Tracking bubble evolution inside a silicic dike

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Abstract: Pressure estimates from rapidly erupted crustal xenoliths constrain the depth of intrusion of the silicic lavas hosting them. This represents an opportunity for tracking magmatic bubble's evolution and quantifying the variation in bubble volume during rapid magma ascent through a volcanic dike just prior to eruption. The petrology, stable-isotope geochemistry and X-ray micro-tomography of dacites containing crustal xenoliths, erupted from a Neogene volcano in SE Spain, showed an increase in porosity from ~1.7 to 6.4 % from ~19 to 13 km depth, at nearly constant groundmass and crystal volumes. This result provides additional constraints for experimental and numerical simulations of subvolcanic magma-crust degassing processes in silicic systems, and may allow the characterization of volcanic eruptive styles based on volatile content.

Direct observation of bubbles in a natural-silicic volcano-laboratory

Combination of petrology, geochemistry and micro-computed-tomography

Bubbles evolution in the magma dike from 19 to 13 km depth

Implications for the volatiles influence in the eruptive processes at higher depths than the usually considered for the volcanic vent

1 **Tracking bubble evolution inside a silicic dike**

2

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26 **ABSTRACT**

27 Pressure estimates from rapidly erupted crustal xenoliths constrain the depth of
28 intrusion of the silicic lavas hosting them. This represents an opportunity for tracking
29 magmatic bubble's evolution and quantifying the variation in bubble volume during
30 rapid magma ascent through a volcanic dike just prior to eruption. The petrology,
31 stable-isotope geochemistry and X-ray micro-tomography of dacites containing
32 crustal xenoliths, erupted from a Neogene volcano in SE Spain, showed an increase
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34 groundmass and crystal volumes. This result provides additional constraints for
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36 processes in silicic systems, and may allow the characterization of volcanic eruptive
37 styles based on volatile content.

38
39 **Keywords:** Volcanic bubbles; Dacite; X-ray micro-tomography; Crustal xenoliths;
40 Conduit depths; Stable Isotopes

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51 **1. Introduction**

52 Processes occurring in the conduit between a magma chamber and the surface
53 may trigger an eruption or alter the characteristics of one already in progress (e.g.
54 Costa et al., 2007; 2009). In this regard, gases and fluids play a crucial. The
55 understanding of how gases behave and influence the eruptive style of volcanoes is
56 mainly based on observations of volcanic products, numerical models, and laboratory
57 experiments (e.g., Eichelberger et al., 1986; Jaupart and Allègre, 1991; Woods and
58 Koyaguchi, 1994; Klug and Cashman, 1996; Okumura et al., 2009; Fiege et al.,
59 2014; Fiege and Cichy, 2015). However, the evolution of magmatic gases and fluids
60 immediately prior to eruption is difficult to understand due to the complexity of
61 observing magmatic activity below the surface. While shallower volcanic vents have
62 been investigated through the analysis and experimental reproduction of pyroclastic
63 products (e.g., Valentine, 2012), interactions between the crust and magma at depth
64 (from c. 4 to 20 km) require further investigation (e.g., Álvarez-Valero et al., 2015;
65 Pla and Álvarez-Valero, 2015). The distribution, size, quantity and morphology of
66 bubbles in acidic magmas prior to eruption is under active research (e.g., Wallace et
67 al., 1995; Gualda and Anderson, 2007; Baker et al., 2012). In particular, a better
68 understanding of bubble behavior at depths greater than 2 km is required in order to
69 constrain the influence of magmatic buoyancy and magma chamber overpressure in
70 driving eruptions (e.g., Malfait et al., 2014), as well as the depth at which melt-gas
71 separation starts via bubble nucleation. The occurrence and dynamics of explosive
72 eruptions mainly depend on the initial volatile content of the magma, the ability of
73 gases to escape during its ascent, the viscosity as proxy for diffusivity of volatile
74 species, solubility vs. composition, and pressure. In order to advance in these
75 aspects, we examine El Hoyazo volcano (a Neogene silicic lava dome in SE Spain)

76 with the aim of finding out: (i) when and where bubbles nucleated under the volcano,
77 and (ii) how the porosity varies along the dike as a function of depth. We explored
78 how magmatic bubbles and volatiles evolve during rapid ascent, and their relations to
79 eruptive potential by integrating (i) visual analysis of dacitic samples using SEM and
80 X-ray micro-computed-tomography (micro-CT) (e.g., Gualda and Rivers, 2006;
81 Polacci et al., 2006; Baker et al., 2012); (ii) petrologic depth estimates of the crustal
82 xenoliths and (iii) stable isotope ratios in xenoliths and dacites, to constrain the open
83 vs. closed nature of the system at the depth within the dike where dacite magma and
84 xenoliths came into contact. We then utilized the magmatic porosities and depths to
85 derive a rate of bubble formation during magma ascent towards eruption at El
86 Hoyazo.

87

88 *1.1. Previous petrological and geochemical results for partially melted crustal*
89 *xenoliths and their host dacites*

90 The Neogene Volcanic Province (NVP) of SE Spain (Fig. 1) contains high-K
91 calc-alkaline and shoshonitic lava series, which are peraluminous. Lithologically
92 these are largely calc-alkaline dacites rich in K₂O, with volatile contents from 2 to 4
93 wt% (mainly H₂O, CO₂) (Zeck, 1970; Benito et al., 1999). These dacites host crustal
94 xenoliths –mainly medium to coarse-grained granulite-facies rocks– with a restitic
95 bulk composition depleted in silica and enriched in aluminium and iron (Zeck, 1970;
96 Cesare et al., 1997; Benito et al., 1999; Duggen et al., 2004). The xenoliths were
97 quenched immediately after eruption (e.g., Zeck, 1970; Cesare et al., 1997; Álvarez-
98 Valero and Waters, 2010), so their microstructures provide a snapshot of the
99 anatectic conditions at depth (e.g., Zeck, 1970; Cesare et al., 1997; Álvarez-Valero
100 and Kriegsman, 2007). El Hoyazo (Fig. 1) was a submarine lava dome with a ~500

101 m crater radius, and is overlain by Miocene reef carbonates. It is a small circular
102 outcrop of c. 0.7 km², which dacites include up to 15% in volume of crustal material
103 (Zeck, 1970). Its atoll geomorphology shows an inner depressed part of mainly
104 dacitic material that corresponds to the old volcanic cone, whereas the top relief is
105 composed by the reef carbonates formed onto the volcanics, which host the crustal
106 xenoliths. Crustal xenoliths and their host dacites are randomly collected within the
107 inner part.

108 The mineralogical and chemical features of the dacite hosting the xenoliths are
109 described, among others, by Zeck (1970) and Benito et al. (1999). The dacite is
110 porphyritic, with >50 vol.% of rhyolitic glassy matrix (both fresh and devitrified),
111 and phenocrysts of mainly plagioclase, biotite, cordierite, and minor orthopyroxene,
112 amphibole, magnetite and ilmenite. Xenocrysts of garnet and quartz are locally
113 present. The xenoliths relevant for this study, i.e. those from the deepest zone
114 detected, are texturally dominated by coarse-grained biotite, elliptical garnet, and
115 large foliated mats of fibrolitic sillimanite, that is locally overprinted by spinel
116 crystals rimmed by melt and minor cordierite. Glass is partially recrystallized to
117 plagioclase, K-feldspar and thin laths of high-Ti biotite. The microtexture of
118 the fibrolitic matrix has a relatively high proportion of glass, and is rimmed by glass
119 against other phases. Spinel porphyroblasts are idioblastic and zoned, both texturally
120 and compositionally (see also Álvarez-Valero and Kriegsman, 2007; Álvarez-Valero
121 and Waters, 2010). The shallowest xenoliths are of the Spl-Crd-M type. The garnet is
122 surrounded by coronas, and the overall paragenesis is Spl-Crd-Pl-Kfs-M.
123 The coronas mimic the outline of the previous garnet, whose breakdown reaction has
124 been described in detail by Álvarez-Valero et al. (2007). Sillimanite occurs as
125 aggregates of fine needles (fibrolite) in the glass.

126 Glass mostly occurs as inclusions in garnet and plagioclase, and as microveins
 127 of devitrified material, as well as intergrown with fibrolite. Melt inclusions exhibit
 128 either rounded or regular, negative-crystal shapes. As outlined by Cesare et al.
 129 (1997), their textural position within host phases is compatible with a
 130 primary trapping (Roedder 1984). Glass is transparent, showing no evidence for
 131 devitrification or crystallization. The bubbles are essentially empty, shrinkage
 132 bubbles, with no detectable Raman-active components. Fluid inclusions are rare and
 133 restricted to biotite-poor xenoliths. These inclusions contain CO₂-dominated fluids
 134 (Cesare et al., 2004).

135 Direct evidence of partial melting and melt extraction in the xenoliths is provided by
 136 the occurrence and high abundance of fresh glass (quenched melt), which occurs as
 137 an interstitial phase in layer-parallel films of the matrix and as both devitrified and
 138 fresh pockets (Zeck, 1970; Cesare et al., 1997). Glass is also enclosed by all
 139 minerals, indicating that all minerals crystallized in the presence of a melt phase,
 140 i.e., during partial melting. The presence of intergranular melt films suggest that melt
 141 was present during grain growth (by incongruent melting reactions), during
 142 subsequent (re)crystallization steps, and after recrystallization of restitic phases had
 143 ceased. Melt in inclusions and intergranular films of xenoliths is chemically different
 144 from the glass of the dacite host, which has lower Al/Si and higher K/Na ratios
 145 (Cesare et al., 1997). Based on mass balance calculations between melt inclusions,
 146 xenoliths and potential metapelitic sources, a high degree (35–60 wt %) of
 147 melt extraction has been estimated at El Hoyazo (Cesare et al., 1997).

148 Numerous isotopic studies have demonstrated that mafic magmas that formed in
 149 the mantle or lower crust interact with felsic mid-crustal magmas and country rock to
 150 produce so-called hybrid magmas (e.g., Benito et al., 1999; Duggen et al., 2004;

151 Leeman et al., 2008). The NVP dacites are the result of mixing between rhyolites
152 extracted from partially melted pelitic crust and basalts derived from mafic
153 underplating of the Betic Cordillera (e.g., Benito et al., 1999; Duggen et al., 2004;
154 Álvarez-Valero and Kriegsman, 2007).

155 Microstructures and age relationships in the xenoliths indicate that they first
156 underwent a stage of migmatization and melt extraction, where the rhyolite mixed
157 with the primary basalt to form the host dacites (Duggen et al., 2004; Álvarez-Valero
158 and Kriegsman, 2007). A second sequence of melt-bearing reaction microstructures,
159 developed in a transiently heated wall-rock profile adjacent to the magma conduit,
160 triggered the collapse of the crustal container walls resulting in the brief residence of
161 xenoliths in a dacitic magma at c. 850°C (Álvarez-Valero and Waters, 2010). The
162 xenoliths experienced rapid transfer from depth to the surface during the eruption,
163 with minutes to hours separating the xenoliths partial melting and their preservation
164 by post-eruptive quenching (Álvarez-Valero and Waters, 2010; Álvarez-Valero et al.,
165 2015; Pla and Álvarez-Valero, 2015).

166

167 **2. Methods**

168 The provenance depths of the crustal xenoliths below El Hoyazo are well
169 constrained at c. 6 and 4.5 Kbar through thermodynamic modeling (i.e. ~ 19 and 13
170 km depth, respectively; Table 1; see also Álvarez-Valero and Waters, 2010 for
171 details of the pressure estimates). These depths define a precise section of the
172 volcanic dike. Hence, in order to track the bubbles evolution at depth along the dike,
173 we selected twelve dacites from within few centimetres of a hosted xenolith that
174 quenched simultaneously. In other words, these dacites host a variety of crustal
175 xenoliths, which served as samples of restite from various known depths (Álvarez-

176 Valero and Waters, 2010) under El Hoyazo volcano (Table 1; Fig. 1). They were
177 analyzed for the stable isotope ratios of hydrogen and oxygen (δD , $\delta^{18}O$). Four of
178 them were selected for X-ray micro-Computed Tomography (micro-CT) and
179 Scanning Electron Microscope (SEM) analysis (samples HY-14-2; HY-14-8; HY-14-
180 9; HY-14-10). Location coordinates are not useful in this studied case as the
181 xenoliths occur randomly distributed within the entire outcrop.

182

183 *2.1. Stable Isotope Analysis*

184 Oxygen isotope analyses of dacites and xenoliths, as well as single xenocrysts, were
185 done at the Servicio General de Analisis de Isotopos Estables, University of
186 Salamanca, Spain, on whole-rock powders by laser fluorination (Clayton and
187 Mayeda, 1963), employing a Synrad 25 W CO₂ laser (Sharp, 1990) and ClF₃ as
188 reagent (e.g., Borthwick and Harmon, 1982). Isotope ratios were measured on a VG-
189 Isotech SIRA-II dual-inlet mass spectrometer. Both internal and international
190 reference standards (NBS-28, NBS-30) were run to check accuracy and precision.
191 Results are reported in $\delta^{18}O$ notation relative to the Vienna Standard Mean Ocean
192 Water (V-SMOW) standard, using a $\delta^{18}O$ value of 9.6‰ for NBS-28 (quartz) for the
193 mass spectrometer calibration. Long-term reproducibility for repeated determination
194 of reference samples was better than $\pm 0.2\text{‰}$ (1σ).

195 D/H ratios were determined on a second SIRA-II mass spectrometer, on H₂ gas
196 obtained by reduction over hot depleted-U of the water released by induction heating
197 of samples, using a vacuum line (Bigeleisen et al. 1952), following the procedures
198 described by Godfrey (1962), with modifications (Jenkin, 1988). Samples were
199 loaded into degassed platinum crucibles that were placed in quartz reaction tubes and
200 heated under vacuum to 125°C overnight to remove any adsorbed H₂O. The yield of

201 evolved gas was used to determine the amount of structural water contained in the
202 sample. Results are reported in δD notation relative to the V-SMOW standard, using
203 a $\delta D = -66.7\text{‰}$ for NBS-30 (biotite) for the mass spectrometer calibration. Long-
204 term reproducibility for repeated determination of reference samples was better than
205 $\pm 2\text{‰}$ (1σ). The fractional extraction and purification of fluids (liquid, gas) from
206 glass inclusions was performed by means of a step-heating device.

207

208 *2.2. X-ray micro-Computed Tomography (micro-CT) and Scanning Electron* 209 *Microscope (SEM)*

210 Samples were imaged by using two different microtomographs; a ScanXmate-
211 D180RSS270 (Comscantecno Co., Ltd.) at Tohoku University, and an Argus
212 (SUINSA Medical Systems) at the University of Salamanca (USal), to characterize
213 the 3D morphology, distribution and volume of bubbles within dacites during ascent
214 from c. 19 to 13 km depth. The tomographic scans were performed at 150 kV and
215 110 μA . The source-to-detector distance was 670.094 mm, while the sample-to-
216 detector distance was 39.747 – 63.403 mm. Each sample was set on a rotation stage,
217 and transmission images were obtained for each 0.18° of rotation, to a total of 2000
218 images. The isotropic pixel edge sizes ranged between 7.53 and 12 μm .

219 Three-dimensional (3D) analyses were reconstructed from the transmission
220 images by using an original software package called “Slice” (Okumura et al., 2008).
221 A representative volume for each sample was selected in order to avoid exposed
222 surfaces and to remove fractures due to sample preparation (see Table 1).

223 The SEM images were segmented to separate bubbles from glass and crystals.
224 The segmentation consists in the choice of a threshold, in order to obtain binary
225 images where the vesicles are isolated from the dacite. For the binarization, we

226 translated CT values to 8 bit values and then made binary images using a threshold
227 value in 8 bit images, which allowed us to separate the vesicles from the dacite.

228 Next crystals in the magma need to be individualized from its glassy matrix. To
229 this end, a pre-segmentation step is usually required to increase the contrast between
230 different phases and to remove background noise, as both crystals and glassy matrix
231 in a sample may have similar X-ray attenuation coefficients (e.g., Zandomeneghi et
232 al., 2010; Voltolini et al., 2011). However, for most of the samples considered (HY-
233 14-10, HY-14-8 and HY-14-2), the contrast between crystals and their embedding
234 matrix was enough as not to require segmentation. Only sample HY-14-9 required
235 application of a bilateral filter (Tomasi and Manduchi, 1998) by using “ImageJ”
236 software (Abramoff et al., 2004), in order to better distinguish the crystals from their
237 glassy matrix, reducing the noise and potential artifacts while preserving edges and
238 the shape of the objects. This type of image processing separated bubbles, glass and
239 crystals, and allowed us to calculate bubble abundances and dacite porosity. We then
240 calculated the bubble number density ($BND = \text{number of bubbles} / \text{volume of glassy}$
241 groundmass) and bubble size distribution (BSD) (Cashman and Mangan, 1994;
242 Hurwitz and Navon, 1994; Mangan and Cashman, 1996; Gardner et al., 1999; Larsen
243 and Gardner, 2000; Blower et al., 2001, 2002; Toramaru, 2006; Bai et al., 2008;
244 Polacci et al., 2008, 2009; Baker et al., 2012; LaRue et al., 2013; Masotta et al., 2014)
245 (see Table 1b). BSD and BND can be used to discern between single or multiple
246 nucleation events during the magma ascent (Bai et al., 2008).

247 In addition, we utilized SEM visual analysis (secondary electron detector) at the
248 Centro de Láseres Pulsados (USal) to check for the presence of bubbles below the
249 detection limit of micro-CT. The SEM used an Extra High Tension of 10.94 kV,
250 Working Distance of 6-17 mm, and Irrigating Probe at 32 pA.

251

252 **3. Results: Stable Isotopes, micro-CT and SEM**

253 Oxygen and hydrogen isotopic ratios in dacite were $\delta^{18}\text{O}=15.4\pm0.2\text{‰}$ and $\delta\text{D}=-$
254 $87.1\pm3.3\text{‰}$ (1σ , $n=4$), respectively. In the xenoliths, whole rock measured values
255 were marginally lighter (although this effect may be attributed to one single sample)
256 and more variable, at $\delta^{18}\text{O} = 15.3 \pm 0.6\text{‰}$ and $\delta\text{D} = - 89.2 \pm 9.4\text{‰}$ (1σ , $n=4$). When
257 mineral separation could be achieved, oxygen isotopic ratios were always higher in
258 the xenoliths than in the host dacite (see Table 2; Fig. 2). The unusually high values
259 of the oxygen isotopic ratios measured are in line with values reported by Benito et
260 al. (1999) for the NVP in general and El Hoyazo in particular.

261

262 *3.1. Shallow samples (i.e., at 13 km depth)*

263 Of the samples available, HY-14-10 corresponds to the shallowest levels.
264 Measured values for the dacite were $\delta\text{D} = -89.2\text{‰}$ and $\delta^{18}\text{O} = 15.1\text{‰}$, while the
265 crustal xenoliths gave $\delta\text{D} = -85.5\text{‰}$ and $\delta^{18}\text{O} = 14.5\text{‰}$ (Table 2). The estimated
266 porosity in the dacites is 6.4% and its BND is 235 mm^{-3} (Table 1; Fig. 3). BSD
267 results indicate a significantly higher number of small size bubbles than those of
268 large size (Fig. 4). SEM images reveal local orientation trends of the bubbles in the
269 dacites (Fig. 3f), as well as irregular geometries (e.g., Fig. 5a, c).

270

271 *3.2. Deep samples (i.e., at 19 km depth)*

272 Sample HY-14-9 comes from the deepest part of the dacitic dyke (Fig. 3, a,b) δD
273 and $\delta^{18}\text{O}$ values are -88.5 and 15.1, respectively. The crustal xenoliths within this
274 sample gave $\delta\text{D} = -102.8\text{‰}$ and $\delta^{18}\text{O} = 15.7\text{‰}$ (Table 2). In the dacite, the porosity
275 is 1.7 % and its BND is 763 mm^{-3} (Table 1; Fig. 3). BSD estimates reveal that the

number of bubbles of small size is higher than those of larger size (Fig. 4). SEM images indicate that deep dacites often show elongated bubble geometries (e.g., Fig. 5e, g).

Simple first order estimates of bubble growth according to results shown in Fig. 4, i.e., how much does a bubble varies (in terms of size) as pressure drops, when other key parameters are in equilibrium (namely e.g., the amount of gas species degassing into the bubble from the surrounding silicate liquid per unit time, itself dependent on the initial volatile composition of the melt, its viscosity and surface tension, and kinetics) hints at an average bubble size enlargement of up to $13 \text{ mm}^3/\text{Kbar}$.

4. Discussion

4.1. Dacite depths: Coeval quenching at the contact with crustal xenoliths

Phase diagram modeling (Álvarez-Valero and Waters, 2010) and numerical simulations (Álvarez-Valero et al., 2015; Pla and Álvarez-Valero, 2015) for El Hoyazo volcano, indicate that the magma ascended from c. 19 to 13 km depth in the range of minutes to hours. Measured isotopic ratios are consistent with this scenario: average $\delta^{18}\text{O}$ and δD are indistinguishable from each other in dacites and their xenoliths (Fig. 2, Table 2). Equilibrium / disequilibrium relations established at depth were preserved during rapid ascent and quenching of the magma, that had no time to reset isotopic ratios, and inherited, therefore, $\delta^{18}\text{O}$ and δD values acquired at high temperature (i.e., c. 850°C) in the magma source region. If anything, there is hint for heavier δD values and lower water contents in shallower samples (HY-14-10) relative to deep ones (HY-14-9), present in both dacites and their xenoliths.

The δD - H_2O isotope systems allow us to determine whether or not gas is able to decouple from melt via open-system degassing (e.g., Taylor et al., 1983; Newman et

al., 1988; Castro et al., 2014). In other words, the isotopic change in a perfectly closed (and theoretical) system exhibits a linear trend by following the mass balance equation of Taylor et al. (1983) where δD decreases moderately for a relatively large decrease in the bulk H_2O in solution in the magma. Comparing the deepest (HY-14-9) with the shallowest (HY-14-10) sample, our results show that δD actually increases from bottom to top, at the time that H_2O contents become similar or slightly lower. If the El Hoyazo system behaved as an open system, different opposite situations would be expected (i.e., either open system water degassing or hydration should result in lower δD ; Taylor et al., 1983; Newman et al., 1988). This indicates that the magmatic system below El Hoyazo did not detect significant additions of external volatiles during magma ascent, nor experienced significant fluxes of volatiles from the surrounding crust.

Local microtextures in the dacites are similar to those in the xenoliths, involving similar mineral assemblages, namely biotite-hercynite-cordierite-melt and garnet-biotite-cordierite-melt (Fig. 1c, d, e). This suggests that the dacites were also partly quenched at those particular depths, i.e., 19 and 13 Km (see also Newmann et al., 1988). Xenolith textures were the result of an anatectic episode, that produced large amounts of melt that escaped from the pelitic country rocks (as described by Zeck, 1970), resulting in migmatization (and quenching) (Álvarez-Valero and Waters, 2010). This event was prior and independent of the transient melting event that rapidly erupted the dacites, transporting all xenolith types to the surface. Therefore we inferred the provenance depth of the dacites, and in turn their measured variation of porosity from 1.7 to 6.4 % between 19 and 13 km depth (Table 1), from the pressure at which their hosted metapelitic xenoliths equilibrated. We state that, otherwise, during the ascent from 19 to 13 km, the different magmatic and porosity

326 microtextures may show evidence of magma fragmentation (e.g. Eichelberger,
327 1995), or been homogenized during their ascent from 13 km depth to the surface.
328 This agrees with recent experimental constraints (e.g., Brugger and Hammer, 2010;
329 Cichy et al., 2011), which demonstrated that changes in mineral assemblage, crystal
330 abundance, variation in mineral compositions and texture are related to isothermal
331 magma ascent and decompression rates.

332 Therefore given that the exposed dacites are not homogenized in terms of porosity
333 in El Hoyazo, we speculate that rapid magma ascent may also have inhibited bubble
334 and crystal nucleation above 13km (e.g. Mangan and Sisson, 2000; Lloyd et al.,
335 2014). In other words, a rapid ascent above 13 km depth may favour a delay in the
336 nucleation process.

337

338 *4.2. Correlation between porosity and depth*

339 The volume of bubbles in an erupted volcanic rock may not necessarily reflect the
340 P-T conditions at which the bubbles formed in the dike, since processes such as
341 outgassing and late-stage crystallization may occur during magma ascent. These
342 processes can promote irregular bubble shapes between the microlites (Fig. 5a) due
343 to the collapse of the bubble. This collapse is promoted by the relaxation timescale
344 that depends on the surface tension of the bubble as well as the melt viscosity. It is
345 widely accepted that when a melt supersaturated with volatiles is depressurized,
346 small clusters of gas molecules nucleate and grow through volatile diffusion in the
347 melt (e.g., Toramaru, 1995; Proussevitch and Sahagian 1996, 1998). We focus on
348 vesicle growth at depth (Fig. 3), not on the initial nucleation process, which is
349 beyond the scope of this contribution. The growth of gas bubbles mainly requires
350 volatile diffusion from the melt into pre-existing bubbles, as growth is kinetically

351 favoured over nucleation when the degree of supersaturation is low enough as not to
352 achieve the necessary nucleation pressure. This also depends on the volatile
353 composition of the melt, its viscosity and surface tension (e.g., Mangan and Sisson,
354 2005; Masotta et al., 2014). Another important driver of bubble growth is the
355 mechanical expansion due to decreasing ambient pressure (e.g., Proussevitch et al.
356 1993; Sparks et al. 1994; Huber et al. 2014).

357 The consistency of groundmass and crystal volumes between 19 and 13 km depth
358 supports that little late-stage crystallization occurred (Table 1, Figs. 3 5), whereas the
359 local orientation trends of the bubbles (Fig. 3e) may be related to the stress
360 conditions at the dike's wall, that were not homogenized at the shallow vent depths.
361 Along the 6 km-dike dacites show similar water contents and microtextures,
362 indicating uniform conditions during magma ascent and degassing (Martel et al.,
363 2000). In addition, the relationship between porosity (Table 1) and dissolved water
364 match the quantitative trends in the multicomponent liquid–gas equilibrium in a
365 silicic system of Papale et al. (2006). Outgassing during rapid feeder dike
366 emplacement is nearly constant at low porosity (Takeuchi et al., 2009), likely due to
367 the opening and healing of fractures on short timescales, followed by densification of
368 the dome dacites, along the shallow parts of the conduits (e.g., Okumura et al.,
369 2010).

370 The presence of crystals (c. 25 % of mainly plagioclase, biotite and hornblende;
371 Figs. 3, 5) may have reduced the activation energy for the nucleation of bubbles, and
372 induced nucleation at 13 km that increased the number of bubbles (confirmed by
373 BND and BSD, Table 1 and Fig. 4). Since crystals are normally present at magma
374 storage conditions in natural magmatic systems, heterogeneous nucleation is often
375 expected (e.g., Mangan et al. 2004), especially in the studied case, where the

376 interstitial melt contains crystals and is not supersaturated (e.g., Hurwitz and Navon
377 1994). Furthermore, the occurrence of bubbles is not systematically related to any
378 particular crystal distribution or type (Fig. 3). Therefore, bubble growth within the
379 El Hoyazo volcano was related to heterogeneous nucleation.

380 Our results indicate that (i) the porosity in the dacites increases upward from 1.7
381 to 6.4% (i.e., from 19 to 13 km depth, respectively), thus yielding a porosity opening
382 rate of 0.78 % /km (see also Table 1), and an average bubble growth of up to 13
383 mm³/Kbar (see also Fig. 4); (ii) BND values of 763 and 235 mm⁻³ for the deep and
384 shallow samples, respectively, are in line with both bubble nucleation clustering
385 around grain boundaries (microlite, xenocryst, xenolith) at higher depths (Figs. 3, 4,
386 5), as well as higher coalescence at 13 km than at 19 km depth. Deep bubble
387 formation contributes to crustal overpressure (e.g., Malfait et al., 2014) and to the
388 lubrication of crystal-rich magmas (e.g., Pistone et al., 2013).

389 Therefore, the presence of small amount of bubbles between 19 and 13 km allows
390 a better understanding of the rapid magma ascent rates obtained from numerical
391 simulations by Álvarez-Valero et al. (2015) and Pla and Álvarez-Valero (2015).
392 Hence, deep bubble formation must be included in evaluations of silicic magma
393 eruptivity (e.g., Takeuchi, 2004; Gottsmann et al., 2009).

394

395 **CONCLUDING REMARKS**

396 - Bubble nucleation and volume in the deep magma dike increase during ascent-
397 driven decompression at a lower rate and to a lower extent than has been described
398 for shallow vents.

399 - Rapidly ascending magma at El Hoyazo volcano experiences negligible late-
400 stage crystallization and bubble nucleation at depths above 13 km below the surface.

- The present porosity rate of 0.78 %/km may be used in silicic subvolcanic systems as an approximation in models and experiments for a better understanding of deep bubble formation, and if necessary, to accurately evaluate magma eruptibility.

- Silicic lava domes hosting crustal xenoliths are extraordinary scenarios to study different bubble snapshots along their evolution during ascent in a deep dike. We provide evidence of how bubbles may behave within the deep silicic dike beneath El Hoyazo. Our results can be used as a proxy for crystallization and bubble formation beneath contemporary volcanoes, and help improving the general knowledge of the vesiculation process at varying depths beneath active volcanoes.

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Figure Captions

Figure 1. (a) Location of El Hoyazo volcano within the NVP and map of the Betic
Cordillera and Rif; (b) Field aspect of a spinel-cordierite-melt (Spl-Crd-M) xenolith
type in the dacitic lava of this volcano. Plane-polarized light microscopy views of:
(c) a xenolith at c. 19 km depth, in contact with its host dacite, which show local
textures of mineral equilibria (e.g. biotite-hercynite-melt) that are also found in the
xenolith; (d) a dacite that show the same chemical reactions and textures as in (c) far
from the contact with the xenolith; (e) a xenolith at c. 13 km depth, in contact with
its host dacite, which show a typical texture in the xenoliths such as garnet xenocryst
with a cordierite-plagioclase-melt corona. Microstructures in the xenoliths reveal a
first stage of migmatization overprinted by a sequence of melt-bearing reaction
microstructures (see the text for details). The microstructural features in the dacites
enhance the possibility of partial quenching at depth.

Figure 2. Stable isotopes analysis of El Hoyazo dacitic magma and crustal xenoliths.
Dacites and their included xenoliths show negligible $\delta^{18}\text{O}$ fractionation between
them, which is typical for rapid subvolcanic decompression occurring at high

temperatures (e.g., O'Neill, 1986). The δD values differentiate the water provenances of dacites and crustal xenoliths.

Figure 3. (a) Scheme (not to scale) of the subvolcanic system at El Hoyazo volcano. Three-dimensional view of samples HY-14-9 (b) and HY-14-10 (c), which were in contact with xenoliths equilibrated at c. 19 and 13 km depth, respectively. Glass and minerals were suppressed for clarity. Bubbles are shaded dark grey (the clearer grey, the higher bubbles concentration). (d) 2D example view of a single microtomographic slice to highlight –in white (e)– the crystals amount vs. groundmass vs. bubbles. Grt: garnet xenocryst; Pl: plagioclase; Hb: hornblende; Bt: biotite. (f) view of a single tomographic slice of sample HY-14-10 to highlight the bubbles orientation within the dacite.

Figure 4. Bubble sizes distribution (BSD) diagram in the dacites HY-14-9 and HY-14-10 (see also Figs. 3, 5 and Table 1). BSD shows that at higher depth the number of small size bubbles is higher than at lower depth. In the same way, BSD also reveals that at shallower depth the number of bubbles is lower but of larger size than at higher depth.

Figure 5. SEM-SEI images of the same dacites of Figure 3 revealing the existence of tiny bubbles and fractures that are below the detection limit of the micro-CT, as well as the high groundmass/microlites ratio that indicates a minor influence of a potential late-stage vesiculation process (see Fig. 3). Bubbles in lava HY-14-10 (shallowest one) show more irregular shapes and larger sizes (e.g., a, c) than in lava HY-14-9 (deepest dacite) with smaller bubbles and more elongated geometries (e.g., e, g). (h) Current bubble after an exsolved fluid inclusion in a biotite crystal. The groundmass and crystals volumes are fairly constant along the vertical studied 6 km section of the

643 dike. The bubbles sizes below the detection limit of the micro-CT indicate that our
644 measurements of the total volumes are minimum.

645 **Table 1. (a)** Representative volumes used for porosities estimation of the dacites
646 along c. 19 and 13 km depth within the dike. **(b)** Microtextural parameters utilized
647 for quantitative analysis of BND and BSD calculations. The bubbles sizes below the
648 detection limit of the micro-CT (Fig. 5) indicate that our measurements of the total
649 volumes are minimum. (*) Álvarez-Valero and Waters, 2010; (**) Álvarez-Valero
650 and Kriegsman, 2007.

651 **Table 2.** Values of the stable isotopes analysis ($\delta^{18}\text{O}$, δD) of El Hoyazo dacitic
652 magma and crustal xenoliths.

653

Figure1

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Figure 1

Figure3

Figure4
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Figure5
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TABLE 1

Lava Sample	Isotropic pixel edge sizes - (µm)	Total imaged volume (mm3)	Total measured volume (mm3)	BUBBLES Porosity (%)	BUBBLES Volume (mm3)	% H2O (± 0.1) calculated (dacite)	% H2O bulk dacite (estimated from XRF)	Xenolith peak P (kbar, 2σ) metamorphism
HY-14-10	9.96	746	746	6.4	35.0	2.3		4.7 ± 0.3 (c. 13 km depth) *
HY-14-8	12.00	966	966	3.1	31.7	up to 4.1	3.7 - 4	5.7 ± 0.3 (5.4 ~ c. 16.2 km depth) *
HY-14-2	12.00	1084	1084	1.7	18.2			5.7 ± 0.3 (6.0 ~ c. 18 km depth) *
HY-14-9	7.53	322	322	1.7	4.76			6.2 ± 0.8 (c. 19 km depth) **

Sample	Analized volume (pixel)	Analized volume (mm3)	Nº Bubbles	GROUNDMASS Volume (mm3)	CRYSTALS Volume (mm3)	CRYSTALLINIT Y (%)	BND (mm-3)
HY-14-10	792x522x701	286	43687	185.68	74.1	28.5	235
HY-14-9	870x1100x700	286	159924	209.64	71.6	25.4	763

Sample	Type	$\delta^{18}\text{O}$ (± 0.15)	δD (± 1)
HY-14-10	dacite (bulk)	15.6	-83.6
HY-14-8	dacite (bulk)	15.3	-91.0
HY-14-2	dacite (bulk)	15.5	-85.2
HY-14-9	dacite (bulk)	15.1	-88.5
HY-14-10	xenolith (bulk)	14.5	-85.5
HY-14-8	xenolith (bulk)	15.6	-81.5
HY-14-2	xenolith (bulk)	15.5	-86.8
HY-14-9	xenolith (bulk)	15.7	-102.8
HY-14-8	cordierite in xenolith	15.5	-78.2
HY-14-9	cordierite in xenolith	15.1	-
HY-14-9	garnet in xenolith	14.0	-
HY-14-9	plagioclase in xenolith	15.1	-
HY-14-7	garnet in dacite	13.5	-80.9
HY-14-8	garnet in dacite	13.6	-
HY-14-9	garnet in dacite	13.7	-
HY-14-9	quartz in dacite	12.5	-