases. plg: plagioclase; ol: olivine; cpx:  
569 clinopyroxene. gl: glass

570

571 **Fig. 3.** a) Core versus rim compositions of modeled olivine macrocrysts from all studied magmatic units.  
572 Brandur, Fontur and Saxi olivines have similar and narrow core compositions and more variable rim  
573 compositions. Þjósárdalshraun, Drekahraun and Veiðivötn record distinct rim compositions with a  
574 narrow range, whereas olivine cores show a larger compositional range. Most macrocrysts plot below  
575 the black 1:1 line, meaning that they are normally zoned. b) Rhodes diagram to test for olivine-liquid  
576 equilibrium. Olivine in equilibrium with coexisting liquid compositions should fall in the grey field  
577 indicative of a  $Kd_{(Fe-Mg)}^{ol-liq} = 0.30 \pm 0.03$  (Roeder and Emslie 1970). Rim compositions of the studied  
578 olivine macrocrysts are close to being in equilibrium with the carrier liquid. Colored bars depict  $1\sigma$   
579 compositional variability uncertainty of the carrier liquid of each sample, where the glass composition  
580 is taken as representative of the carrier liquid. The large variation of the Þjósárdalshraun carrier liquid  
581 is due to its microcrystalline nature.

582

583 **Fig. 4.** Representative backscattered electron (BSE) images (a,d,g,j,m,p), concentration profiles with  
584 model fits (b, e, h, k, n, q) and stereographic plots (c, f, i, l, o, r) of studied olivine macrocrysts from all  
585 localities. White arrows in the BSE photos indicate the position of the analytical traverse. Black dashed  
586 lines indicate the assumed initial profile. Red lines are best fit models for the measured compositional  
587 gradient. For each macrocryst the calculated timescale (days) is also reported. The orientation of  
588 crystallographic axes with respect to the direction of the EPMA traverse (red cross) is shown in the  
589 stereographic plots. Analytical error of EPMA data is based on  $2\sigma$  for multiple standard analyses.

590

591 **Fig. 5.** Fe-Mg diffusion modeling timescale results for a) Brandur, b) Fontur, c) Saxi, d) Þjórsárdalshraun,  
592 e) Drekahraun and f) Veiðivötn 1477. Early-Holocene units (Brandur, Fontur and Saxi tephra cones)  
593 record longer and more variable timescales up to 1300 days, compared to other magmatic units.  
594 Timescale uncertainties have been calculated following the procedure described by Kahl et al. (2015).  
595 Olivines found in the gabbroic nodules and in the tephra samples are shown with different markers.  
596 n=number of modeled olivine macrocrysts for each locality.

597

598 **Fig. 6.** Kernel density estimate plots with bandwidth 0.1, showing calculated timescales for a) early-  
599 Holocene units, b) middle-Holocene units and c) historical unit. Early-Holocene units record the largest  
600 timescale variation among all localities, with a main distribution peak between 200-400 days. n=number  
601 of olivine macrocrysts modeled for each locality.

602

603 **Fig. 7.** a) Olivine core compositions plotted versus melt inclusion (MI) OPAM equilibration pressures.  
604 Early-Holocene macrocrysts are separated into olivines from tephra samples (upwards-pointing  
605 triangles) and olivines from gabbroic nodule samples (inverted triangles). Most of the MIs are hosted  
606 in olivine macrocrysts from Brandur (n=10), Fontur (n=3) and Saxi (n=5). A statistically significant linear  
607 correlation ( $R^2=0.70$ ) between MI OPAM pressure, MI composition and olivine Fo is observed among  
608 macrocrysts from early-Holocene units. As they move upwards in the crust, the composition of crystals  
609 and melts becomes more evolved, suggesting storage and evolution across a stacked-sills system. Error  
610 bar refers to the OPAM standard error of estimate (SSE=1.3 kbar). Grey symbols are olivine-hosted MIs  
611 from Caracciolo et al. (2020). b) Olivine diffusion timescales vs MI OPAM pressures within the same

612 macrocrysts. Olivines that have crystallized in deep-seated regions record similar timescales to olivines  
613 formed at mid-crustal levels. Symbols colored according to melt inclusion Mg#.

614

615 **Fig. 8.** Schematic cartoon (not to scale) summarizing processes operating underneath the Bárðarbunga-  
616 Veiðivötn volcanic system in the early-Holocene (a-b) and in the middle-Holocene to present time (c-  
617 d). Our observations suggest the presence of a multi-tiered magmatic system in which crystals are  
618 gradually transported to shallower levels and exposed to more evolved compositions. The main storage  
619 reservoir(s) is located at 7-10 km depth (Caracciolo et al. 2020). a) In the early-Holocene, the mid-crustal  
620 storage zone is composed of a packed and compact mush fabric, containing predominantly primitive  
621 crystals which are transferred throughout stacked-sills at different depths. b) Around 200-400 days  
622 before the eruption, relatively evolved melts located in the middle-crust intruded into the mushy  
623 reservoir(s), marking the onset of olivine equilibration. This intense melt injection, possibly triggered  
624 by rifting events and/or enhanced by increased early-postglacial melt production rates, caused  
625 loosening and mobilization of the packed mush pile(s). The incorporation of macrocrysts and crystalline  
626 gabbroic nodules in the carrier liquid and subsequent vertical transport occurred over long and variable  
627 timescales (~200-400 days). c) Macrocrysts are more evolved in composition and they are more likely  
628 to have formed at mid-crustal levels (1-4 kbar), with no evidences of deep-seated storage reservoirs  
629 (Caracciolo et al. 2020). Since the middle-Holocene, the mid-crustal storage reservoir(s) is composed of  
630 a less rigid mush fabric with relatively evolved crystals being more abundant compared to the early  
631 Holocene. d) Once evolved melt pocket(s) intruded the reservoir, the mush was quickly remobilized  
632 and macrocrysts ascended to the surface within short timescales (days up to a few months). Different  
633 color gradients of crystals and melt layers point out a variation of composition with time and

634 throughout the crust. However, different colors do not reflect real crystal or melt populations. ol:  
635 olivine; plg: plagioclase; cpx: clinopyroxene.

636

637 **Fig. 9.** Estimated timescales in the Bárðarbunga-Veiðivötn volcanic system for different eruptions as a  
638 function of time and macrocryst cargo. Subglacial and early-Holocene units record highly variable  
639 crystal mush disaggregation timescales with the most probable estimates on the order of a few months  
640 to one year. Subglacial and early-Holocene units have the largest macrocryst content (up to 45 vol %)   
641 among all studied localities. Middle-Holocene and historical units record narrow timescale variations in  
642 the range of a few months at most. These more recent magmatic units are associated with relatively  
643 small macrocryst load. The denser mush fabric associated with subglacial and early-Holocene units  
644 could be responsible for the observed longer timescales between melt injection, mush mobilization and  
645 eruption. Most probable timescales for the Skuggafjöll eruption are between 50 and 400 days. Data for  
646 Skuggafjöll are taken from Mutch et al. (2020) and Neave et al. (2014). Sku= Skuggafjöll, B=Brandur,  
647 F=Fontur, S=Saxi, Th= Þjórsárdalshraun, Dr= Drekahraun, V1477= Veiðivötn 1477.

648

649

650 **Fig. 10.** Compilation of crystal mush disaggregation to eruption timescales derived from modeling  
651 diffusive re-equilibration of compositional zoning in magmatic macrocrysts. We report all crystal mush  
652 disaggregation timescale ranges from Iceland (light green field) along with available literature data for  
653 mid-ocean ridge settings (light yellow field). In each case, the light-shaded bar represents the full range  
654 of calculated diffusion times while the inner black bar indicates the location of the most probable band  
655 in a KDE distribution within the time interval. Diffusion timescales modeled in this work are indicated

656 with colored bars. Eruptions within the Bárðarbunga volcanic system are outlined by the black square  
657 and ordered according to time from subglacial to historical units. The volcanic eruptions are listed below  
658 the horizontal axis. Note that the timescale on the vertical axis is logarithmic. From left to right:  
659 Skuggafjöll (Mutch et al. 2020); Brandur, Fontur, Saxi, Þjórsárdalshraun, Drekahraun and Veiðivötn 1477  
660 (this study); Holuhraun 2014-2015 (Hartley et al. 2018); Laki 1783-84 (Hartley et al. 2016);  
661 Eyjafjallajökull 500-720 ka (Nikkola et al. 2019); Eyjafjallajökull 2010 (Pankhurst et al. 2018); Serocki  
662 Volcano (Costa et al. 2010); Cost Rica Rift (Costa et al. 2010); East Pacific Rise 2005-06 (Moore et al.  
663 2014).  $n_{ol}$  = number of olivine crystals modeled for diffusion kinetics;  $n_{plg}$  = number of plagioclase  
664 crystals modeled for diffusion kinetics. <sup>a</sup> Diffusion chronometry calculated with a different method:  
665 timescales are based on H<sup>+</sup> re-equilibration between plagioclase (n=70) and olivine-hosted (n=9) melt  
666 inclusions with a more hydrous melt. <sup>b</sup> Authors do not specify timescale ranges. For those eruptions,  
667 the interval between crystal mush disaggregation and eruption is  $\leq 1.5$  years (Serocki Volcano),  $<10$   
668 days (Costa Rica Rift) and  $<2$  days (East Pacific Rise).

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670 References 2:

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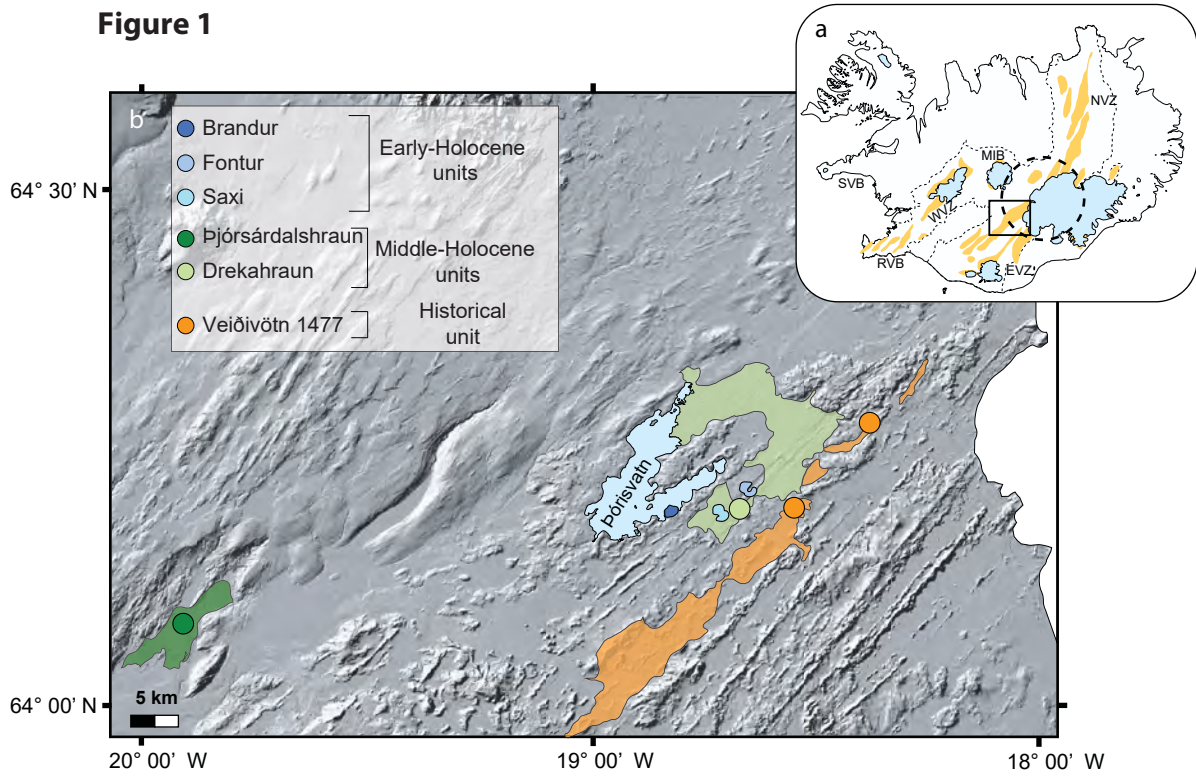
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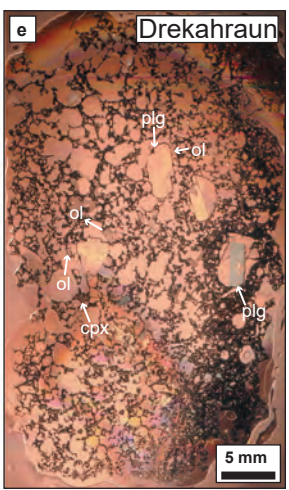
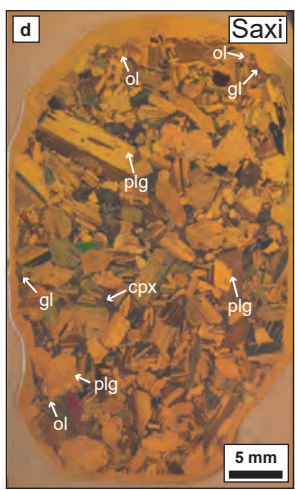
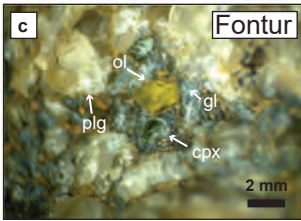
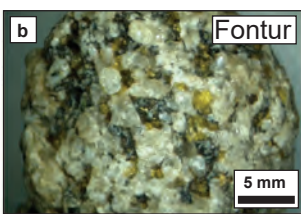
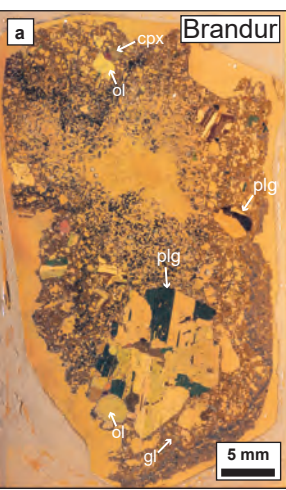
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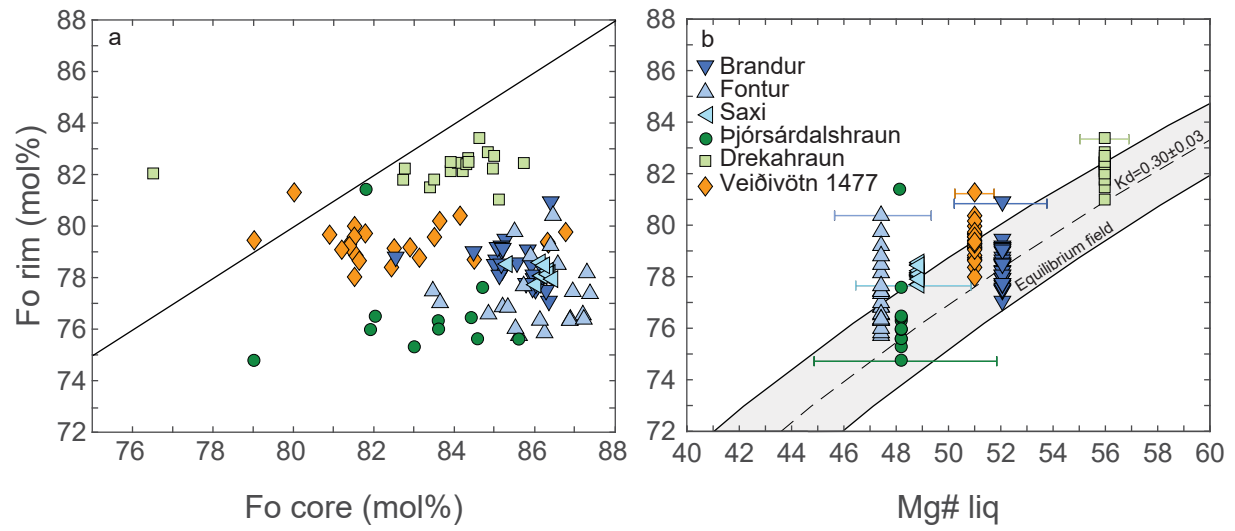
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**Figure 1**



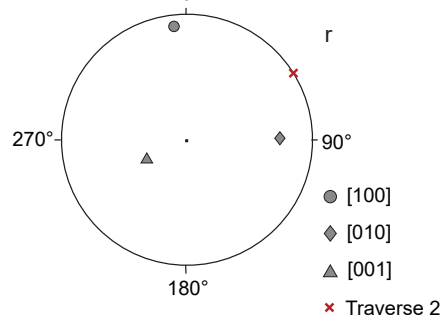
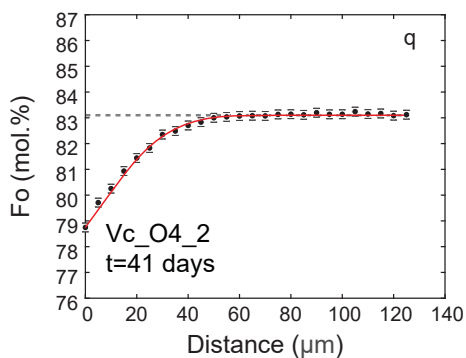
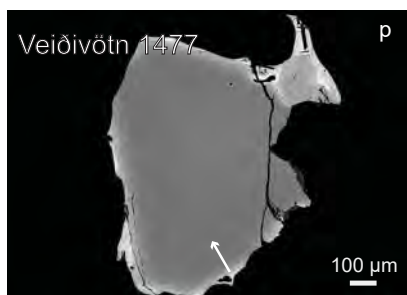
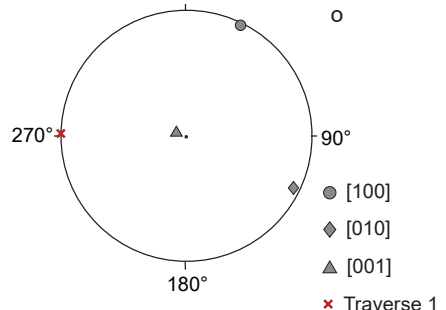
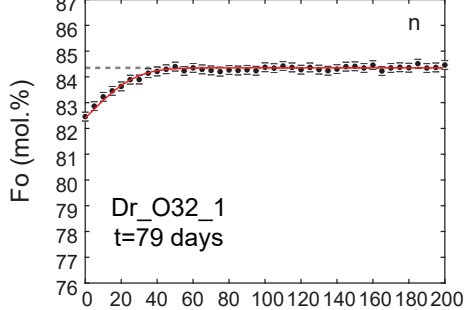
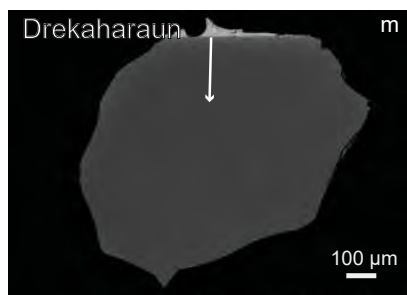
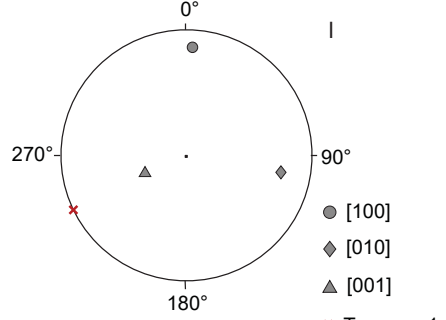
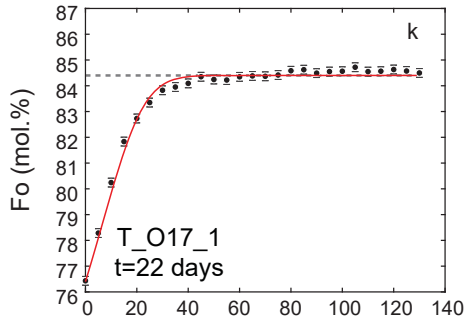
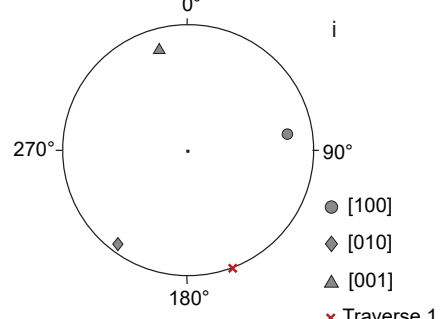
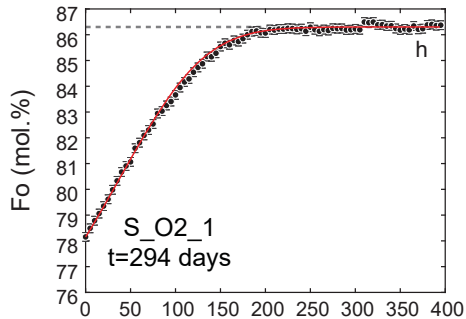
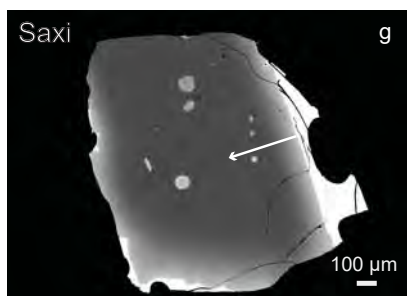
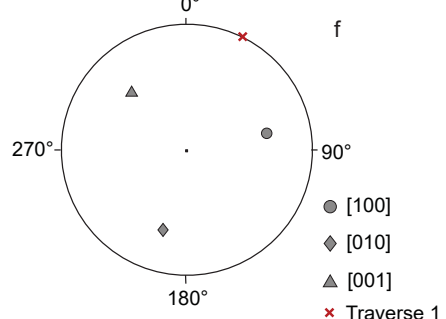
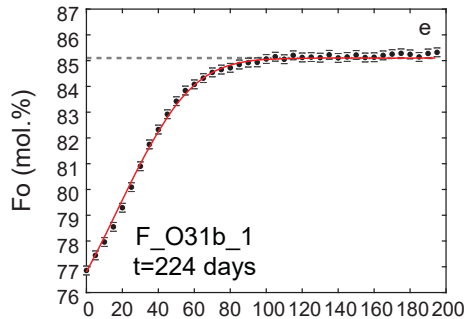
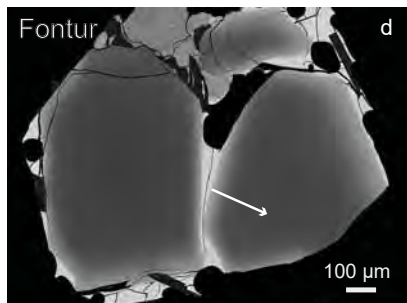
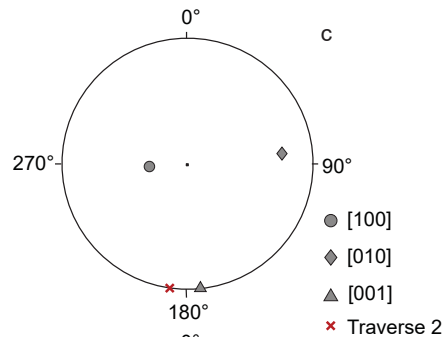
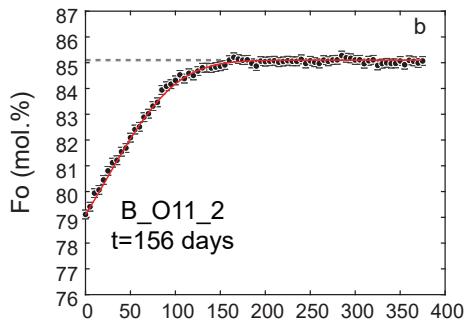
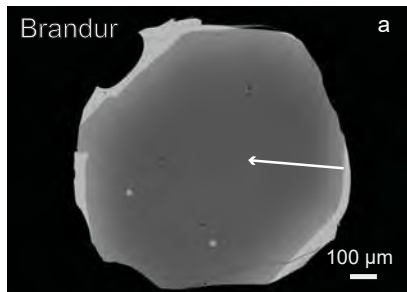


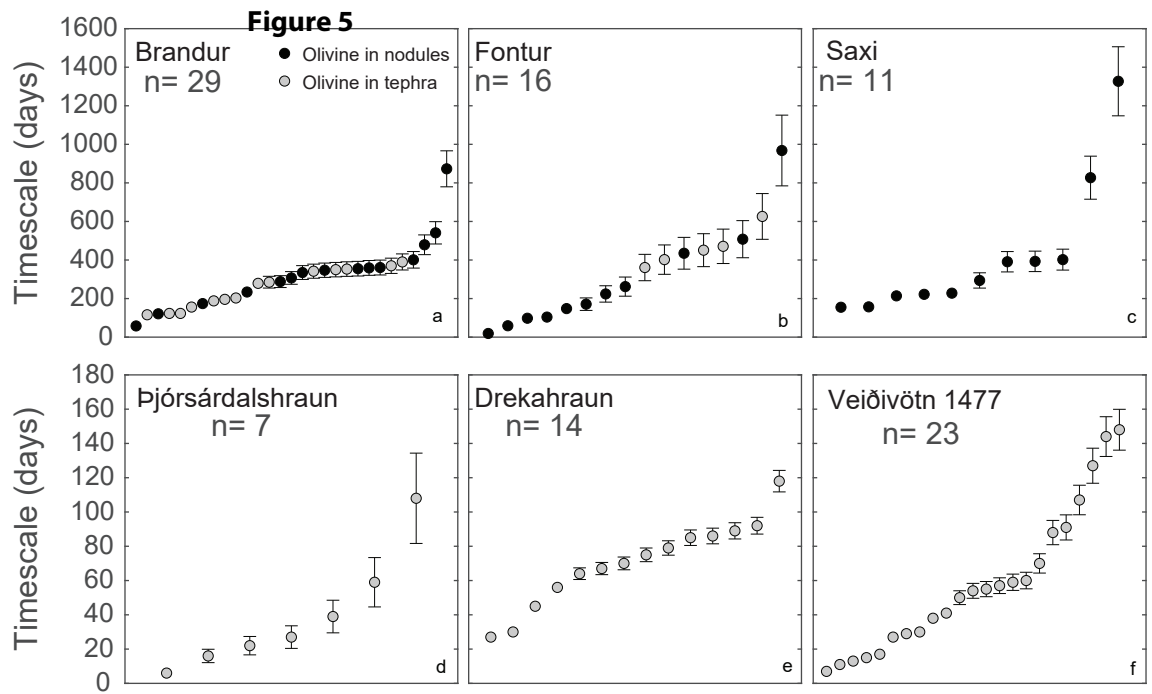


**Figure 3**



**Figure 4**



**Figure 5**

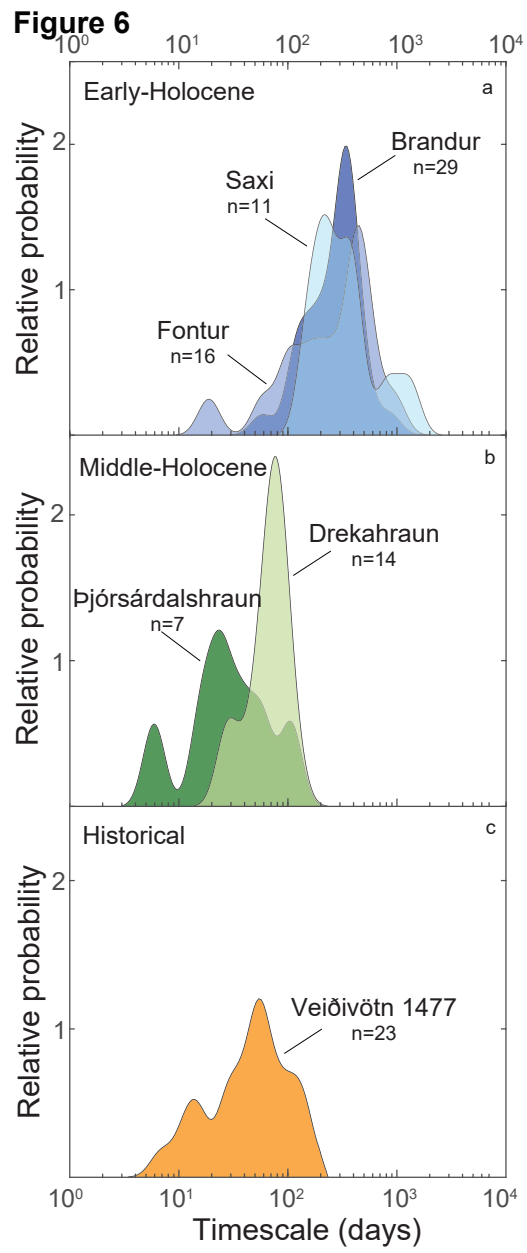
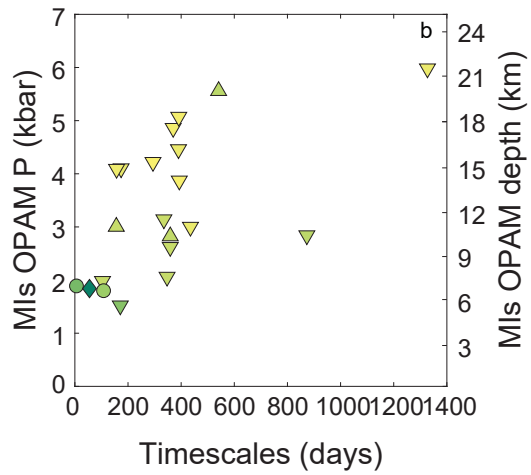
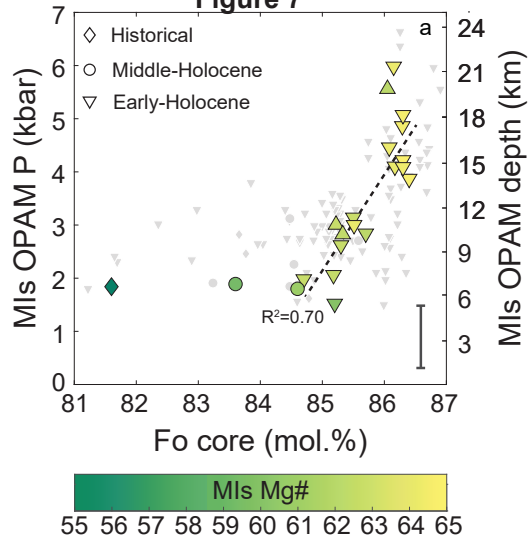
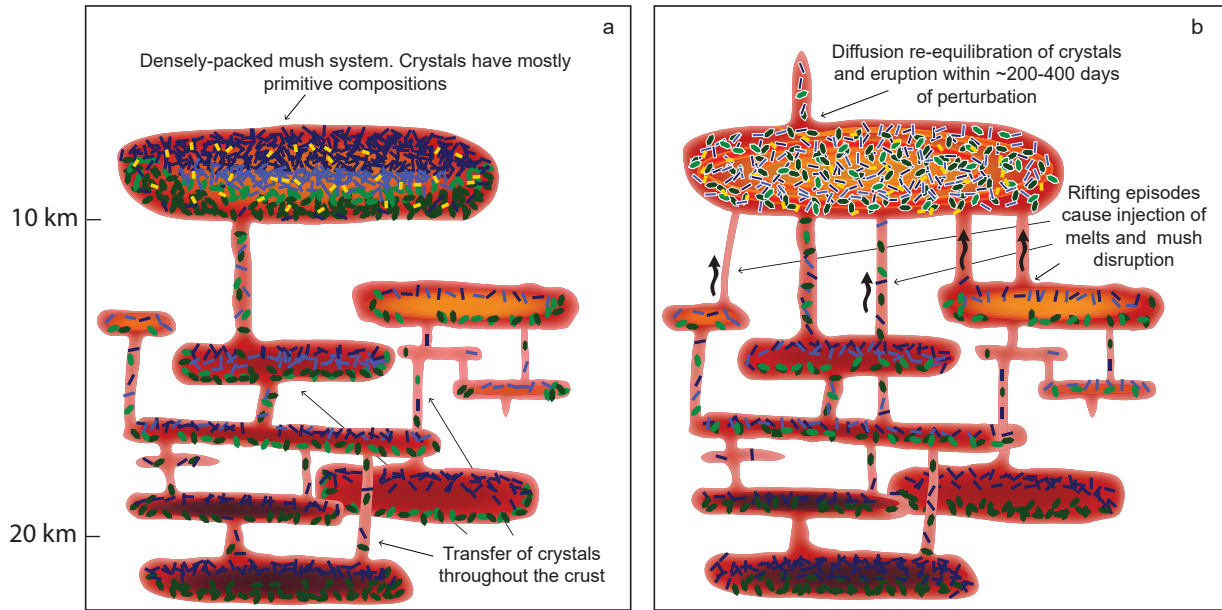


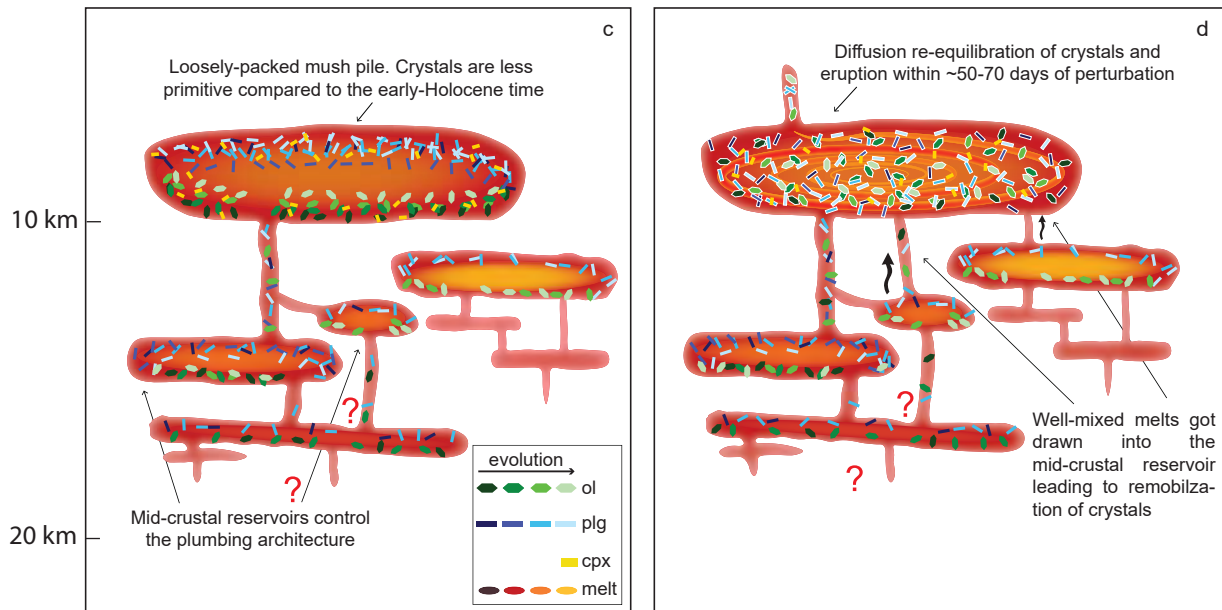
Figure 7



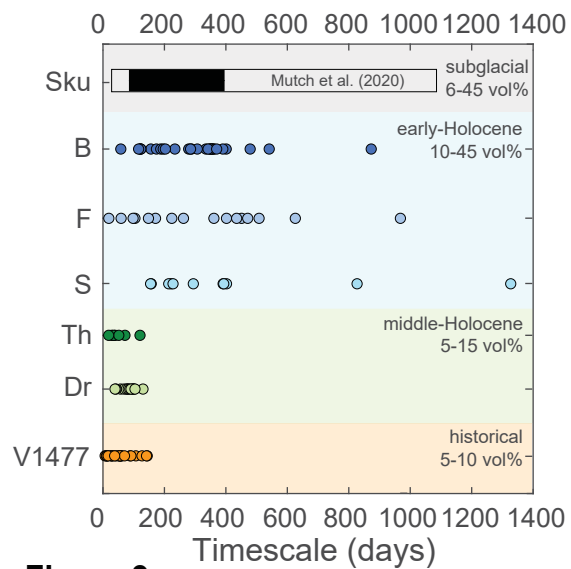
## Early-Holocene



## Middle-Holocene to present time

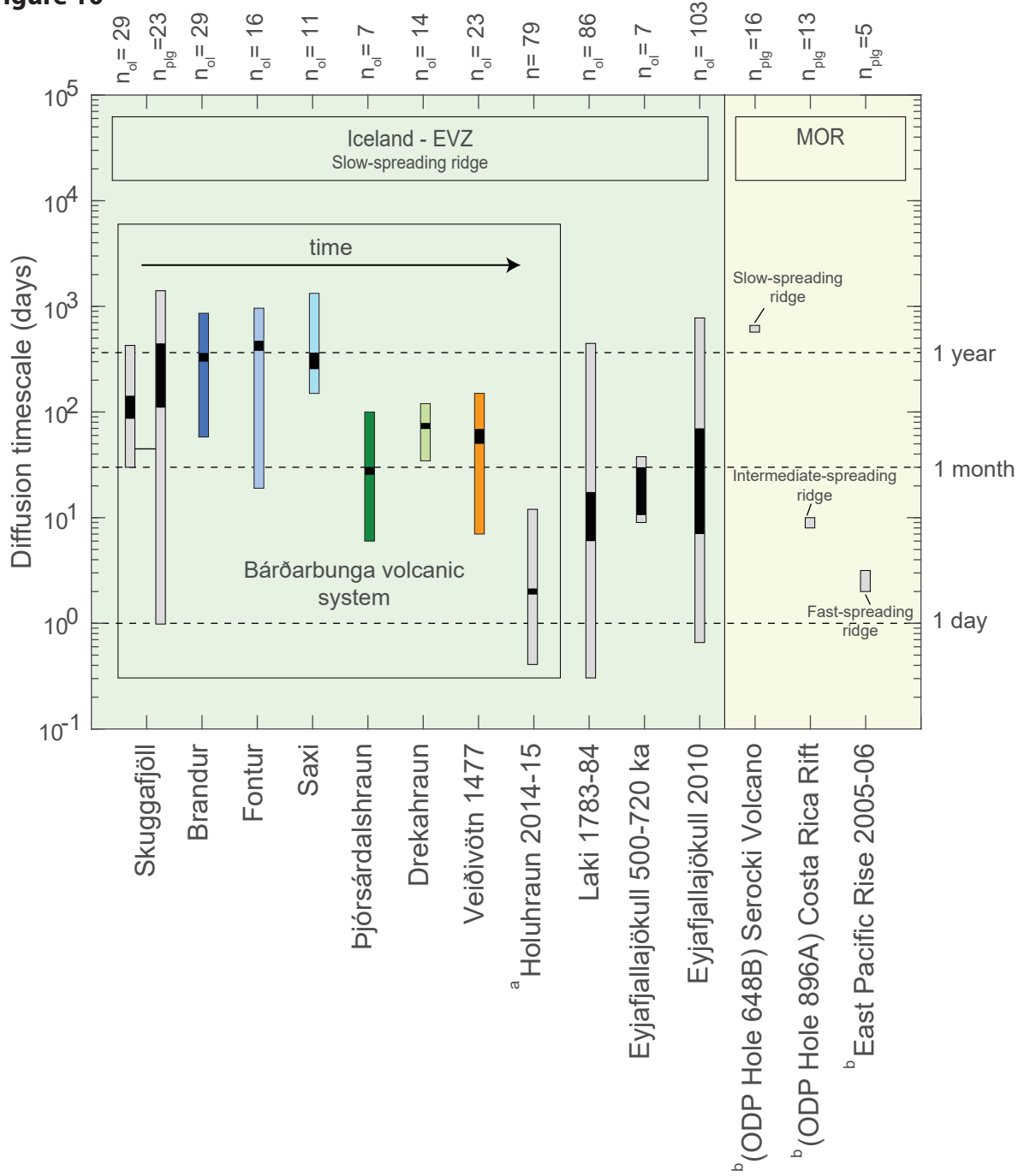


**Figure 8**



**Figure 9**

**Figure 10**



**Table 1.** Number of olivine crystals, chemical profiles and timescale (days) result for each magmatic unit.

Unit	Age	Olivine	Chemical	Modelled	Timescale		Temperature*		Pressure*	
		grains	profiles	olivines	<i>average</i>	<i>st.dev.</i>	<i>Mean</i>	$\sigma$	<i>Mean</i>	$\sigma$
Brandur	Early-Holocene	31	44	29	300	158	1170	4	2.2	0.7
Fontur	Early-Holocene	21	31	16	332	241	1161	7	2.2	0.6
Saxi	Early-Holocene	13	13	11	419	339	1163	5	2.3	0.7
Þjórsárdalshraun	Middle-Holocene	11	16	7	40	32	1160	9	1.9	0.7
Drekahraun	Middle-Holocene	19	27	14	70	24	1174	2	2.6	0.8
Veiðivötn 1477	1477 AD	26	42	23	58	41	1167	3	1.9	0.6

\*Modelling input pressure (kbar) and temperature (°C) data from Caracciolo et al. (2020)