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# 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, stableisotope 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
33	in porosity from ~1.7 to 6.4 % from ~19 to 13 km depth, at nearly constant
34	groundmass and crystal volumes. This result provides additional constraints for
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## 51 **1. Introduction**

52 Processes occurring in the conduit between a magma chamber and the surface may trigger an eruption or alter the characteristics of one already in progress (e.g. 53 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 (from c. 4 to 20 km) require further investigation (e.g., Álvarez-Valero et al., 2015; 64 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 largelly calc-alkaline dacites rich in K<sub>2</sub>O, with volatile contents from 2 to 4 93 wt% (mainly H<sub>2</sub>O, CO<sub>2</sub>) (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

m crater radius, and is overlain by Miocene reef carbonates. It is a small circular
outcrop of c. 0.7 km<sup>2</sup>, which dacites include up to 15% in volume of crustal material
(Zeck, 1970). Its atoll geomorphology shows an inner depressed part of mainly
dacitic material that corresponds to the old volcanic cone, whereas the top relief is
composed by the reef carbonates formed onto the volcanics, which host the crustal
xenoliths. Crustal xenoliths and their host dacites are ramdonly collected within the
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 and compositionally (see also Álvarez-Valero and Kriegsman, 2007; Álvarez-Valero 120 121 and Waters, 2010). The shallowest xenoliths are of the Spl-Crd-M type. The garnet is 122 surrounded by coronas, and the overall parageniesis is Spl-Crd-Pl-Kfs-M. 123 The coronas mimic the outline of the previous garnet, whose breakdown reaction has been described in detail by Álvarez-Valero et al. (2007). Sillimanite occurs as 124 125 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 CO <sub>2</sub> -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

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

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

and Kriegsman, 2007). A second sequence of melt-bearing reaction microstructures,

developed in a transiently heated wall-rock profile adjacent to the magma conduit,

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**

168The 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

- analyzed for the stable isotope ratios of hydrogen and oxygen ( $\delta D$ ,  $\delta^{18}O$ ). Four of
- 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

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 CO<sub>2</sub> laser (Sharp, 1990) and ClF<sub>3</sub> 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  $\pm 0.2\%$  (1 $\sigma$ ).

195 D/H ratios were determined on a second SIRA-II mass spectrometer, on H<sub>2</sub> 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

- 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  $125^{\circ}$ C overnight to remove any adsorbed H<sub>2</sub>O. The yield of

201 evolved gas was used to determine the amount of structural water contained in the

sample. Results are reported in  $\delta D$  notation relative to the V-SMOW standard, using

 $\Delta \delta D = -66.7$  % for NBS-30 (biotite) for the mass spectrometer calibration. Long-

term reproducibility for repeated determination of reference samples was better than

 $\pm 2 \%$  (1 $\sigma$ ). 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

the 3D morphology, distribution and volume of bubbles within dacites during ascent

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,

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  $\mu$ m.

219 Three-dimensional (3D) analyses were reconstructed from the transmission

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

surfaces and to remove fractures due to sample preparation (see Table 1).

The SEM images were segmented to separate bubbles from glass and crystals.

The segmentation consists in the choice of a threshold, in order to obtain binary

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 = number of bubbles / 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 detection limit of micro-CT. The SEM used an Extra High Tension of 10.94 kV, 249 250 Working Distance of 6-17 mm, and Irrigating Probe at 32 pA.

251

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

Oxygen and hydrogen isotopic ratios in dacite were  $\delta^{18}O=15.4\pm0.2\%$  and  $\delta D=-$ 253 254  $87.1\pm3.3\%$  (1 $\sigma$ , n=4), respectively. In the xenoliths, whole rock measured values 255 were marginally lighter (although this effect may be attributed to one single sample) and more variable, at  $\delta^{18}O = 15.3 \pm 0.6\%$  and  $\delta D = -89.2 \pm 9.4\%$  (1 $\sigma$ , n=4). When 256 257 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. Measured values for the dacite were  $\delta D = -89.2\%$  and  $\delta^{18}O = 15.1\%$ , while the 264 crustal xenoliths gave  $\delta D = -85.5\%$  and  $\delta^{18}O = 14.5\%$  (Table 2). The estimated 265 porosity in the dacites is 6.4% and its BND is 235 mm<sup>-3</sup> (Table 1; Fig. 3). BSD 266

results indicate a significantly higher number of small size bubbles than those of

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

and  $\delta^{18}$ O values are -88.5 and 15.1, respectively. The crustal xenoliths within this

sample gave  $\delta D = -102.8\%$  and  $\delta^{18}O = 15.7\%$  (Table 2). In the dacite, the porosity

is 1.7 % and its BND is 763 mm<sup>-3</sup> (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 mm<sup>3</sup>/Kbar.

285

## 286 4. Discussion

287 4.1. Dacite depths: Coeval quenching at the contact with crustal xenoliths

288 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:

292 average  $\delta^{18}$ O and  $\delta$ D are indistinguishable from each other in dacites and their

293 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,  $\delta^{18}$ O and  $\delta$ D values acquired at high

temperature (i.e., c. 850°C) in the magma source region. If anything, there is hint for

297 heavier  $\delta D$  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.

299 The  $\delta D$ -H<sub>2</sub>O 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

301 al., 1988; Castro et al., 2014). In other words, the isotopic change in a perfectly 302 closed (and theoretical) system exhibits a linear trend by following the mass balance 303 equation of Taylor et al. (1983) where  $\delta D$  decreases moderately for a relatively large 304 decrease in the bulk H<sub>2</sub>O in solution in the magma. Comparing the deepest (HY-14-305 9) with the shallowest (HY-14-10) sample, our results show that  $\delta D$  actually 306 increases from bottom to top, at the time that H<sub>2</sub>O contents become similar or 307 slightly lower. If the El Hoyazo system behaved as an open system, different 308 opposite situations would be expected (i.e., either open system water degassing or 309 hydration should result in lower  $\delta D$ ; Taylor et al., 1983; Newman et al., 1988). This 310 indicates that the magmatic system below El Hoyazo did not detect significant 311 additions of external volatiles during magma ascent, nor experienced significant 312 fluxes of volatiles from the surrounding crust. 313 Local microtextures in the dacites are similar to those in the xenoliths, involving 314 similar mineral assemblages, namely biotite-hercynite-cordierite-melt and garnet-315 biotite-cordierite-melt (Fig. 1c, d, e). This suggests that the dacites were also partly 316 quenched at those particular depths, i.e., 19 and 13 Km (see also Newmann et al., 317 1988). Xenolith textures were the result of an anatectic episode, that produced large 318 amounts of melt that escaped from the pelitic country rocks (as described by Zeck, 319 1970), resulting in migmatization (and quenching) (Álvarez-Valero and Waters, 320 2010). This event was prior and independent of the transient melting event that 321 rapidly erupted the dacites, transporting all xenolith types to the surface. Therefore 322 we inferred the provenance depth of the dacites, and in turn their measured variation 323 of porosity from 1.7 to 6.4 % between 19 and 13 km depth (Table 1), from the 324 pressure at which their hosted metapelitic xenoliths equilibrated. We state that, 325 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,

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

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

in El Hoyazo, we speculate that rapid magma ascent may also have inhibited bubble

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 the336 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 P-T conditions at which the bubbles formed in the dike, since processes such as 340 341 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

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).

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

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

the opening and healing of fractures on short timescales, followed by densification of

the dome dacites, along the shallow parts of the conduits (e.g., Okumura et al.,

369 2010).

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

induced nucleation at 13 km that increased the number of bubbles (confirmed by

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

area 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

1994). Furthermore, the occurrence of bubbles is not systematically related to any

378 particular crystal distribution or type (Fig. 3). Therefore, bubbles 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
- to 6.4% (i.e., from 19 to 13 km depth, respectively), thus yielding a porosity opening
- rate of 0.78 % /km (see also Table 1), and an average bubble growth of up to 13
- $383 \text{ mm}^3/\text{Kbar}$  (see also Fig. 4); (ii) BND values of 763 and 235  $\text{mm}^{-3}$  for the deep and

384 shallow samples, respectively, are in line with both bubble nucleation clustering

- around grain boundaries (microlite, xenocryst, xenolith) at higher depths (Figs. 3, 4,
- 5), as well as higher coalescense at 13 km than at 19 km depth. Deep bubble
- 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
- 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).
- 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

Bubble nucleation and volume in the deep magma dike increase during ascentdriven decompression at a lower rate and to a lower extent than has been described
for shallow vents.

- Rapidly ascending magma at El Hoyazo volcano experiences negligible late-

400 stage crystallization and bubble nucleation at depths above 13 km below the surface.

401 - The present porosity rate of 0.78 %/km may be used in silicic subvolcanic 402 systems as an approximation in models and experiments for a better understanding of 403 deep bubble formation, and if necessary, to accurately evaluate magma eruptibility. 404 - Silicic lava domes hosting crustal xenoliths are extraordinary scenarios to study 405 different bubble snapshots along their evolution during ascent in a deep dike. We 406 provide evidence of how bubbles may behave within the deep silicic dike beneath El 407 Hoyazo. Our results can be used as a proxy for crystallization and bubble formation 408 beneath contemporary volcanoes, and help improving the general knowledge of the vesiculation process at varying depths beneath active volcanoes. 409

410

411

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603	Figure Captions
604	Figure 1. (a) Location of El Hoyazo volcano within the NVP and map of the Betic
605	Cordillera and Rif; (b) Field aspect of a spinel-cordierite-melt (Spl-Crd-M) xenolith
606	type in the dacitic lava of this volcano. Plane-polarized light microscopy views of:
607	(c) a xenolith at c. 19 km depth, in contact with its host dacite, which show local
608	textures of mineral equilibria (e.g. biotite-hercynite-melt) that are also found in the
609	xenolith; (d) a dacite that show the same chemical reactions and textures as in (c) far
610	from the contact with the xenolith; (e) a xenolith at c. 13 km depth, in contact with
611	its host dacite, which show a typical texture in the xenoliths such as garnet xenocryst
612	with a cordierite-plagioclase-melt corona. Microstructures in the xenoliths reveal a
613	first stage of migmatization overprinted by a sequence of melt-bearing reaction
614	microstructures (see the text for details). The microstructural features in the dacites
615	enhance the possibility of partial quenching at depth.
616	Figure 2. Stable isotopes analysis of El Hoyazo dacitic magma and crustal xenoliths.
617	Dacites and their included xenoliths show negligible $\delta^{18}$ O fractionation between
618	them, which is typical for rapid subvolcanic decompression occurring at high

Zandomeneghi, D., Voltolini, M., Mancini, L., Brun, F., Dreossi, D., Polacci, M.,

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temperatures (e.g., O'Neill, 1986). The  $\delta$  D values differentiate the water

620 provenances of dacites and crustal xenoliths.

621 Figure 3. (a) Scheme (not to scale) of the subvolcanic system at El Hoyazo volcano.

622 Three-dimensional view of samples HY-14-9 (b) and HY-14-10 (c), which were in

623 contact with xenoliths equilibrated at c. 19 and 13 km depth, respectively. Glass and

624 minerals were suppressed for clarity. Bubbles are shaded dark grey (the clearer grey,

625 the higher bubbles concentration). (d) 2D example view of a single

626 microtomographic slice to highlight –in white (e)– the crystals amount vs.

627 groundmass vs. bubbles. Grt: garnet xenocryst; Pl: plagiclase; Hb: hornblende; Bt:

biotite. (f) view of a single tomographic slice of sample HY-14-10 to highlight the

629 bubbles orientation within the dacite.

630 Figure 4. Bubble sizes distribution (BSD) diagram in the dacites HY-14-9 and HY-

631 14-10 (see also Figs. 3, 5 and Table 1). BSD shows that at higher depth the number

632 of small size bubbles is higher than at lower depth. In the same way, BSD also

633 reveals that at shallower depth the number of bubbles is lower but of larger size than

634 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

637 as the high groundmass/microlites ratio that indicates a minor influence of a potential

638 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

640 (deepest dacite) with smaller bubbles and more elongated geometries (e.g., e, g). (h)

- 641 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
- 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
- and Kriegsman, 2007.
- **Table 2.** Values of the stable isotopes analysis ( $\delta^{18}$ O,  $\delta$ D) of El Hoyazo dacitic
- 652 magma and crustal xenoliths.







Figure4 Click here to download high resolution image





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		$4.7 \pm 0.3$ (c. 13 km depth) *
HY-14-8	12.00	966	966	3.1	31.7		3.7 - 4	$5.7 \pm 0.3$ (5.4 ~ c. 16.2 km depth) *
HY-14-2	12.00	1084	1084	1.7	18.2	up to 4.1		$5.7 \pm 0.3$ (6.0 ~ c. 18 km depth) *
HY-14-9	7.53	322	322	1.7	4.76			$6.2 \pm 0.8$ (c. 19 km depth) **
		-	-					

Sampla	Analized volume	Analized volume		GROUNDMASS	CRYSTALS CRYSTALLINIT		DND (mm 2)
Sample	(pixel)	(mm3)	IN Bubbles	Volume (mm3)	Volume (mm3)	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	Туре	$\boldsymbol{\delta^{18}O}~(\pm~0.15)$	<b>δD</b> (± 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	-