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REVISION 2

Timescales of magma storage and migration recorded by olivine crystals in basalts of the March-April 2010 eruption at Eyjafjallajökull volcano, Iceland

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Abstract

The early eruptive phase of the 2010 eruption at the Fimmvörðuháls Pass, east of Eyjafjallajökull volcano, produced poorly evolved basalts with mildly alkaline affinity, and benmoreitic tephra were emitted during the second explosive phase from the summit vent of the volcano. In this study, textural features and chemical zoning preserved in olivine crystals of the early erupted basalts have been used in order to define the timescales of differentiation processes and magma ascent before the eruption. These lavas contain a mineral assemblage constituted by olivine (Fo₇₀₋₈₈) and plagioclase (An₅₇₋₈₃) in rather similar proportions with scarce clinopyroxene and opaque oxides. Olivine occurs as euhedral or embayed crystals characterized by different core compositions and zoning patterns. Three main olivine populations have been found, namely crystals with: a) wide Fo₈₈ cores with normal zoning towards narrow rims (P1); b) ~Fo₈₁ cores with either no zoning or slight reverse zoning patterns towards the rims (P2); c) ~Fo₇₇ cores with reverse zoning at the rims (P3). The

28 olivine reverse zoning indicates that these poorly evolved magmas experienced mixing
29 processes in addition to limited fractional crystallization at different levels of the plumbing
30 system. Timescales of transfer dynamics before the eruption have been estimated through Fe-
31 Mg diffusion modelling on these olivine populations. The olivine-melt equilibration through
32 diffusion was triggered by interaction of magmas differing in their evolutionary degree. P1
33 and P2 crystals recorded a first event of interaction in a ~22-km-deep reservoir that took place
34 about one month before the emission of the analysed products. Only part of P2 crystals
35 records reverse zoning due to interaction with more basic magma bearing P1 crystals (which
36 consequently develop normal zoning), suggesting fast timescales of magma mixing that
37 prevented the complete homogenization. A second mixing event, that is evident in the P3
38 olivines, occurred at shallower levels (5-6 km of depth) ~15 days before the emplacement and
39 can be considered the triggering mechanism leading to the eruption at the Fimmvörðuháls
40 Pass. Integration of our timescales with seismic data relative to the hypocentre migration
41 indicate rates of magma ascent throughout the deep plumbing system of ~0.01 m/sec. This
42 study provides evidence that magmas emitted at Eyjafjallajökull volcano, and more in general
43 at similar other volcanic systems in ocean ridge settings, can undergo complex processes
44 during their storage and transport in the crust, chiefly due to the presence of a multi-level
45 plumbing system.

46

47 **Keywords:** Eyjafjallajökull, olivine, diffusion modeling, magma mixing, ascent dynamics.

48

49

Introduction

50 Recent studies chiefly based on geophysical considerations have profoundly changed the
51 view of a single-level, molten system of reservoirs distributed beneath the mid ocean ridges
52 (e.g., Dunn et al., 2000, and references therein). Mush zones constituted of liquid regions of

53 variable dimensions and other portions characterized by various degrees of crystallization are
54 thought more plausible to describe the physical configuration of magma storage zones
55 beneath ocean ridges. The idea of a multi-level plumbing system, articulated through distinct
56 paths of magma ascent, is more pronounced for important volcanic systems above ocean
57 ridges characterized by central activity. The best examples of such kind are the Icelandic
58 shield volcanoes (e.g., Katla, Eyjafjallajökull, Askja, Krafla, Grimsvötn, Bardarbunga).
59 Magma transfer dynamics from the source zones up to the surface and eruption at these
60 volcanic systems are not always easily understood, and requires that chemical and physical
61 constraints are first made for the magma reservoirs (location, sizes, timescales of magmatic
62 processes etc.).

63 The presence of zoning in crystals is evidence for changes of the chemical and/or physical
64 conditions of the magmatic system such as melt composition, oxygen fugacity, temperature,
65 pressure, volatile contents during their growth history (Wallace and Bergantz, 2002; 2004;
66 2005; Ginibre et al., 2007; Streck, 2008; Viccaro et al., 2010; Kahl et al., 2011; 2013).
67 Furthermore, diffusion chronometry applied to compositionally zoned crystals has proved to
68 be valuable for understanding either the reasons for or the timescales that lead to such
69 changes (e.g. Zellmer et al., 1999; Costa et al., 2003; Costa and Chakraborty, 2004; Costa et
70 al., 2008; Costa and Morgan, 2010; Morgan et al., 2004; Morgan and Blake, 2006; Kahl et al.,
71 2011; 2013). As the crystals record physical and chemical changes of the magmatic
72 environment, the intra-crystalline diffusion starts and the chemical gradient among different
73 zones of the crystal is reduced over time. The extent to which the zoning will be equilibrated
74 by diffusion depends on many parameters such as the mineral structure, the elements involved
75 in the process, temperature and time. Thus, the zoning patterns preserved in minerals could be
76 used to reconstruct the evolutionary processes that magmas underwent throughout the

77 plumbing system and the duration of storage and transport in different environments, which
78 together are among the most important goals of the modern volcanology.

79 In this study, the textural features and chemical zoning preserved in olivine crystals have
80 been used to track the pre-eruptive history of basaltic magmas feeding the early phase of the
81 2010 eruption of Eyjafjallajökull volcano at the Fimmvörðuháls Pass. Estimations on
82 timescales of magmatic processes acting beneath the Eyjafjallajökull volcano have been
83 obtained by modeling the Fe-Mg diffusion observed in olivine chemical profiles. Our results
84 provide elements to understand the nature and timing of magmatic processes before the
85 eruption of primitive basalts in Iceland that, although infrequent, can ~~have~~ present a
86 significant hazard due to the limited timespan occurring between the beginning of magma
87 ascent and the final magma extrusion.

88

89 **The Eyjafjallajökull volcano**

90 **Geological setting and volcanism**

91 Eyjafjallajökull volcano rises to a height of 1666 m at the southwestern end of the East
92 Volcanic Zone (EVZ), which at present represents the most volcanically active region in
93 Iceland (Fig. 1a). The EVZ is a propagating SW-NE trending rift located outside the main
94 zone of spreading (the axial rift). This area includes 30 volcanic systems that, on the whole,
95 account for ~79% of the total volume of erupted magmas in Iceland during the last 11
96 centuries. However, most of the emitted magma is accommodated by the active systems of
97 Katla, Grimsvötn, Hekla and Bardarbunga-Veidivötn (Fig. 1a; Thordarson and Larsen, 2007).
98 The EVZ is dominated by emission of tholeiitic magmas in its northeastern segment, whereas
99 mildly alkaline magmas characterize the southwestern segment, which is currently
100 propagating southwesterly (Thordarson and Larsen, 2007 and references therein).

101 Eyjafjallajökull has been constructed through sub-glacial eruptions and its edifice is
102 completely covered by an ice cap. Historic activity of Eyjafjallajökull has been characterized
103 by long-lasting periods of quiescence, commonly interrupted by initial hydromagmatic
104 explosions evolving to eruptive episodes with Strombolian to sub-Plinian styles. Prior to the
105 2010 AD eruption, the historical record includes a radial fissure eruption dated ~920 AD and
106 two small-volume summit eruptions at 1612 AD and 1821-1823 AD, which were identified
107 through chrono-stratigraphic studies and analysis of historic chronicles (Thordarson and
108 Larsen, 2007; Dugmore et al., 2013 and references therein). Extensive intrusions at 4.5 and
109 6.5 km of depth, with estimated volumes of $10\text{-}17 \times 10^6 \text{ m}^3$ and $21\text{-}31 \times 10^6 \text{ m}^3$ respectively,
110 occurred beneath the Eyjafjallajökull volcano in 1994 and 1999, as revealed by InSAR
111 observations, GPS geodetic measurements, and optical tilt leveling (Pedersen and
112 Sigmundsson, 2004; 2006; Sturkell et al. 2003; Sigmundsson et al., 2010).

113 All the historic eruptions of Eyjafjallajökull were more or less contemporaneous with
114 eruptions or evidence of magma movements [i.e., glacier floods and several small
115 earthquakes] at the nearby Katla volcano (~25 km to the east). This trend held during the 2010
116 eruption at Eyjafjallajökull, as demonstrated by several seismic swarms beneath Katla in the
117 2011-2012 period. Sturkell et al. (2003) also argued that magma movements took place at
118 shallow levels beneath Katla in 1999 at the same time of a magma intrusion beneath
119 Eyjafjallajökull, both evidenced by GPS and seismic data.

120

121 **The March-May 2010 eruption**

122 The 2010 event at Eyjafjallajökull volcano was characterized by two distinct eruptive
123 phases: an initial effusive flank eruption, characterized primarily by eruption of lava flows at
124 the Fimmvörðuháls Pass (Fig. 1b), and a second explosive summit eruption producing ash-
125 tephra fall-out that had heavy impact on air-traffic in a large part of Europe. Three months of

126 intense seismicity and deformation preceded the onset of the flank eruption on March 20,
127 2010 (Gudmundsson et al., 2010; Sigmundsson et al., 2010; Sigmarsson et al., 2011, and
128 references therein). Over the previous 18 years, several signals of volcanic unrest were
129 detected through geodetic and geophysical investigations, such as earthquakes and flank
130 deformation attributed to magma injections beneath the volcano. Precursory signals, recorded
131 since late December 2009, were the prelude of the opening on March 20, 2010 of a ~300 m
132 long fissure at Fimmvörðuháls Pass, between the Eyjafjallajökull and the Myrdalsjökull
133 glaciers (Figs. 1b and 2). The eruption produced lava fountains up to 150-m-high and limited
134 tephra fall-out. On March 31, a new fissure opened with an angle of 50° from that of March
135 20 (trending N15°W), where on April 2 the eruptive activity moved. The activity produced a
136 northward expanding small a'a lava flow field, filling deep gullies excavated on the flanks of
137 the Eyjafjallajökull volcano and forming spectacular lava falls (Fig. 2). Effusions from these
138 fissures continued until April 12, with an average magma emission rate of ~13 m³/s
139 (Sigmundsson et al., 2010). This led to the formation of two small scoria cones at the active
140 vents and a lava field of basalts with mildly alkaline affinity (Sigmarsson et al., 2011; Keiding
141 and Sigmarsson, 2012; Moune et al., 2012). At the end of the flank eruption, the estimated
142 total volume of emitted magma was approximately 25×10^6 m³ (Sigmundsson et al., 2010 and
143 references therein).

144 After just two days of rest, the volcanic activity started again in the early morning of April
145 14 at the central crater of Eyjafjallajökull volcano, preceded by a seismic swarm with
146 hypocentres fast moving from 7 km depth toward the ice-capped summit of the volcano
147 (Tarasewicz et al., 2012). The magma-ice interaction gave rise during the first days to a
148 hydromagmatic, highly explosive central eruption that produced a ~10 km-high ash plume
149 (Sigmundsson et al., 2010 and references therein; Sigmarsson et al., 2011). Thereafter, the
150 hydromagmatic activity decreased and evolved to purely magmatic after less than one week.

151 Main peaks of the explosive activity were recorded at the onset of the summit eruption and
152 later on 5-6 May, with emission rates on the order of 10^6 kg/s (Sigmarsson et al., 2011). The
153 sustained magma discharge continued until May 22 with emission of fine ash and dust with
154 benmoreitic-trachytic composition.

155

156 **Sampling and analytical methods**

157 A total of 16 volcanic rock samples was collected from the last-emitted products of the
158 lava flow field developed between March 20-April 12 2010 during the eruption at the
159 Fimmvörðuháls Pass, east of the summit of Eyjafjallajökull volcano (Figs. 1b and 2). Samples
160 are from the activity of April 10-12, just before the end of the eruption. Polished thin sections
161 have been made for petrographic and textural analysis ~~on~~ of olivine crystals as well as for in
162 situ microanalysis. High-contrast back-scattered electron (BSE) images, elemental x-ray maps
163 and microanalytical data were obtained at the Dipartimento di Scienze Biologiche,
164 Geologiche e Ambientali of Catania, Italy, on ~50 selected olivine crystals representative of
165 the identified dimensional classes and textures ~~by~~ using a Tescan Vega-LMU scanning
166 electron microscope equipped with an EDAX Neptune XM4-60 energy dispersive system,
167 which is characterized by an ultra-thin Be window, coupled with an EDAX WDS LEXS
168 (wavelength dispersive low energy x-ray spectrometer) calibrated for light elements.
169 Operating conditions were set at 20 kV accelerating voltage and ~8 nA beam current for
170 obtaining high-contrast BSE images. At the same operating conditions, elemental x-ray maps
171 have been acquired with dwell time of 500 μ s for 128 frames (total time for acquisition of
172 each x-ray map around 4.5 hours). Operating conditions for the analysis of major element
173 abundances in olivine and the other mineral phases were set at 20 kV accelerating voltage and
174 0.2 nA beam current. Repeated analyses on internationally certified Fo-rich olivine and glass

175 internal standards during the analytical runs ensure precision for all the collected elements on
176 the order of 3-5% (all data available in the ESM 1).

177

178 **Characteristics of the volcanic rocks**

179 **Petrography and chemistry of minerals**

180 Volcanic products erupted during the first phase of the 2010 eruption at Eyjafjallajökull are
181 basalts (Sigmarsson et al., 2011; Keiding and Sigmarsson, 2012; Moune et al., 2012) with
182 rather homogeneous petrographic features. Differences are mainly related to groundmass
183 textures and the modal abundance of mineral phases. Samples are mildly porphyritic (15 to 20
184 vol.% of phenocrysts) and highly vesiculated; phenocrysts mostly consist of plagioclase and
185 olivine in similar proportions, together making up 80-85 vol.% of the total phenocryst content
186 (Fig. 3a). The mineral assemblage also includes scarce clinopyroxene (~10-15 vol.%) and
187 opaque oxides (<5 vol.%). Phenocrysts generally occur as single crystals with variable grain
188 sizes. Glomerophytic structures involving olivine, plagioclase and clinopyroxene are also
189 present. The groundmass is vitrophyric to hyalopilitic with plagioclase microlites as the
190 predominant phase (Fig. 3a).

191 Plagioclase crystals vary in size from large crystals (up to 2400 μm) to micro-phenocrysts
192 (~200 μm) with labradorite (An_{53}) to bytownite (An_{83}) compositions (Fig. 3a; Table ESM 1).
193 The medium-size (500-1500 μm) plagioclase generally occurs as euhedral crystals, tabular in
194 shape and rather homogeneous in composition. Analyses under the polarized light microscope
195 do not reveal significant optical zoning. Also skeletal, swallow-tailed microlites are present in
196 the analysed sections. The largest grains commonly exhibit a coarsely-sieved core, embedded
197 by oscillatory-zoned outer rims. Phenocrysts affected by extensive dissolution (rounded
198 edges) or with sieve-textures at the rim have been also found.

199 Olivines are present as large euhedral to anhedral phenocrysts up to ~4.5 mm in size (Fig.
200 3a-d). Most of the largest grains are strongly destabilized with extensive embayments that
201 extend deeply into the crystal core; these embayments do not show reaction rims. Smaller
202 crystals (from ~200 to 600 μm) are generally euhedral, though some of them have slight
203 embayed morphology. Olivine core compositions vary from Fo_{88} to Fo_{77} (see Table ESM 1).
204 In this regard, rim-to-rim major element profiles allowed three main crystal populations to be
205 identified. Details of textural features and zoning patterns of the three populations are
206 described in the following section.

207 Clinopyroxenes mostly occur as small euhedral phenocrysts ranging in size from 200 to
208 600 μm , rarely found as large single crystals (up to 1200 μm in size) or in aggregates with
209 plagioclase and olivine. Their compositions are in the range Wo_{38-45} , En_{40-51} , Fs_{12-16} (see Table
210 ESM 1).

211 Two opaque oxides have been identified with compositions either of titaniferous-magnetite
212 or Cr-spinel (Table ESM 1). The former is from subhedral to euhedral, with sizes up to few
213 tens of μm , whereas the latter commonly occurs as anhedral crystals, up to 200 μm in size,
214 enclosed in the largest olivine crystals with Fo_{88} and Fo_{81} .

215

216 **Textural and compositional features of olivine crystals**

217 Major element analyses were performed along rim-to-rim profiles on olivine crystals with
218 sizes ranging from ~400 μm to 2500 μm . Traverses crosscut the centre of the crystal, with
219 step of 7-13 μm between each analysed spot. Three main olivine populations have been
220 identified on the basis on their core compositions, namely: 1) the first population (P1) refers
221 to olivine with Fo_{88} cores (Fig. 3b); 2) the second population (P2) pertains to olivine with
222 ~ Fo_{81} cores (Fig. 3c); 3) the third population (P3) is constituted by olivine cores at ~ Fo_{77} (Fig.
223 3d; Table ESM 1).

224 The P1 olivines ($\sim\text{Fo}_{88}$) are resorbed with extensive embayments (Fig. 4). They are rather
225 uncommon (2-3 individuals per thin section) and typical of the largest phenocryst (900-2500
226 μm across). P1 olivines display nearly flat compositional profiles at their wide cores and
227 normally zoned rims, decreasing to Fo_{70} at the outermost rim (50-80 μm).

228 Olivines belonging to P2 and P3 include euhedral or embayed large crystals with size
229 ranging from 900 to 1500 μm or smaller olivines ($\sim 600 \mu\text{m}$). P2 crystals constitute the most
230 abundant population, being $\sim 70\%$ of the olivine phenocrysts observed in the analysed rocks
231 (Fig. 5). These olivines show two distinct zoning patterns: a) rather constant $\sim\text{Fo}_{81}$ core
232 composition and normally zoned rims, which is the dominant zoning pattern in P2; b) reverse
233 zoning with slightly increasing Fo toward the rim (up to $\sim\text{Fo}_{83}$), which occurs less commonly
234 than normal zoning. Independently from the zoning pattern type, all the P2 crystals record a
235 sudden drop to Fo_{73} in the outermost envelopes (last 30-50 μm).

236 All the P3 olivine crystals have core compositions at $\sim\text{Fo}_{77}$ with a marked reversely zoning
237 toward the rims (Fig. 6). Variable ΔFo in the reverse zoned portion of crystals leads to define
238 two sub-populations: P3a) with minor variations ($\sim\Delta\text{Fo}_5$), shifting the rim composition from
239 $\sim\text{Fo}_{77}$ to $\sim\text{Fo}_{82}$; P3b) with slightly more consistent changes ($\sim\Delta\text{Fo}_8$) that produce $\sim\text{Fo}_{85}$
240 envelopes. The outermost rims are normally zoned with decreasing Fo content to $\sim\text{Fo}_{70}$.

241

242 **Storage zones and processes**

243 Compositional differences of the three populations of olivine crystals, which were found in
244 the volcanic rocks emitted at the Fimmvörðuháls Pass during the 2010 eruption, necessarily
245 imply the presence of three different magmatic environments characterised by changes of one
246 or more of the above mentioned chemical-physical parameters. Various studies of Fe-Mg
247 partitioning between olivine and basaltic liquids showed that the $^{Ol/Liq}K_{\text{DFe-Mg}}$ varies little with
248 temperature and melt composition, being rather constant at 0.30 under pressures $< 2\text{-}3 \text{ GPa}$

249 (Roeder and Emslie, 1970). Conversely, the pressure dependence of $^{O/Liq}K_{DFe-Mg}$ is more
250 marked, in a way that it is positively correlated with pressure (Herzberg and O'Hara, 1998;
251 Putirka, 2005; Toplis, 2005). This means that olivines crystallizing from liquids rather similar
252 in their temperature and composition evolve to lower Fo contents under decreasing pressure.

253 The physical and chemical conditions of crystallization for the three olivine populations
254 have been constrained through two-step thermodynamic modelling using the MELTS code
255 (Ghiorso and Sack, 1995; Asimow and Ghiorso, 1998). A mantle-equilibrated composition of
256 the 2010 Eyjafjallajökull lavas has been chosen as the starting liquid and considered the
257 parental liquid for all the three populations, even though we have evidence for open-system
258 processes (i.e., magma mixing). However, mixing occurred between magmas similar in their
259 compositions, which means that errors deriving from the use of a single, evolving
260 composition through fractional crystallization modelled by MELTS are restricted. Effects due
261 to the rather limited fractionation of these magmas have been removed by adding ~10-15% of
262 Fo₈₈ olivine until the calculated liquid has the crystallizing olivine with Fo₈₈, representative of
263 equilibrium with a primary magma at mantle conditions (P1 olivines). In the first step of
264 simulation, we have tried to reproduce the physical-chemical conditions of crystallization for
265 the P2 olivines (~Fo₈₁) using the range of physical-chemical parameters defined through
266 geothermobarometry by Keiding and Sigmarsson (2012) for the crystallization of these
267 basalts, i.e.: pressure between 600 and 650 MPa, temperature between 1161°C and 1174°C,
268 H₂O between 0.5 and 0.9 wt.%. For this reason, parameters have been set as follows for the
269 first step of simulation: pressure between 630 and 600 MPa (representative of crystallization
270 at 24-23 km of depth for a crust density of 2.8 g/cm³; cf. Tarasewicz et al., 2012), temperature
271 between 1180°C and 1160°C, H₂O = 0.7 wt.%. Several attempts ($n > 100$) have been done,
272 crossing the above-defined X - P - T parameters. However, P2 olivines (~Fo₈₁), coexisting with
273 orthopyroxene and clinopyroxene of augitic composition, starts to crystallize at pressure of

274 615 MPa only if temperature is raised at 1210°C and fO_2 fixed at the NNO buffer ($\sim 3.5 \times 10^{-3}$
275 Pa). Lower temperatures or more oxidizing redox conditions do not allow crystallization of P2
276 olivines and the associated paragenesis. Assuming that more fayalitic olivines (such as P3
277 olivines at $\sim Fo_{77}$) are produced through crystallization from a slightly more evolved liquid
278 and/or at different physical conditions, there is need of a further simulation at changed
279 chemical-physical conditions. The final liquid resulting from the first step of simulation has
280 been used as starting liquid in the second step. Even in this case, several attempts ($n > 100$)
281 have been done, crossing various X - P - T conditions. The crystallization of $\sim Fo_{77}$ olivines from
282 the starting liquid composition can be reproduced only decreasing pressure and temperature at
283 more oxidizing redox conditions and slightly higher water contents. For the second step of
284 simulation, parameters have been therefore set as follows: pressure between 170 and 120 MPa
285 (representative of crystallization at 6-4.5 km of depth for a crust density of 2.8 g/cm³; cf.
286 Tarasewicz et al., 2012), temperature between 1200-1140°C, fO_2 at the QFM buffer and H₂O
287 = 0.9 wt.% (cf. Keiding and Sigmarsson, 2012). Under these conditions, P3 olivines ($\sim Fo_{77}$)
288 starts to crystallize at pressure of 145 MPa, temperature of 1150°C and $fO_2 \sim 1.7 \times 10^{-4}$ Pa.

289 The cores of the three olivine populations are flat, i.e., they do not present any
290 compositional zoning over widths of hundreds of μ m. This behaviour could reflect residence
291 for significant periods of time at high temperature, allowing complete equilibration through
292 Fe-Mg diffusion prior to the rim overgrowth. Indeed, different zoning features at the rim of
293 the three populations indicate different magmatic histories before the eruption. P1 olivines
294 show peculiar features, as they are namely: a) resorbed with extensive embayments; b) rather
295 scarce (2-3 crystals per thin section); c) typical of the largest phenocrysts (900-2500 μ m); d)
296 the most magnesian (Fo_{88}) of the entire dataset; e) with nearly constant core compositions and
297 normally zoned rims (Fig. 4). The core characteristics are typical of unperturbed growth from
298 a melt (hereafter called M1) almost equilibrated with the mantle. The presence of these

299 crystals suggests that pre-eruptive dynamics have been influenced in some way by the ascent
300 of the M1 magma from the source zones.

301 P2 olivines ($\sim\text{Fo}_{81}$) with normal zoning are the most abundant ($\sim 70\%$) in the emitted
302 products. Their crystallization occurred from a magma (hereafter called M2) slightly more
303 evolved than M1. The subordinate reversely zoned envelopes in some P2 olivines (up to
304 $\sim\text{Fo}_{83}$) reflect interaction with a magma necessarily more basic, whose olivine crystallizing at
305 equilibrium is more forsteritic than Fo_{83} (Fig. 5). The Fo decrease at the rim of P1 olivines
306 coupled with Fo increase around P2 crystal cores suggest that the mixing process involved
307 M1 and part of M2 in the magma reservoir at the mantle-crust boundary, which is located at
308 ~ 22 km b.s.l. beneath the Eyjafjallajökull volcano (Tarasewicz et al., 2012 and references
309 therein; Fig. 7). Such deep magma reservoirs are usually present underneath central volcanoes
310 in different areas of Iceland, especially in relation to the most active portions of the rift
311 branches (Searle, 2013 and references therein). Although the precise size of this reservoir
312 beneath Eyjafjallajökull is not known, its bottom and top have been constrained respectively
313 at depths of 24 and 21 km on the grounds of the location of the micro-earthquake foci
314 occurred between March 6 and May 31, 2010 (Tarasewicz et al., 2012; Fig. 7). The result of
315 mixing between M1 and M2 is a hybrid magma (hereafter called M3; Fig. 7), which carried
316 during its ascent the P1 and P2 olivines. We infer that the mixing process within the deep
317 reservoir had very fast timescales because most of the P2 olivines did not register any reverse
318 zoning. This feature could be attributed to lack of complete homogenization of the magma
319 volume involved in the mixing process probably due to the rapid magma ascent towards the
320 surface. This prevented the development of compositional changes in part of the crystals.

321 Compositional zoning data support the occurrence of a second mixing event at shallower
322 depth. End-members of this second interaction could be M3 and a more evolved magma
323 stored in the crust (hereafter called M4), as suggested by the more fayalitic core compositions

324 of P3 olivine crystals ($\sim\text{Fo}_{77}$) with respect to P1 and P2 (Fig. 6). Magma mixing is markedly
325 evident at the rim of P3 olivines, with Fo contents growing from Fo_{77} at the cores to Fo_{82} -
326 Fo_{85} towards the rims (Fig. 6). The presence of the inferred M4 reservoir is supported by
327 InSAR observations, GPS geodetic measurements, and optical tilt leveling data by several
328 authors, who provide evidence of two 4.5 and 6.5 km deep intrusions beneath the
329 Eyjafjallajökull volcano that took place in 1994 and 1999 respectively (Fig. 7; Pedersen and
330 Sigmundsson, 2004; 2006; Sturkell et al. 2003; Sigmundsson et al., 2010). In the next section,
331 time constraints to the above mentioned magma dynamics are provided.

332

333 **Timescales of magma recharge and transfer**

334 In order to assess timescales of intrusion and open system processes such as magma
335 mixing events before the 2010 Eyjafjallajökull flank eruption, the approach of Costa et al.
336 (2008) and Costa and Morgan (2010) was adopted to model the diffusion-controlled re-
337 equilibration of the Fe-Mg zoning of olivine. In numerical implementations, concentrations
338 are described as a function of space (x) and time (t) and are directly related to values of the
339 diffusion coefficients (D) along the measured profile. Thus, time evolution of the initial
340 concentration distribution of the crystal can be traced until a new profile that fit the observed
341 concentration profile is obtained. Due to the strong diffusion anisotropy, crystals were
342 preliminarily selected to minimize uncertainties on time determination related to the section
343 orientation with respect to the fast diffusion direction (c-axis), following the selection criteria
344 listed in Costa and Chakraborty (2004) and Shea et al. (2015). The modelling has been
345 performed by using Fick's Law-based diffusion equations with concentration dependent
346 diffusion coefficients ($D_{\text{Fe-Mg}}$); we assumed the measured concentration at the rim of the
347 crystals as boundary conditions (see Table ESM 2). The expression for calculating the Fe-Mg
348 diffusion coefficients along [001] is from Dohmen and Chakraborty (2007). Coefficients were

349 calculated at $T = 1210^{\circ}\text{C}$, $P = 615 \text{ MPa}$ and $f\text{O}_2$ at $\sim 3.5 \times 10^{-3} \text{ Pa}$ for the P1 and P2
350 populations, and at $T = 1150^{\circ}\text{C}$, $P = 145 \text{ MPa}$ and $f\text{O}_2$ at $\sim 1.7 \times 10^{-4} \text{ Pa}$ for the less forsteritic
351 P3 crystals (Table 1). Such parameters have been chosen in accordance with the initial T , P
352 and $f\text{O}_2$ values of crystallization derived through our MELTS simulations, and are also
353 consistent with the range of crystallization conditions defined for the early basaltic products
354 of the 2010 eruption and the depths of magma storage beneath the volcano as described above
355 (cf. Keiding and Sigmarsson, 2012; Tarasewicz et al., 2012 and references therein).
356 Anisotropy-corrected diffusivity along the direction of the profile (D_{trav}) was obtained taking
357 into account how the compositional traverses were oriented with respect to the olivine
358 crystallographic a, b and c axes (respectively coinciding with the optical indicatrix axes X, Y
359 and Z measured by conoscopic observations under a polarizing optical microscope equipped
360 with a Zeiss 4 axis universal stage) to further improve the accuracy and precision on time
361 calculations (cf. Costa and Chakraborty, 2004; Shea et al., 2015). After application of all the
362 correction criteria (i.e. influence of sectioning, diffusion anisotropy, crystal morphology), the
363 model replicates diffusion times with a high degree of precision, although the number crystals
364 suitable for calculation is considerably reduced (Table 1; ESM 2). Uncertainties are mainly
365 related to the temperature and oxygen fugacity determination. A temperature uncertainty of
366 $\pm 10^{\circ}\text{C}$ yields errors of 1 day (2σ) on the timescale estimates; $f\text{O}_2$ variation of one order
367 magnitude produces errors of ca. 3 days (2σ). Conversely, the pressure dependence is small,
368 with time oscillations of ca. 1 hour for pressure changes of 10 MPa, at constant temperature
369 and $f\text{O}_2$.

370 The chemical records preserved in olivine cores of the basalts erupted at Fimmvörðuháls
371 Pass are evidence of at least three compositionally distinct magmatic environments where
372 crystals grew close to equilibrium. Later, at the rims, they developed chemical gradients as a
373 consequence of changed conditions. Awareness of the magmatic process causing

374 disequilibrium in olivines is important to set the initial composition before re-equilibration of
375 the system through diffusion. The high and constant Fo content at the P1 crystal core implies
376 an early growth stage in a primitive host magma under unperturbed chemical and physical
377 conditions. The decreased Fo content at the rims of the P1 crystals has been attributed to
378 interactions with more evolved magmas during the intrusion at crustal levels, in order to
379 achieve the new chemical and physical equilibrium at lower Fo. Therefore, through the Fe-
380 Mg diffusion model we tried to estimate the timescale of this early intrusion and mixing at
381 crustal depth (Fig. 4). The presence of at least two magma storage zones beneath
382 Eyjafjallajökull volcano has been recognized through the zoning patterns preserved in the P2
383 and P3 olivine crystals (Figs. 5 and 6). Indeed, the core compositions at Fo₈₁ (P2) and Fo₇₇
384 (P3) suggest their growth in different magma environments, from melts more evolved than
385 that of the P1 crystals. Reverse Fo zoning at the rim of both populations reflects mixing
386 consequent to mafic recharge. These two recharging events produced chemical gradients at
387 the crystal rims, and are considered responsible for triggering diffusion. Modelling of the
388 chemical zoning in P2 and P3 olivines leads then to define timing of these mixing events
389 (Figs. 5 and 6).

390 Based on our calculations, the chemical zoning of P1 crystals requires ~34 days to re-
391 equilibrate through diffusion; this result matches well the timescale obtained for the diffusion
392 event in P2 crystals (30 days on average), suggesting an interaction between the P1 and P2
393 host magmas about one month before the emission of the analysed products on April 10-12
394 (Figs. 4 and 5; Table1). The micro-earthquake locations provided by Tarasewicz et al. (2012)
395 confirm that a magma intrusion occurred at 21-24 km of depth about one month before the
396 2010 flank eruption at Eyjafjallajökull (Fig. 7). The more evolved P3 olivines record
397 timescales of re-equilibration that are significantly shorter than those obtained for P1 and P2
398 crystals (Fig. 6). Time estimations indicate that the second mixing event recorded by rims of

399 the P3 olivines occurred about two weeks before magma emission at the Fimmvörðuháls Pass
400 (see Table1). Even in this case, micro-earthquake locations corroborate the assumption that
401 the second event of interaction took place between 6.5 and 4.5 km of depth and that this final
402 event of magma mixing was the triggering mechanism for the intrusion of an E-W-oriented
403 dyke from the shallow storage zone up to the Fimmvörðuháls Pass (Fig. 7; cf. Tarasewicz et
404 al., 2012).

405 Considering that the distance covered by the intruding magma between the sites of the two
406 mixing processes is ~17 km and that the obtained timescales for the two mixing events are
407 ~30 and ~15 days respectively, the calculated average magma ascent rate is in the order of
408 0.01 m/sec. These ascent rates refer to magma migration in the deep levels of the
409 Eyjafjallajökull plumbing system, and are well in the range expected for basaltic liquids (e.g.,
410 Rutherford, 2008, and references therein; Armienti et al., 2013). Evaluation of the ascent rate
411 at shallow levels is more complex due to the influence of several variables (i.e., gas
412 exsolution, conduit dynamics, fracture opening etc.). In this regard, literature data for other
413 basaltic volcanoes indicate extrusive ascent rates, especially for the early explosive phase of
414 the eruption, up to one order of magnitude higher than those derived for migration in the deep
415 levels of the plumbing system (cf. Rutherford, 2008, and references therein).

416

417

Implications

418 The Fe-Mg diffusion modelling in olivine chemical profiles found in products of the 2010
419 eruption at the Fimmvörðuháls Pass (Eyjafjallajökull volcano, Iceland) have been used to
420 reconstruct the pre-eruptive history of the emitted magmas. Our approach has led to the
421 comprehension of the dynamics of magmatic processes and of their timescales prior to the
422 eruption. All the data available support the idea of a multi-level storage system beneath the
423 Eyjafjallajökull volcano, where a poorly evolved magma ascending to the surface experienced

424 multiple events of mixing with other magmas slightly different in their evolutionary degree.
425 The equilibration through diffusion in three olivine populations, as a consequence of the
426 interactions, constrains the timescales of transfer dynamics from the deep levels of the
427 plumbing system (21-24 km of depth) towards the shallow reservoirs (4.5-6.5 km of depth) at
428 ascent rates estimated at 0.01 m/sec. Our study elucidates the physical configuration of
429 volcanic systems grown in ocean ridge settings, emphasizing the possibility that magmas
430 undergo complex processes during their storage and transport in the crust. Since the evidence
431 of the first intrusion, the magma upraises from deep levels of the feeding system to the
432 surface in a short timespan; this means that injection of primitive magmas in the deep
433 reservoirs can trigger significant eruptions in rather short times.

434

435

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440

441

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553

554 **List of figure captions**

555 Figure 1 – a) Schematic map of Iceland with composition of the volcanic rocks belonging to
556 different neo-volcanic zones. b) Digital Elevation Model of the Eyjafjallajökull and Katla
557 volcanoes and the area of Fimmvörðuháls Pass (source www.lmi.is/en/okeypis-kort/).

558

559 Figure 2 – A and B) Scoria cones and a' a lava flow field at Fimmvörðuháls Pass; C) Part of
560 the lava flow field at the Fimmvörðuháls Pass with top of the Eyjafjallajökull volcano on
561 the background; D) Lava flow field at the Fimmvörðuháls Pass with the Myrdalsjökull
562 glacier on the background.

563

564 Figure 3 – a) Photo taken under the polarized light microscope (crossed Nicols) showing the
565 petrographic features of a representative basalt emitted during the 2010 eruption at the
566 Fimmvörðuháls Pass; b) c) d) BSE images taken at the scanning electron microscope of
567 representative P1, P2 and P3 olivine crystals respectively.

568

569 Figure 4 – X-ray map for Mg, rim-to-rim zoning profile (white line on the X-ray map) and Fe-
570 Mg diffusion modelling for a representative P1 olivine crystal, with ~Fo₈₈ core and normal

571 zoning at the rim, found in the basalts emitted at the Fimmvörðuháls Pass during the 2010
572 eruption. The figure shows the rim-to-rim profile for the forsterite concentration (Fo%)
573 measured on the E8_O15 olivine (circles). The black dashed line indicates the initial
574 concentration profile prior to the diffusion, whereas the red line reveals the best-fit
575 diffusion model for the observed zoning profile. The stereographic plot indicates the
576 angular relations between the a, b and c crystallographic axes in olivine and the directions
577 of the measured rim-to-rim traverse.

578

579 Figure 5 – X-ray map for Mg, rim-to-rim zoning profile (white line on the X-ray map) and Fe-
580 Mg diffusion modelling for a representative P2 olivine crystal, with $\sim\text{Fo}_{81}$ core and reverse
581 zoning at the rim, found in the basalts emitted at the Fimmvörðuháls Pass during the 2010
582 eruption. The figure shows the rim-to-rim profile for the forsterite concentration (Fo%)
583 measured on the E8_O11 olivine (circles). The black dashed line indicates the initial
584 concentration profile prior to the diffusion, whereas the red line reveals the best-fit
585 diffusion model for the observed zoning profile. The stereographic plot indicates the
586 angular relations between the a, b and c crystallographic axes in olivine and the directions
587 of the measured rim-to-rim traverse.

588

589 Figure 6 – X-ray map for Mg, rim-to-rim zoning profile (white line on the X-ray map) and Fe-
590 Mg diffusion modelling for a representative P1 olivine crystal, with $\sim\text{Fo}_{77}$ core and marked
591 reverse zoning at the rim, found in the basalts emitted at the Fimmvörðuháls Pass during
592 the 2010 eruption. The figure shows the rim-to-rim profile for the forsterite concentration
593 (Fo%) measured on the E15_O15 olivine (circles). The black dashed line indicates the
594 initial concentration profile prior to the diffusion, whereas the red line reveals the best-fit
595 diffusion model for the observed zoning profile. The stereographic plot indicates the

596 angular relations between the a, b and c crystallographic axes in olivine and the directions
597 of the measured rim-to-rim traverse.

598

599 Figure 7 – Cartoon of the petrological evolution leading to the 2010 eruption at the
600 Fimmvörðuháls Pass and at the summit of Eyjafjallajökull volcano. Moho discontinuity is
601 reported at ca. 22 km b.s.l. (cf. Tarasewicz et al., 2012 and references therein). Blue dots
602 represent the locations of earthquake hypocentres during the period March 6 - May 31,
603 2010. The magmatic history starts with ascent from the mantle of the primitive magma
604 (M1) carrying P1 olivines (with Fo₈₈ cores). Considering that the analysed products have
605 been erupted between April 10 and 12, 2010, the diffusion modelling of P1 olivines dates a
606 first event of intrusion of M1 into a deep magma environment ~34 days before the
607 emission that coincide with the first detected earthquakes at 23-26 km of depth. At this
608 depth, M1 magma mixed with the residing slightly more evolved magma (M2) that bears
609 P2 olivines (with Fo₈₁ cores). Mixing between M1 and M2 produced a new magma (M3),
610 as testified by reverse Fo zoning (up to Fo₈₃) in some P2 crystals. Diffusion modelling
611 dates this mixing event between 28 and 33 days before the emission on April 10-12, 2010.
612 M3 magma started its ascent carrying P1 and P2 olivines and mixed at 4.5-6 km b.s.l. with
613 a residing magma (M4) already intruded during 1994 and 1999 (Pedersen and
614 Sigmundsson, 2004; 2006; Sturkell et al. 2003; Sigmundsson et al., 2010) that bears P3
615 olivines (with Fo₇₇ cores). This mixing event, testified by reverse Fo zoning (Fo₈₂₋₈₅) in P3
616 olivines, is dated by diffusion modelling between 12 and 18 days before the emission of
617 the April 10-12 lavas. Result of this second mixing event is the final hybrid magma that
618 intruded through an E-W-oriented dyke and was erupted at the Fimmvörðuháls Pass (M5)
619 between March 20 and April 12, 2010. The trachyandesitic magma reservoir that mixed

620 with less differentiated magma on April 12 (Sigmarsson et al., 2011) is reported in the
621 figure.

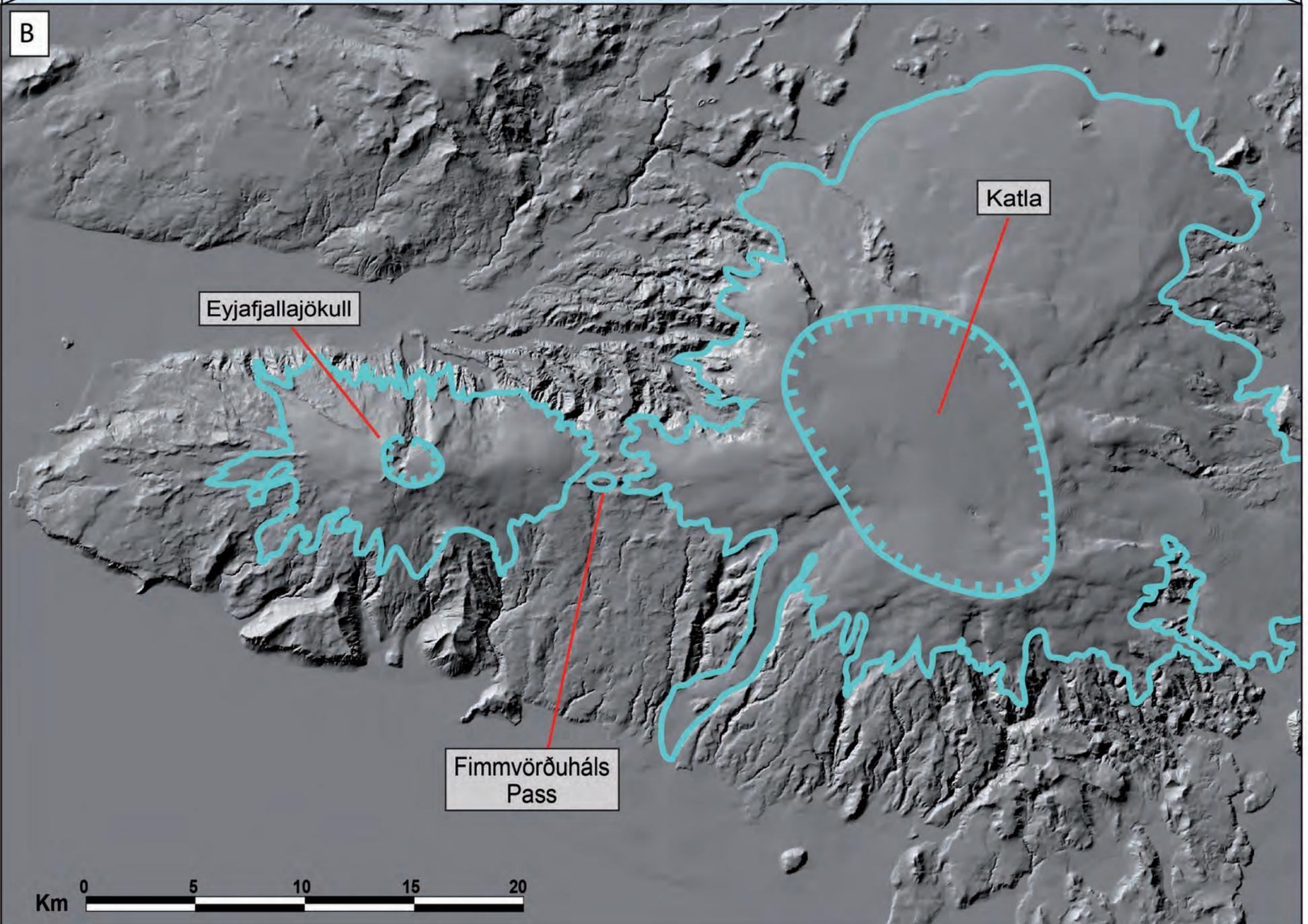
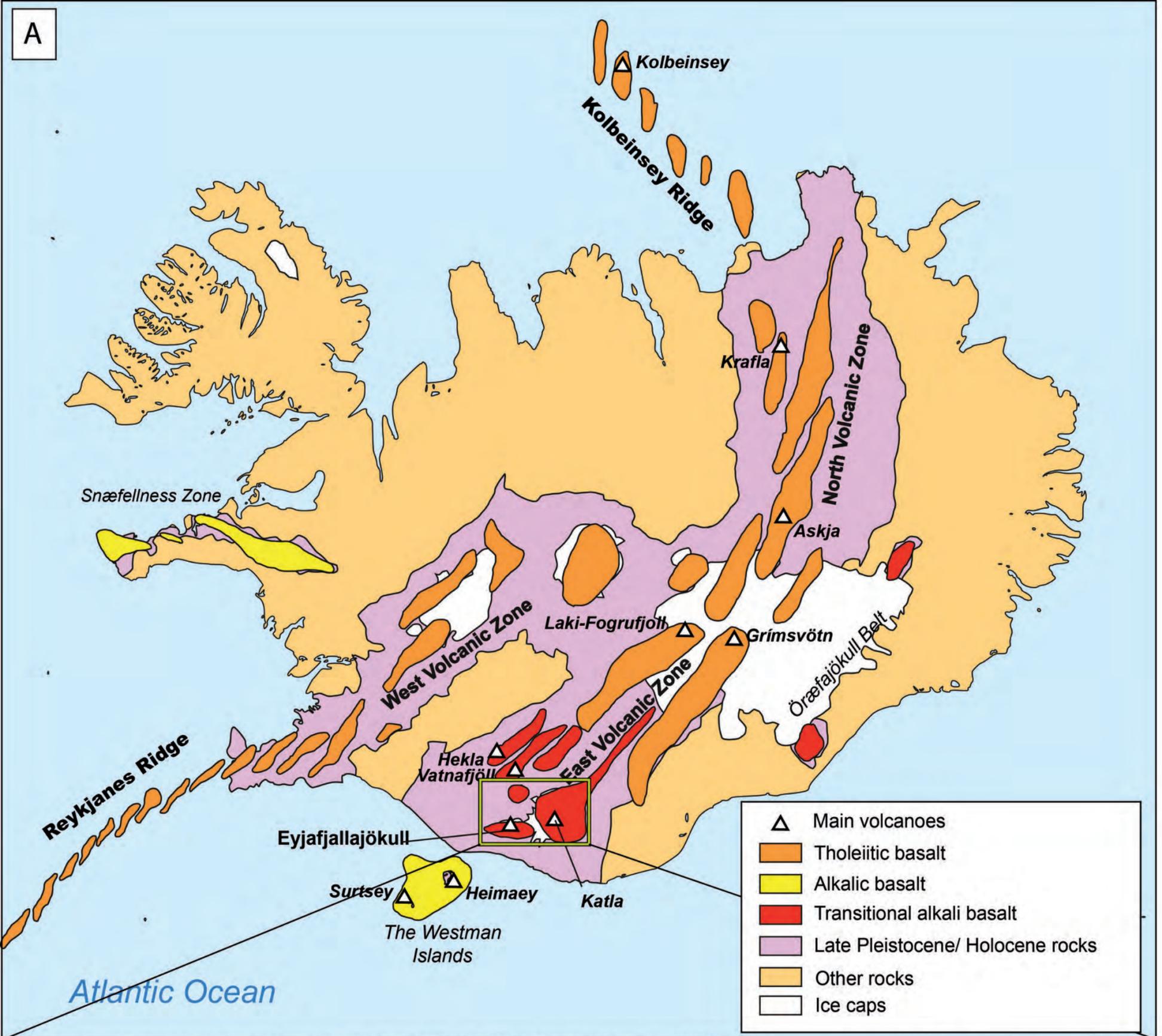
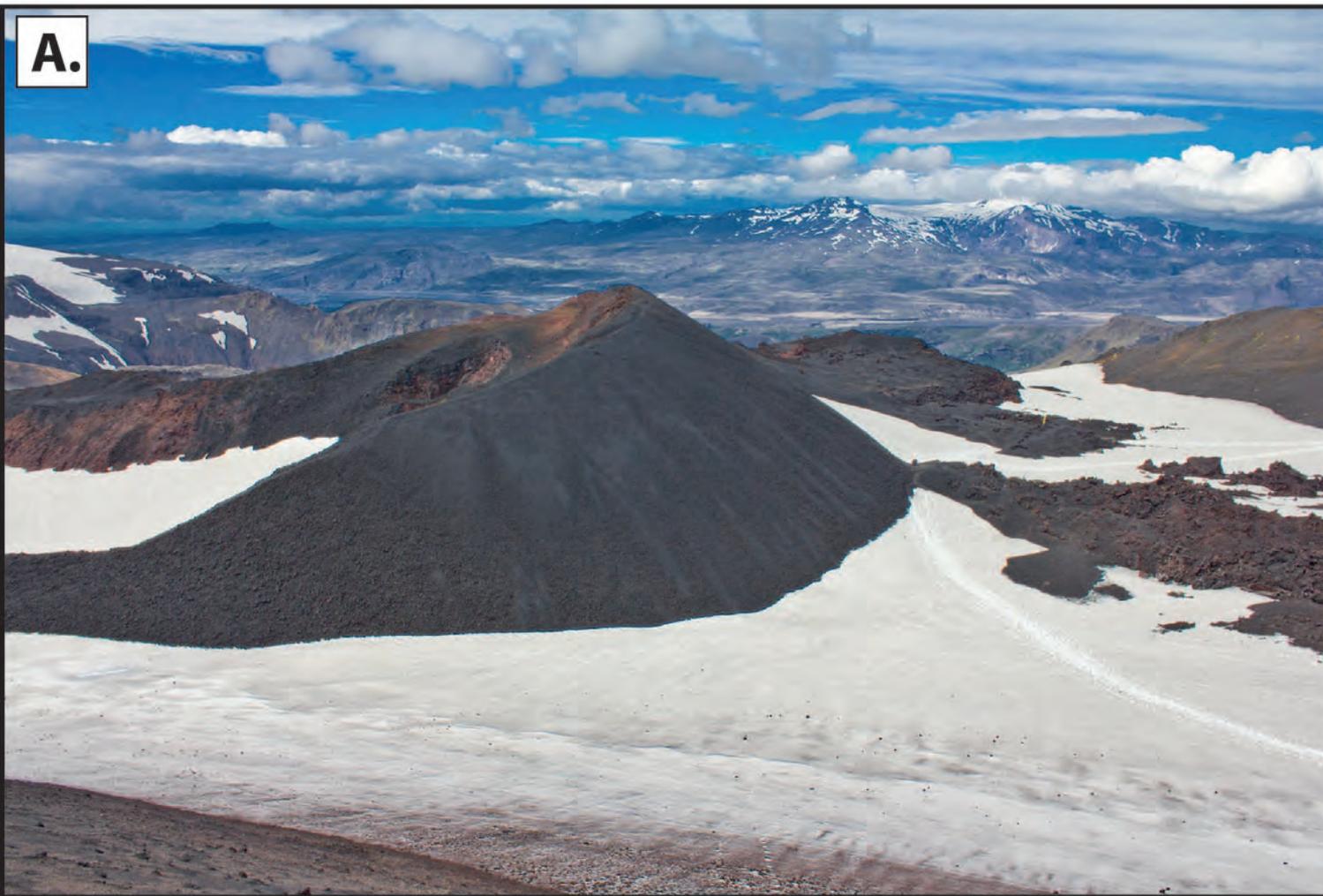


Figure 2



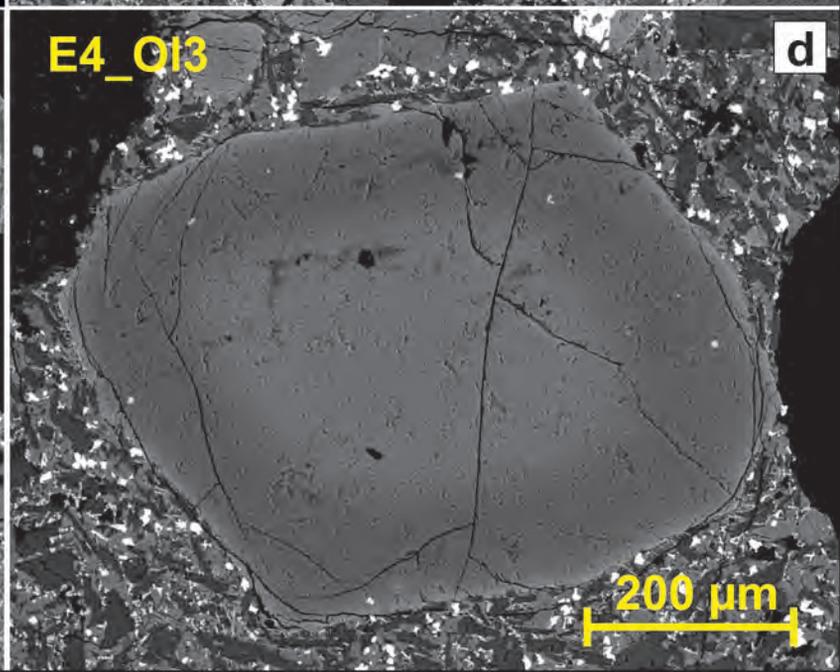
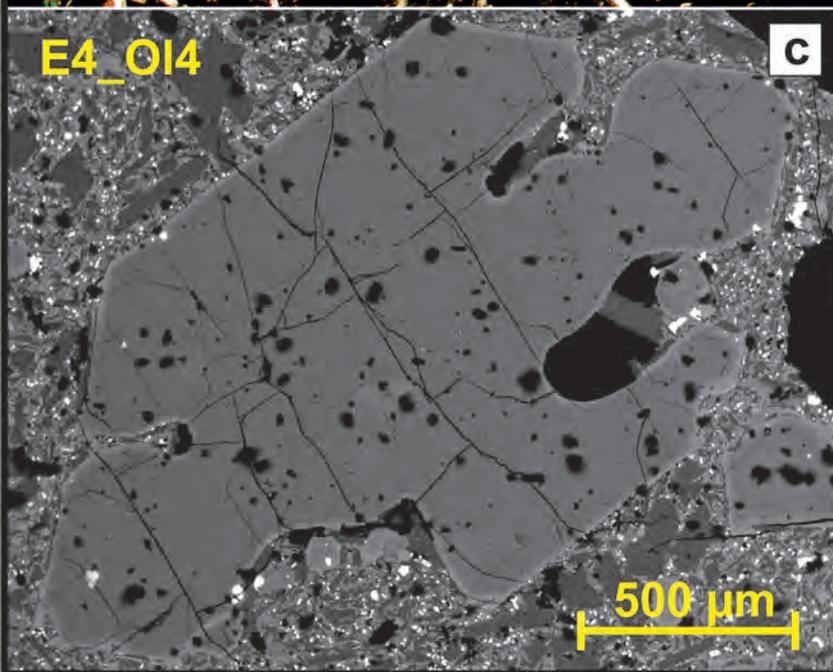
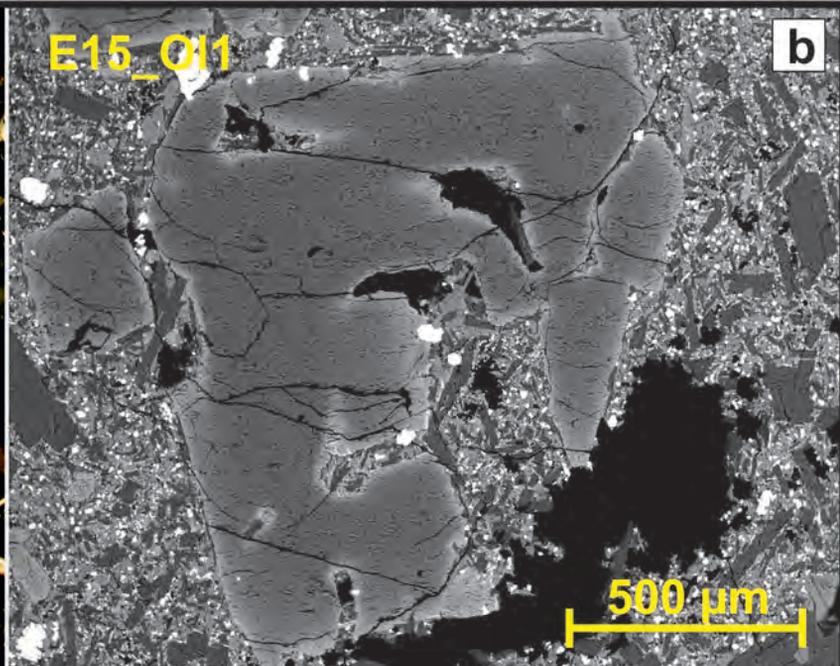
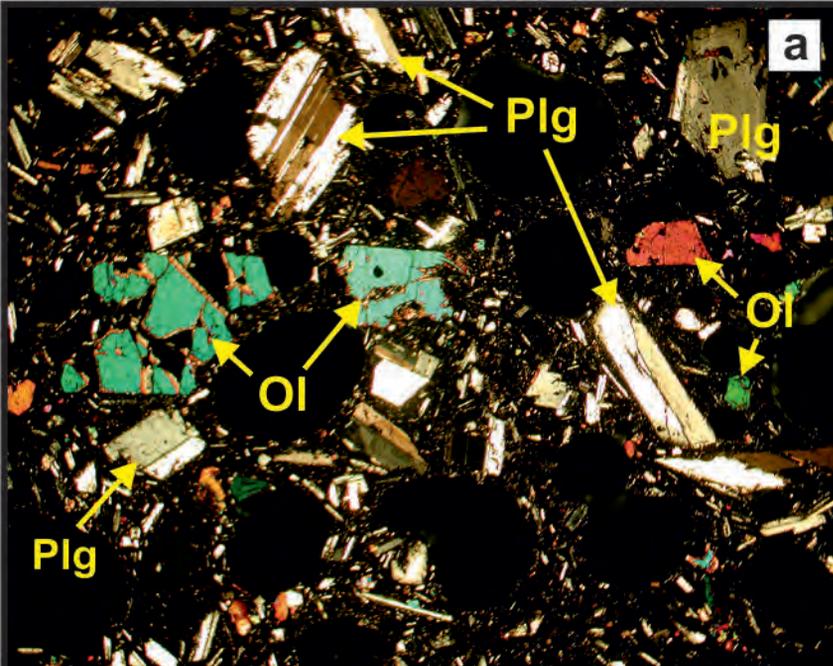


Figure 3

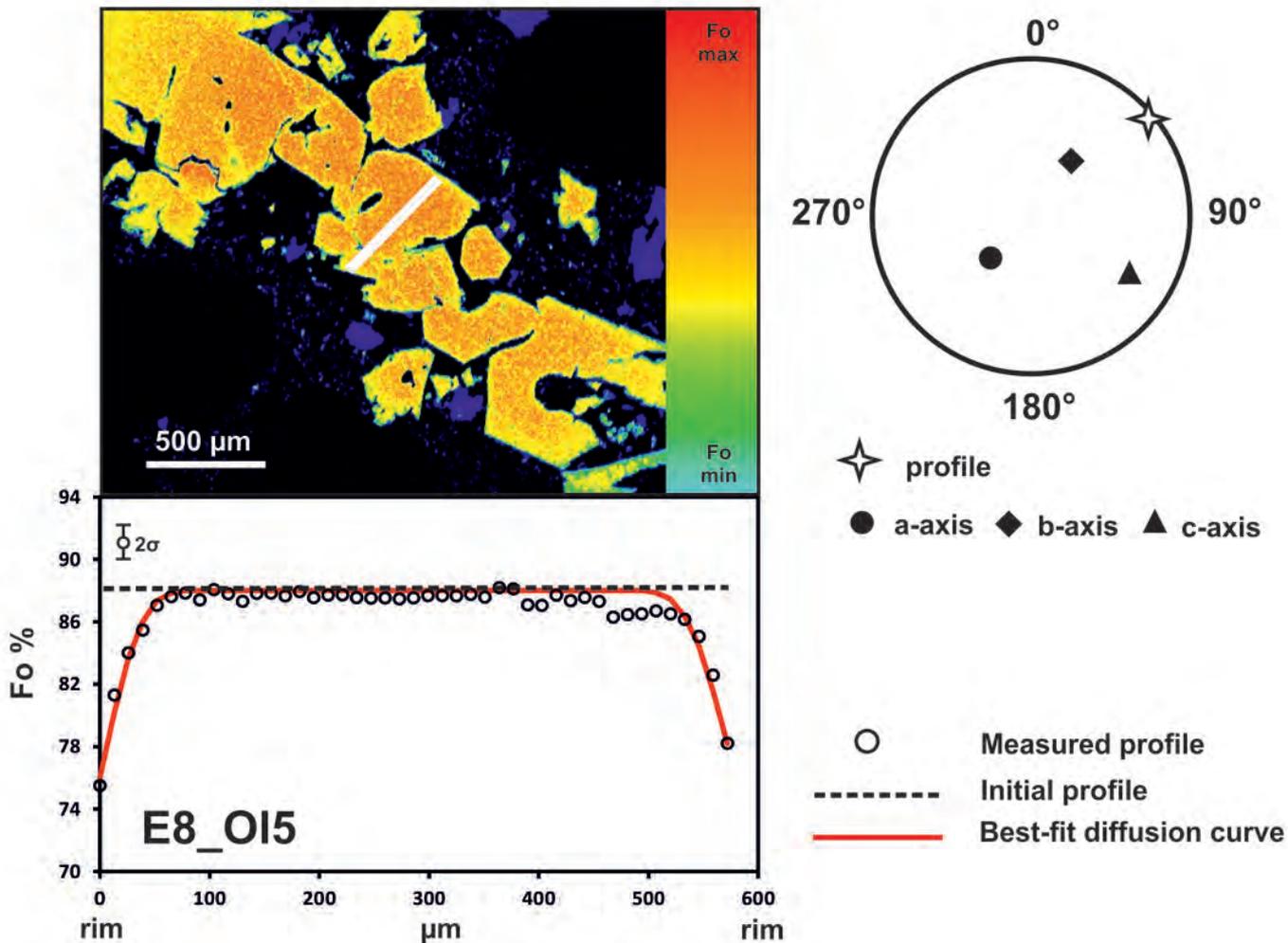


Figure 4

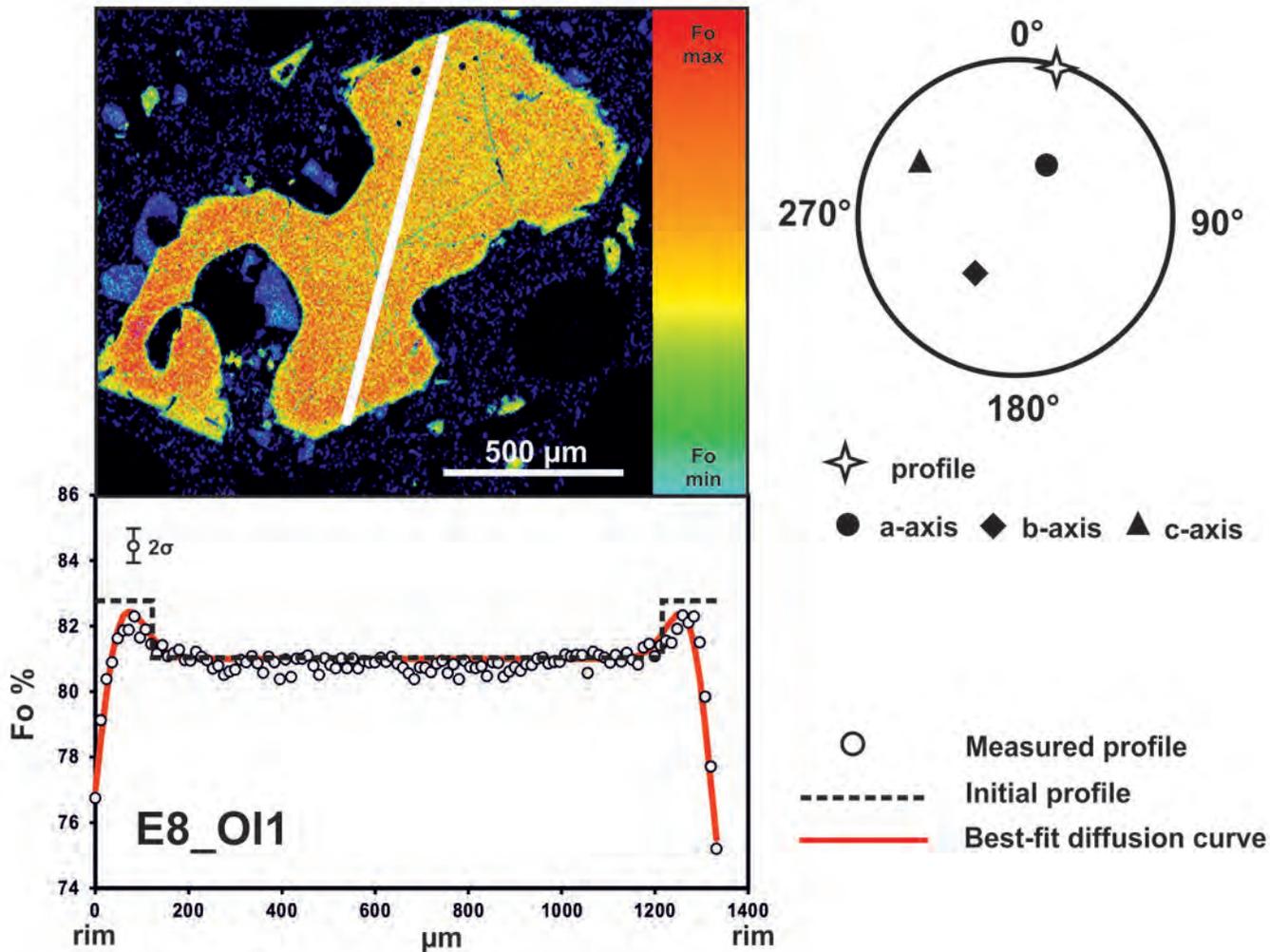
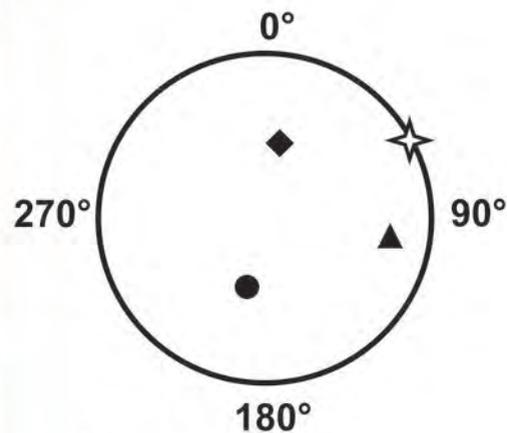
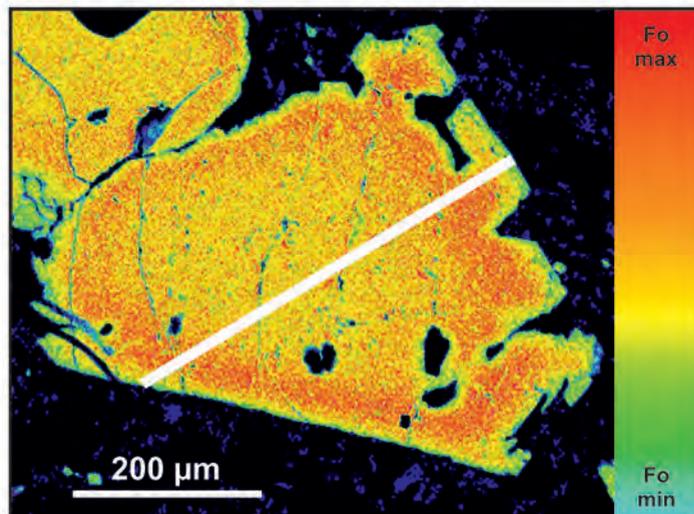
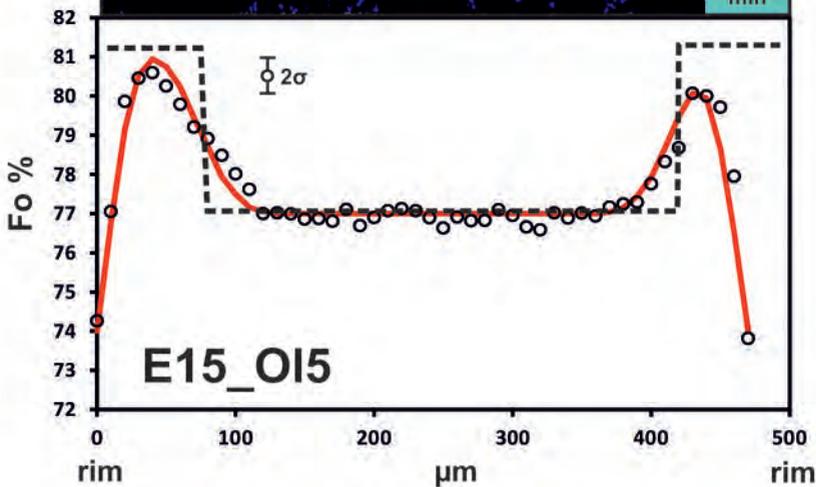


Figure 5



- ☆ profile
- a-axis ◆ b-axis ▲ c-axis



- Measured profile
- Initial profile
- Best-fit diffusion curve

Figure 6

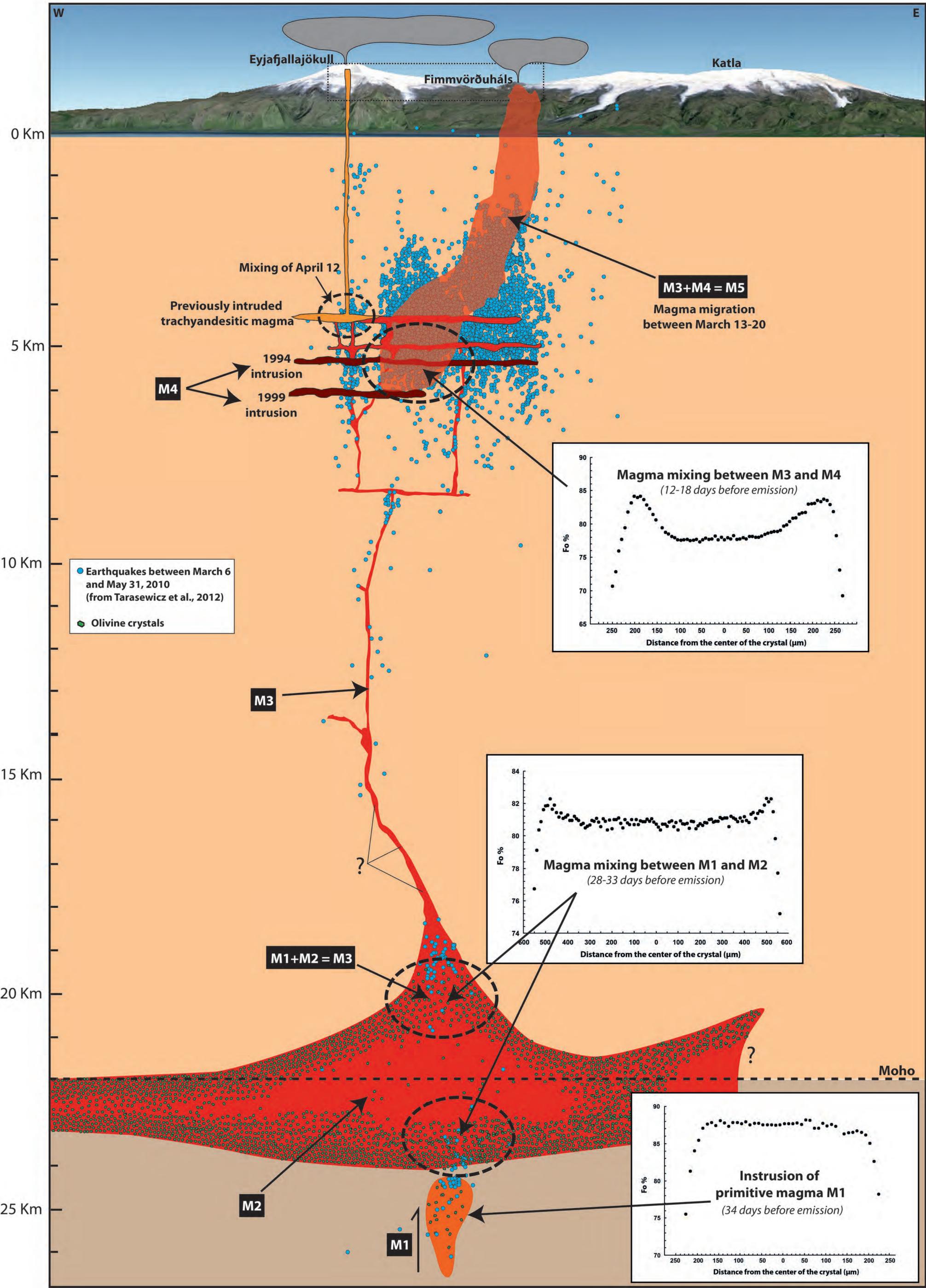


Table 1 - Calculated timescales of re-equilibration and main parameters used for modeling the Fe-Mg diffusion of selected olivine crystals from the 2010 basaltic eruption at Eyjafjallajökull.

| Population | Olivine | Core composition (Fo %) | Temperature (°C) | Pressure (Kbars) | fO_2 (Pa) | D_{trav} ($\mu m^2 \cdot s^{-1}$) | Average Time (days) |
|------------|---------|----------------------------|---------------------|---------------------|----------------------|--|------------------------|
| P1 | E8_O15 | 88 | 1210 | 6.15 | $3.55 \cdot 10^{-4}$ | $1.31 \cdot 10^{-4}$ | 34 |
| P2 | E4_O12 | 81 | 1210 | 6.15 | $3.55 \cdot 10^{-5}$ | $4.61 \cdot 10^{-4}$ | 33 |
| P2 | E8_O11 | 81 | 1210 | 6.15 | $3.55 \cdot 10^{-5}$ | $2.94 \cdot 10^{-4}$ | 28 |
| P3 | E4_O15 | 77 | 1150 | 1.45 | $2.19 \cdot 10^{-5}$ | $1.05 \cdot 10^{-4}$ | 18 |
| P3 | E15_O15 | 77 | 1150 | 1.45 | $2.19 \cdot 10^{-5}$ | $1.63 \cdot 10^{-4}$ | 18 |
| P3 | E15_O17 | 77 | 1150 | 1.45 | $2.19 \cdot 10^{-5}$ | $2.95 \cdot 10^{-4}$ | 12 |
| P3 | E3_O11 | 78 | 1150 | 1.45 | $2.19 \cdot 10^{-5}$ | $2.35 \cdot 10^{-4}$ | 18 |
| P3 | E15_O16 | 78 | 1150 | 1.45 | $2.19 \cdot 10^{-5}$ | $1.00 \cdot 10^{-4}$ | 16 |
| P3 | E8_O14 | 79 | 1150 | 1.45 | $2.19 \cdot 10^{-5}$ | $1.05 \cdot 10^{-4}$ | 12 |