

1 **The onset of an eruption: selective assimilation of hydrothermal minerals**
2 **during pre-eruptive magma ascent of the 2010 summit eruption of**
3 **Eyjafjallajökull volcano (Iceland)**

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19 dynamics.

20
21 **Abstract**

22 The complex processes occurring in the initial phases of an eruption are often recorded in the products of its
23 opening stage, which are usually characterized by small volume and limited dispersal, and thus generally poorly
24 studied. The 2010 eruption of Eyjafjallajökull (Iceland) represents a unique opportunity for these investigations
25 thanks to the good preservation of tephra deposits within the ice/snow pack. A detailed geochemical investigation
26 on the glassy groundmass of single ash clasts disclosed a population of fragments with unusual high ⁸⁷Sr/⁸⁶Sr (up
27 to 0.70668) for Icelandic magmatism, and anomalous elemental composition with respect to most of the juvenile
28 material of the eruption. This suggests that during its rise, before intruding into the ice cover, magma at a dyke tip
29 selectively assimilated hydrothermal minerals with seawater-related, high-Sr isotopic ratios (zeolites, silica phases,

30 anhydrite) hosted in altered volcanic/epiclastic rocks. According to the observed precursory seismicity, only
31 restricted to few hours before the onset of the eruption, this process could have accompanied subcritical aseismic
32 fracture opening during the days before the eruption, possibly related to stress corrosion-cracking processes, which
33 enhanced the partial dissolution/melting and subsequent selective assimilation of the host rocks.

34

35 **Introduction**

36 Studies of explosive eruptions are generally based on the products of their paroxysmal phases, recognised and
37 correlated at the scale of the whole dispersal area (Nakamura, 1964; Thorarinsson, 1954; Booth et al., 1978; Fisher
38 and Schmincke, 1984; Cas and Wright, 1987; Wilson, 1993; Cioni et al., 2015). Conversely, in most cases the very
39 initial products of an eruption, often related to transient phases of weak intensity, are not studied in detail, as they
40 are not easily recognized in the field due to the limited dispersal and to the fact that they are generally confined to
41 very proximal sites, often not easily accessible. However, the information that might be derived from these
42 products can be of fundamental importance to highlight timing and dynamics of the processes involved in the
43 initial stages of magma ascent to the surface (Cioni et al., 2000; Cashman and Hoblitt, 2004; Meletlidis et al.,
44 2012; Druitt, 2014). The study of initial deposits of an eruption can thus help the interpretation of its precursory
45 signals, which mostly detected in the form of ground deformation or seismicity (e.g. Sparks, 2003; Dzurisin, 2007;
46 Chouet and Matoza, 2013). These signals are related to pre-eruptive shallow magma storage and movements,
47 involving the final fracturing of rocks beneath the eruptive vent and the associated decompressional path of
48 magma to the surface. Consequently, a record of these processes can be preserved in the compositional and
49 textural features of the first erupted material.

50 Here, a large dataset of the textural features and the glassy groundmass elemental/isotopic compositions of
51 selected juvenile clasts is presented, in order to check possible micro-scale compositional heterogeneities within
52 the products of the opening phase of the 2010 Eyjafjallajökull eruption (Iceland). The results are interpreted in
53 terms of processes accompanying the initial phases of magma ascent and outbreak to the surface.

54 The 2010 eruption is particularly suitable for the investigation of its opening phase thanks to the perfect
55 preservation of the deposits emplaced during the first part of the explosive event, which represent 3-4 hours of
56 subglacial activity during which magma melted its way through the ice-filled summit caldera. The tephra clasts of
57 this stage were packed within the snow/ice after emplacement and strongly contrast with those of the following
58 ash-producing activity in terms of morphology and texture, as well as for their geochemical and isotopic signature.

59

60 **1. The 2010 summit eruption: brief chronology of events**

61 The 14 April - 22 May 2010 summit eruption of Eyjafjallajökull volcano (Iceland) was characterized by a large
62 production of ash that was mainly dispersed towards the east and south-east; the ash cloud caused significant
63 damage to vegetation and various economic sectors in Iceland, and reached the southern parts of Europe leading to
64 the most dramatic global disruption of air traffic since the WWII (Miller, 2011; O'Regan, 2011). The climactic
65 explosive activity was preceded (20 March-12 April) by a fissural basaltic eruption, characterized by lava
66 fountains and lava flows located on the eastern flank of Eyjafjallajökull, outside of the caldera border. After 2 days
67 of quiescence of seismic and eruptive activity, a summit eruption started on 14 April and continued, with variable
68 intensity, for 39 days (Gudmundsson et al., 2012). Seismicity preceded by only few hours the onset of the summit
69 eruption and was mainly concentrated in two main clusters at 1-2 km and 4-5 km, along a subvertical pipe below
70 the summit (Tarasewicz et al., 2012). The 14 April - 22 May activity has been divided into three main phases
71 (Gudmundsson et al., 2012; Bonadonna et al., 2013; Cioni et al., 2014):

72 - *Phase I*: A seismic swarm started at 22:29 UTC on 13 April, increasing in intensity during the following few
73 hours. A subglacial eruption possibly started around 01:15 UTC of 14 April, marked by a gradual increase in low
74 frequency tremor. The subglacial phase came to an end before 05:55 UTC, when a white, vapour-laden plume was
75 observed above the summit (ICAO report, 2012). Activity during this “opening” stage likely started along a fissure
76 several hundred meters long, as suggested by the alignment of at least 4 ice-melting cauldrons observed from the
77 first airborne synthetic aperture radar images taken by the Icelandic Coast Guard (Magnusson et al., 2012). The
78 intensity of the summit eruption gradually increased during the day, and culminated with the emergence at 18:30
79 UTC of a 9-10 km-high, dark ash-laden plume; the mass discharge rate reached about $1 \times 10^6 \text{ kg s}^{-1}$ and the activity
80 was characterized by steam-dominated and ash-poor plumes rising from a series of vents aligned along a 2 km-
81 long, north-south oriented fissure. Flash floods (jökulhlaups) were immediately triggered, forcing the evacuation
82 of around 800 people. Phase I was dominated by a continuous series of frequent (seconds to minutes) pulses,
83 mainly driven by the interaction of the magma with the ice-melting water and ash slurry inside the caldera
84 (Gudmundsson et al., 2012). This phase lasted until the early hours of 18 April, with more energetic pulses
85 generating sustained ash plumes on 14, 16 and 17 April.

86 - *Phase II* of the eruption began on 18 April and was characterized by a significant drop in the intensity of
87 explosive activity (mass discharge rate around $10^4 - 10^5 \text{ kg s}^{-1}$), and by the onset of lava emission, which lasted
88 until 4 May. With the exception of a strong explosive pulse on 28 April, low-level (typically below 5 km high),

89 ash-poor plumes characterized this phase. Seismic activity remained at prevalent shallow levels during Phase I and
90 II, and volcanic tremor intensified in correspondence of the onset of lava effusion during Phase II.

91 - *Phase III*: intense explosive activity renewed from 5 May, generating large amounts of fine-grained tephra that
92 were injected into the atmosphere up to 10 km above sea level and dispersed over Europe. During this phase
93 seismic activity migrated downward up to 30 km depth, concentrating in some clusters aligned on a nearly vertical
94 path under the volcano, suggesting the progressive involvement of magma stalling at depth (Tarasewicz et al.,
95 2012). After 18 May, activity progressively declined, outlining the final part of the eruption which ended on 22
96 May 2010.

97 The total volume of tephra emitted during 39 days of activity has been estimated at 0.32 km³ (0.19 km³ DRE) and
98 only about 5% of the volume reached the continental Europe; about 70% of material was produced during the first
99 phase (0.27 km³ or 0.15 km³ DRE).

100

101 **2. Tephra sampling and study methods**

102 An extensive tephra sampling was carried out in the first days of May 2010, between 2 and 56 km from the vent,
103 east and southeast of the volcano (see Bonadonna et al., 2011, Bagnato et al., 2013 and Cioni et al., 2014, for
104 further sampling details). Tephra emplaced during Phase I was collected from the primary fallout deposits packed
105 and interlayered in the ice (section EJ13, located ≈4.5 km east from the crater, Fig. 1). The basal layer (EJ13A) is a
106 2 cm-thick, coarse ash deposit; the intermediate tephra deposit (EJ13B) is a 5 cm-thick layer of coarse ash with
107 scattered lapilli and the upper layer (EJ13C) is a fine-grained, 10 cm-thick, faintly stratified, ash deposit (Fig. 2).
108 Sample EJ13A, in particular, was attributed to the opening stage of Phase I (14 April), whereas layers EJ13B and
109 EJ13C to the peak of activity of Phase I (14-18 April), during which most of the tephra sequence was emplaced
110 (Bonadonna et al., 2011, Cioni et al., 2014). Samples of Phase II (EJ13T and EJ07) were collected directly on the
111 glacier, at 4.5 and 20 km from the vent, respectively. Tephra of Phase III was collected approximately 9 km east of
112 the crater on May 5, while falling from the plume (EJ14T, EJ15). On the same day, scoria lapilli (EJ27) were also
113 collected around the crater area.

114 Major and trace element contents and Sr isotope ratios of tephra were determined on whole-rock samples, on fresh
115 matrix glasses of single juvenile ash clasts randomly collected in the 0.71-2 mm grain-size interval, and on
116 plagioclase phenocrysts. Nd isotopes were analyzed on whole-rocks. Although our goal was the characterization of
117 clasts from the opening stage (sample EJ13A), clasts collected from the following phases were also analysed in
118 order to highlight the main differences among samples. In particular, in order to check possible magma

119 compositional heterogeneities at the micro-scale, Sr isotope analyses on single ash clasts (for a total of 24
120 analyses) from the different eruptive phases were performed collecting micro-samples of matrix glass by the *in-*
121 *situ* microdrilling technique (Fig. 3). Using the same method, $^{87}\text{Sr}/^{86}\text{Sr}$ values were also determined *in-situ* on 1
122 mm-sized loose plagioclase phenocrysts (17 analyses) of different compositions (An_{12-84}) rimmed by juvenile
123 glass, in order to check crystal/liquid isotopic equilibrium. Volatile contents in the groundmass of 6 obsidian ash
124 clasts from EJ13A sample were measured by synchrotron Fourier Transform InfraRed (FTIR) microspectroscopy.
125 Clasts used for microanalysis were accurately selected in order to study juvenile fragments characterized by fresh
126 matrix glass and low microlite content; chemical analyses were always performed on glass portions far enough
127 from phenocrysts and, as far as we could, microlites. Methods are reported in the Analytical Procedure section of
128 the Supporting Information, together with Tables S1-S3 that summarize the complete dataset.

129

130 **3. Morphology and textures of the juvenile ash clasts**

131 The juvenile component ejected during the different phases of the eruption shows a large morphological and
132 textural variability also at the scale of the single eruptive phase (Dellino et al., 2011; Cioni et al., 2014).

133 Four main clast types were identified in the juvenile ash fraction (Cioni et al., 2014): spongy finely vesicular
134 (SFV), coarsely vesicular (CV), and fluidal (FL) clasts, characterized by moderate to high vesicularity, and blocky
135 clasts (BL), showing incipient to moderate vesicularity and a glass-dominated groundmass (Fig. 2). The four types
136 of clasts also differ by their internal texture (amount of microlites, amount and shape of vesicles; Cioni et al.,
137 2014). Relative abundance of the different juvenile clasts varies between the studied samples: SFV, CV and FL
138 clasts are usually dominant (>80 vol%), although with different proportions in the different phases, except only for
139 the opening stage (sample EJ13A), where BL clasts reach 40 vol%. BL clasts are very rare in the following phases
140 (Fig. 2). These are generally subequant, with 12-40 vol% of spherical to tubular vesicles, obsidian-luster
141 appearance and very low crystallinity of the groundmass. Plagioclase, clinopyroxene and minor oxides represent
142 the mineral groundmass phases, regardless of the texture and crystallinity of the clasts and of the eruption phase
143 (Cioni et al., 2014).

144

145 **4. Geochemistry of the juvenile ash clasts**

146 Whole-rocks are mainly benmoreites, with some dacitic samples (Table S1) similar to the less alkaline glass
147 compositions reported by Keiding and Sigmarsson (2012).

148 Matrix glasses have silica contents between 55 and 68 wt%, as also reported by recent works (Sigmarsson et al.,
149 2011; Borisova et al., 2012; Gudmundsson et al., 2012; Keiding and Sigmarsson, 2012). There is no strong
150 correlation between glass composition, clast type and stratigraphic position of the sample (Figs. 2, 4). Some clasts
151 (mainly BL-type) from products of the opening stage (sample EJ13A), however, show anomalous compositions
152 characterized by higher silica contents (up to 65.5 wt%) compared with those of Phase I (59-62.5 wt%; Fig. 4 and
153 Table S2). Although major elements usually display good correlations with silica, these samples plot outside the
154 general Al₂O₃ - SiO₂ negative correlation (Fig. 4c), having higher alumina contents. The same clasts also contain
155 lower K₂O, MgO, V and Sc and higher Na₂O in respect with other glasses with comparable silica content from the
156 successive eruptive phases (Fig. 4; Table S2).

157 Trace element contents of matrix glass are generally well inter-correlated and overlap the compositional trends of
158 literature data (Sigmarsson et al., 2011; Borisova et al., 2012) (Fig. 4; Table S2), although Sigmarsson et al. (2011)
159 report larger compositional variations (e.g., Th: 3.6-10.9 ppm, Rb: 26.0-80.5 ppm). Samples EJ13A and, to a lesser
160 extent, EJ13B, show a larger scatter of Rb, Ba and U abundances than the other trace elements; U, in particular, is
161 also more enriched in most of the clasts. BL clasts showing anomalous major element composition are also
162 generally enriched in U, Rb and Ba (Fig. 4 and Table S2).

163 ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd of whole-rocks are virtually constant within the analytical errors (Fig. 5; Table S2); they
164 are similar to those already reported for this eruption (Sigmarsson et al., 2011; Borisova et al., 2012) and fall in the
165 typical isotope range of the Icelandic magmatism (⁸⁷Sr/⁸⁶Sr: 0.7026-0.7037; Sigmarsson et al., 2008) (Fig. 5).
166 ⁸⁷Sr/⁸⁶Sr were also analyzed *in-situ* on matrix glasses of single clasts, selected based on different texture and
167 composition. Results indicate that clasts from samples EJ13C and EJ15 have quite constant ⁸⁷Sr/⁸⁶Sr and similar to
168 the whole-rock values; conversely, glasses from samples of the opening stage (EJ13A) and paroxysmal Phase I
169 (EJ13B) show a larger Sr-isotopic variability, ranging from typical values recorded in Icelandic fresh volcanic
170 products up to much higher values never found in Iceland magmatism (Fig. 5). Significantly, these high values are
171 measured in the compositionally anomalous BL clasts of the opening stage, especially in the glass of clast D2 from
172 sample EJ13A (hereafter *sample-[clast]*, e.g. EJ13A-[D2]) which attains a value of 0.70668, remarkably unusual
173 for Icelandic magmatism. Considering that the blank contamination level during analyses resulted in negligible
174 contribution (≤1%), and therefore not able to affect the third decimal digit of isotopic data (details in the
175 Analytical Procedure section of the Supporting Information), the obtained high Sr isotope values have a real
176 significance. Only one (EJ13B-[C6]) out of 5 clasts from the paroxysmal Phase I has ⁸⁷Sr/⁸⁶Sr significantly higher

177 than Icelandic isotopes (Fig. 5), although this clast, belonging to the finely vesicular group, is not characterized by
178 anomalous elemental composition (Figs. 4, 5).

179 *In-situ* Sr-isotopes of plagioclase crystals with variable anorthite content are low, constant and similar to the
180 whole-rock values (0.70324-0.70331), ranging from 0.70321 to 0.70335. A core-rim pair from a plagioclase with
181 normal zoning and sieved core resulted in isotopic equilibrium (Fig. 5; Table S3), confirming that isotope ratios
182 did not change during crystallization.

183 Groundmass glasses of six BL clasts from EJ13A have low water content in residual glasses, averaging at 0.54
184 wt% (0.35- 0.72 wt%); CO₂ content in these glasses was below the detection limit of the apparatus.

185

186 **5. Discussion**

187 Presented data point out some interesting characteristics of the juvenile ash clasts emitted during the opening stage
188 of the 2010 Eyjafjallajökull summit eruption. These include:

189 - few clasts from the initial stage are characterized by high ⁸⁷Sr/⁸⁶Sr values and anomalous elemental compositions
190 (higher SiO₂, Al₂O₃, Na₂O, U, Rb and lower MgO, K₂O) compared with the other products of the eruption;

191 - the majority of clasts from the following phases of the eruption (irrespective of their textural features) have the
192 typical isotopic compositions of Icelandic rocks (⁸⁷Sr/⁸⁶Sr: 0.7026-0.7037; Sigmarsson et al., 2008);

193 - most of the anomalous glass shards have similar textural features, being blocky, glassy, and nearly microlite-free
194 fragments (BL type);

195 - differently from glass shards, plagioclase phenocrysts do not reveal any isotopic anomaly.

196 We interpret these characteristics as related to the processes occurring during magma ascent immediately
197 preceding the onset of the eruption, and they are used to infer the modalities and relative timing of magma rise.

198

199 **5.1. Anomalous compositions of the juvenile clasts from the opening stage**

200 The remarkable unusual composition of some clasts from the opening stage can be interpreted as the result of the
201 occurrence of transient, local-scale phenomena during the final ascent of the magma. The strongly anomalous
202 isotopic composition measured in the BL clasts is particularly interesting. Firstly, we can exclude that the high Sr
203 isotopes of these glass shards is related to assimilation of Icelandic fresh oceanic crust, due to the low ⁸⁷Sr/⁸⁶Sr that
204 characterizes these rocks (Sigmarsson et al., 2008). As a consequence, the only way to increase radiogenic Sr
205 content in magmas up to the measured values is to assimilate components derived by interaction with seawater
206 strontium, having ⁸⁷Sr/⁸⁶Sr of 0.70920 (Elderfield and Greaves, 1981). These components could be represented by:

207 i) bulk oceanic crust altered by seawater circulation; ii) carbonate marine sediments; iii) alteration minerals formed
208 by seawater-related hydrothermal fluids, where marine Sr can be particularly concentrated. Among the three
209 options, we exclude the assimilation of sediments because, despite the high $^{87}\text{Sr}/^{86}\text{Sr}$ (e.g., Sveinbjörnsdóttir et al.,
210 1993), this cannot justify the elemental compositional variability observed in the anomalous clasts. Furthermore,
211 with respect to the other products, these clasts do not show anomalous CaO contents (Table S2), as expected if
212 assimilation of carbonate sediments had occurred.

213 In order to understand which component contaminated a part of the firstly erupted magma and to explain the
214 $^{87}\text{Sr}/^{86}\text{Sr}$ and Sr contents of the anomalous glasses, we performed quantitative calculations, considering a process
215 of simple contamination instead of assimilation + fractional crystallization (AFC) or any other more complex
216 process involving crystallization. The exclusion of AFC or other processes is due to the low, Sr-isotope ratios
217 measured in all the analysed plagioclase phenocrysts, which are also equal to those of most of the 2010 host
218 magmas (Fig. 5; Tables S1-S3), and close to the average values measured for Icelandic magmas. Melt
219 contamination without involvement of mineral phases clearly indicates that crystal fractionation was not
220 contemporaneous with magma contamination. In particular, the two similar isotope ratios found in core and rim of
221 a plagioclase phenocryst (Fig. 5; Table S3) suggest that phenocryst crystallization occurred separately from any
222 assimilation process (in the opposite case, a higher Sr isotope in the rim would be expected).

223 Because the simple contamination process is described by mixing equation, we performed mixing calculation
224 between low Sr-radiogenic magmas and components with seawater Sr-isotope ratios (Fig. 6a, b). The matrix glass
225 of EJ13A-[D2] has high Sr-isotopes but relatively low Sr content compared with glasses of lower silica
226 abundances (Fig. 6a); thus, the assimilated component leads to increase isotopes, without increasing Sr content.
227 We can reasonably assume that the original Sr concentration in the uncontaminated magma was in the range
228 measured for EJ13A glasses (190-367 ppm; Fig. 6a) and whole rock (359 ppm; Table S1). Accordingly,
229 calculations indicate that, depending on the starting Sr content, the compositions of the EJ13A-[D2] and EJ13A-
230 [b1+C1] clast can be achieved by high assimilation degrees (50-70 wt% and 15-55 wt%, respectively) of possible
231 components with 185-260 ppm and 95-360 ppm of Sr, respectively (Fig. 6b). A mixing line (Sr contents of magma
232 and contaminant: 210 and 240 ppm, respectively) fits both the EJ13A-[D2] and EJ13A-[b1+C1] compositions,
233 indicating assimilation degrees of 55% and 20%, respectively. We suggest this is the most suitable model able to
234 explain the shallow level magma contamination in the opening stage (Fig. 6b). It is noteworthy that the amount of
235 assimilation would increase considering contaminants with Sr-isotopes lower than the seawater value.

236 These results allow excluding the seawater-altered bulk oceanic crust as possible contaminant, due to its $^{87}\text{Sr}/^{86}\text{Sr}$
237 much lower than 0.70920 (generally lower than 0.706, rarely reaching 0.707 even in palagonite; Menzies and
238 Seyfried, 1976; Elderfield and Greaves, 1981; Staudigel and Hart, 1983; Kawahata et al., 2001) and also
239 commonly lower than $^{87}\text{Sr}/^{86}\text{Sr}$ of the EJ13A-[D2] glass.

240 Selective assimilation of alteration minerals formed by seawater-derived hydrothermal fluids, where these
241 minerals are the only crustal components melted by the hot magmas (Kitchen, 1985), seems therefore the most
242 reasonable explanation for constraining the anomalous compositions of clasts erupted during the opening stage.
243 Secondary mineral phases, possibly hosted in veins and vacuoles of the basaltic basement (e.g., Weisenberger and
244 Selbekk, 2009), can allocate radiogenic Sr from seawater, attaining high Sr-isotopes up to the seawater values
245 (e.g., Humphris and Bach, 2005; Marks et al., 2010). They also have specific compositions able to change the
246 geochemical characteristics of the assimilating magmas towards those observed in the anomalous glasses. The
247 assimilated minerals should account for the SiO_2 , Al_2O_3 , and Na_2O increase and the MgO , V and Sc decrease in
248 the high $^{87}\text{Sr}/^{86}\text{Sr}$ glass shards, as well as the nearly constant (or negligible decrease) K_2O and CaO contents (Fig.
249 4; Table S2). The composition of the contaminating material (black boxes in figures 4a,c,e) has been tentatively
250 quantified by inverse mixing calculation (Table S4 in the Electronic Supporting Information), using the hybrid
251 composition of EJ13A-[D2] clast, the assimilation degree of 55 wt% resulted by the most suitable Sr vs $^{87}\text{Sr}/^{86}\text{Sr}$
252 model (Fig. 6b: mixing line with the contaminant at 240 ppm of Sr), and the most appropriate composition of
253 assimilating magma deduced by geochemical trends of Figs. 4 and 6a. Specifically, the silica content of the latter
254 magma has been approximately estimated from the Sr vs. SiO_2 negative correlation (Fig. 6a) fixing Sr content at
255 210 ppm (Fig. 6b); then, the other element contents have been assessed by their trends with silica (Fig. 4; Table
256 S4). Among the variety of hydrothermal minerals (e.g., Alt et al., 1986; Jakobsson and Moore, 1986; Deer et al.,
257 1992; Weisenberger and Selbekk, 2009), the association in different proportions of zeolites (mainly analcite for 38
258 wt% and phillipsite for 31 wt%, with 2% of chabasite), silica phases (24 wt%) and few amounts of apophyllite (5
259 wt%; Table S4) could be the most suitable assimilated mineral assemblage for explaining the previously calculated
260 contaminant compositions of the EJ13A-[D2] clast glass (Table S4) (Marriner et al., 1990; Franzson et al., 2008;
261 Weisenberger and Selbekk, 2009). We can also speculate that the similar Al_2O_3 , SiO_2 and Na_2O contents and
262 lower Sr isotopes of EJ13A-[b1+C1] and EJ13A-[C3] clast glasses (Figs. 4, 5), in respect with the EJ13A-[D2]
263 clast glass, are linked to the expected compositional variability of the zeolite assemblage, both in elements and
264 isotopes. These glasses could have indeed assimilated a lower amount of prevailing analcite + silica phases
265 (having higher Al_2O_3 , SiO_2 and Na_2O contents) or, more probable, a similar amount of an analogue zeolite

266 assemblage with lower $^{87}\text{Sr}/^{86}\text{Sr}$ (around 0.707) than seawater. In the latter case, the contamination degree and the
267 assimilated hydrothermal mineral assemblage would remain the same for all the anomalous glasses but $^{87}\text{Sr}/^{86}\text{Sr}$ of
268 the contaminant would not be constant at the seawater value as we could effectively expect. Hence, the elemental
269 and isotopic compositions of the anomalous clasts can be reasonably achieved by selective assimilation of
270 hydrothermal secondary components, justifying the deviations from the compositional trends of the other glasses
271 (Figs. 4, 5).

272 Considering trace elements and their large variability in zeolites (e.g., Rb: 5-120 ppm, Ba: 40-2400 ppm; Wood et
273 al., 1976; Deer et al., 1992; Pickhardt et al., 2000; Weisenberger and Selbekk, 2009), we can hypothesise the
274 occurrence of assimilated minerals with particularly high Rb, Ba, U, and low Sr contents. This assumption is based
275 on trace element contents of the EJ13A anomalous glasses, together with the scatter of U, Rb and Ba abundances
276 in the products erupted at the beginning of the eruption (EJ13A and EJ13B; Figs. 2, 4).

277 Analyses carried out on tephra clasts from the overlaying stratigraphic layer of Phase I (sample EJ13B) show that
278 only one clast (EJ13B-[C6]) has slightly higher $^{87}\text{Sr}/^{86}\text{Sr}$ than the isotopic variability of Icelandic rocks (Fig. 5).
279 Glass from the same clast also shows a significantly different composition than those of the EJ13A-[D2] and
280 EJ13A-[b1+C1] glasses, particularly containing higher Sr and lower silica and Na_2O contents. These evidences
281 may suggest that a small amount of EJ13B magma, possibly less evolved than the assimilating EJ13A magma, was
282 slightly contaminated by a different crustal component in respect to that involved in the EJ13A anomalous glasses.
283 In Fig. 6b, a mixing model shows that the EJ13B-[C6] composition could be explained by only 4% assimilation of
284 a possible component with quite high Sr abundances (900 ppm), starting from Sr content of the EJ13B whole rock
285 (294 ppm). Based on these considerations, we may suppose this magma assimilated a different zeolite assemblage,
286 possibly with also the involvement of anhydrite, having high Sr contents and Sr isotopes (Kawahata et al., 2001;
287 Humphris and Bach, 2005; Rubio et al., 2005; Marks et al., 2010).

288

289 **5.2. Significance of BL clasts**

290 The textural features of BL clasts in the deposits of the opening stage of the eruption suggest mechanisms of
291 hydromagmatic fragmentation (completely lacking in the following activity) and magma granulation by
292 quenching, possibly occurred when magma firstly intruded at the base of the ice cover (Cioni et al., 2014).
293 Accordingly, the low vesicularity of BL clasts and the absence of microlites also suggest that they possibly
294 represent fragments of the sill/dyke body quenched at contact with ice (Magnusson et al., 2012; Cioni et al., 2014).
295 Pressure at which quenching occurred can be derived from the dissolved water concentration measured in BL glass

296 (Tuffen et al., 2007; Stevenson et al., 2009; Tuffen et al., 2010). Using the solubility model of Papale et al. (2006)
297 and a magma temperature of 1040°C (Keiding and Sigmarsson, 2012), results suggest that degassing stopped at
298 pressures corresponding to an average water depth of 380 m (ice thickness: 414 m). This well agrees with the
299 caldera glacier thickness (200-400 m; Magnusson et al., 2012) and indirectly suggests that, in the hypothesis that
300 degassing was interrupted by ice-magma contact, the preceding syn-eruptive decompressional degassing occurred
301 close to equilibrium (no water in excess). We suggest here that BL clasts are the product of non-explosive
302 fragmentation of the vanguard magma intersecting the ice cover during the opening stage of the eruption, and
303 erupted to the surface when the gas/water/pyroclast mixture broke its way through the summit glacier.

304

305 **5.3. Magma ascent dynamics during the opening phase**

306 The “typical Icelandic” values of Sr-isotopes of the analysed plagioclase phenocrysts, in strong disequilibrium
307 with the values measured in the glass of anomalous BL clasts, suggest a late-stage contamination during magma
308 ascent. This occurred after phenocryst crystallization and involved only a very small portion of the erupting
309 magma, as suggested by the low amount of anomalous clasts (Figs. 4,5; Table S2). Given the occurrence of
310 juvenile fragments with high Sr-isotopic values only in the deposits of the initial stage of the eruption, and the
311 absence of isotopic equilibrium between this glass and phenocrysts, we propose that contamination involved only
312 the vanguard portion of the ascending dyke while it opened its way to the surface (Fig. 7), after phenocryst
313 crystallization.

314 Assimilation could have been favoured by the relatively low magma supply rate to the ascending dyke, which
315 possibly characterized the precursory phases of the eruption, as previously supposed on the basis of syn-eruptive
316 decompressional degassing probably occurring close to equilibrium. In this case, 1:1 proportions of magma to
317 contaminant (i.e., up to 55% of assimilated crustal components) could be consistent with interaction between the
318 head (and walls) of an opening dyke and the host rock. In this process, even assimilation of small amounts
319 (compared to the entire volume of the erupted magma in the opening phase) of hydrothermal minerals hosted in
320 the wallrock could have significantly contaminated the ascending magma, because only the small magma portion
321 at the magma/wallrock contact of the dyke tip is involved in the process, without chemical re-equilibration with
322 the inner and lower portions of the dyke which, in turn, continuously supplied the heat needed to assimilation (Fig.
323 7). These transient processes interesting only a small, well confined magma portion can be only detected when
324 single clasts from the opening phase products are separately analysed.

325 To constrain the main physical parameters of this magma contamination process, we have roughly calculated the
326 volume of contaminated magma and of related assimilated rocks, in order to hypothesize the volume balance
327 between magma and assimilated material. A first order approximation for the total erupted material during the
328 opening stage can be derived by the few available data on thickness of the EJ13A deposit, obtained tracing a
329 nearly circular isopach for each available point (Fig. 1) and using the methods of Pyle (1989) and Legros (2000).
330 The estimated deposit volume ($3.5 \times 10^6 \text{ m}^3$; average of the values from the two methods) can be converted to dense
331 rock equivalent (DRE) of $1.2 \times 10^6 \text{ m}^3$, using a magma density of 2500 kg m^{-3} and an average deposit density of
332 900 kg m^{-3} (as measured in similar deposits; Williams, 1983; Coltelli et al., 1998), corresponding to a magma mass
333 of $3.1 \times 10^9 \text{ kg}$ (Table S5 in the Electronic Supporting Information). The low percentage of anomalous clasts, which
334 are approximately 20% of the BL type clasts (Figs. 4,5; Table S2), and hence about 8% of the whole material
335 erupted during the opening stage, suggests a mass of contaminated magma of $2.5 \times 10^8 \text{ kg}$ (Table S5). This mass is
336 used to constrain the volume of wallrock involved in the contamination process discussed above. An assimilation
337 degree of 50 wt%, chosen among the highest values calculated by Sr-isotopic modelling, results in a mass of
338 assimilated mineral paragenesis of about $1.3 \times 10^8 \text{ kg}$. In a selective assimilation, where only the alteration minerals
339 are digested by the uprising magma (Kitchen, 1985), the volume of altered bulk rock involved in the
340 contamination process depends by its degree of alteration (i.e., percentage of hydrothermal minerals), which can
341 largely vary. In any case, using a moderate alteration degree of 20 wt% (e.g., Weisenberger and Selbakk, 2009) we
342 estimate a mass of involved bulk rock of $6.3 \times 10^8 \text{ kg}$ (corresponding to a volume of $3.1 \times 10^5 \text{ m}^3$ assuming a density
343 of 2000 kg m^{-3} for an altered basement rock; Table S5) being aware of the fact that this estimation is largely
344 conservative in the case of a more pervasively altered wallrock. Assuming that this mass was possibly confined in
345 the contact zone with the dyke walls, we can calculate its thickness on the base of hypothesised dyke geometry.
346 Direct observations of magma intercepting the glacier during the first stages of the eruption suggest a NNE-SSW
347 oriented fissure a few hundred meters long, which possibly propagated from a maximum depth of 5 km (based on
348 seismic data; Tarasewicz et al., 2012; Magnusson et al., 2012). Accordingly, confining the estimated volume of the
349 bulk rock interested by selective assimilation along a dyke assumed to have a length between 50-200 m and depth
350 between 1000-5000 m, we calculate a thickness of the shell of wallrock involved in the assimilation ranging from
351 0.15 to 3 m. This thickness represents a highly conservative estimation, as it could even be smaller in case of a
352 more altered wallrock or of an assimilation degree lower than 50%, consistent with the modelling of intermediate
353 Sr-isotope ratios such as those of EJ13A-[b1+C1] glasses (Fig. 6). Thus, these calculations indicate that the
354 amount of assimilated material in the opening stage system is at least one order of magnitude lower respect to the

355 total erupted magma (Table S5), confirming a negligible thermal effect on magma. An independent confirmation
356 of an important interaction of the ascending magma with hydrothermally altered rocks derives from a study of the
357 ash erupted during the different phases of the eruption (Paque et al., 2016). Interestingly, the amount of zeolites
358 and minor smectites observed in the ash was greater during the first phase of the eruption and progressively
359 decreased during the following phases, possibly following the formation of a stable conduit system.

360 The abundant data on seismic activity preceding the beginning of the summit eruption can give some hints about
361 the timing of the assimilation event. Seismicity occurred only few hours before the onset of the summit eruption,
362 possibly related to the final fracturing of host rocks and consequent rapid magma ascent, and was mainly
363 concentrated in two main clusters at 1-2 km and 4-5 km along a subvertical pipe below the summit (Tarasewicz et
364 al., 2012). This time is probably too short for the opening and magma invasion of a dyke connecting the main
365 magma reservoir (4-5 km depth; Gudmundsson et al., 2012), suggesting that the upward migration of the dyke tip
366 from the main shallow reservoir around 4 km up to 1-2 km could have occurred aseismically at least in the two
367 days of seismic quiet separating the end of the lateral eruption and the onset of the summit eruption. A viable
368 mechanism for rock weakening and aseismic subcritical crack growth can be represented by stress corrosion, as
369 already recognized at other volcanoes (Anderson and Grew, 1977; Atkinsons, 1984; Kilburn and Voight, 1998;
370 Vinciguerra, 1999). This mechanism of chemical reaction between country rocks and fluids (possibly exsolving
371 magmatic fluids and geothermal fluids) is able to fracture rocks at stresses much smaller than their theoretical
372 strength. Stress corrosion has been considered as a possible mechanism generating intermediate to short-term
373 seismic quiescence or variation in seismic rate (Main and Meredith, 1991). Accordingly, at Eyjafjallajokull, the
374 process of selective assimilation of hydrothermal minerals by the vanguard magma could have started before the
375 seismic crisis that heralded the eruption; hot acidic fluids exsolved during pre-eruptive magma ascent might have
376 enhanced the selective dissolution of altered rocks, making available for magma contamination suitable portions of
377 melted/dissolved hydrothermal minerals through a complex process of fluid/melt interaction. The absence of
378 isotopic equilibrium between the small contaminated portion of the ascending magma and its phenocrysts,
379 however, confirms that the process of stress corrosion/assimilation possibly occurred on a short timescale (few
380 days?) , after phenocrysts crystallization; the days-long seismic quiescence which preceded the onset of the
381 summit eruption could so be considered an indirect measure of the minimum length of this time interval.

382

383 **6. Conclusive remarks**

384 Tephra clasts erupted during the very first (hours-lasting) phase of the 2010 Eyjafjallajökull eruption were
385 immediately packed within the ice/snow cover and thus preserved after the eruption.

386 We performed microanalyses on matrix glass of single clasts from this opening phase; the obtained chemical
387 dataset allowed to detect transient processes occurring during the final part of magma rise. In particular, results
388 suggest that:

389 1) dense, obsidian, blocky clasts (BL) are abundant during the opening stage of Phase I; some of these
390 (approximately 20%) show anomalous elemental compositions with respect to the material of the following phases
391 of the eruption, as well as unusual high $^{87}\text{Sr}/^{86}\text{Sr}$ values, even for Icelandic magmatism;

392 2) based on quantitative mixing calculations, we propose that the very first erupted magma was contaminated by
393 selective assimilation of hydrothermal minerals (e.g., zeolites, quartz, \pm apophyllite, \pm anhydrite) present in veins
394 or vacuoles of basement rocks altered by seawater-derived fluids from which the high Sr-isotopes are derived;

395 3) this contamination occurred at shallow levels, during the slow ascent of a vanguard magma body in the pre-
396 eruptive phase. Assimilation affected only a small portion of the ascending magma (possibly the magma hosted in
397 the upward migrating tip of the intruding dyke), which, once reached the ice cover, rapidly quenched and
398 fragmented to form BL clasts. These were finally ejected as soon a cauldron opened in the glacier and the eruptive
399 column started, and were emplaced together with vesicular fragments in the basal deposit;

400 4) mechanisms of stress corrosion driven by exsolved acidic fluids at the tip of the migrating dyke could have
401 enhanced the dissolution of shallow-level hydrothermal minerals of altered rocks and the final magma
402 contamination. Such a process has been often discussed and reported as the possible cause of dyke propagation
403 before an eruption. We suggest that the occurrence of magma contamination could represent an important side-
404 effect of this important process, and that it should deserve more detailed studies in the future;

405 5) The presented data confirms the importance of studying in detail the products of the very initial phases of an
406 eruption, as they can give important hints on the processes accompanying the final magma ascent and outbreak to
407 the surface, giving information on timing and modalities of these processes.

408

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554

555 **Figure captions**

556

557 **Figure 1** - General map with sampling sites. Green dots refer to samples used in this study. Red diamonds
558 represent other surveyed sites during the fieldwork. Dispersal of the opening phase deposit is also shown with

559 hatched lines (thickness in cm). Geographic coordinates of sampling sites are: EJ13 UTM 27V 572791 7056448,
560 EJ13T UTM 27V 572791 7056448, EJ07 UTM 27V 588031 7051116, EJ14T UTM 27V 570354 705628, EJ15
561 UTM 27V 577553 7053608, EJ27 UTM 27V 566401 7057469.

562
563 **Figure 2** - Stratigraphic sequence packed within the ice with samples (EJ13A, EJ13B, EJ13C, EJ07, EJ15) and
564 relative abundances (vol.%) of different clast types within each sample (BL=blocky-obsidian clast; SFV=spongy
565 finely vesicular clast; CV=coarsely vesicular clast; FL=fluidal clast). Back-scattered electron microscopy images
566 of different clast types are also shown. The base of the sequence also shows tephra of basaltic lava fountains from
567 the effusive fissural eruption occurred on the East flank of Eyjafjallajökull volcano from 20 March to 12 April
568 2010. Please note that the picture of the tephra sequence do not correspond exactly to EJ13 location but was taken
569 ≈2 km eastwards, where also the basaltic layer was preserved (photo by A. Hoskuldsson; geographic coordinates
570 27V 576791 7057007 UTM).

571
572 **Figure 3** - Comparison between back-scattered electron microscopy images of clasts before (left) and after (right)
573 *in-situ* microdrilling.

574
575 **Figure 4** - Inter-elemental diagrams for matrix glass compositions of ash clasts from samples EJ13A and EJ13B,
576 reported according to their different morphology (BL=blocky-obsidian clast; CV=coarsely vesicular clast,
577 SFV=finely vesicular clast; *n.d.*=not determined). For comparison, glass compositions of EJ13C (Phase I) and
578 Phase II - III samples are also reported as variation field. Clast names show those with anomalous high Sr isotopes
579 (see Fig. 5). Crosses in figures b, d, f represent data reproducibility on trace elements as 2 standard deviations
580 from the mean. Lines in figures a, c, e show examples of mixing calculation between the assimilating magma (full
581 circles) and the contaminating material (boxes); the latter composition has been tentatively quantified by inverse
582 mixing calculation (Table S4 in the Electronic Supporting Information), using the hybrid composition of EJ13A-
583 [D2] clast explained by $^{87}\text{Sr}/^{86}\text{Sr}$ model (see Fig. 6) through an assimilation degree of 55 wt% (see text and Table
584 S4 for other explanations). The composition of the anomalous clast with higher silica content (down triangle),
585 where Sr isotopes and trace elements are not determined, is probably explained by increasing the quartz/zeolite
586 proportion in the assimilated material.

587

588 **Figure 5** - $^{87}\text{Sr}/^{86}\text{Sr}$ vs major element diagrams for matrix glasses of single clasts from the analyzed samples
589 (EJ13A, EJ13B, EJ13C for Phase I, EJ15 for Phase III). *In-situ* isotopes (glass and plagioclase) have been
590 analyzed by microdrilling technique. Error bars are inside the symbols. Some matrix glasses (e.g. b1+C1) from
591 distinct, but geochemically similar, clasts have been combined to achieve the micro-milled glass sufficient for
592 allowing Sr-isotope analyses.

593

594 **Figure 6** - Sr contents versus silica (a) and $^{87}\text{Sr}/^{86}\text{Sr}$ (b) diagrams for matrix glasses of single clasts. Symbols and
595 compositional field are as in Figures 4 (a) and 5 (b). Error bars are inside the symbols. The best fits of mixing
596 calculations are reported in figure (b) as mixing lines between uncontaminated magmas, having low Sr-isotopes
597 and different Sr contents, and assimilated hydrothermal minerals assumed to have $^{87}\text{Sr}/^{86}\text{Sr}$ of seawater (Elderfield
598 and Greaves, 1981). The variable Sr contents of the contaminating mineralogical paragenesis are calculated for
599 each assumed Sr abundance in the uncontaminated magmas, using for the latter the minimum (190 ppm) and
600 nearly maximum (350 ppm) values and the more suitable value (210 ppm) that better fits two clast glass
601 compositions. Transversal bars along the lines with numbers indicate contamination degrees (in %).

602

603 **Figure 7** - Simplified sketch of selective assimilation of hydrothermal mineral paragenesis present in altered rocks
604 by the opening stage magma during its early ascent towards the surface. Minerals dissolution was possibly
605 enhanced by stress corrosion mechanism driven by exsolved acidic fluids at the tip of the migrating dyke. When
606 magma reached the ice cover rapidly quenched (to form blocky clasts) and assimilation stopped due to fast ascent.
607 *Legend* – Solid (1) and melted (2) hydrothermal minerals in veins and vacuoles; 3: release of exsolved acidic
608 fluids.

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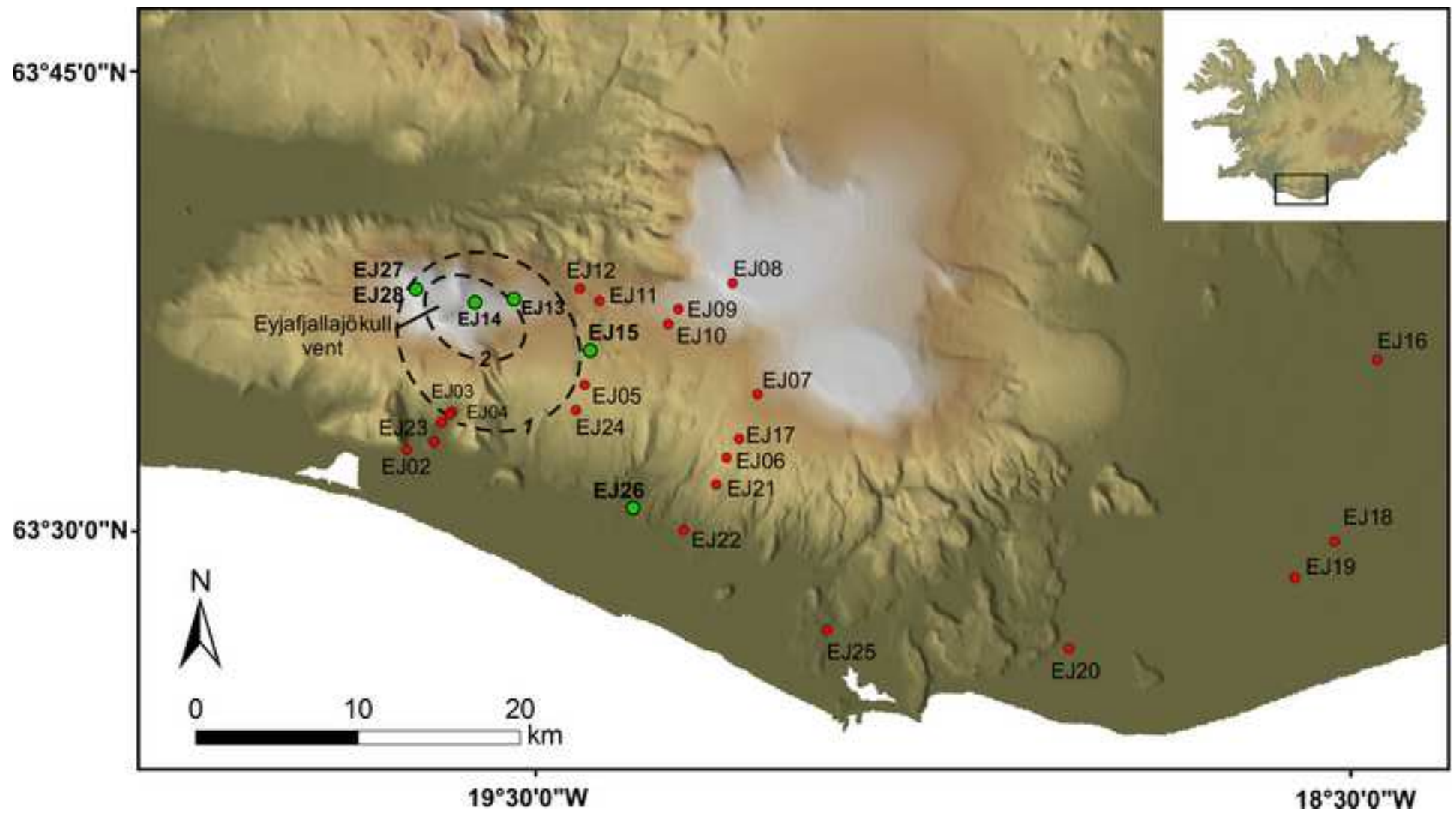


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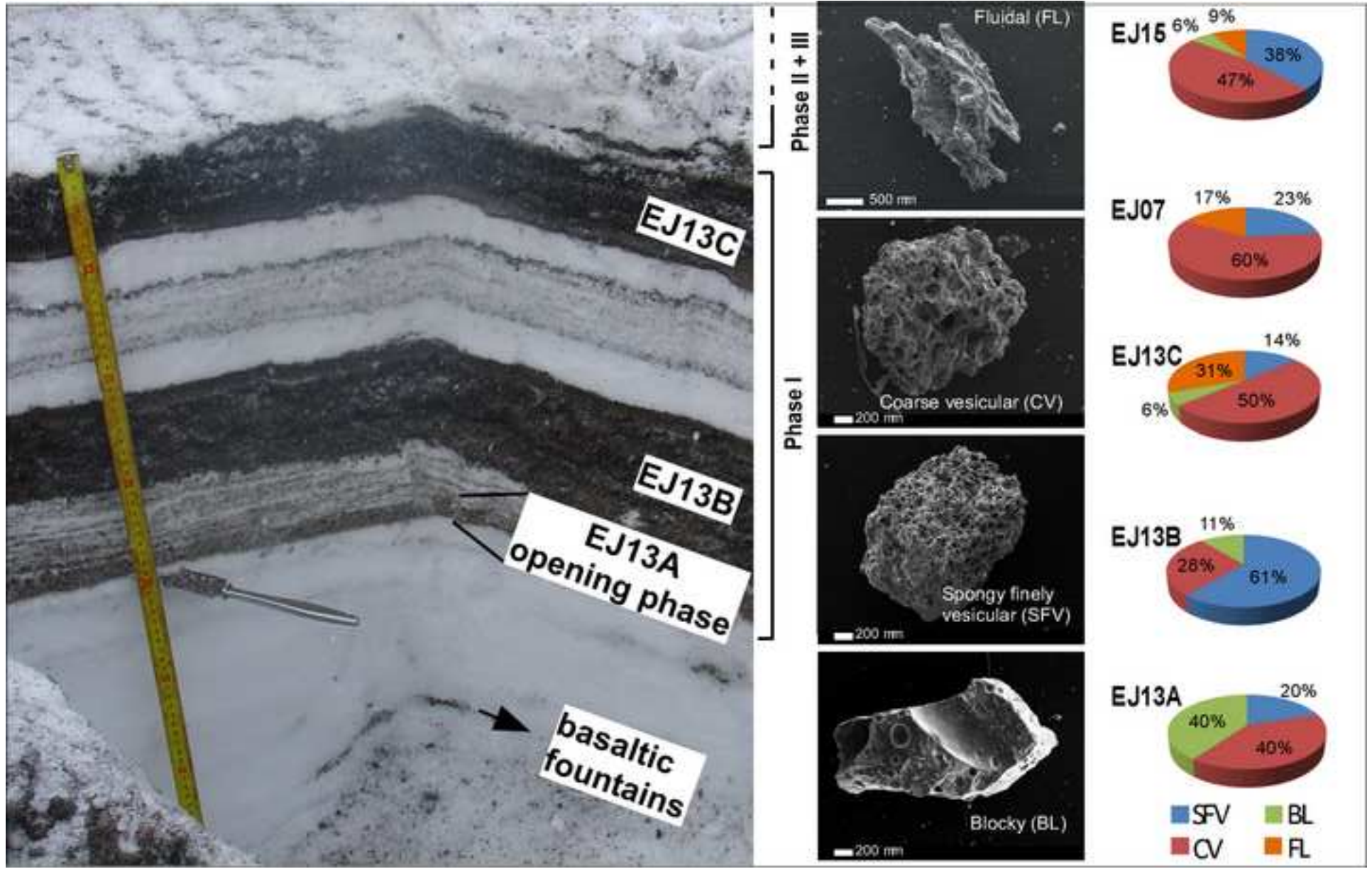


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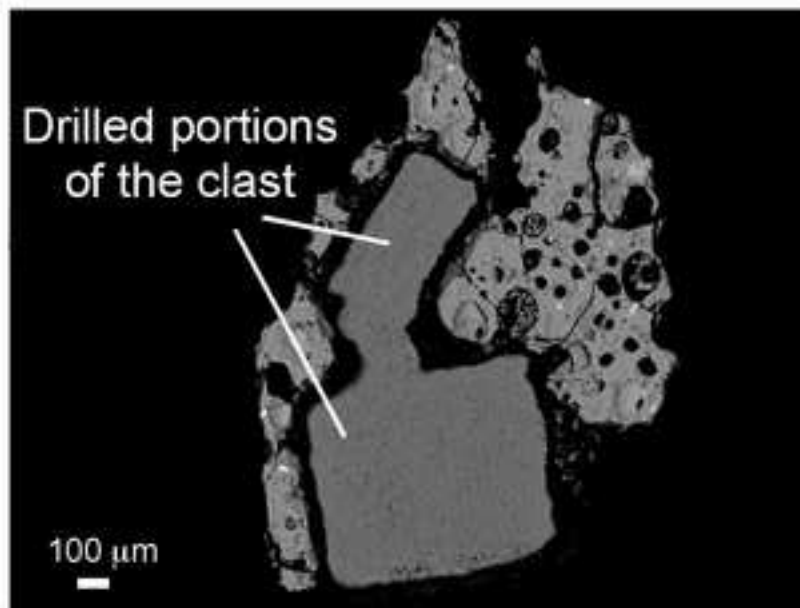
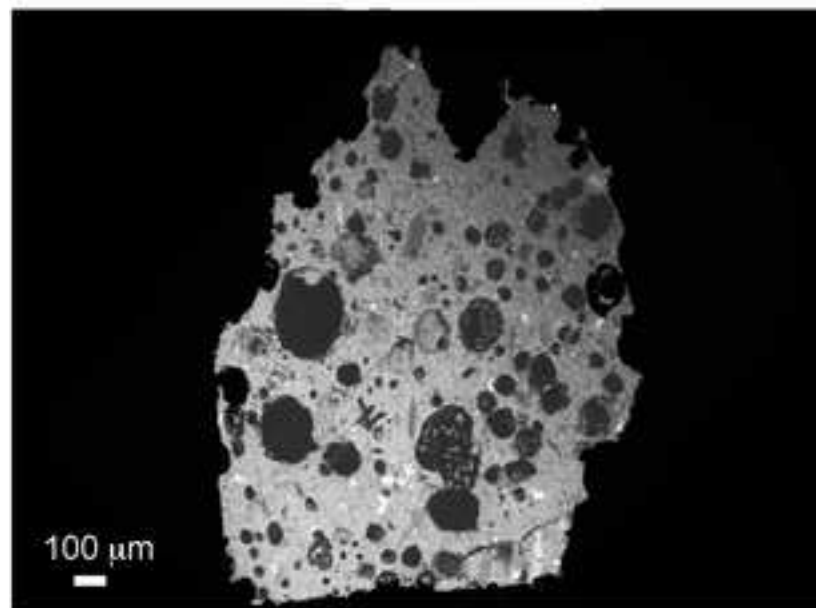
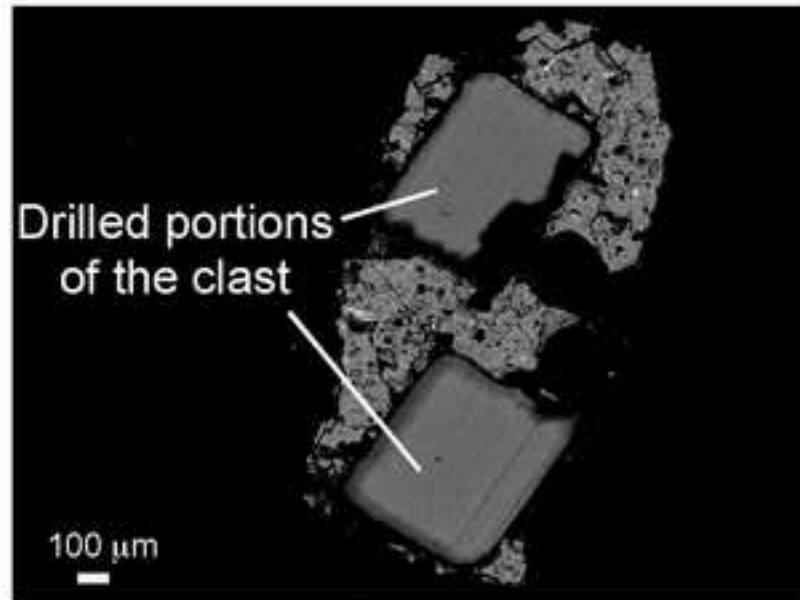
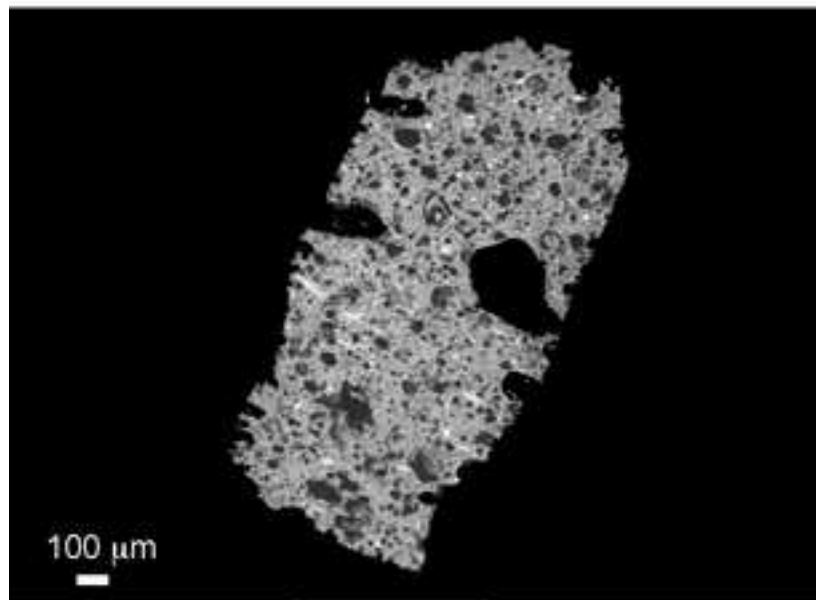


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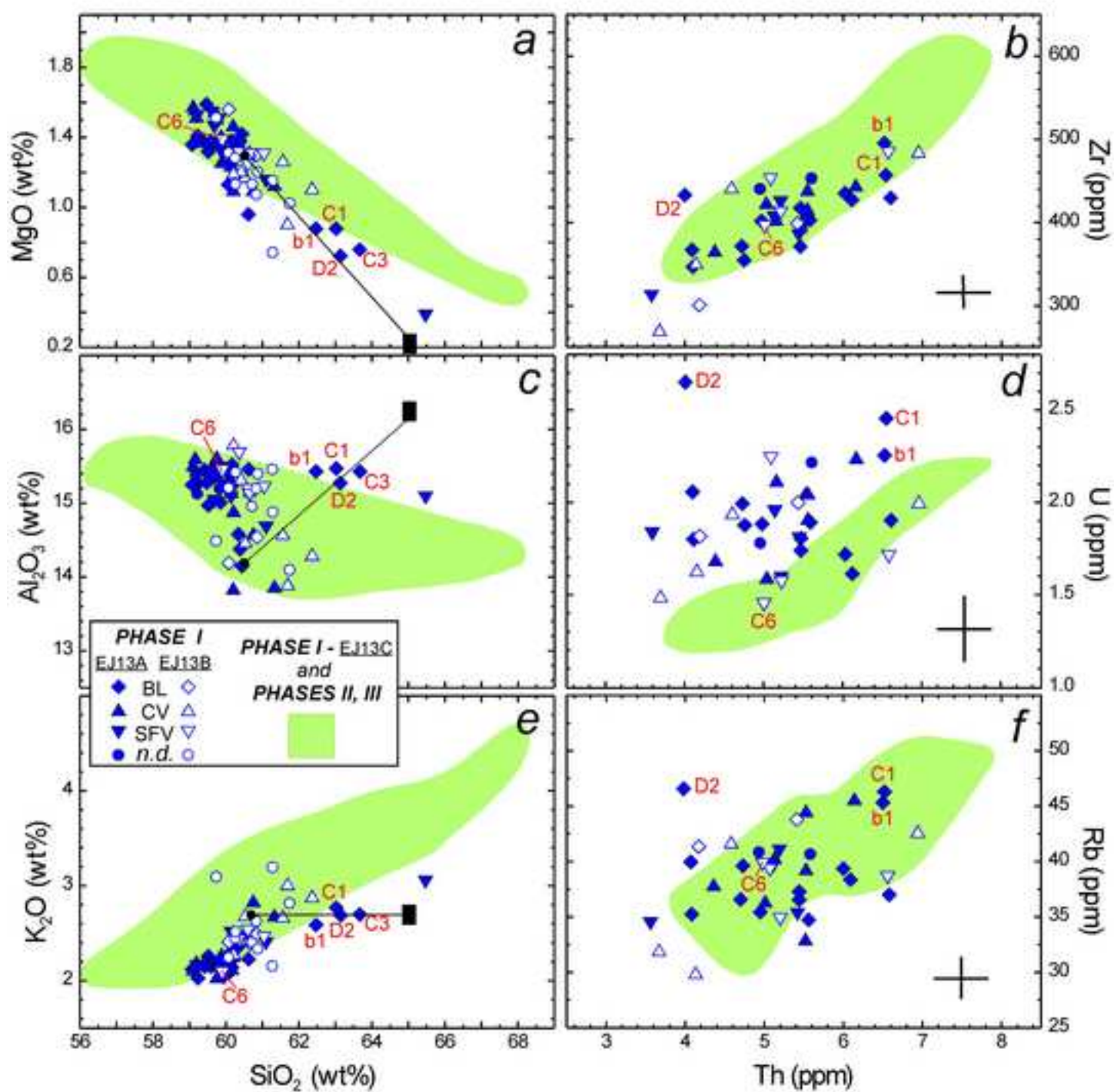


Figure 5

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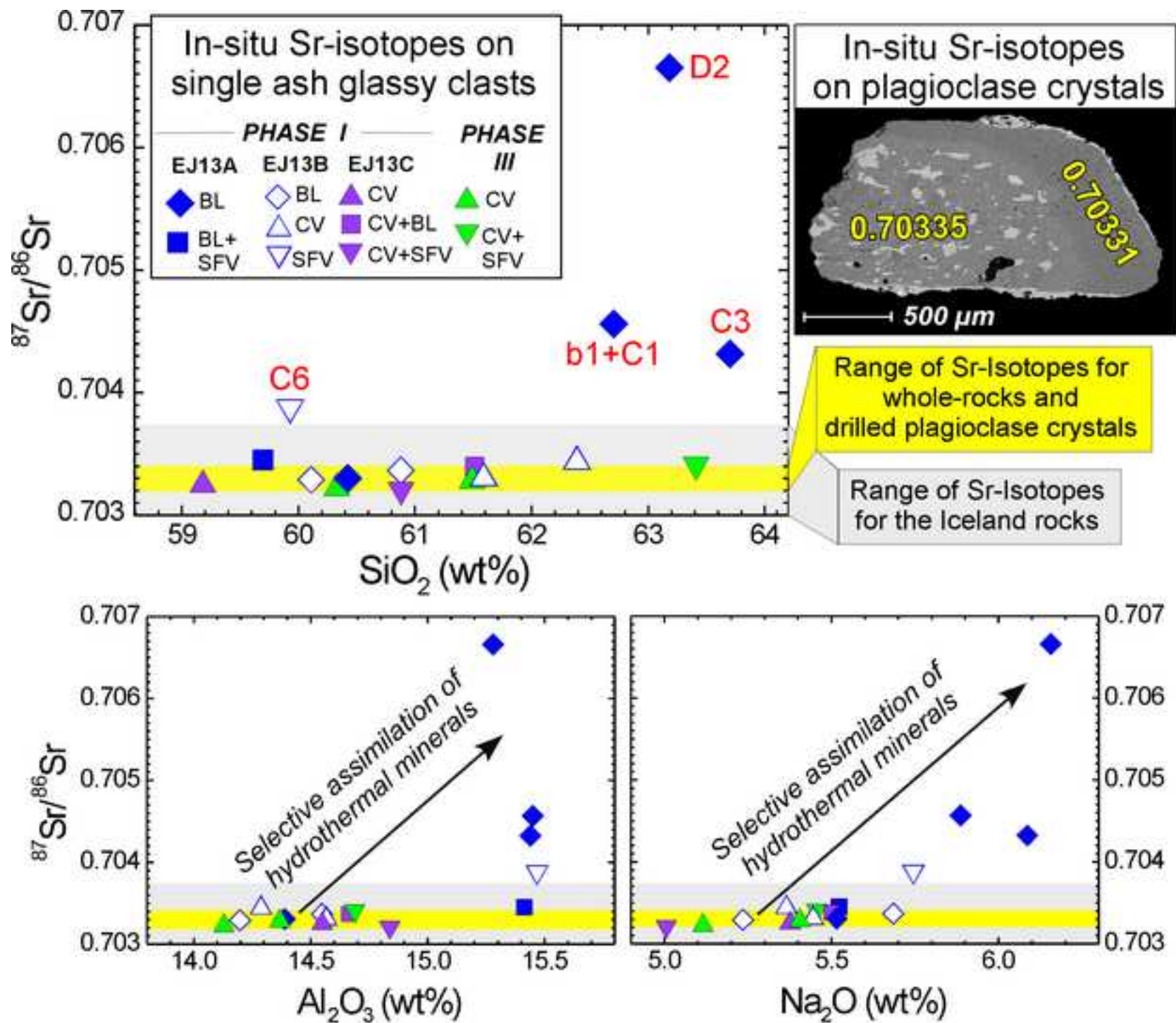


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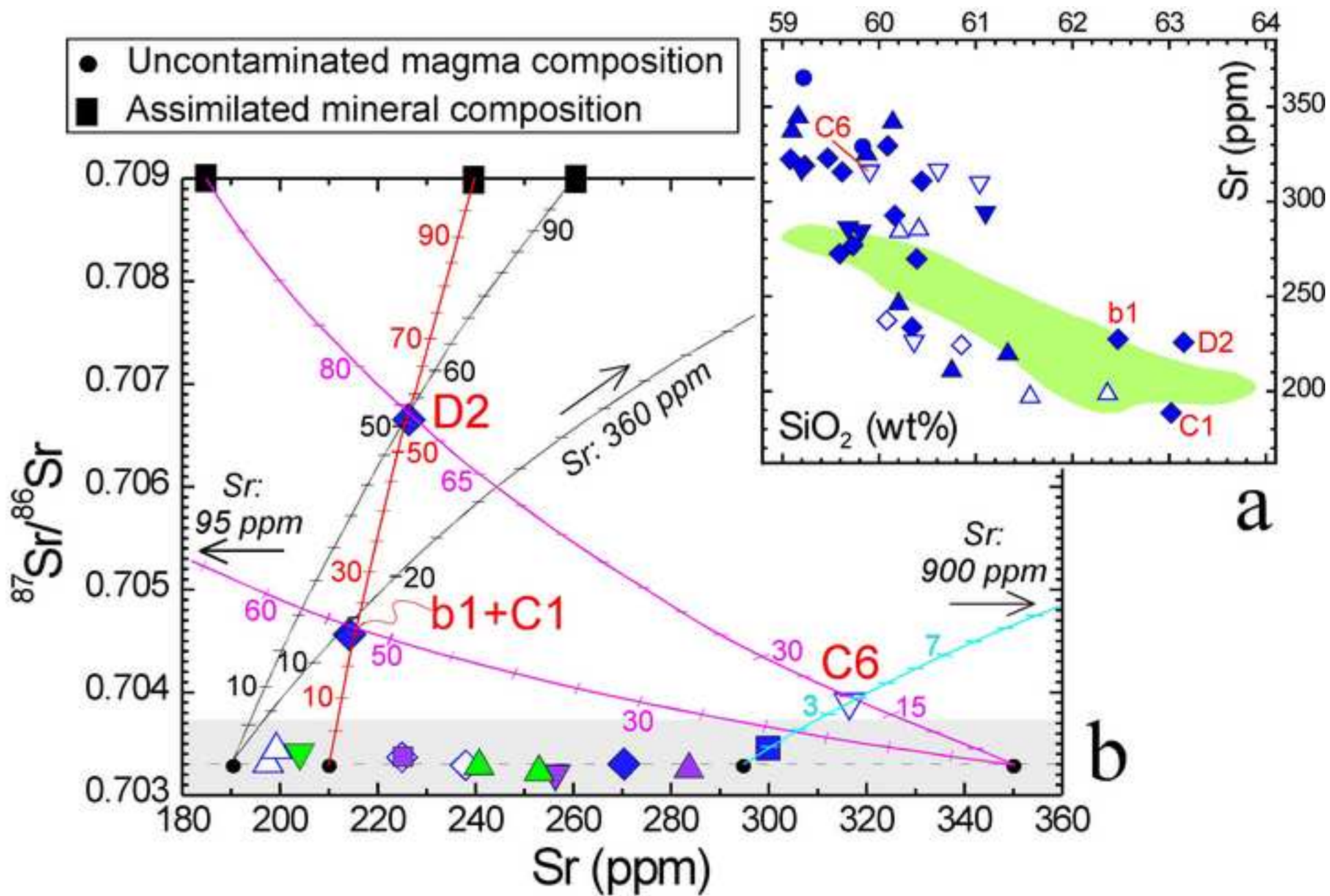


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