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## Constraining pre-eruptive magma conditions and unrest timescales during the Monte Nuovo eruption (Campi Flegrei, Southern Italy): integrating textural and CSD results for experimental and natural trachy-phonolites

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<b>Abstract:</b>	<p>We present crystallization experiments concerning a broad range of growth conditions of alkali feldspar and sodalite in trachy-phonolite magma composition during their later evolution state. Our results include: i) textural data and mineral assemblage of synthetic samples, ii) feldspar kinetics and growth rate estimates, and iii) textural data, mineral abundances and Crystal Size Distribution trend of samples representative of the Monte Nuovo eruption (1538 AD, last in the Campi Flegrei, Southern Italy). Experiments reproduced the texture and feldspar content of natural products indicating that the kinetics data can provide insights into processes within the volcanic system shortly before and during this small-magnitude eruption, and, particularly, about magma ascent timescale. Based on the bulk of the results we suggest that the groundmass crystallization of Monte Nuovo magma started between 7 and 4 km of depth (<math>\leq 200</math> MPa) at a temperature between 825 and 840 °C (close to the liquidus of alkali feldspar). The crystallization kinetics of alkali feldspar and the absence of sodalite in most of the natural samples indicate that the magma ascent was faster between 2 and 3 km (50-70 MPa) before to reach the fragmentation zone, during the first phases of the eruption. The crystallization time of the magma allows us to definitively constrain that the magma reached the fragmentation level in few days to several hours. Here we also show that a small decrease in pressure could induce a dramatic increase of crystallinity with associated rheological changes able to modify the eruption style. Thus, the products from the later phases of the Monte Nuovo eruption are more crystalline and contain sodalite in response to the reaching a shallower fragmentation depth.</p>
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3 **experimental and natural trachy-phonolites**

4

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27        **Abstract**

28        We present crystallization experiments concerning a broad range of growth conditions of alkali  
29        feldspar and sodalite in trachy-phonolite magma composition during their later evolution state. Our  
30        results include: i) textural data and mineral assemblage of synthetic samples, ii) feldspar kinetics  
31        and growth rate estimates, and iii) textural data, mineral abundances and Crystal Size Distribution  
32        trend of samples representative of the Monte Nuovo eruption (1538 AD, last in the Campi Flegrei,  
33        Southern Italy). Experiments reproduced the texture and feldspar content of natural products  
34        indicating that the kinetics data can provide insights into processes within the volcanic system  
35        shortly before and during this small-magnitude eruption, and, particularly, about magma ascent  
36        timescale. Based on the bulk of the results we suggest that the groundmass crystallization of Monte  
37        Nuovo magma started between 7 and 4 km of depth ( $\leq 200$  MPa) at a temperature between 825 and  
38        840 °C (close to the liquidus of alkali feldspar). The crystallization kinetics of alkali feldspar and  
39        the absence of sodalite in most of the natural samples indicate that the magma ascent was faster  
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45        the Monte Nuovo eruption are more crystalline and contain sodalite in response to the reaching a  
46        shallower fragmentation depth.

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48        **Keywords:** alkali feldspars; trachytic melts; crystallization kinetics; CSD; Monte Nuovo; Campi  
49        Flegrei

50 1. Introduction

51 1.1 The aim of the study

52 Alkali feldspar is an abundant crystal phase in evolved alkaline rocks (phonolites, alkali  
53 rhyolites), and is widespread in the Campania Province (Piochi et al. 2005). Its occurrence as  
54 microlites in the pumice and scoria groundmass potentially provides information on the timescale of  
55 magma migration within the crust, specifically from the magma chamber to the fragmentation level  
56 (Marsh 1988; Cashman and Marsh 1988; Marsh 1998). This information may be unraveled by  
57 studying the size distribution of microlites pre- and syn-eruptively crystallized and quenched in the  
58 matrix of juvenile fragments. Since the crystals present in an igneous rock and the observed  
59 variations in both their composition and texture reflect the integrated pressure (P) - temperature (T)  
60 - composition (X) - time (t) history of the magma from which they formed, it is possible to link  
61 textural observations with experimentally derived data for rates of crystal growth ( $Y_L$ ) and crystal  
62 number density ( $N_a$ ) for the specific mineral phase and the undercooling ( $\Delta T$ ) values of the parental  
63 melt. In fact, this allows us to gain insights into magmatic processes and their time-scales using  
64 textural observation.

65 However, there are few studies about crystallization kinetics on trachy-phonolitic melts (e.g.,  
66 Iezzi et al. 2008; Arzilli and Carroll 2013). In this study we present results for 16 new  
67 crystallization experiments on hydrous trachytic melt, which complement the previous experimental  
68 work of Arzilli and Carroll (2013). All of the experiments use the same single-step (cooling and  
69 decompression) method to reproduce the trachytic melt evolution in response to instantaneously  
70 applied thermodynamic driving force (i.e., undercooling,  $\Delta T = T_{\text{liquidus}} - T_{\text{experimental}}$ ).

71 The experiments were conducted at different pressures and  $\Delta T$ , in order to highlight the differences  
72 in crystallinity between high and low pressure (and thus melt H<sub>2</sub>O content). The preliminary study  
73 of Arzilli and Carroll (2013) focused mainly on the influence of  $\Delta T$  and time on the nucleation and  
74 growth of alkali feldspar. They showed the occurrence of several nucleation events of alkali  
75 feldspar in short times (hours), with dominance of nucleation at large  $\Delta T$ . Here we utilize our

76 experiments and those performed by Arzilli and Carroll (2013) to study how the crystallization may  
77 change as a function of  $P_{H_2O}$  (proportional to melt water content for the water-saturated conditions  
78 investigated) and  $\Delta T$  (induced by cooling and/or decompression). Our study is focused in  
79 understanding how fast the crystallinity of a trachytic magma could change after small variation of  
80 pressure. These experiments provide constraints on the P-T conditions of the trachytic to phonolitic  
81 melts during their ascent to the surface. Therefore, differently from Arzilli and Carroll (2013), here  
82 we use the experiments to better understand the volcanological and magmatic processes during the  
83 Monte Nuovo eruption in the Campi Flegrei (Southern Italy; Fig. 1). This eruption is particularly  
84 interesting because it occurred after 3000 years of volcanic quiescence and following a period of  
85 ground level movements and seismicity with characteristics comparable to the recent bradyseisms  
86 in the Campi Flegrei area (Parascandola 1947; Del Gaudio et al. 2010), as described by historical  
87 chronicles (Guidoboni and Ciucciarelli 2011). Constraining the timing of magma movements in the  
88 subsurface shortly before the eruption is crucial to unravel the significance of phenomena affecting  
89 the Campi Flegrei during volcanic quiescence. The eruption was already the object of similar  
90 studies (D’Oriano et al. 2005; Piochi et al. 2005) that, nevertheless, were limited by the lack of  
91 crystallization kinetic data for trachytic-phonolitic melt compositions (D’Oriano et al. 2005; Piochi  
92 et al. 2005). In order to constrain the conditions and the timescales of magmatic processes, we focus  
93 on the crystallization kinetics of alkali feldspar in trachytic melts and new crystal size distribution  
94 (CSD) data for groundmass feldspars in the natural samples. This choice is in line with several  
95 studies (e.g. Geschwind and Rutherford 1995; Hammer and Rutherford 2002; Couch et al. 2003;  
96 Armienti et al. 2007; Martel 2012) that used crystallization kinetics and CSD of feldspar (mainly  
97 plagioclase) to unravel shallow dynamics in pre- and syn-eruptive conditions at several calc-  
98 alkaline volcanoes; our new data allow similar investigation using the abundant alkali feldspar  
99 crystals in the Campi Flegrei system (e.g., Piochi et al. 2005; 2008 and reference therein).

100

101 1.2 Volcanological and petrological background

102 The 1538 Monte Nuovo eruption (Fig. 1a-e) was characterized by relatively low intensity and  
103 magnitude (Di Vito et al. 1987; D’Oriano et al. 2005; Piochi et al. 2005, 2008). Similar to the  
104 majority of volcanic eruptions of the last 14.9 ka b.p. in the area (see Rosi et al. 1983; Di Vito et al.  
105 1999; Isaia et al. 2009), it occurred from a monogenic vent and extruded  $<0.1 \text{ km}^3$  of magma (Lirer  
106 et al. 1987; Rosi and Sbrana 1987; Mormone et al. 2011; Smith et al. 2011) with a K-rich, phono-  
107 trachytic composition (Fig. 2) that generated sparsely porphyritic magmas, with typically alkaline  
108 feldspars, and variably vesicular pumice and scoria fragments (e.g., Rosi and Sbrana 1987; Piochi et  
109 al. 2008; D’Oriano et al. 2005). However, the Monte Nuovo eruption seems to be an exceptional  
110 event in the Campi Flegrei history, given that in the past 14.9 ka Campi Flegrei volcanism mostly  
111 concentrated in epochs of intense activity, alternating with quiescence periods of thousands of  
112 years; in such epochs eruptions occurred with a decadal frequency (Di Renzo et al. 2011; Smith et  
113 al. 2011). Instead, the Monte Nuovo event was the last and occurred after a volcanic quietness of  
114  $\sim 3000$  years.

115 The eruption occurred in an area characterized by persistent fumarolic activity and by  
116 bradyseisms since AD 1450 (Parascandola 1947; Lirer et al. 1987; Guidoboni and Ciucciarelli  
117 2011) (Fig. 1d). Uplift episodes (bradyseisms) occurred in 1503 and 1511 AD (Fig. 1d) inducing  
118 the local Authorities to work on the reconstruction of earthquake damaged buildings and to  
119 establish the ownership of the new land created. However, strong and continuous seismicity was  
120 described since the middle of AD 1536 with a significant progressive intensification about 3 months  
121 before the eruption (see table 2 in Guidoboni and Ciucciarelli 2011; see also Fig. 1d,e). In the  
122 middle of September, 1536, the ground deformations focused in a small zone where a vent opened  
123 along the coastal sector of the Averno Lake (Fig. 1a). Up to 4.5 meters of uplift occurred 1 day  
124 before the eruption, with the sea recession of about 400 m. Fractures at the surface with hot  
125 groundwater outflow, flames, and sudden uplift of a sunken area preceded the explosion of smoke  
126 and incandescent material that destroyed Le Tripergole Village. The eruption started in the night  
127 between September 29<sup>th</sup> and 30<sup>th</sup> (Guidoboni and Ciucciarelli 2011) and the first phreato-magmatic

128 phase produced pumiceous lapilli-bearing ash deposits that built the cone, whereas the final activity  
129 generated scoria deposits. The first main deposit (LM in D’Oriano et al. 2005, and Unit I-II in Di  
130 Vito et al. 1987) is composed of coarse to fine ash levels, being coarser-sized and lapilli-bearing in  
131 the lowermost levels (Unit I) where they are intercalated with pumice or scoria lenses or beds  
132 (Boivin et al. 1982). Block sags are common, while juvenile fragments are often sub-rounded,  
133 associated to infrequent obsidian-like clasts and lava and tuff lithics. This deposit was emplaced by  
134 pyroclastic flow mechanism. On October 3, after two days of diminishing violence, explosions  
135 produced two dark scoriae layers intercalated with a gray ash bed (UM1 or Unit III, in D’Oriano et  
136 al. 2005 and Di Vito et al. 1987, respectively). The final phase, after 3 days of eruptive quiescence,  
137 produced a scoria unit by flow (UM2 in D’Oriano et al. 2005) or/and fallout (Unit IV in Di Vito et  
138 al. 1987) mechanism.

139

## 140 2. Methods: synthetic and natural products

### 141 2.1 Synthetic products: starting material and experimental strategy

142 We complete the experiments of Arzilli and Carroll (2013) to obtain a wider data set of feldspar  
143 crystallization conditions involving growth under cooling, decompression and  
144 “cooling+decompression” experiments. The starting material (Table 1) is the same obsidian used in  
145 previous runs; it was sampled within the Breccia Museo Unit, a pyroclastic breccia associated with  
146 the Campanian Ignimbrite eruption dated at c.a 39 ka (Melluso et al. 1995; Fedele et al. 2008). The  
147 sample is known as ZAC in the literature (Di Matteo et al. 2004; Fabrizio and Carroll 2008;  
148 Calzolaio et al. 2010; Arzilli and Carroll 2013). It is sub-aphiric with <10% by volume of crystals.  
149 Phenocrysts are mostly alkali feldspar with minor plagioclase, clinopyroxene, biotite and magnetite,  
150 in order of decreasing abundance; apatite and titanite are accessory phases. The composition is  
151 trachytic, near the trachyte-phonolite border in the TAS diagram (Di Matteo et al. 2004). It is  
152 similar but not identical to the Monte Nuovo products in terms of whole-rock and glassy matrix  
153 chemistry (Fig. 2; Table 1); also it presents slight differences for content of phenocrysts (<10%



154 ZAC vs. 3% Monte Nuovo) and mineral assemblage (much abundance of pyroxene and biotite in  
155 ZAC). Despite these small differences, we use ZAC for which the phase diagram (Fabbrizio and  
156 Carroll 2008; Arzilli and Carroll 2013) allowed us to choose accurately the experimental conditions  
157 relevant for our study the feldspar crystallization kinetic. Also, Piochi et al. (2008) proposed similar  
158 composition for the magma systems feeding eruptions at the Campi Flegrei.

159 To complete the experimental data set of Arzilli and Carroll (2013), we have performed 16 new  
160 crystallization experiments. Particularly, we extend cooling and degassing experiments in water-  
161 saturated conditions towards higher pressure (experimental final pressure,  $P_f = 100, 150$  and  $200$   
162 MPa) and lower pressure ( $P_f = 30$  and  $50$  MPa) within a range of temperatures between  $750$  and  $850$   
163 °C (Table 2). The initial conditions of the experiments were above the liquidus of the alkali  
164 feldspar. The experiments were carried out using Cold Seal Pressure Vessels (CSPV) at the  
165 Geology Division (University of Camerino); details on CSPV facilities, capsule preparation and  
166 experimental methods are described in Arzilli and Carroll (2013).

167

## 168 2.2 Natural products

169 Analyzed pumice and scoria fragments are from the Lower and Upper Members of the Monte  
170 Nuovo formation (Fig. 1a, b, c), respectively. Fragments and eruption levels were selected on the  
171 basis of previous textural and chemical characterization (D’Oriano et al. 2005; Piochi et al. 2005;  
172 Piochi et al. 2008) with the aim to analyze in major detail the textural and petrological variability in  
173 products representative of the eruption phase, including new X-ray diffraction data.

174 Pumices are mostly angular clasts, yellowish in colour and with sub-spherical vesicles, from few  
175 to several centimetre in dimension. Other pumices are characterized by alternating yellow and  
176 brown portions and are referred to as banded pumices. Scoriae are mostly blackish clasts, generally  
177 dm-sized, containing heterogeneous vesicles and often a more vesicular core and less vesicular  
178 margin, with elongated vesicles in some pumice fragments. The pumice and scoria fragments  
179 contain <3% of phenocrysts including mostly feldspars, subordinate magnetite and rare spinel. The

180 vesicularity and groundmass crystallization degree is variable (Fig. 3) (see also, D’Oriano et al.  
181 2005; Piochi et al. 2005; Piochi et al. 2008). Microcrystallinity is determined by alkali-feldspars  
182 (anorthoclase and sanidine). It varies with vesicularity: microlites are less abundant and smaller in  
183 size in highly vesicular pumice compared with the scoria clasts. Overall, the products are denser  
184 and more crystalline upwards in the sequence, as observed by D’Oriano et al. (2005) and Piochi et  
185 al. (2005). Bands in pumice fragments are strongly parallel to each other and are associated with  
186 variations in groundmass texture: brown bands being more crystalline and less vesicular than  
187 yellowish bands (Piochi et al. 2008). The microlites have a general acicular shape with larger sizes  
188 compared with those in scoriae; some of them are curved, following the shape of adjacent vesicle,  
189 suggesting that they grew concomitantly with vesicle (Piochi et al. 2005; Piochi et al. 2008). In  
190 pumices, microlites are nearly-to-strongly aligned, and microlites and color bands share the same  
191 alignment.

192 Early fragmentation caused by magma-water interaction during magma ascent produced the  
193 pumices at the base of sequence (samples LM inf d and LM inf l; Fig. 1a). This is texturally (Fig.  
194 3) testified by i) the vesicle walls that are broken and show shocking cracks, ii) their vesicularity up  
195 to 80% higher than in other pumices and scoriae; and iii) the lower groundmass crystallinity. All  
196 these associated to higher water content with respect to products higher in the sequence (Piochi et  
197 al., 2008). Textural variation of brown pumices (LMC3; Fig. 1c) and scoriae (MN4top; Fig. 1b and  
198 Fig. 3b) mostly resulted from the variable magma outgassing occurring in relation to the magma  
199 ascent dynamics (Piochi et al. 2008) during the proceeding of the eruption (D’Oriano et al. 2005;  
200 Piochi et al. 2005).

201

### 202 3. Analytical methods

#### 203 3.1 SEM and image analysis

204 Electron microscope images were taken for both synthetic and natural samples in order to obtain  
205 more details on the textural features and mineralogical assemblage (Figs 3, 4, 5). Back scattered

206 electrons (BSE) images were collected for experimental samples using JEOL JSM-6390LA FE-  
207 SEM (at the Institute of Geochemistry and Petrology, ETH Zurich) and JEOL JSM-6500F -  
208 upgraded to 7000 series – FEG at the INGV of Rome. More details about image analysis procedure  
209 and the calculation method of crystallization kinetics are provided by Arzilli and Carroll (2013).  
210 BSE images of pumices and scoriae of Monte Nuovo were acquired through a LEO 430 SEM at  
211 “Istituto Geomare Sud – CNR” of Napoli, Italy and a ZEISS SUPRA 35 at the “Dipartimento di  
212 Ingegneria dell’Informazione” of the II Università di Napoli in Aversa, Italy, operating at 10 keV,  
213 and a work distance of 15 mm and 7–12 mm, respectively.

214

### 215 3.2 CSD analysis

216 The fragments from the selected natural samples were studied through CSD in order to estimate  
217 the magma residence time, knowing the relation between feldspar population density, the sizes and  
218 the specific growth rates of alkali feldspar. CSD studies provide quantitative information on relation  
219 between crystal population density and crystal length for a population of crystals. The linear  
220 relation provides estimates of time scales of magmatic processes (Marsh, 1988; Cashman and  
221 Marsh, 1988). The slope of the correlation is equal to  $-1/(\text{growth rate} * \text{residence time})$  and the  
222 intercept is equal to the nucleation density.

223 Crystal dimensions and abundances of each size population were recovered through accurate  
224 image analyses by using BSE images and Adobe Photoshop ® software package. The crystals were  
225 manually segmented because the contrast between trachytic glass and feldspar microlites is weak.  
226 We were able to define the contour of each single crystal also in agglomerate structures. However,  
227 for highly crystalline samples, the geometry of each crystal was affected by some interpretation.  
228 Several elaborations were made to define different crystal geometry in agglomerates but they  
229 provided similar results. The reconstructed image was processed through ImageJ software (NIH  
230 Inage; Abramoff et al. 2004; Schneider et al. 2012) package to quantify data on crystallinity  
231 percentage and sizes. The relation between crystal population density and crystal length for a

232 population of crystals were finally obtained employing the CSD Corrections 1.3 program (Higgins  
233 2000; Higgins 2002).

234

### 235 3.3 X-ray diffraction analysis

236 Selected whole-rocks and related hand-picked shards from the same sampled units used for CSD  
237 were examined by powder XRD. Samples were powered in an agate mortar and analyzed by X-ray  
238 diffraction method with the aim to further characterize mineralogy and semi-quantitative phases  
239 estimation, on larger amount of sample than that studied on thin section.

240 X-ray powder diffraction (XRPD) patterns were obtained by using a Panalytical X'Pert Pro  
241 diffractometer of the Istituto Nazionale di Geofisica e Vulcanologia, sezione Osservatorio  
242 Vesuviano (Naples, Italy), as illustrated in Mormone et al. (2014). Operating conditions were:  
243 CuK $\alpha$  radiation, 40 kV, 40 mA, 2 $\theta$  range from 3 to 100 $^\circ$ , equivalent step size 0.0179 $^\circ$  2 $\theta$ , equivalent  
244 counting time 298.09 s per step. The X'Pert High Score Plus 2.2 d software allows qualitative and  
245 semi-quantitative data of the analysed natural powders. Semi-quantitative XRD analysis was carried  
246 out on 7 samples by using the X'Pert High Score Plus 3.0e d package interface as detailed in the  
247 Supplementary Material 1.

248

## 249 4. Results

### 250 4.1 Experimental samples: crystallization kinetics and textural features

251 The experiments performed in this study are characterized by several phases: alkali feldspar,  
252 clinopyroxene, Fe-Ti oxide, biotite, sodalite, glass and vesicles (Table 3). The sodalite (Table 3) is  
253 always generated at pressure  $\leq 50$  MPa, and it occurs at high crystal fraction of alkali feldspar ( $\phi$ ).

254 The experiments were texturally analyzed in order to study the crystallization kinetics of the  
255 alkali feldspar, quantifying the number density ( $N_a$ ), crystallinity ( $\phi$ ), crystal size ( $L$ ) and growth  
256 rates ( $Y_L$ ) of alkali feldspar (Table 3). The experiments between 50 and 200 MPa show that the  
257 number density of alkali feldspar ranges from  $10^4$  and  $10^6$  cm $^{-3}$  (Table 3) and the order of

258 magnitude of  $N_a$  increase with decreasing pressure. The experiments are characterized by a wide  
259 range of crystallinities of alkali feldspar ( $\phi$ ) which vary between 0.01 and 0.95. The experiments at  
260 high pressure (100, 150 and 200 MPa) and short experimental duration (7200 – 28800 s)  $\phi$  change  
261 from 0.01 to 0.32 (Table 3). Whereas, for the same pressures but at long duration (57600 s)  $\phi$  is ~  
262 0.42. The experiments at 50 MPa and  $15 \leq \Delta T \leq 40$  °C,  $\phi$  ranges between 0.29 and 0.55 Table 3.  
263 Several samples (see D69, D78, D21 and D62 in the Table 3) show that low pressure (30-50 MPa),  
264 H<sub>2</sub>O content (1.7-2.7 wt % H<sub>2</sub>O) and  $40 \leq \Delta T \leq 115$  °C favor an extreme crystallization of alkali  
265 feldspar ( $0.72 \leq \phi \leq 0.95$ ). Therefore, strongly intergrown textures were favored between 30 and 50  
266 MPa, characterized by a large quantity of tiny crystals (tabular and acicular crystal intergrowth)  
267 with very low abundance of glass (Fig. 4a, b, c, i). Alkali feldspars have commonly tabular and  
268 elongated rectangular shapes (Figs 4a, b, c, i and 5b, c, d) at the different experimental conditions.  
269 Instead, spherulitic alkali feldspar crystals are more common at higher  $P_f$  ( $=P_{H_2O}$ ), and they are  
270 present in the same way both in cooling and decompression experiments. Furthermore, although the  
271 difficulty to obtain reliable chemical analysis, it was possible to establish that 1) alkali feldspars  
272 have less than 8 mol% of An and 39 to 71 mol% of Or and 2) glasses are trachytes.

273 The growth rates ( $Y_L$ ) of alkali feldspar range from  $10^{-8}$  to  $10^{-7}$  cm/s (Table 3), showing a  
274 sensible decreasing at long experimental duration (57600 s), probably related to a closer approach  
275 to equilibrium at longer times.

276

#### 277 4.2 Natural samples: textural, XRPD and CSD data

278 The dominant alkali-feldspar mineral phase is present in various percentages, with nearly glassy  
279 matrix (Fig. 3e) to very high abundance (Fig. 3b). X-ray diffraction patterns (see Supplementary  
280 Material 1) allow evaluating the relative proportion of anorthoclase and sanidine and the accessory  
281 minerals present in the various samples (Table 4) providing information additional to the that  
282 derived by the optical and electron microscope investigations (Fig. 3). The anorthoclase abundance  
283 is higher in the highly crystalline brown pumices (71-83%) and scoriae (55-60%). These products

284 also show small % of sodalite (~3 and ~6%, respectively). Biotite contributes to the XRPD patterns  
285 of several samples with relative abundance <0.2-0.6%. Note that the biotite is absent in the scoriae  
286 erupted at the end of the eruption. Furthermore, the XRPD semi-quantitative data on whole-rock  
287 and related separated shards of each sample indicate comparable mineral phase percentage.  
288 Therefore, the contribute of phenocrysts is not significant and the XRPD can be used to asses  
289 microlite abundance, as also it results by comparing semi-quantitative data by XRPD  
290 (Supplementary materials 1) and BSE images (Piochi et al., 2008).

291 Alkali feldspars are generally aligned in the bands and, sometimes, clustered in spherulitic clots.  
292 No significant contortion characterizes the bands. The size and abundance of alkali feldspar  
293 determined in natural samples allow us to obtain several measurements of CSDs (Fig. 6). In  
294 agreement with previous analysis (D’Oriano et al. 2005; Piochi et al. 2005; Mastrolorenzo and  
295 Pappalardo 2006), the size distributions of microlites appear to define two approximately linear  
296 trends, a steeper one for crystals smaller than about 0.6 mm, and another, more shallow slope for  
297 larger crystals; trends for different samples are nearly parallel. However, our CSDs are more  
298 accurate in comparison to those in the literature that show comparable intercept values, but lower  
299 crystal size as the consequence of smaller area investigated by previous authors (Piochi et al.,  
300 2005). The stratigraphically lower pumices show a simple CSD with a unique growth trend, while  
301 scoriae are characterized by two trends that show two timing of crystal growth (Fig. 6). Therefore,  
302 the smaller microlites define CSD curves with slopes that range from -22 to -8 and an intercept at  
303  $10^{-11} \text{ mm}^{-2}$ ; instead the curves for larger microlites have slopes between -2 and -7 and extrapolated  
304 intercepts at  $10^{-9}$ - $10^{-11} \text{ mm}^{-2}$  (Table 5). Lower slope values are generally determined in the lower  
305 vesicular brownish bands and black scoria (c2s 9-12 – 1 and MN4 top 06 – 1, respectively, in Table  
306 5); whereas higher slopes are from the highly vesicular yellow glass.

307

## 308 5. Discussion

### 309 5.1 The rapid change of crystallinity: the role of $P_{\text{H}_2\text{O}}$ and $\Delta T$

310 The cooling experiments performed by Arzilli and Carroll (2013) show that  $\phi$  varies between  
311 0.05 and 0.64, instead, here we show that  $\phi$  could be ranged from 0.01 to 0.93 a certain conditions.  
312 This can be also observed in decompression experiments:  $\phi$  shows similar variation, from 0.13 to  
313 0.76 for “decompression + cooling” experiments and from 0.09 and 0.77 for isothermal  
314 decompression (Arzilli and Carroll 2013). Here we show that in the latter kind of experiment  $\phi$   
315 reaches 0.95 at 30 MPa and long duration (14 h). Instead, the abundances of clinopyroxene and  
316 magnetite remain low (0.01 to 0.10) for this feldspar-dominated bulk composition (Arzilli and  
317 Carroll, 2013).

318 The abundance and morphologies of alkali feldspar are linked to  $P_f (= P_{H_2O})$ , water content,  $\Delta T$   
319 and experimental duration (as observed by Arzilli and Carroll, 2013). By combining our results with  
320 those of Arzilli and Carroll (2013) we show that cooling experiments at 50 and 70 MPa can produce  
321 higher  $\phi$  than those at 100, 150 and 200 MPa (Fig. 7a, b). The relation between  $N_a$  and  $\Delta T$  as a  
322 function of  $P_f$  for cooling experiments (Fig. 7c) show that at 70 and 100 MPa,  $N_a$  increases as  $\Delta T$   
323 increases, whereas for experiments at 50 MPa,  $N_a$  is almost constant with  $\Delta T$ . The main evidence is  
324 that at low  $\Delta T$ ,  $N_a$  increases as  $P_f$  decreases and the nucleation process is favored at low pressure,  
325 with lower melt water contents. The diagram in Fig. 7d shows also that  $N_a$  is almost constant with  
326 the time, confirming that  $P_f (= P_{H_2O})$  and  $\Delta T$  play a crucial role on the nucleation of alkali feldspar,  
327 producing either high or low crystallized rocks over the limited temperature range investigated.

328 The decompression experiments show the same relations observed for cooling experiments. The  
329 high crystallinity textures are produced at 30 and 50 MPa (Fig. 8a, b). The order of magnitude of  $N_a$   
330 for experiments at 30 and 50 MPa is up to three times higher than experiments at 70, 100 and 150  
331 MPa (Fig. 8c), increasing with decreasing water content. The exceptions are the samples at 50 MPa  
332 ( $\Delta T = 25 \text{ }^\circ\text{C}$ ) and that at 70 MPa ( $\Delta T = 14 \text{ }^\circ\text{C}$ ), which have a low value of  $N_a$  similar to the  
333 experiments at 150 MPa (Fig. 8c). Probably, small  $\Delta T$  could slow down the nucleation process,  
334 favoring the growth. As shown above from cooling experiments, the orders of magnitude of  $N_a$  is

335 almost constant over time, while low pressures facilitate greater nucleation of alkali feldspar (Fig.  
336 8c,d). The latter diagram highlights also that similar  $\Delta T$  ( $\sim 40$  °C) can produce different orders of  
337 magnitude of  $N_a$ , confirming that a strong nucleation of alkali feldspar is induced at low pressure.

338 Although we have studied the isobaric cooling and isothermal decompression processes, we have  
339 not distinguished the concept of undercooling ( $\Delta T$ ) and effective undercooling ( $\Delta T_{\text{eff}}$ ), the effective  
340 undercooling related to decompression process at constant temperature (Hammer and Rutherford,  
341 2002) because, as shown by Arzilli and Carroll (2013), under the same experimental conditions,  
342 both processes produce similar crystallization kinetics. Furthermore, we performed "cooling +  
343 decompression" experiments where it is difficult to separate  $\Delta T$  and  $\Delta T_{\text{eff}}$ . The comparison between  
344 cooling and decompression experiments shows that at similar conditions, in terms of  $P_f$ ,  $\Delta T$  and  
345 superheating, cooling and decompression experiments produce similar  $N_a$  and  $\phi$ . This may mean  
346 that the distinction between  $\Delta T$  (related to cooling process) and  $\Delta T_{\text{eff}}$  is useless when a single fast  
347 step of cooling and decompression occurs.

348 The nucleation of large amount of crystals (tabular and acicular crystal intergrowth) was favored  
349 at low pressure (Figs 4, 7 and 8) and not necessarily at low  $\Delta T$ , instead it was reduced at high  
350 pressure (Figs 5, 7 and 8), highlighting that a small decrease in pressure can induce a dramatic  
351 increase of crystallinity.

352

## 353 5.2 Experimental vs natural materials

354 The experiments produced material texturally comparable with natural products, although the  
355 pyroxene occurrence represents the most important discrepancy. This may depend on the starting  
356 experimental conditions as pyroxenes are already contained in ZAC, and the oxygen fugacity of the  
357 CSPV ( $\sim \text{NNO}+1$  log units) may also be different in comparison to that of the Monte Nuovo system.

358 The alkali feldspar morphologies of natural samples (Fig. 3) are similar to in the experimental  
359 ones (Figs 4 and 5). Experimental alkali feldspars are characterized by elongated rectangular  
360 crystals and they occur in clots (Fig. 4) or isolated in the glass (Fig. 5), as in the natural pumices.



361 Both types of decompression experiments at 50 MPa produced similar textures to natural scoriae,  
362 with high crystallinity and aligned tabular or elongated rectangular crystals (see Figs 3b and 5c),  
363 also including magnetite and sodalite crystals (Table 4). These highly crystalline charges also show  
364 vesicles with irregular margin similarly to the natural scoriae. Furthermore, the Or content of the  
365 synthetic alkali feldspars and those of natural ones are similar (D’Oriano et al. 2005; Piochi et al.  
366 2005; Piochi et al. 2008).

367 Diagrams in Fig. 9a and b allow quantitatively comparing experimental and natural crystalline  
368 textures. Fig. 9a shows a relation between  $N_a$  and  $\phi$  obtained through cooling experiments, where  $\phi$   
369 increases as growth dominates the crystallization process. Instead,  $N_a$  increases as nucleation  
370 dominates the process. In particular,  $N_a$  increases as  $P_f$  (therefore water content) decreases (Fig.  
371 9a), showing that  $P_{H_2O}$  and the water content have an important effect on textural evolution. Low  
372 experimental pressure and low water content seems to enhance nucleation density, thus producing  
373 more alkali feldspar nuclei. The diagram also suggests that natural alkali feldspars of the glassy  
374 pumices must crystallize at  $\geq 70$  MPa, based on comparison of relative nucleation densities.  
375 Sodalite absence and biotite occurrence in these samples support such barometric conditions. In  
376 agreement with Mastrolorenzo and Pappalardo (2006), glassy pumices are characterized by low  $N_a$   
377 of alkali feldspar. Moreover,  $\phi$  of natural alkali feldspars is between 0.01 and 0.30, suggesting a  
378 crystallization event at low  $\Delta T$ , therefore crystallization temperature could be close to the liquidus  
379 of alkali feldspar. Instead, data from more crystal-rich pumice and scoria resemble most closely  
380 experimental samples produced at 70 MPa pressure, under less than 69 °C and 39 °C of  $\Delta T$  (Arzilli  
381 and Carroll, 2013), respectively. Nevertheless, these experiments do not reproduce sodalite.  
382 Notably, the natural sample with  $\phi \sim 0.95$  (Fig. 9a; D’Oriano et al. 2005) could be reproduced at  
383 high  $\Delta T$  ( $>60$  °C), in agreement with the experiments performed in this study (D69, D78 and D62).  
384 The holocrystalline texture probably requires low pressures (between 50 and 30 MPa) and more  
385 time for crystal growth, supporting a possible origin involving magma stagnation in the conduit (in  
386 agreement with D’Oriano et al. 2005). Hence, the sodalite occurrence should be due to the high

387 crystallinity and the enrichment of Cl in the residual melt (Carroll 2005).

388 The results of “decompression + cooling” and isothermal decompression experiments are shown  
389 separately in Table 3, but for the discussion they are included together to have a wider range of  
390 potential pre- and syn-eruptive conditions. Decompression experiments are characterized by two  
391 populations of samples which have differences in nucleation density (Fig. 9b). The first population  
392 at low pressure ( $30 \text{ MPa} < P_f < 50 \text{ MPa}$ ) has values of  $N_a$  between  $10^6$  and  $10^7 \text{ cm}^{-2}$ , exceeding  
393 values observed in the natural samples. In contrast, the second group of higher P experiments ( $100$   
394  $\text{MPa} < P_f < 150 \text{ MPa}$ ), has values of  $N_a$  between  $10^4$  and  $10^5 \text{ cm}^{-2}$ , and closely resemble  
395 observations on natural scoria and pumices. The experiments at 50 MPa (charge D81), 70 MPa  
396 (charge D82), 100 MPa (charge D80) and 150 MPa (charge D55) are characterized by  $N_a = \sim 10^5$   
397  $\text{cm}^{-2}$  and  $\phi$  between  $\sim 0.10$  and  $\sim 0.30$  (see Table 3 and the results of Arzilli and Carroll, 2013),  
398 therefore comparable with poorly crystallized natural pumice. As a whole, the  $\phi$  of alkali feldspar =  
399  $0.01$ - $0.30$  in the pumice fragments implies  $\Delta T < 50 \text{ }^\circ\text{C}$ , the increase up to  $0.40$ - $0.60$  in the scoriae, as  
400 well as the highest one at  $0.95$  (D’Oriano et al. 2005), requires higher  $\Delta T$  and low pressure. In  
401 particular, scoriae trend toward experiments at  $\leq 70 \text{ MPa}$  in terms of  $N_a$  and  $\phi$ , as well as containing  
402 traces of sodalite.

403 Experimental results suggest that crystallization of natural alkali feldspar occurred: i) between  
404  $200$  and  $70 \text{ MPa}$  and, mostly  $150$ - $100 \text{ MPa}$  (low  $\Delta T$ ), at temperature between  $750 \text{ }^\circ\text{C}$  and  $825 \text{ }^\circ\text{C}$ , in  
405 water saturated cooling conditions or/and ii) at  $> 50$ , and mostly  $150 \text{ MPa}$ , at temperature between  
406  $750$  and  $840 \text{ }^\circ\text{C}$ , if melt decompressed. Furthermore, the absence or presence of sodalite crystals  
407 indicate that natural glassy samples register  $P \geq 50 \text{ MPa}$ , whereas lower pressure corresponds to the  
408 more crystalline brown pumices and scoriae. Thus, we constrain: the crystallization in the brown  
409 pumice and scoriae occurred over a longer time compared with pumice sample crystallization.  
410 Furthermore, for water saturated conditions and in the  $200$ - $50 \text{ MPa}$  pressure range, temperature  
411  $\geq 850 \text{ }^\circ\text{C}$  means that conditions are always above the alkali feldspar liquidus. Therefore, natural

412 alkali feldspar crystallization between 200 and 150 MPa needs temperature of 750-775 °C, or 800-  
413 825 °C at 100 to 70 MPa, or 840-860 °C at 50 MPa.

414

### 415 5.3 Magma residence time deduced from growth rates and CSDs

416 Measurements of Crystal Size Distribution (CSD), when combined with experimental growth  
417 rate data for alkali feldspar ( $Y_L$ ), can help us to better constrain the time of magma crystallization  
418 just before fragmentation (e.g., Marsh 1988; Cashman and Marsh 1988; Higgins 1996; Armienti et  
419 al. 2007). Following Piochi et al. (2005) and D'Oriano et al. (2005), these times may correspond to  
420 the time of magma crystallization in the chamber (cooling-dominated conditions) or to magma  
421 movements in the volcanic conduit (decompression-dominated conditions).

422 We use new data on both CSDs (Fig. 6) and feldspar growth rate in recalculating crystallization  
423 times ( $t_r$ ) for the Monte Nuovo magma. The estimated growth rate ( $Y_L$ ) mostly varies between  
424  $1.1 \cdot 10^{-8}$  and  $7.2 \cdot 10^{-7}$  cm/s (considering  $Y_L$  obtained in this study and from Arzilli and Carroll,  
425 2013), with an isolated higher value at  $1.2 \cdot 10^{-6}$  cm/s and the mean  $Y_L$  equal to  $2.21 \cdot 10^{-7}$  cm/s  
426 (Table 5). Because the crystal growth rate varies by up to two orders of magnitude, significant  
427 variations of the magma timing must be expected. Therefore, the crystallization time is from few  
428 months to few hours. However, the mean  $Y_L$  suggests that magma was stored for a few days in the  
429 subsurface and the ascent lasted a few hours. Notably, the lower CSD slopes, i.e., the higher  
430 residence times, characterize the most crystalline matrices. This is in agreement with the feldspar  
431 chemistry detected by XRPD and reproduced in longer experiments (Supplementary Material 1).

432

### 433 5.4 Implication for magma dynamics

434 Experimental results suggest that the crystallization of alkali feldspars in magma erupted during  
435 the Monte Nuovo eruption started between <200 and 100 MPa when temperature was close to the  
436 liquidus of alkali feldspar (at least 825 °C). This is also supported by the few biotite in our natural  
437 products (Supplementary Materials 1); in fact, previous experimental work and the present results

438 (see charges D20, D15, D17, D25 in Tables 1 and 2) on trachytic melts indicate the stability field of  
439 biotite at pressure >135 MPa (Fabbrizio and Carroll 2008). Based on these results, we constrain the  
440 pre-eruptive magma storage at a depth allowing the biotite crystallization, i.e., >4 km, comparable  
441 or slightly higher than hypothesized in Piochi et al. (2005). Furthermore, our experimental data  
442 indicate that the early erupted Monte Nuovo magma (LM unit) fragmented before reaching 50-70  
443 MPa (~2 km of depth). Pre-eruptive pressure estimates (50-70 MPa) suggested by our results are in  
444 agreement with the pre-eruptive  $fH_2O$  of phlegraean magma at ~70 MPa estimated by Fabbrizio et  
445 al. (2009), implying a repetitive behaviour for the volcanic system. Low  $\Delta T$  can be suggested for  
446 the early erupted products, in agreement with previous inferences (D'Oriano et al., 2005). Our  
447 results indicate lower pressures for the later erupted magma (i.e., scoriae in the Upper Member)  
448 registers. The high groundmass crystallinity with alkali feldspar as prevalent phase and the sodalite  
449 occurrence in these later erupted fragments (Table 4) suggest effective undercoolings of several  
450 hundreds °C (Fig. 9) and pressure of crystallization below 50 MPa. Notably, Mastrolorenzo and  
451 Pappalardo (2006) also placed the fragmentation level of phlegraean volcanoes between 10 and 30  
452 MPa, but without distinction among eruptive phases. Decompression was the main process  
453 controlling the textures of the Monte Nuovo products, in agreement with occurrence of microlites  
454 that are curved following the morphology of adjacent vesicles, suggesting that they grew  
455 concomitantly with vesicles (Piochi et al., 2005; Piochi et al. 2008). In particular, our results  
456 support the idea that open-system degassing promoted massive crystallization in later erupted  
457 magma (Fig. 3), moving the melt towards the most evolved compositions observed (D'Oriano et al.,  
458 2005; Piochi et al., 2008).

459 The magma ascent from the storage ( $\leq 135$  MPa) to fragmentation ( $\leq 50$  MPa) pressures occurred  
460 rapidly, with timescales on the order of hours suggested by our CSD results (see Table 5). Fabbrizio  
461 and Carroll (2008) also suggested that the travel of the biotite-bearing phlegraean magma from  
462 reservoir at depth of c. 7 km (~200 MPa) to the surface was rapid, based on dissolution rate data of  
463 biotites (hours to a maximum of 1-2 days; Fabbrizio and Carroll, 2008). Low ascent rate results in

464 dissolution of biotite. Therefore, the magma ascent rate is estimated in 400-100 m/h from a depth  
465 of 4-5 km (biotite-stable). Magma storage of some days in the shallow crust is indicated by the  
466 occurrence of some phenocrysts (that forms slope  $>-10$ , Table 5, Fig. 6), whereas the smaller-sized  
467 crystals (and the CSD with higher slopes; Table 5 and Fig. 6) were able to crystallize in few hours.  
468 These results better constrain previous reconstruction of this eruption (Piochi et al. 2005; D’Oriano  
469 et al. 2005). The detected CSD variation reflects the textural features of samples, resulting from  
470 small differences in rates of magma ascent, crystallization, and degassing (see also D’Oriano et al.  
471 2005; Piochi et al. 2005, 2008) during eruption. The LM vesicular pumices (early erupted; Fig. 1)  
472 register the magmatic fragmentation generated by magma-water interaction following gas  
473 expansion; while syn-eruptive outgassing-induced crystallization produced brown pumices and  
474 scoriae (later erupted magma; Fig. 1) in the course of the eruption. The most crystalline and less  
475 vesicular scoriae contain lower amount of H<sub>2</sub>O than LM microlite-poor and vesicle-rich pumices  
476 (Piochi et al. 2008).

477 The Phlegraean trachyte can rapidly crystallize large crystal fractions of alkali feldspar, over  
478 small pressure-temperature intervals, as shown in this experimental study, and this can strongly  
479 modify the rheology of a trachytic magma, favoring rapid changes in eruptive style, if not the  
480 ending of the eruption. The obtained results allow us to constrain the magma processes producing  
481 the change in the eruptive style from pumice to scoria type deposits, due to an increase of magma  
482 viscosity (Caricchi et al., 2008; Vona et al. 2013), quantitatively supporting the idea of D’Oriano et  
483 al. (2005) about the plug generation in the volcanic conduit in the last eruptive phase. In fact,  
484 values of  $\phi$  between 0.80 and 0.90 observed in the natural scoriae of Monte Nuovo produced in this  
485 phase requires higher  $\Delta T$  and lower P, as suggested from the experiments. Therefore, the same  
486 experimental data indicates lowering water content, corroborating hypothesis of open degassing  
487 conditions during the scoria eruption (D’Oriano et al. 2005; Piochi et al. 2008). The abundance of  
488 crystals in the scoria deposits reaching values higher than 90% (D’Oriano et al. 2005) may be  
489 related to high  $\Delta T$  and low  $P_f (=P_{H_2O})$  or long residence of the magma in shallower part ( $P_f < 50$

490 MPa) of the conduit, as shown from our results (see samples D69, D78 and D62).  $\Delta T$  can enhance  
491 crystal fraction by increasing crystal nucleation rate, changing the magma rheology toward more  
492 viscous behaviour as crystal content increases. This is in agreement with evidence showing that in  
493 a closed magmatic system undergoing continuous decompression and cooling, crystal could only  
494 become larger and more numerous with time (Wolf and Eichelberger 1997). Furthermore, Melnik  
495 (2000) shows that the high viscosity value of the magma in the conduit is probably responsible for  
496 increasing pressurization, and our results justify a potential overpressure in the conduit, which may  
497 have triggered the final vulcanian explosion of Monte Nuovo.

498 Finally, the coexistence of pumices with different vesicle and microlite textures (for example,  
499 samples LM inf 1, LM in d and LM c3) suggests a complex process of magma ascent dynamics.  
500 Mingling between magmas that reached the surface through different pathways and pressure-  
501 temperature-time conditions in the subsurface or, following D’Oriano et al. (2005), the horizontal  
502 zoning of magma conditions in the conduit are possibilities to be further investigated.

503

## 504 6. Conclusion

505 The comparison between experimental data and data from natural samples furnishes a set of P-T-  
506 H<sub>2</sub>O content conditions for trachytic magmas valid for the Monte Nuovo eruption, at the Campi  
507 Flegrei. The cooling and decompression experiments allowed reproducing textures observed in  
508 pumices and scoriae produced during the 1538 AD eruption. First, the obtained results indicate that  
509 the juvenile pyroclastic components i) represent the magma at or near the fragmentation surface and  
510 ii) register the magma condition within the volcanic conduit. Also, this eruption requires a water-  
511 saturated magma stored at around 135 MPa, near the liquidus temperature of alkali feldspar; its  
512 ascent toward the surface lasted a few hours to several days. Months of magma crystallization are  
513 registered in some pyroclastic fragments. Therefore, the intense seismicity, gas emissions and  
514 ground deformations described by the historical chronicles (see Guidoboni and Ciucciarelli 2011  
515 and references therein) in the months before the eruption are phenomena related to the volcanic

516 unrest. Our results, instead, suggest that the low seismicity and gas emissions that affected the  
517 Campi Flegrei area prior to at least 1530-1536 AD are not directly connected to the unrest and  
518 should be related either to the deeper magmatic system or to the hydrothermal environment. Our  
519 results do not furnish direct information concerning what triggered the eruption at Monte Nuovo.

520 As a whole, this experimental study offers a set of pressure, temperature and undercooling (water  
521 saturated) conditions, useful to constrain the timescale and to better understand the magma behavior  
522 of numerous phlegraean eruptions, starting from the texture of their eruptive products. In fact, alkali  
523 feldspar is the main crystal phase in the phlegraean rocks and, as shown for the Monte Nuovo  
524 eruption, its crystallization could be started between 7 and 4 km depth ( $\leq 200$  MPa) and proceed in  
525 the conduit, where the magma ascent was faster and texture was effectually frozen in, located  
526 mostly between 2 and 3 km (50-70 MPa). The sodalite absence in most of the natural samples  
527 implies that magmas generally fragmented before reaching 30-50 MPa (pressure for sodalite  
528 stability) or magmas stored at these levels only occasionally were erupted. Furthermore, the  
529 temperature of the magma, in pre-eruptive conditions, is most likely between 825 and 840 °C (close  
530 to the liquidus of alkali feldspar). Notably, this study allows us to tentatively forecast the pre-  
531 eruptive magma conditions and timescales of future events in the Campi Flegrei area.

532

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543

544

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664

665 **Figure captions**

666

667 Figure 1: The Monte Nuovo tuff cone location (a) with image of the lowermost ash- to sand-  
668 levels and the uppermost scoria deposits (b and c, respectively). The diagrams show the timing of  
669 the phenomena preceding the eruption since 1470 AD (d) and 1538 (e), on the basis of Guidoboni  
670 and Ciucciarelli (2011). The d and e diagrams indicate the time of occurrence for seismicity  
671 (vertical bars), flow output (cloud) and ground deformation (arrows and dotted lines, right  
672 concavity meaning inflation and viceversa). The relation between phenomena and outcrops is also  
673 indicated. The onset for microlite growth as suggested by CSD is also indicated. The brown pumice  
674 and scoria samples location is indicated by black boxes in panels a and b. The sampling site of the  
675 early erupted yellow pumices of LM inf d and LM inf l is in panel a.

676

677 Figure 2: TAS (Le Bas et al. 1986), on the left, and Alkali vs. CaO, on the right, diagrams for the  
678 Monte Nuovo natural glasses and ZAC bulk rock. Data on natural glass and whole-rocks from  
679 Piochi et al. (2008), Rosi and Sbrana (1987), D’Oriano et al. (2005).

680

681 Figure 3: I have changed contrast-maybe better to see phases Groundmass texture of natural  
682 pumice (samples: c1 1-7, c2s 9-12, infl 8-12, infsc 02-05) and scoriae (samples: MN3/1, MN4top)  
683 from base upwards in the stratigraphic sequence. Samples MN3/1 (a) and MN4top (b) are two  
684 scorias upwards in the sequence. Note the cusped shape of vesicle and the magnetite occurrence in  
685 the most crystallized and low-vesicular MN4top. Samples c1 1-7 (c) and c2s 9-12 (d) evidence the  
686 groundmass crystallinity and vesicularity feature of a yellowish and a brown pumice, respectively,  
687 within the main pyroclastic unit. Note the microlite alignment and the curved shape of several  
688 crystals. Rare microlites and high vesiculation of pumice at the base of the flow sequence. In  
689 particular, sample infl 8-12 (e) shows the cracks (white arrows) produced by water/magma  
690 interaction. Sample infsc 02-05 (f) is a banded pumice within the main pyroclastic unit

691 characterized by low crystal abundance and higher vesiculation of yellowish band (on the left) with  
692 respect of the adjacent brown sector. White bars are 100  $\mu\text{m}$ .

693

694 Figure 4: Backscattered SEM images of textures obtained at low pressure (50 and 30 MPa) from  
695 isobaric cooling, “decompression+ cooling” and isothermal decompression experiments. Cooling  
696 experiments: (a) D33 ( $P_f = 50 \text{ MPa}$ ,  $\Delta T = 25 \text{ }^\circ\text{C}$ ); (b) D10 ( $P_f = 50 \text{ MPa}$ ,  $\Delta T = 40 \text{ }^\circ\text{C}$ ); (c) D69 ( $P_f$   
697  $= 50 \text{ MPa}$ ,  $\Delta T = 115 \text{ }^\circ\text{C}$ ). “Decompression+ cooling” experiments: (d) D45 ( $P_f = 50 \text{ MPa}$ ,  $\Delta T = 115$   
698  $^\circ\text{C}$ ); (e) D43 ( $P_f = 50 \text{ MPa}$ ,  $\Delta T = 115 \text{ }^\circ\text{C}$ ); (f) D47 ( $P_f = 30 \text{ MPa}$ ,  $\Delta T = 140 \text{ }^\circ\text{C}$ ). Isothermal  
699 decompression experiments: (g) D57 ( $P_f = 50 \text{ MPa}$ ,  $\Delta T = 40 \text{ }^\circ\text{C}$ ); (h) D60 ( $P_f = 30 \text{ MPa}$ , ;  $\Delta T = 65$   
700  $^\circ\text{C}$ ); (i) D62 ( $P_f = 30 \text{ MPa}$ ,  $\Delta T = 65 \text{ }^\circ\text{C}$ ). The samples D33, D10, D69 and D62 were performed in  
701 this study, the other ones are data of Arzilli and Carroll (2013).

702

703 Figure 5: Backscattered SEM images of textures obtained at high pressure (200, 150, 100 and 70  
704 MPa) from isobaric cooling, “decompression+ cooling” and isothermal decompression experiments.  
705 Cooling experiments: (a) D1 ( $P_f = 200 \text{ MPa}$ ,  $\Delta T = 20 \text{ }^\circ\text{C}$ ); (b) D15 ( $P_f = 150 \text{ MPa}$ ,  $\Delta T = 17 \text{ }^\circ\text{C}$ ); (c)  
706 D6 ( $P_f = 100 \text{ MPa}$ ,  $\Delta T = 19 \text{ }^\circ\text{C}$ ). “Decompression+ cooling” experiments: (d) D17 ( $P_f = 150 \text{ MPa}$ ,  
707  $\Delta T = 17 \text{ }^\circ\text{C}$ ); (e) D55 ( $P_f = 150 \text{ MPa}$ ,  $\Delta T = 42 \text{ }^\circ\text{C}$ ); (f) D51 ( $P_f = 100 \text{ MPa}$ ,  $\Delta T = 69 \text{ }^\circ\text{C}$ ). Isothermal  
708 decompression experiments: (g) D80 ( $P_f = 100 \text{ MPa}$ ,  $\Delta T = 19 \text{ }^\circ\text{C}$ ); (h) D82 ( $P_f = 70 \text{ MPa}$ , ;  $\Delta T = 14$   
709  $^\circ\text{C}$ ); (i) D82 ( $P_f = 70 \text{ MPa}$ ,  $\Delta T = 14 \text{ }^\circ\text{C}$ ). The samples D15, D6 and D17 were performed in this  
710 study, the other ones are data of Arzilli and Carroll (2013).

711

712 Figure 6: CSDs of early erupted low-crystalline and highly vesicular pumice (left), brown  
713 pumices intermediate in the stratigraphic sequence (central diagram) and scoriae (right diagram).  
714 Note the occurrence of two microlites population and the similarity of slopes of smaller microlites.

715

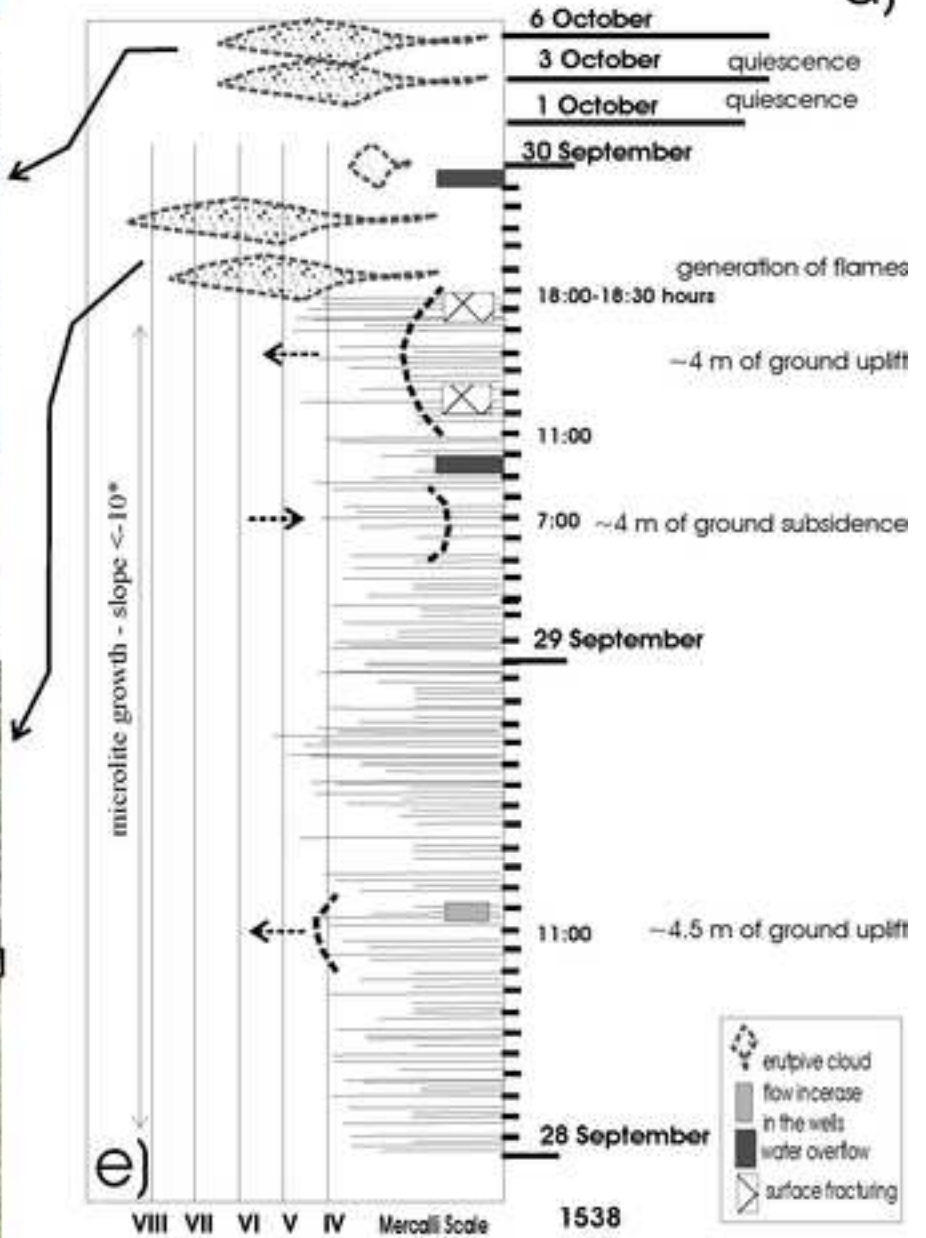
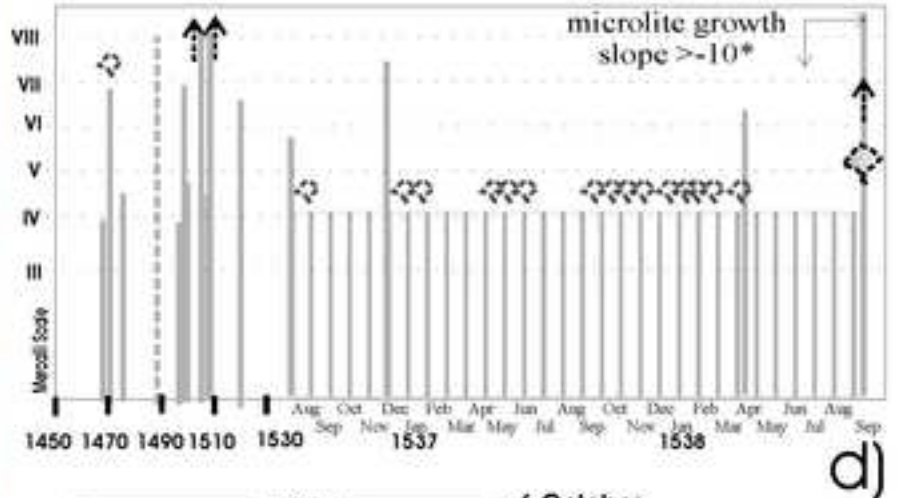
716 Figure 7: The relation of  $\phi$  and  $N_a$  with  $P_f$ ,  $\Delta T$  and  $t_{exp}$  in cooling experiments. (a) Relation  
717 between crystal fraction of alkali feldspar ( $\phi$ ) and  $\Delta T$  as a function of  $P_f$ . (b) Relation between  
718 crystal fraction of alkali feldspar ( $\phi$ ) and  $t_{exp}$  as a function of  $P_f$  and  $\Delta T$ . (c) Relation between  $N_a$  and  
719  $\Delta T$  as a function of  $P_f$ . (d) Relation between  $N_a$  and  $t_{exp}$  as a function of  $P_f$  and  $\Delta T$ .

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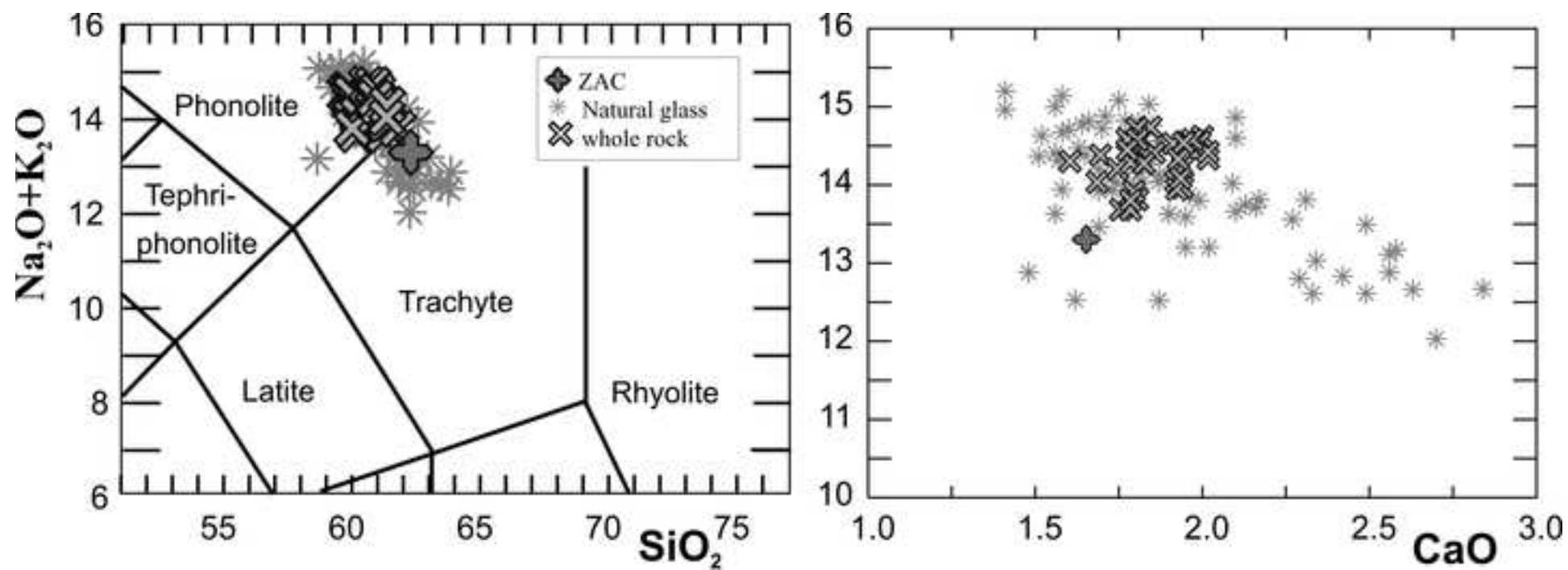
721 Figure 8: The relation of  $\phi$  and  $N_a$  with  $P_f$ ,  $\Delta T$  and  $t_{exp}$  in “decompressio+cooling” and isothermal  
722 decompression experiments. (a) Relation between crystal fraction of alkali feldspar ( $\phi$ ) and  $\Delta T$  as a  
723 function of  $P_f$ . (b) Relation between crystal fraction of alkali feldspar ( $\phi$ ) and  $t_{exp}$  as a function of  $P_f$   
724 and  $\Delta T$ . (c) Relation between  $N_a$  and  $\Delta T$  as a function of  $P_f$ . (d) Relation between  $N_a$  and  $t_{exp}$  as a  
725 function of  $P_f$  and  $\Delta T$ .

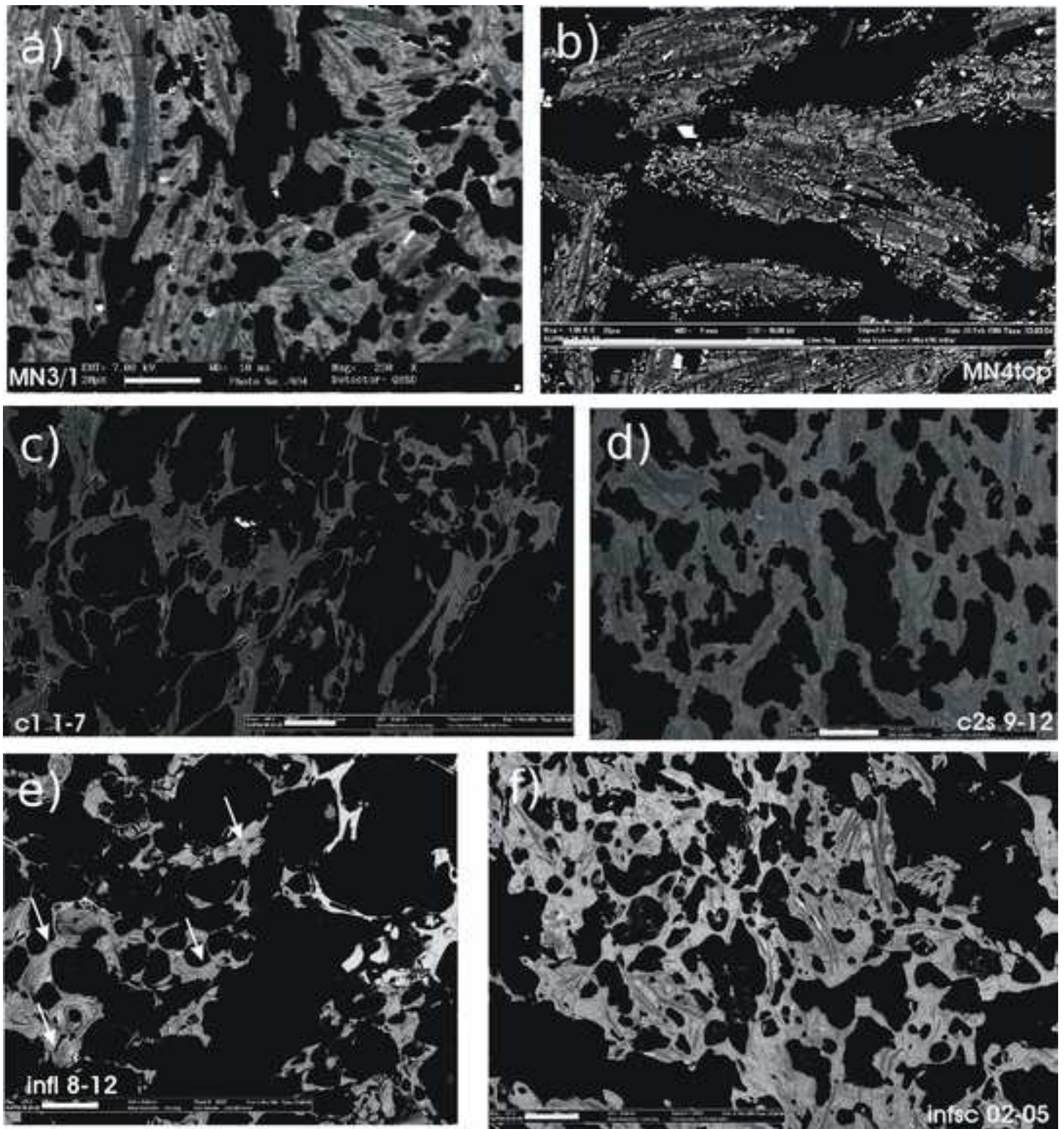
726

727 Figure 9:  $N_a$  vs  $\phi$ . a) Relation between  $N_a$  (nucleation density) and  $\phi$  (crystallinity) as a function  
728 of  $P_f$  and  $\Delta T$  for cooling experiments and natural samples of Monte Nuovo (pumices and scoriae);  
729 b) relation between  $N_a$  and  $\phi$  as a function of  $P_f$  and  $\Delta T$  for both types of decompression  
730 experiments and natural samples of Monte Nuovo (pumices and scoriae).

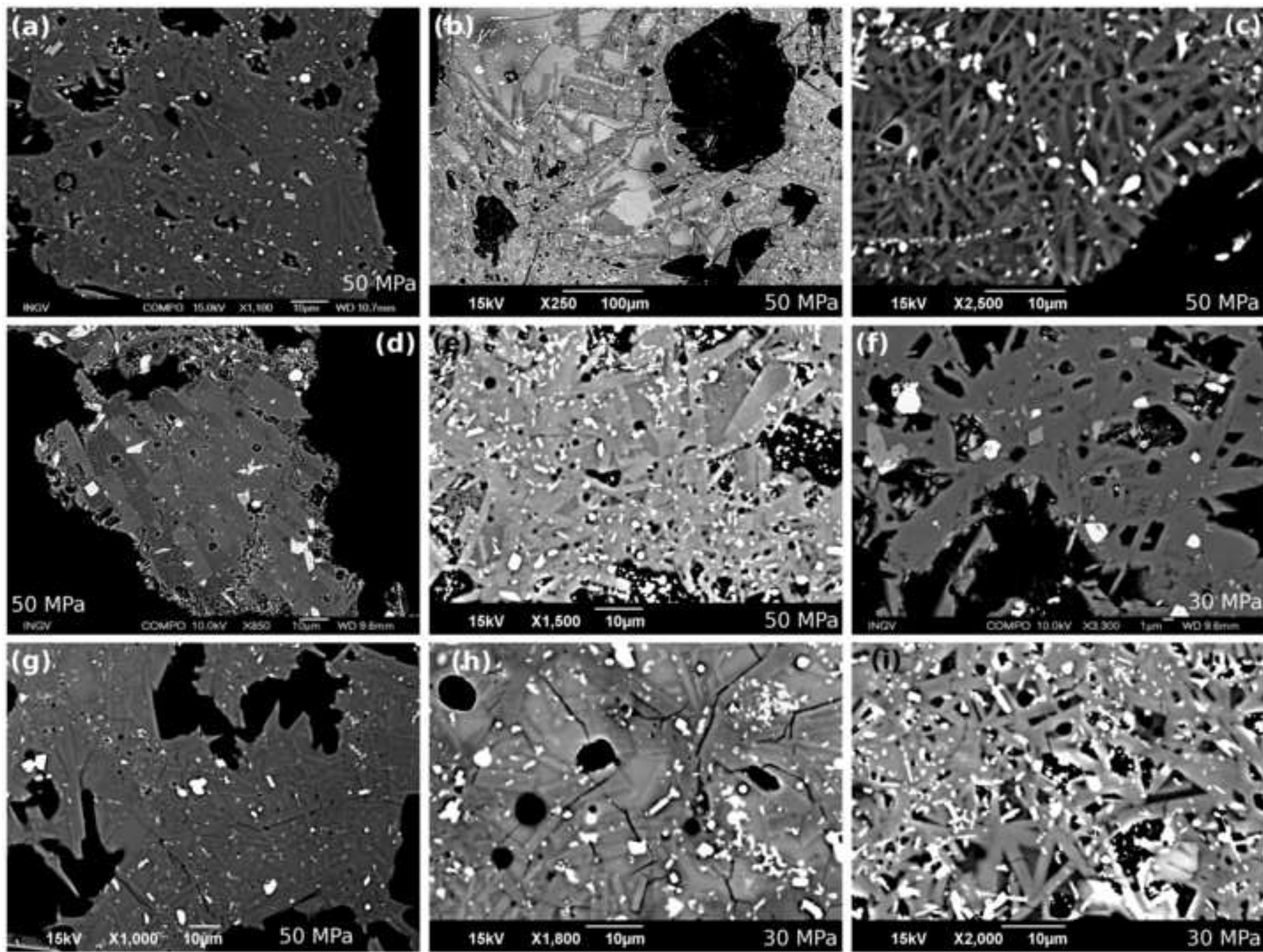




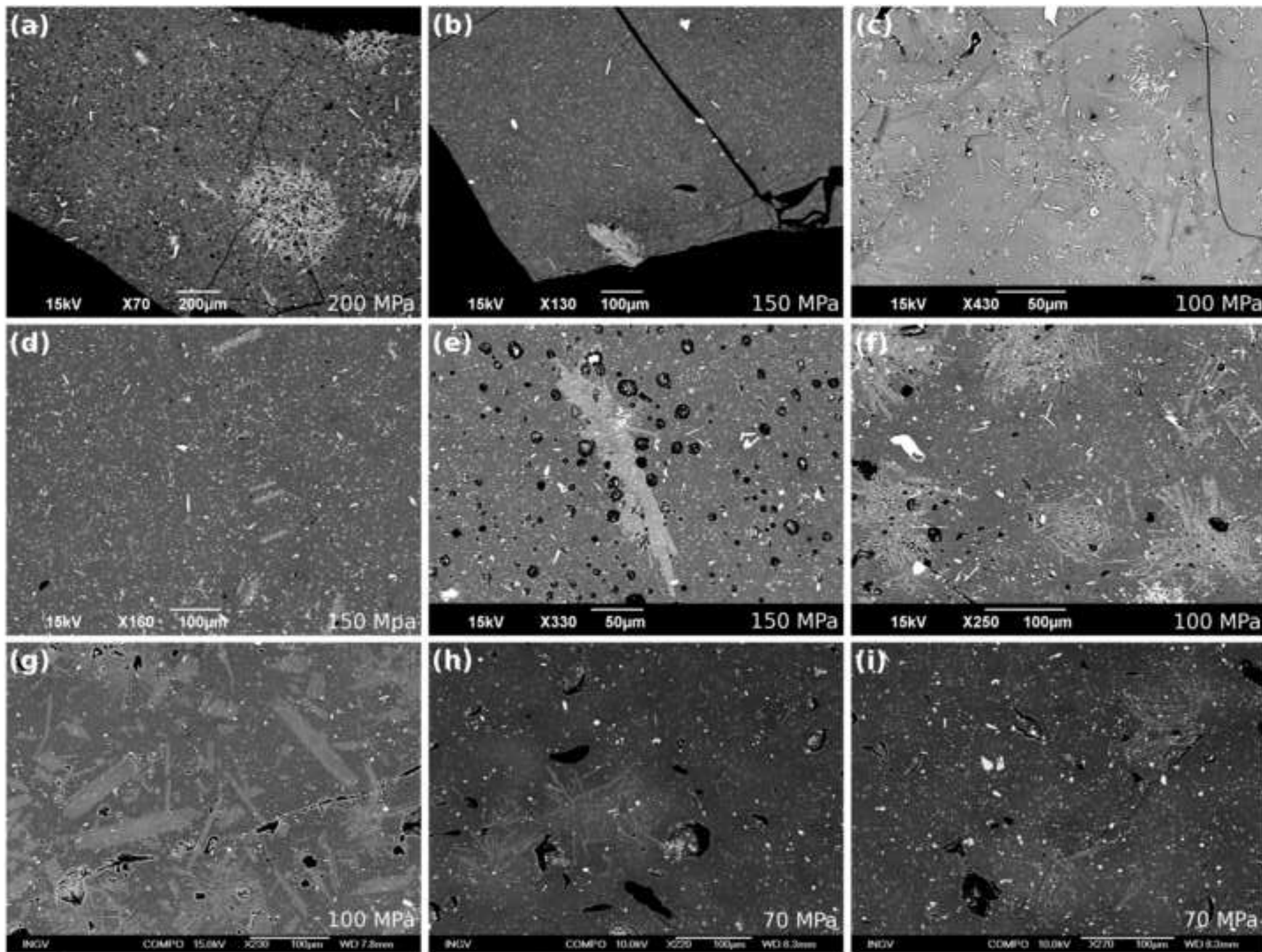


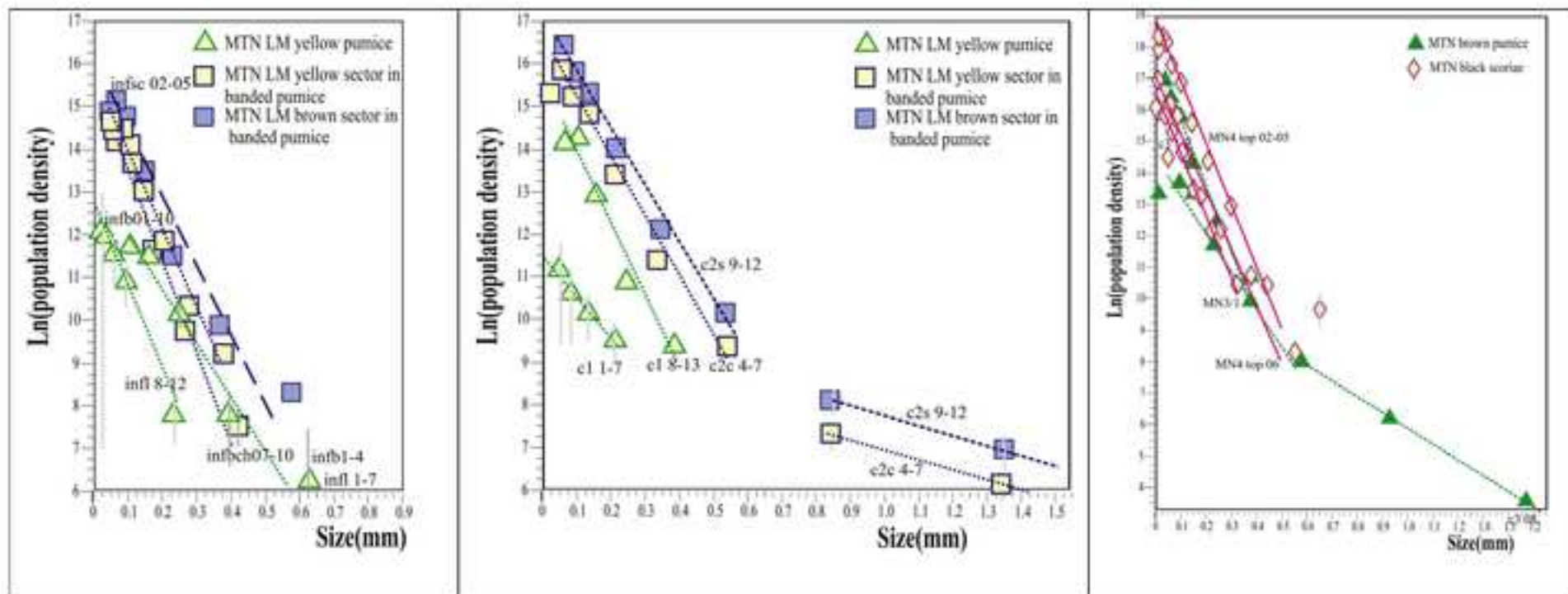


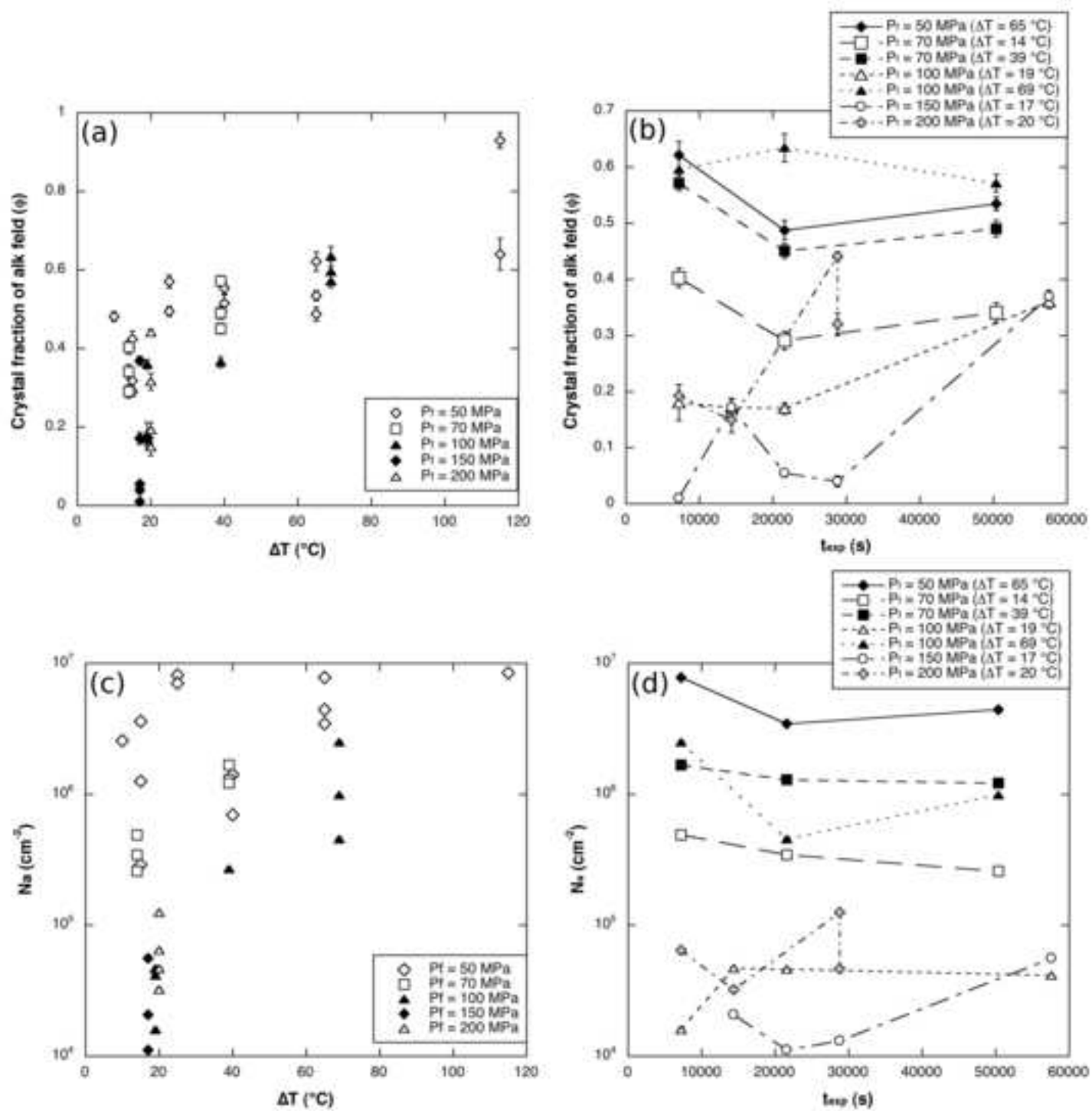


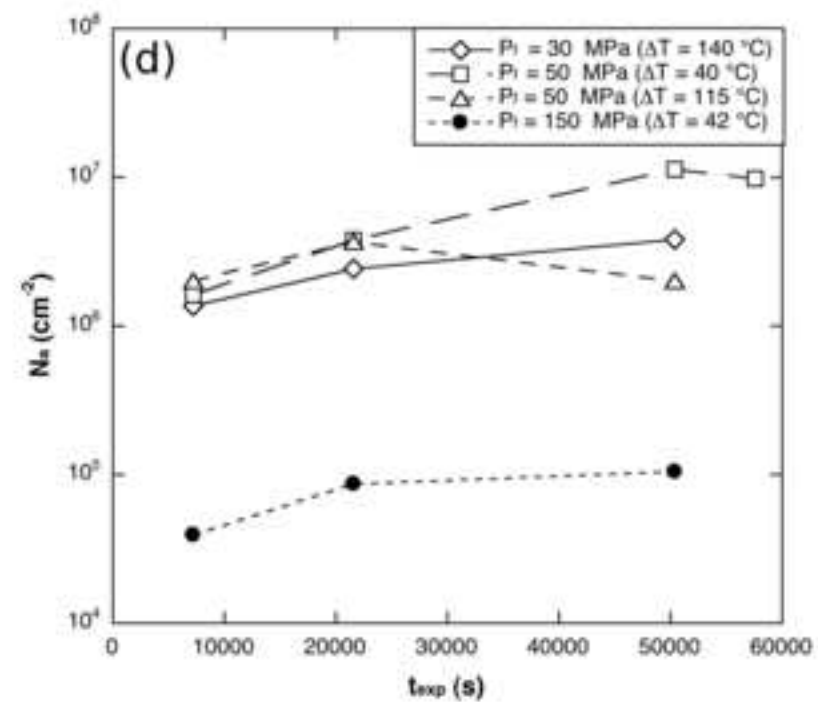
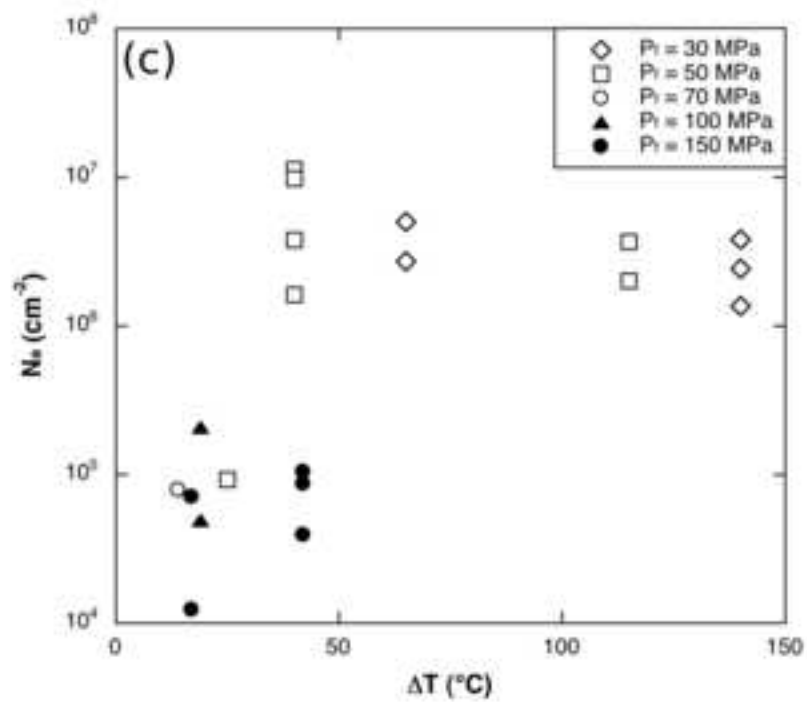
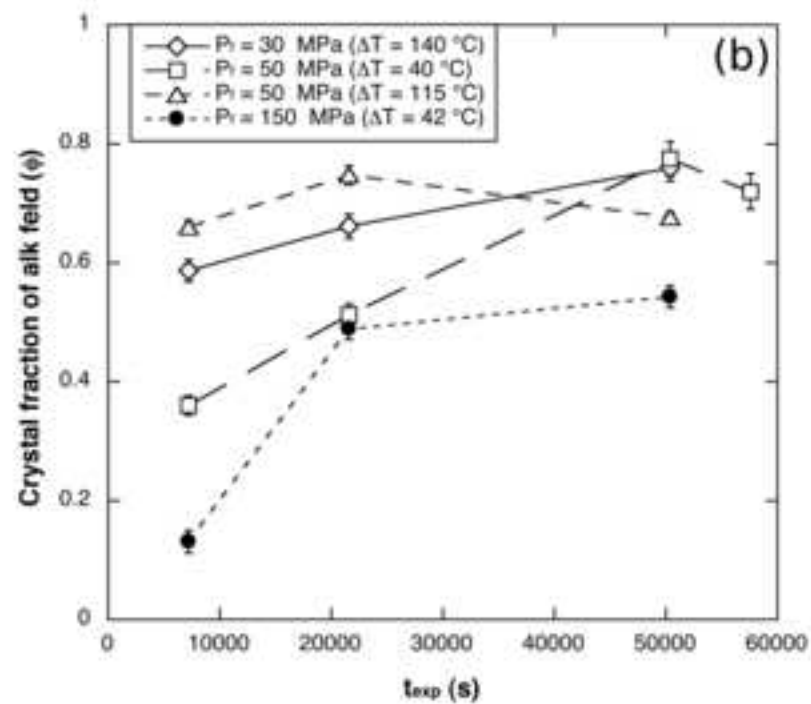
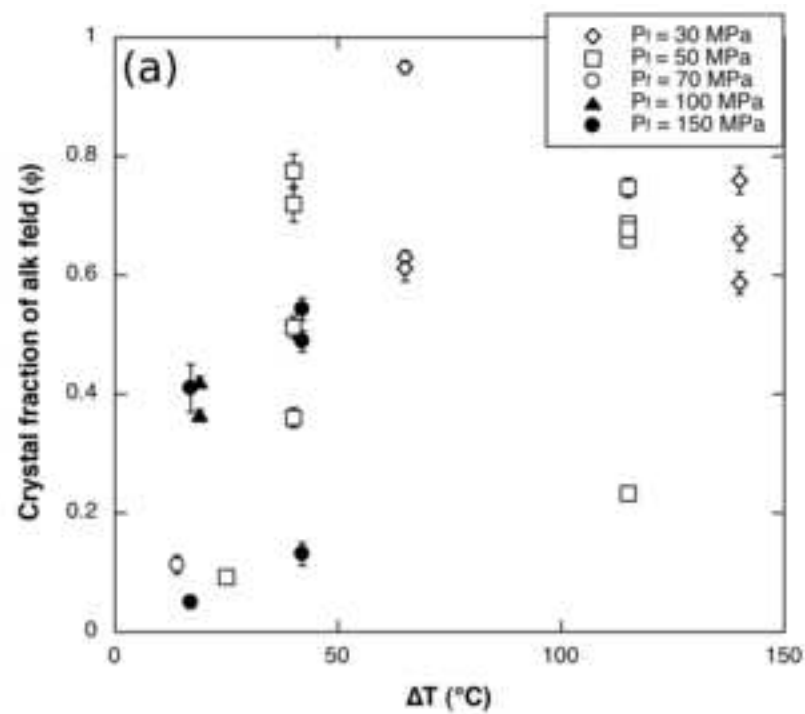














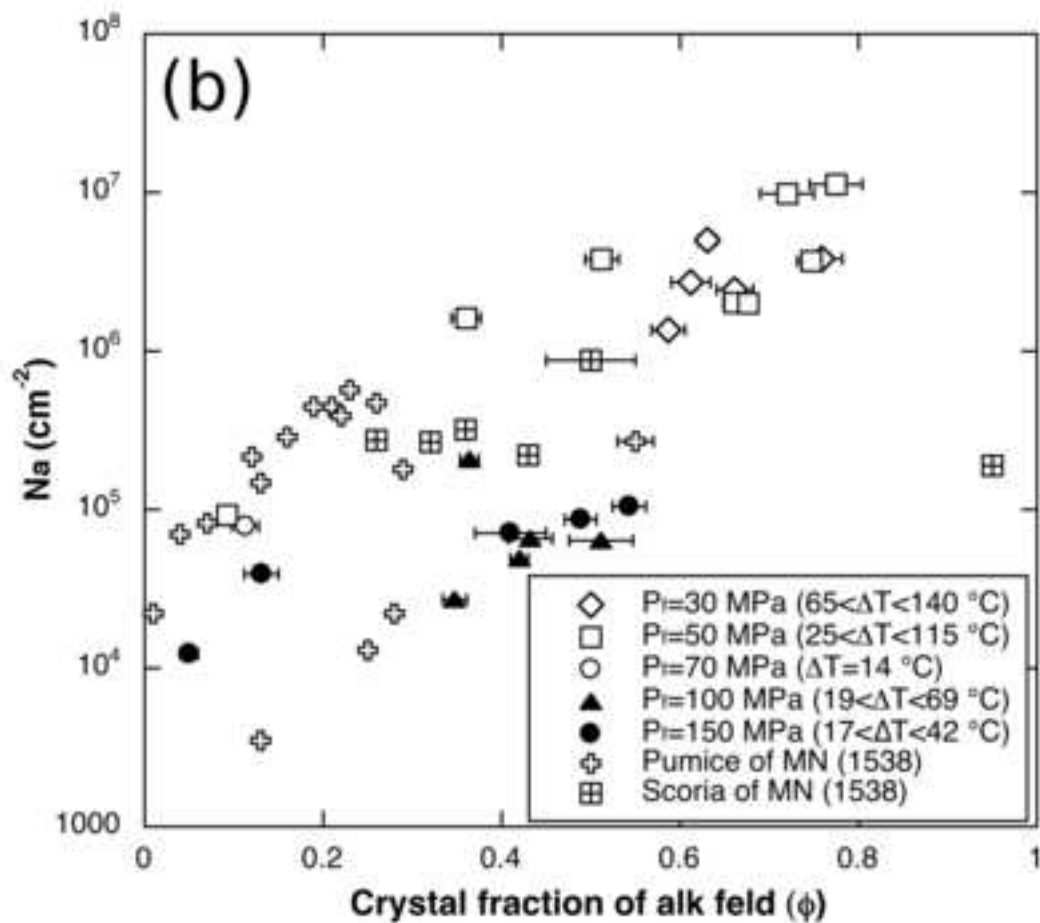
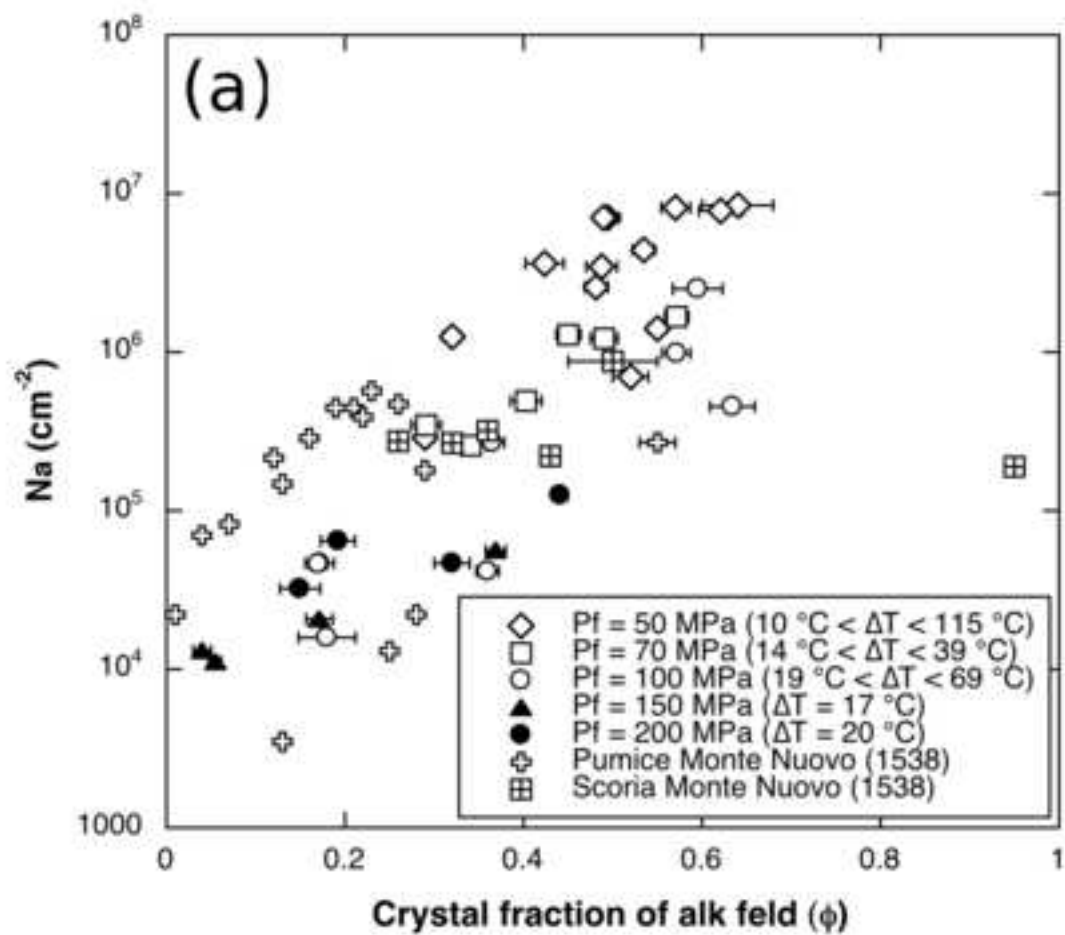




Table 1: Chemical composition of ZAC (from Di Matteo et al. 2004) and average for natural samples from units selected for this study (see Piochi et al., 2008 for not H<sub>2</sub>O-free normalized data). FeO\* = total iron as FeO.

oxide (wt%)	ZAC	LM inf	LMc	MN4
SiO <sub>2</sub>	62.18	60.76	60.77	63.71
TiO <sub>2</sub>	0.45	0.42	0.43	0.31
Al <sub>2</sub> O <sub>3</sub>	18.70	19.09	19.39	19.91
FeO*	3.19	3.05	3.05	1.63
MnO	0.27	0.27	0.27	0.07
MgO	0.23	0.23	0.24	0.12
CaO	1.65	1.89	1.94	1.55
Na <sub>2</sub> O	6.16	7.09	6.82	6.67
K <sub>2</sub> O	7.14	7.17	7.08	6.04
P <sub>2</sub> O <sub>5</sub>	0.02	0.03	0.02	0.01
Total	100.00	100.00	100.00	100.00

Table 2. Experimental conditions for “isothermal cooling”, “decompression + cooling” and “isothermal decompression” experiments.

sample	T <sub>i</sub> (°C)	T <sub>f</sub> (°C)	P (MPa)	t <sub>m</sub> (s)	t <sub>exp</sub> (s)	*H <sub>2</sub> O (wt. %)	ΔT (°C)	-ΔT (°C)	
<i>Isobaric cooling experiments</i>									
D20	900	750	200	72000	28800	7	20	131	
D15	900	775	150	7200	7200	6	17	108	
D13	880	775	150	7200	28800	6	17	88	
D6	880	800	100	7200	21600	5.2	19	61	
D11	900	850	50	7200	7200	2.7	15	35	
D12	900	850	50	7200	14400	2.7	15	35	
D33	900	840	50	7200	14400	2.7	25	35	
D10	900	825	50	7200	28800	2.7	40	35	
D9	900	825	50	7200	57600	2.7	40	35	
D69	880	750	50	7200	21600	2.7	115	15	
D78	880	750	50	7200	50400	2.7	115	15	
sample	T <sub>i</sub> (°C)	T <sub>f</sub> (°C)	P <sub>i</sub> (MPa)	P <sub>f</sub> (MPa)	t <sub>m</sub> (s)	t <sub>exp</sub> (s)	*H <sub>2</sub> O (wt. %)	ΔT (°C)	-ΔT (°C)
<i>Decompression + cooling experiments</i>									
D17	825	775	200	150	72000	21600	6	17	56
D25	825	775	200	150	72000	57600	6	17	56
D29	825	800	200	100	7200	57600	5.2	19	56
D21	825	825	200	50	7200	57600	2.7	40	56
<i>Isothermal decompression experiments</i>									
D62	825	825	150	30	3600	50400	1.7	65	33

note: T<sub>i</sub> = initial (or melting) temperature; T<sub>f</sub> = final (or experimental) temperature; P<sub>i</sub> = initial (or melting) pressure; P<sub>f</sub> = final (or experimental) pressure; t<sub>m</sub> = melting time; t<sub>exp</sub> = experimental time; ΔT = the undercooling degree; -ΔT = the superheating degree. \*H<sub>2</sub>O (wt %) was calculated using the polynomial fit by Di Matteo et al. (2004).

Table 3. Experimental results of nucleation and growth of alkali feldspar during “isothermal cooling”, “decompression + cooling” and “isothermal decompression” experiments.

sample	$N_a$ (cm <sup>-2</sup> )	$\phi$	L (cm)	$Y_L$ (cm/s)	phases
<i>Isobaric cooling experiments</i>					
D20	4.6E+04(1)	0.32(0.02)	0.0260(0.0095)	4.5E-07(9)	gl-af-cpx-mt-bt-bl
D15	n.d.	0.01	n.d.	n.d.	gl-af-cpx-mt-bt-bl
D13	1.31E+04(1)	0.04(0.01)	0.0116(0.0020)	2.0E-07(4)	gl-af-cpx-mt-bt-bl
D6	4.62E+04(1)	0.17(0.01)	0.0182(0.0032)	2.8E-07(4)	gl-af-cpx-mt-bl
D11	2.90E+05(4)	0.29(0.01)	0.0107(0.0035)	7.4E-07(2)	gl-af-cpx-mt-bl
D12	1.26E+06(1)	0.32(0.01)	0.0096(0.0019)	3.3E-07(7)	gl-af-cpx-mt-bl
D33	7.1E+06(1)	0.49(0.01)	0.0041(0.0006)	1.4E-07(2)	af-cpx-mt-gl-bl
D10	6.97E+05(2)	0.52(0.02)	0.0116(0.0020)	2.0E-07(3)	af-cpx-mt-gl-bl
D9	1.41E+06(2)	0.55(0.01)	0.0045(0.0011)	3.9E-08(9)	af-cpx-mt-gl-bl
D69	n.d.	0.93(0.02)	n.d.	n.d.	af-cpx-mt-gl-bl-sod
D78	n.d.	0.93(0.02)	n.d.	n.d.	af-cpx-mt-gl-bl-sod
<i>Decompression + cooling experiments</i>					
D17	1.23E+04(1)	0.05(0.01)	0.0092(0.0022)	2.13E-07(8)	gl-af-cpx-mt-bt-bl
D25	7.11E+04(6)	0.41(0.04)	0.0174(0.0032)	1.51E-07(3)	gl-af-cpx-mt-bt-bl
D29	4.92E+04(5)	0.42(0.01)	0.0185(0.003)	1.6E-07(3)	gl-af-cpx-mt-bl
D21	9.82E+06(3)	0.72(0.03)	0.004(0.0012)	3.5E+08(2)	af-cpx-mt-gl-bl
<i>Isothermal decompression experiments</i>					
D62	n.d.	0.95(0.01)	n.d.	n.d.	af-cpx-mt-gl-bl-sod

note:  $N_a$  = nucleation density;  $\phi$  = volume fraction; L = maximum length;  $Y_L$  = maximum growth rate. The value in parentheses is the standard deviation of the mean value. n.d. = not determined phases presents because the contrast between several phases is not sufficient to distinguish them in BSE images.

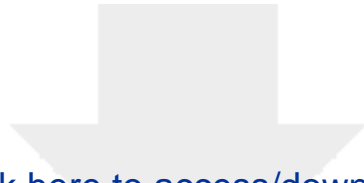
Table 4: Mineral phase abundances (wt%) in the Monte Nuovo products as determined by semi-quantitative analyses of the XRPD data (see Supplementary Material 2).

<b>sample</b>	<b>Anorthoclase</b>	<b>Sanidine</b>	<b>Sodalite</b>	<b>Biotite</b>	<b>Magnetite</b>
<b>LM C3 wr</b>	70.9	24.8	3.4	0.6	0.4
<b>LM C3 sh</b>	82.6	14.2	2.8	0.2	0.3
<b>LM inf l wr</b>	35.9	63.7	-	0.5	-
<b>LM inf l sh</b>	34.9	64.9	-	0.2	-
<b>LM inf d sh</b>	39.8	59.6	-	0.5	0.1
<b>MN4wr</b>	59.5	33.6	5.8	-	1.1
<b>MN4 sh</b>	54.9	39.2	5.6	-	0.3

Note: Goodness of Fitt (GOF) <3; refer to text for analytical procedures; XRPD patterns and crystallographic index phase in the Supplementary Material 2.

Table 5: Crystal growth rates ( $Y_L$ ) as derived by experiments and related residence time ( $t_r$ ) as calculated through the equation:  $t_r = (-1/Y_L * \text{slope})$ . Slopes are from the CSD diagrams in Fig. 7.

sample	slope	intercept (mm)	tr (h) - mean	tr (h) - min	tr (h) - max	tr(days) - max
infl 8-12	-20.8	12.8	6.1	1.1	121.6	5.1
infb 01-10	-21.6	15.7	5.8	1.1	117.2	4.9
infl 1-7	-12.3	13.2	10.2	1.9	204.8	8.5
infb 1-4	-15.6	16.2	8.0	1.5	161.7	6.7
infsc 02-05	-18.5	16.0	6.8	1.3	136.4	5.7
infb ch07-10	-16.1	13.3	7.8	1.4	156.5	6.5
c1 1-7	-9.7	11.3	13.0	2.4	261.1	10.9
c1 8-13	-17.0	15.8	7.4	1.4	148.9	6.2
c2c 4-7 - 1	-2.3	9.3	54.7	10.1	1099.5	45.8
c2s 9-12 - 1	-2.3	10.0	55.6	10.2	1117.4	46.6
c2c 4-7 - 2	-8.1	16.8	15.6	2.9	313.2	13.1
c2s 9-12 - 2	-13.7	17.3	9.2	1.7	184.4	7.7
MN4 top 06 - 1	-6.7	11.0	18.9	3.5	378.8	15.8
MN4 top 06 - 2	-12.9	14.7	9.7	1.8	195.1	8.1
c3 1-2	-19.6	16.3	6.4	1.2	128.6	5.4
c3 1-2	-20.4	17.5	6.2	1.1	123.7	5.2
MN4top 02-05	-20.0	19.0	6.3	1.2	126.3	5.3
Experimental Growth rate			mean $Y_L =$	mean $Y_L =$	mean $Y_L =$	
		cm/s	2.21E-07	1.20E-06	1.10E-08	
		mm/h	7.96E-03	4.32E-02	3.96E-04	



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