Integrating field, textural and geochemical monitoring to track eruption triggers and dynamics: a case-study from Piton de la Fournaise

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19 Abstract

20 The 2014 eruption at Piton de la Fournaise (PdF), La Réunion, which occurred after 41 months of quiescence, began with surprisingly little precursory activity, and was one of the 21 22 smallest so far observed at PdF in terms of duration (less than 2 days) and volume (less than $0.4 \times 10^6 \text{ m}^3$). The pyroclastic material was composed of golden basaltic pumice along with 23 fluidal, spiny-iridescent and spiny-opaque basaltic scoria. Density analyses performed on 200 24 lapilli reveal that while the spiny-opaque clasts are the least dense (1600 kg/m³) and most 25 crystalline (55 vol. %), the golden pumices are the least dense (400 kg/m³) and crystalline (8 26 vol. %). The connectivity data indicate that the fluidal and golden (Hawaiian-like) clasts have 27 more isolated vesicles (up to 40 vol. %) than the spiny (Strombolian-like) clasts (0-5 vol. %). 28 These textural variations are linked to primary pre-eruptive magma storage conditions. The 29 golden and fluidal fragments track the hotter portion of the melt, in contrast to the spiny 30 fragments and lava that mirror the cooler portion of the shallow reservoir. Exponential decay 31 of the magma ascent and output rates through time revealed depressurization of the source 32

during which a stratified storage system was progressively tapped. Increasing syn-eruptive 33 degassing and melt-gas decoupling led to a decrease in the explosive intensity from early 34 fountaining to Strombolian activity. The geochemical results confirm the absence of new 35 input of hot magma into the 2014 reservoir and confirm the emission of a single, shallow, 36 differentiated magma source, possibly related to residual magma from the November 2009 37 eruption. Fast volatile exsolution and crystal-melt separation (second boiling) were triggered 38 by deep pre-eruptive magma transfer and stress field change. Our study highlights the 39 40 possibility that shallow magma pockets can be quickly reactivated by deep processes without 41 mass or energy (heat) transfer and produce hazardous eruptions with only short term elusive 42 precursors.

Key words: Piton de la Fournaise, Hawaiian activity, Strombolian activity, shallow reservoire, texture,
petrology, geochemistry

45 1. Introduction

46 A detailed characterization and understanding of eruptive dynamics and of processes driving and modulating volcano unrest is crucial in monitoring active volcanoes and fundamental for 47 forecasting volcanic eruptions (Sparks, 2003). Many studies suggest that eruptive phenomena 48 are strongly dependent on the physico-chemical properties of ascending magma in the conduit 49 (e.g., temperature, viscosity, porosity, and permeability) (e.g. Sparks, 1978; Rust and 50 51 Cashman, 2011; Gonnermann and Manga, 2013; Polacci et al., 2014). Integrating petrographic, chemical and textural data can thus provide critical information to constrain 52 both the pre-eruptive storage conditions, and the processes related to magma ascent, degassing 53 and cooling (e.g., reference in Table 1 in Gurioli et al., 2015). This multidisciplinary approach 54 55 is of even greater importance in the monitoring of volcanoes which emit relatively uniform magma compositions over time, like basaltic volcanoes (e.g. Di Muro et al., 2014; Gurioli et 56 al., 2015; Coppola et al., 2017). As a result, monitoring of textures, and petrochemical 57 properties of lava fragments and pyroclasts is now routinely carried out on a daily basis at 58 59 active volcanoes such as Kilauea, Etna, and Stromboli (e.g., Taddeucci et al., 2002; Thornber et al., 2003; Polacci et al., 2006; Swanson et al., 2009; Colo' et al., 2010; Houghton et al., 60 61 2011; 2013; 2016; Carey et al., 2012; 2013; Lautze et al., 2012; Andronico et al., 2013a; b; 2014; Corsaro and and Miraglia, 2014; Di Muro et al., 2014; Gurioli et al.; 2014; Eychenne et 62 al., 2015; Leduc et al., 2015; Kahl et al., 2015). In the past, time series of petrographic and 63 geochemical data have been measured for PdF basalts and particularly for effusive products. 64

The aim of these datasets was to constrain the spatial and temporal evolution of magma for 65 66 one of the most active basaltic volcanoes of the world (e.g. Albarède et al., 1997; Vlastélic et al., 2005; 2007, 2009; Boivin and Bachèlery, 2009; Peltier et al., 2009; Schiano et al., 2012; 67 Lénat et al., 2012; Di Muro et al., 2014; 2015; Vlastèlic and Pietruszka, 2016). However, this 68 type of approach has seldom been coupled with detailed textural studies at PdF and instead 69 70 has mostly focused on crystal textures and crystal size distribution (Welsch et al., 2009; 2013; Di Muro et al., 2014; 2015). Moreover, only sporadic data exist on the textures of pyroclasts 71 72 ejected by the eruptions at PdF (Villemant et al., 2009; Famin et al., 2009; Welsch et al., 2009; 73 2013; Michon et al., 2013; Vlastélic et al., 2013; Di Muro et al., 2015; Morandi et al., 2016; 74 Ort et al., 2016).

75 Within this paper, we present a multidisciplinary textural, chemical and petrological approach to quantify and understand the short-lived 2014 PdF eruption. This approach 76 77 combines detailed study of the pyroclastic deposit (grain size and componentry) with bulk texture analysis (density, vesicularity, connectivity, permeability, morphology, vesicle 78 79 distribution and crystal content) and a petro-chemical study (bulk rock, glass, minerals, melt inclusions) of the same clasts. This integrated approach has now been formalized within the 80 81 French National Observation Service for Volcanology (SNOV), as routine observational (DynVolc), Dynamics of Volcanoes, (http://wwwobs.univ-82 systems bpclermont.fr/SO/televolc/dynvolc/) and GazVolc, Observation des gaz volcaniques, 83 (http://wwwobs.univ-bpclermont.fr/SO/televolc/gazvolc) to provide data for the on-going 84 activity at PdF (Harris et al., 2017). 85

In spite of being the first of a series of eruptions, the June 2014 event was preceded by 86 87 only weak inflation and by a rapid increase in number of shallow (< 2 km below volcano summit) volcano tectonic earthquakes that happened only 11 days before the eruption (Peltier 88 et al., 2016). The eruptive event was dominantly effusive, lasted only 20 hours and emitted a 89 very small volume of magma (ca. $0.4 \times 10^6 \text{ m}^3$, Peltier et al., 2016), which makes this event 90 one of the smallest, in terms of duration and volume, observed at PdF up to now. In addition, 91 92 the eruption started during the night and very little direct observation exists for the first few hours of the activity, when the lava effusion was associated with very weak fountaining 93 activity and Strombolian explosions. 94

This eruption occurred just outside the southern border of the summit Dolomieu caldera, at the top of the central cone of PdF (Fig. 1). This is a high risk sector because of the high number of tourists. Identification of precursors of this kind of activity represents an important challenge for monitoring systems (Bachélery et al., 2016).

99 Therefore this eruption represents an ideal context to apply our multidisciplinary 100 approach, with the aim of addressing the following key questions:

why was such a small volume of magma erupted instead of remaining 101 (i) endogenic? 102

what caused the rapid trigger and the sudden end to this small volume 103 (ii) eruption? 104

which was the source of the eruption (shallow versus deep, single versus 105 (iii) 106 multiple small magma batches)?

what was the time and space evolution of the eruptive event?

(iv) 107

(v)

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what was the ascent and degassing history of the magma?

109 Furthermore, this eruption provides an exceptional opportunity to study processes leading to the transition from mild Hawaiian (<20 m high fountains, following the nomenclature 110 111 proposed by Stovall et al., 2011) to Strombolian activity (<10 m high explosions), whose products are little modified by post-fragmentation processes because of the very low intensity 112 113 of the activity.

114 2 The 2014 activity

2.1 Precursory activity 115

116 The 20 June 2014 summit eruption represents the first eruption at PdF after 41 months of quiescence. The last eruption had been on 9 December 2010, with a shallow (above sea level) 117 intrusion on 2 February 2011 (Roult et al., 2012). From 2011, the deformation at PdF was 118 constant with two distinct types of behaviour: (i) a summit contraction of a few centimetres 119 every year (Fig. 1d), and (ii) a preferential displacement of the east flank at a rate of 1-3 120 centimetres per year (Brenguier et al., 2012; Staudacher and Peltier, 2015). The background 121 122 microseismicity was very low (< 5 shallow events/day below volcano summit) and lowtemperature summit intracaldera fumaroles emitted very little sulphur (H₂S or SO₂) and 123 carbon (CO₂) (Di Muro et al., 2016). After 41 months of rest, a new intense cycle of activity 124 (June 2014, February 2015, May 2015, July 2015, August-October 2015; May 2016; 125 September 2016; January 2017 and July 2017) began with surprisingly little and ambiguous 126 precursory activity. 127

The 2014 summit eruption started during the night of June 20/21, at 21h35 GMT 128 (0h35 local time) and ended on June 21 at 17h09 GMT (21h09 local time), after less than 20 129 130 hours of dominantly effusive activity. The volcano reawakening was preceded, in March and

April 2014, by deep (15-20 km below sea level) eccentric seismicity and increase in soil CO₂ 131 flux below the western volcano flank, 15 km NW of the volcano summit (Liuzzo et al., 2015; 132 Boudoire et al., 2017). Background micro-seismicity and inflation of the central cone 133 increased progressively starting on 9 June 2014. Weak inflation recorded on both distal and 134 summit baselines (Fig. 1d) suggest that deep (below sea level) magma up-rise was 135 pressurizing the shallow (above sea level) magma storage system (Peltier et al., 2016). On 136 June 13, 17 and 20, three shallow (hypocentres located above sea level) intense seismic crises 137 occurred below the summit Dolomieu caldera (Fig. 1), with hundreds of events located in a 138 narrow depth range between 1100 and 2100 metres below the volcano summit. These seismic 139 crises consisted of swarms of low magnitude (M: 1-2) volcano tectonic events which 140 141 increased in number from the first to the third crisis. On June 20, seismicity increased progressively and a final seismic crisis started at 20h20 GMT, only 75 minutes before the 142 143 eruption. This last seismic crisis was coupled with acceleration in the deformation of the summit area, which began only 60 minutes before the eruption. Interestingly, only slight 144 145 inflation of the central cone (< 2 cm of dilatation) was detected 11 days before the 2014 eruption with a maximum of 1 cm and 1.6 cm enlargement at the summit and the base of the 146 cone, respectively (Peltier et al., 2016 and Fig. 1d). A moderate increase in CO₂ and H₂S 147 emissions from summit intracaldera fumaroles was detected starting on June 2, but only very 148 minor SO₂ emissions occurred before the eruption (mostly on June 7 and 15, unpublished 149 data). Therefore, the acceleration in both geophysical and geochemical parameters was mostly 150 related to the late phase of dyke propagation towards the surface just before the eruption. 151 Following the end of the June 20-21 eruption, a long-term continuous inflation of the edifice 152 153 began, at a moderate rate, and mostly at the base of the volcano. More than one year after this first eruption, the long-term deformation trends showed that the 2014 eruption marked a kink 154 between the deflation trend which followed the caldera-forming 2007 eruption (Staudacher et 155 al., 2009) and the currently ongoing continuous inflation trend (Fig. 1d, and Peltier et al., 156 157 2016; Coppola et al., 2017).

158 2.2 Chronology of the events

We reconstructed the chronology of the events by combining a distribution map of the fissures, pyroclastic deposits and lava flows (Fig. 1) with a review of available images and videos extracted from the observatory data base, the local newspapers, and web sites (Fig. 2). The 2014 eruption occurred at the summit and on the SE slopes of the Dolomieu Caldera (Figs. 1a, 1b and 1c) and evolved quickly and continuously over 20 hours. The full set of fractures opened during a short period of time (minutes) and emitted short (<1.7 km long)
lava flows (Fig. 1 and Figs. 2c and 2d). Feeding vents were scattered along a 0.6 km long
fissure set (Fig. 1a) and produced very weak (low) Hawaiian to Strombolian activity (Fig. 2).

Fissures opened from west to east, initially sub-parallel to the southern border of 167 Dolomieu caldera and then propagated at lower altitude (Fig. 1). The summit part of the 168 fractures (ca. 2500 m asl, Western Fracture, WF in Fig. 1) emitted only small volumes of lava 169 and pyroclasts. This part of the fracture set was active only during the first few hours of the 170 eruption, at night. The eastern part of the fractures (Upper Fracture, UF in Fig. 1) descended 171 172 to lower altitude (between 2400 and 2300 m asl, Middle Fracture, Fig. 1) along the SE flank of the summit cone and emitted most of the erupted volume. As often observed in PdF 173 174 eruptions, the activity progressively focused on a narrow portion of the fractures at low altitude and finally on a single vent located at the lower tip of the fracture system (Main Vent, 175 176 at 2336 m asl, MV in Figs. 1, 2). The first in situ observations in the morning of June 21 (ca. 04h00 GMT) showed that weak Strombolian activity (Figs. 2a and 2b) was focused on a 177 178 narrow segment of the lower fractures and that a'a lavas had already attained the elevation of 1983 m asl (0.2 km before maximum runout, Fig. 2c). A small, weak gas plume was also 179 180 blowing northwards. A single sample of partially molten lava was collected from the still active lava front and partially water quenched (REU140621-1, Table S1, Fig. 2d). During 181 most of June 21, the activity consisted of lava effusion in three parallel lava streams (Fig. 2c) 182 merging in a single lava flow (Fig. 2e) and mild-weak "Strombolian" explosions at several 183 closely spaced spots along the lower part of the feeding fracture. At 13.00 (GMT), only weak 184 explosions were observed within a single small spatter cone (Figs. 2f and 2g). Most of the 185 lava field was formed of open channel a'a lavas. The total volume of lava was estimated by 186 MIROVA service (https://www.sites.google.com/site/mirovaweb/home), with the use of the 187 MODIS images and the analyses of the flux from the spectral properties, to be within 0.34 + -188 $0.12 \times 10^6 \text{ m}^3$. (Coppola et al., 2017). Satellite derived volume estimates are consistent with 189 independent photogrammetric estimates (0.4 \pm 0.2 x 10⁶ m³; Peltier et al., 2016) and rank the 190 191 2014 eruption at the lower end of the volume range typically emitted by PdF (Roult et al., 2012). 192

193 3. Methodology

194 **3.1 Sampling strategy**

Apart from the sample from the front of the still active lava flow (Fig. 2d), all other samples 195 196 were collected in two phases: 3 days (pyroclasts on June 24, Fig. 3a and Table S1) and 11 days after the eruption (lavas on July 2, Table S1), and three months later (pyroclasts from the 197 MV, Fig. 1, on November 18 and Table S1). June 24 samples were collected both from the 198 main fractures (WF and UF, Fig. 1a), the MV and the active lava flow (Fig. 1 and Table S1). 199 Twenty five scoriaceous bombs and lapilli (REU140624-9a-1 to REU140624-9a and 200 REU140624-9b-6 to REU140624-9b-25, in Table S3) were collected from the discontinuous 201 202 deposit (Fig. 3d) emplaced at the WF site (Fig. 1a), active only at the beginning of the 203 eruptive event. Because of the short duration of the activity at the WF, the scoria fragments on 204 the ground were scarce (Fig. 3d). The strategy was to collect a sample that was formed by the 205 largest available number of clasts that was representative of this discrete deposit (REU140624-9 in Table S1). From the UF (Fig. 1a) only one big scoria was collected 206 207 (REU140624-13, Table S1) that broke in five parts, allowing us to measure its vesiculated core and the dense quenched external part (REU140624-13-a to REU140624-13-e, in Table 208 209 S3). In contrast, the sustained and slightly more energetic activity at the lower tip of the 210 fractures, at the MV site, built a small spatter cone (Fig. 2) and accumulated a continuous, 211 small volume deposit (Fig. 3a) of inversely graded scoria fallout (Figs. 3b and 3c). This deposit is 10 cm thick at 2 m from the vent and covers an area of about $\sim 1000 \text{ m}^2$. For this fall 212 deposit we collected two bulk samples, one from the base (within the lower 5 cm, 213 REU141118-6 in Table S1) and the other from the top (within the upper 5 cm, REU140624-3, 214 in Table S1), for the grain size (Fig. 3c) and componentry analyses. The sample at the base 215 216 was collected in November because on June 24 the loose proximal lapilli blanket was still very hot (405 °C; thermocouple measurement) and fumaroles with outlet temperatures in the 217 range 305-60 °C were sampled all along the fractures several weeks after the eruption (Fig. 1b 218 219 and Table S1). These latter geochemical data are not presented in this paper. We selected 103 220 fragments from the coarse grained bulk deposit within the upper 5 cm of the scoria fall out deposit (Fig. 3b) at MV (REU140624-3-1 to REU140624-3-103, in Table S3) for density, 221 222 connectivity, permeability, petrological and geochemical analysis. In addition, in November 2014, more than 200 clasts (comprising the REU141118-1 to REU141118-5 samples, Table 223 224 S1) of similar size were collected, both close to the MV and in the 'distal' area (30 metres away from the MV site) to complete the particle bulk texture analyses and the chemical 225 226 analyses.

227 **3.2 Grain size and componentry**

We performed grain size analyses on the two bulk samples collected from the MV, following 228 229 the procedure of Jordan et al. (2015) (Table S2). The samples were dried in the oven at 90° C and sieved at $\frac{1}{2}$ phi intervals in the range of -5 ϕ to 4 ϕ (Fig. 3c); the data are also shown in 230 full phi for comparison with the deposits of the 2010 PdF fountaining episode (Hibert et al., 231 2015; Fig. 3f). Sieving was carried out by hand and for not longer than three minutes to avoid 232 breaking and abrasion of the very vesicular and fragile clasts. For the scattered scoria sampled 233 from the WF (Figs. 1, 3d and 3e), we followed the grain size strategy proposed in Gurioli et 234 al. (2013). Within this procedure we sampled each fragment and we recorded the weight and 235 236 the three main axes (a being the largest, b, and c). To allow comparison with the sieving grain 237 size analyses (Inman, 1952), we used the intermediate b axis dimension to obtain $\varphi = -\log_2 b$.

238 Following the nomenclature of White and Houghton (2006) the componentry analysis is the subdivision of the sample into three broad components: i) juvenile, ii) non-juvenile 239 240 particles, and iii) composite clasts. The juvenile components are vesicular or dense fragments, as well as crystals, that represent the primary magma involved in the eruption; non-juvenile 241 242 material includes accessory and accidental fragments, as well as crystals that predate the eruption from which they are deposited. Finally, the composite clasts are mechanical mixtures 243 of juvenile and non-juvenile (and/or recycled juvenile) clasts. In these mild basaltic 244 explosions, the non-juvenile component is very scarce, so we focused on the juvenile 245 component that is characterized by three groups of scoria: (i) spiny-opaque, (ii) spiny-glassy, 246 and (iii) fluidal, along with golden pumice (Fig. 4). The componentry quantification was 247 performed for each grain size fraction between -5 ϕ to 0.5 ϕ (Figs. 5a and 5b), where a 248 249 binocular microscope was used for the identification of grains smaller than -1 phi (Table S2).

In the following, we will use the crystal nomenclature of Welch et al. (2009), with the strictly descriptive terms of macrocrysts (> 3 mm in diameter) mesocrysts (from 0.3 to 3 mm in diameter), and microcrysts (<0.3 mm in diameter). Regarding the June 2014 products, these ranges of size may however change in comparison to the December 2005 products studied by Welsch et al. (2009).

255 3.3 Particle bulk texture (density, porosity, connectivity, permeability) and microtexture

For each sample site (WF, UF and MV, Fig. 1a), we selected all the available particles within the 8-32 mm fraction for density/porosity, connectivity and permeability measurements (Table S3). This is the smallest granulometric fraction assumed to be still representative of the larger size class in terms of density (Houghton and Wilson, 1989; Gurioli et al., 2015), and has been used in previous textural studies (e.g., Shea et al., 2010). In addition, this size range is ideal

for vesicle connectivity measurements (e.g. Formenti and Druitt, 2003; Giachetti et al., 2010; 261 262 Shea et al., 2012; Colombier et al., 2017a, b). Density of juvenile particles was measured by the water-immersion technique of Houghton and Wilson (1989), which is based on 263 Archimedes principle. A mean value for the vesicle-free rock density was determined by 264 powdering clasts of varying bulk densities, measuring the volumes of known masses using an 265 Accupyc 1340 Helium Pycnometer, then averaging. The same pycnometer was also used to 266 measure vesicle interconnectivity for each clast using the method of Formenti and Druitt 267 (2003) and Colombier et al. (2017a). Permeability measurements were performed on five 268 269 clasts: two golden pumices, one fluidal, one spiny glassy and one opaque scoria, all collected 270 from the MV (Table S3). Following Colombier et al. (2017a), the clasts were cut into 271 rectangular prisms to enable precise calculation of the cross-sectional area, which is required to calculate permeability. These prisms were then embedded in a viscous resin, which was left 272 273 to harden for 24 h. The sample surface had been previously coated with a more viscous resin 274 and then wrapped with parafilm to avoid intrusion of the less viscous resin inside the pores. 275 The coated samples were placed with a sample holder connected to a permeameter built at Laboratoire Magmas et Volcans (LMV, France) following Takeuchi et al. (2008). The 276 measurements were performed at atmospheric pressure (i.e. without confining pressure) and 277 the samples were measured at a range of gas flow rates and upstream air pressures to create a 278 curve that could be fitted using a modified version of Darcy's Law, the Forchheimer equation, 279 to solve for viscous (k_1) and inertial permeabilities (k_2) (Rust and Cashman, 2004, Lindoo et 280 al. 2016 and Colombier et al. 2017). 281

Vesicle size distribution was performed following the method of Shea et al. (2010) and 282 283 Leduc et al. (2015), while the total crystallinity, the percentages for both crystal phases (plagioclase and clinopyroxene) and size-populations (meso and microcrysts) were calculated 284 using the raw data from FOAMS program (Shea et al 2010) and the CSDcorrections program 285 of Higgins (2000) and the CSDslice data base (Morgan and Jerram 2006) to have the 286 percentage of crystals in 3D with the corrected assumption for shape. We performed these 287 288 analyses on eight clasts picked up from each component-density distribution (stars in Figs. 6a and 6b). The choice of the clasts was made mostly on the typologies, rather than on each 289 290 density distribution, in order to avoid the analysis of clasts with transitional characteristics. 291 For example, two golden pumice fragments were selected from the largest clasts that were the 292 less dense and didn't break, even if the values in vesicularity were similar. A larger number of fluidal fragments were chosen (even if the density distribution was unimodal) because this 293 294 typology of clasts was the most abundant and was emitted all along the active fracture, so we

did our best in order to study products representative of the WF, the UF and the MV activities.

- Only one spiny glassy and one spiny opaque were selected, because they were emitted only at
- 297 the MF. A full description of the textural measurements all performed at LMV, as well as the

raw data of these measurements are available at DynVolc Database (2017).

299 3.4 Bulk geochemistry

For the determination of the bulk chemistry (Table S4 and Fig. 7) of the different pyroclasts 300 we selected the largest pyroclasts of golden pumice and the largest fluidal, spiny glassy and 301 spiny opaque scoriae (Table S4). We also analyzed two fragments of lava, from the beginning 302 and the end of the eruption (Table S4). Samples were crushed into coarse chips using a steel 303 jaw crusher and powdered with an agate mortar. Major and trace element compositions were 304 analyzed using powder (whole rock composition). In addition, for a sub-set of pyroclasts, 305 glass chips (2-5 mm in size) were hand-picked under a binocular microscope and analyzed 306 separately for trace elements. For major element analysis, powdered samples were mixed with 307 308 LiBO₂, placed in a graphite crucible and melted in an induction oven at 1050 °C for 4.5 minutes, resulting in a homogeneous glass bead. The glass was then dissolved in a solution of 309 deionized water and nitric acid (HNO₃), and finally diluted by a factor of 2000. The final 310 solutions were analyzed by ICP-AES. Trace element concentrations were analysed following 311 312 a method modified from Vlastélic et al. (2013). About 100 mg of sample (powder and chip) were dissolved in 2 ml of 28M HF and 1 ml of 14M HNO₃ in teflon beaker for 36 hours at 313 70° C. Solutions were evaporated to dryness at 70° C. The fluoride residues were reduced by 314 repeatedly adding and evaporating a few drops of concentrated HNO₃ before being fully 315 dissolved in ca. 20 ml of 7M HNO₃. These solutions were diluted by a factor of 15 with 316 0.05M HF (to reach rock dilution factor of ca. 4000) and trace element abundances were 317 determined by quadrupole ICPMS (Agilent 7500). The analyses were performed in plasma 318 robust mode (1550 W). The reaction cell (He mode) was used to reduce interference on 319 masses ranging from 45 (Sc) to 75 (As). The signal was calibrated externally (every 4 320 samples) with a reference basaltic standard (USGS BHVO-2) dissolved as for the samples and 321 322 using the GeoRem recommended values (http://georem.mpch-mainz.gwdg.de/). For elements that are not well characterized in literature (As, Bi, Tl), or which show evident heterogeneity 323 (e.g. Pb) in BHVO-2 powder, the signal was calibrated using the certified concentrations of a 324 synthetic standard, which was also repeatedly measured. The external reproducibility (2σ 325 326 error) of the method is 6% or less for lithophile elements and 15% or less for chalcophile elements. 327

328 **3.5 Glass and crystal chemistry**

Spot analyses of matrix glass and crystal composition (Table S5) were carried out using a 329 Cameca SX100 electron microprobe (LMV), with a 15 kV acceleration voltage of 4 nA beam 330 331 current and a beam of 5 µm diameter for glass analyses. However, for the spiny opaque scoria, characterized by abundant crystals with rapid growth textures, a voltage of 8 nA beam 332 333 current and a beam of 10 µm diameter were used. For this latter sample, 10 analyses per 334 sample were performed due to the heterogeneity within the highly crystallised glass (Fig. 8a), while for the other samples 6 analyses per sample were enough to characterize the clean 335 homogeneous glass. For crystal analysis, a focused beam was used. For the characterization of 336 the meso- and micro-crysts, due to their small size, only two to three measurements were 337 performed, one at the edge, one in the middle and one at the core of the crystals, to check for 338 possible zonation. 339

340 **3.6 Melt inclusions**

Melt inclusions (MIs; Table S6, Figs. 8b and 9) were characterized in the olivine mesocrysts from the three groups of scoriae (fluidal, spiny glassy and spiny opaque), but not in the pumice group, because crystals were too rare and small to be studied for MIs.

Olivine crystals were handpicked under a binocular microscope from the 100-250 and 344 250- 600 µm grain size fractions of crushed tephra. Crystals with MIs were washed with 345 acetone, embedded in epoxy and polished individually to generate adequate exposure of the 346 MIs for in situ electron probe microanalysis. The MIs are spherical to oblate in shape and 347 range in size from 10 to 200 µm. Some of the MIs contain shrinkage bubbles but all of those 348 studied are totally deprived of daughter minerals. Major elements were measured on a 349 Cameca SX-100 microprobe at LMV (Table S6). For major elements, the larger MIs were 350 analyzed with a spot diameter of 10-20 µm and sample current of 8 nA, whereas the smaller 351 MIs were analyzed with a beam of 5 µm and a sample current of 4 nA. The results are given 352 in Table S6, and analytical details and uncertainties are listed in Óladóttir et al. (2011) and 353 Moune et al. (2012). 354

355 **4 Results**

4.1 Deposit texture (grain size, componentry, morphology) and petrological description
of the samples

The pyroclastic deposits at the WF and UF sites (Fig. 1a) are formed by scattered homogeneous smooth fluidal (Figs. 3d) bombs and lapilli scoria. The average dimension of the fragments is around 4 cm (maximum axis) with bombs up to 10 cm and scoria lapilli up to 2 cm in size (Fig. 3e).

At the MV, the reversely graded deposit (Fig. 3b) is made up of lapilli and bombs, with 362 only minor coarse ash (Fig. 3c). The lower 5 cm at the base are very well-sorted and show a 363 perfect Gaussian distribution with a mode at 4 mm (Fig. 3c). In contrast, the grain size 364 365 distribution of the upper 5 cm is asymmetrical with a main mode coarser than 22 cm and a 366 second mode at 8 mm (Fig. 3c). This upper deposit is negatively skewed due to the abundance of coarse clasts. The dataset shows a similarity between the grain size distributions of the 367 368 basal tephra ejected from the 2014 MV and the ones for the lava fountaining of the 2010 summit event (Fig. 3f and Hibert et al., 2015). On the contrary, the top of the 2014 fall differs 369 370 from fountain deposits, being coarser and polymodal, and it is ascribed to dominantly 371 Strombolian activity (Fig. 3f).

372 In terms of componentry of the deposits, four types of clasts were distinguished (Fig. 4): (i) golden pumice, (ii) smooth or rough fluidal scoriae, (iii) spiny glassy scoria, (iv) spiny 373 opaque scoria. The pumices are vesicular, low-density fragments, characterized by a golden to 374 light brown color, sometimes with a shiny outer surface (Fig. 4a). They are usually rounded in 375 shape. Golden clasts studied for textures contain a few microcrysts of plagioclase (up to 0.1 376 mm in diameter), clinopyroxene up to 0.05-0.06 mm in diameter, and small olivine up to 0.03377 mm in diameter (Fig. 4), together with large areas of clean, light brown glass. The fluidal 378 scoria fragments have dark, smooth or rough shiny surfaces (Fig. 4b). They can be more or 379 380 less elongated in shape and have spindle as well as flattened shapes. The fluidal fragments are characterized by rare mesocrysts of plagioclase and clinopyroxene and microcrysts of 381 plagioclase, clinopyroxene and olivine (Fig. 4b). The spiny glassy fragments are dark, spiny 382 scoria that range in shape from subrounded to angular (Fig. 4c). These fragments contain 383 abundant glassy areas, while the spiny opaque fragments lack a glassy, iridescent surface. 384 385 Both groups of spiny clasts are characterized by the presence of dark and light brown glass. The spiny opaque fragments are the densest fragments and have the largest amount of 386 387 crystals. They contain, as the most abundant phase, relatively large meso- and micro-crysts of 388 plagioclase, up to 3 mm long, together with meso- and micro-crysts of clinopyroxene and 389 olivine (Figs. 4c and 4d). In the dark portions of their matrix, tiny fibrous microcrysts of olivine + clinopyroxene + plagioclase + Fe-Ti oxides occur. The spiny glassy fragments have 390 391 the same crystal populations as the spiny opaque ones, but their plagioclases are much smaller

and attain a maximum length of only 0.3 mm. Clusters of plagioclase and clinopyroxene are 392 present in both the spiny opaque and the spiny glassy fragments, as well as rare macrocrysts 393 of olivine. The olivine macrocrysts exhibit the typical compositional (Fo 84.2) and 394 petrographic features of olivine phenocrysts described in previous studies (Clocchiatti et al., 395 1979; Albarede and Tamagnan, 1988; Bureau et al., 1998a and b; Famin et al., 2009; Welsch 396 et al., 2013). They are automorphic, fractured with oxides (mostly chromite) and melt 397 inclusions (Fig. 4c). Fluidal and pumice fragments studied for textures contain rare 398 399 macrocrysts and mesocrysts of olivine, and the crystals are essentially microcrysts. The 400 pumice and some fluidal fragments have lower contents of microcrysts than some fluidal and spiny fragments, with the latter having the highest microcryst content (Table S4). For 401 402 comparison two fragments of lava have been analyzed as well (Table S3). The lava fragments are poorly vesiculated and completely crystalline (Fig. 4e). The lava contains the same 403 404 paragenesis of crystals described in the spiny opaque fragments, with the main difference that its matrix is completely crystallized and constituted mostly by well-formed plagioclase up to 405 406 800 microns and clinopyroxene up to 500 microns. Scarce, smaller olivines, are also present Ubiquitous tiny rounded Fe-Ti oxides provide evidence of post emplacement crystallization. 407

The componentry results are reported in Figure 5 for the MV deposits; being the 408 deposits from the WF and UF characterized exclusively by fluidal clasts (Fig. 3). At the base 409 of the MV deposit, the coarse fraction of the deposit is rich in golden and fluidal components 410 that represent more than 60-70 vol. % (Figs. 5a and 5b). The proportion of the two groups is 411 similar. In contrast, in the upper, coarse grained fall deposit, the clasts bigger than 8 mm are 412 dominated by the spiny scoria fragments, while the fraction smaller than 8 mm show a 413 dramatic increase in the golden and fluidal fragments, with the fluidal ones always more 414 abundant than the golden ones (Figs. 5a and 5b). Abundant low-density, golden, coarse lapilli 415 pumice and bombs have been found scattered laterally up to 30 metres from the main axis and 416 were not found in the proximal deposit. On the basis of the high amount of pumice in the 417 lower part of the deposit, we correlate the large, low-density clasts with the base of the 418 419 proximal deposit, and consequently we interpret them as material emitted at the beginning of the June 2014 eruptive event. 420

421 **4.2** Particle density, porosity, connectivity, permeability and micro-texture

Density analyses performed on 200 coarse lapilli reveal a large variation in density values
from 390 kg/m³ to 1700 kg/m³ with a median value at 870 kg/m³ (Table S3). The fragments
collected from the MV have a bimodal density distribution, with a main population of low-

density fragments having a mode at 800 kg/m³, and a second and denser population centered 425 at 1400 kg/m³ (Fig. 6a). The golden and fluidal fragments form the lower-density population 426 and the spiny fragments are dominant in the denser population (Fig. 6a). For these samples 427 there is a marked correlation between porosity and morphology, so that the spiny-opaque 428 clasts are the densest (up to 1600 kg m⁻³, with a vesicularity of 45 vol. %) and the golden 429 pumice are the least dense (minimum density of 390 kg m^{-3} with a vesicularity of up to 86 vol. 430 %; with a Dense Rock Equivalent density of 2880 kg m⁻³). The fluidal fragments collected at 431 the WF (Fig. 1b), have a density range from 700 to 1400 kg m⁻³ and a mode at 1000 kg m⁻³ 432 (Fig. 6b). The five fragments from the only bomb collected at the UF are characterized by two 433 distinct density values, the low density one (700-800 kg m⁻³) refers to the core of the sample, 434 while the high density one (1400-1500 kg m⁻³) represents the quenched external rim of the 435 bomb. Finally, the two fragments of lava show the highest density values at 1800 and 2150 kg 436 m⁻³. This last value is one of the highest found in the lava collected from 2014 up to 2017 (see 437 Fig. 13 in Harris et al., 2017 and unpublished data). 438

439 In all these samples, the increase in vesicularity correlates with an increase in the amount of small (0.1 mm), medium (0.5-1 mm) and large (up to 4 mm) vesicles. In the fluidal 440 441 clasts, these vesicles have a regular rounded or elliptical shape and are scattered throughout 442 the sample. The low-density pumices are often characterized by the presence of a single, large central vesicle (10 - 15 mm) with the little vesicles and a few medium vesicles distributed all 443 around it (Fig. 4). The spiny glass texture is characterized by a lower amount of small vesicles 444 than in the pumice and by the presence of mostly medium sized vesicles, while the spiny 445 opaque has more irregular shape, very large (up to 10 mm) vesicles with a small and a 446 medium sized bubble population. In the spiny glass samples, the glass is more or less brown, 447 with the dark brown portions being the ones with the lowest vesicle content and the highest 448 microcrysts content. The opaque samples have a central, very dark glass portion, with low 449 vesicle content, and a more vesicular glassy portion at the outer edges (Fig. 4). The two 450 fragments of lava are poorly vesiculated (Fig. 6a) and characterized by large, irregular 451 vesicles (up to 5 mm in diameter). Clusters of small vesicles (up to 0.1 mm) are scattered 452 between the large ones. 453

The vesicle size distribution (VSD in Fig. 4) histograms are characterized by a decrease in percentage of vesicles from the golden to the lava as well as an increase in coalescence and or expansion signatures in the spiny fragments, marked by the increasing of the large vesicles population (Figs 4c and 4d). This trend is also marked by the decrease in number of vesicle per unit of volume (N_v , Fig. 4) from the golden to the lava. Finally, the

trend is also mirrored by the total percentage of crystals (calculated in 3D, Fig. 4 and reported 459 460 for each sample in Table S3) that increases with the increase of density of the clasts, from a minimum of 8 vol. % for the golden up to 55 vol. % for the spiny opaque scoria, and 100 vol. 461 % for the lava (Fig. 4). Mesocrystals, formed mostly by the same proportion of plagioclase 462 and clinopyroxenes, are absent or very scarce in the golden and fluidal fragments, while they 463 reach their maximum values, up 21 vol. % in the spiny opaque fragment. The population of 464 microcrystals is mostly constituted by plagioclases that range from a minimum of 6 vol. % in 465 the golden, up to 23-25 vol. % in the spiny fragments and to 64 vol. % in the lava. 466

467 The connectivity data (Fig. 6c) also indicate that the fluidal and golden clasts have a larger amount of isolated vesicles (up to 40 vol. %) with respect to the spiny products. The 468 469 fluidal clasts from the WF are the most homogeneous with an average percentage of isolated vesicles around 30 vol. %. In contrast, both the pumice and the fluidal fragments from the 470 471 MV, characterized by higher values of porosity (> 75%), have a wide range in percentage of isolated vesicles (between 20 and a few vol. %). The fragments of the bomb collected at the 472 473 UF are consistent with a vesiculated core characterized by scarce isolated vesicles and the quenched rind that has 30 vol. % of isolated vesicles. Finally the spiny fragments have the 474 lowest content of isolated vesicles (0-5 vol. %). Despite the presence of these isolated 475 vesicles, all the samples shear high values of permeability, with the Darcian (viscous, K_1) 476 permeability values ranging from 10⁻¹¹ to 10⁻¹⁰ m² (Fig. 6d and Table S3). The graph of 477 vesicularity versus K₁ shows a slightly increase in permeability with vesicularity, being the 478 golden pumice the most permeable among the samples and the spiny glassy fragment the least 479 permeable. The three samples collected from the February 2015 eruption fit this trend. 480 However, the densest spiny opaque scoria of the 2014 eruption shares the high permeability 481 482 value of the golden pumice.

483 **4.3 Chemistry of the products**

Major and trace element concentrations of whole-rock and hand-picked glass samples are 484 reported in Table S4. Whole rock major element composition is very uniform (e.g., 485 6.5<MgO<6.7 wt%) and well within the range of Steady State Basalts (SSB), the most 486 common type of basalts erupted at PdF (Albarède et al., 1997). However, compatible trace 487 elements, such as Ni and Cr, are at the lower end of the concentration range for SSB 488 (<100ppm) indicating that the June 2014 eruption sampled relatively evolved melts. Ni and Cr 489 generally show higher concentrations in 2014 bulk rocks (79<Ni<92ppm and 71<Cr <87ppm) 490 compared to the 2014 glass chips (66<Ni<73ppm and 54<Cr <59ppm for all but two chips). 491

In the Cr vs Ni plot (Fig. 7a), whole rocks plot to the right of the main clinopyroxene +/-492 493 plagioclase-controlled melt differentiation trend. This shift reflects the addition of Ni-rich olivine (Albarède and Tamagnan, 1988). We estimate that the Ni excess results from the 494 occurrence of a low amount (0.7 to 1.3 wt%) of cumulative olivine in whole rocks, consistent 495 with thin section observations. The composition of olivine macrocrysts (ca. Fo84) is too 496 magnesian to be in equilibrium with the low-MgO evolved composition of the 2014 magma. 497 Using our estimate for the amount of cumulative olivine, we recalculate the olivine-corrected 498 MgO content of the 2014 magma at 6.2 wt%. The June 2014 melt is thus only moderately 499 depleted in compatible elements compared to the previous eruption of December 2010 500 (MgO~6.6 wt%, Ni~80 ppm, Cr~120 ppm). Conversely, the June 2014 melt is significantly 501 502 depleted in compatible elements compared to the earlier November 2009 eruption, which sampled relatively primitive magmas (average MgO~7.7 wt%, Ni~135 ppm, Cr~350 ppm) 503 504 (Fig. 7a). The 2014 evolved composition plots at the low-Ni-Cr end of PdF historical differentiation trend (Albarède and Tamagnan, 1988), near the composition of lavas erupted 505 506 on 9 March 1998 after 5.5 years of quiescence (1992-1998). Note that olivine accumulation at PdF generally occurs in melt having ca.100 ppm Ni (Albarède and Tamagnan, 1988). Olivine 507 accumulation in evolved melts (Ni < 70 ppm) seems to be a distinctive feature of many small 508 post-2007 eruptions (e.g. this event and the three 2008 eruptions, see Di Muro et al., 2015). 509

A closer inspection of Ni-Cr variability in June 2014 whole rock samples (Fig. 7b) reveals that scoria from the WF (140624-9b-6, Table S4) and early erupted lavas (1406-21-1, Table S4) have the lowest amount of olivine (<0.9%) whereas scoria from the UF (140624-13a) and late erupted lavas (140324-12) have a slightly higher amount of olivine (>1.2%). This is consistent with the general trends observed at PdF of olivine increase from the start to end of an eruption (Peltier et al., 2009).

The so called "olivine control trend" in Ni-Cr space cannot be explained either by 516 addition of pure olivine, which contains less than 500 ppm Cr (Welsch et al., 2009; Salaün et 517 al., 2010; Di Muro et al., 2015), or by the addition of olivine plus pyroxene (which would 518 519 require ca. 50% pyroxene with 970 ppm Ni and 4800 ppm Cr, see Fig. 7 caption). Instead, addition of olivine hosting ca. 1% Cr-spinel (with 25 wt.% Cr) accounts for data and 520 521 observations, and is consistent with crystallization of olivine and Cr-spinel in cotectic 522 proportions (Roeder et al., 2006). The fact that some samples (golden pumice) plot off the 523 main, well-defined array, can be explained either by addition of more or less evolved olivine crystals (within the range of Fo 80-85 measured in June 2014 samples) and/or slight 524 525 variations ($\pm 0.02\%$) in the proportion of Cr-spinels (Fig. 7b).

The glass chemistry of the four clast types allows us to correlate porosity and oxide 526 527 contents and shows an increase in MgO from the spiny opaque to fluidal and golden fragments (Fig. 8a). Consistent with petrological and textural observations, the spiny opaque 528 is the most heterogeneous type of clast in terms of glass composition (Fig. 8). The glassy 529 portion at the edge of the clast is similar to the spiny glass, while the interior, characterized by 530 dark areas rich in tiny fibrous microcrysts, shows scattered glass compositions with very low 531 MgO content as well as a decrease in CaO (Fig. 8). We attribute the significant variation in 532 533 glass composition within the different components to variable degrees of micro-crystallisation 534 as the bulk chemistry of all clasts is very similar and globally homogeneous.

535 4.4 Melt inclusions

536 MI analyses must be corrected for post-entrapment host crystallisation at the MI - crystal 537 interface. We used a Kd = $(FeO/MgO)_{ol} / (FeO/MgO)_{melt} = 0.306$ (Fisk et al., 1988; Brugier, 538 2016) and an average $Fe^{3+}/\Sigma Fe_{total}$ ratio of 0.11 (Bureau et al., 1998a; Di Muro et al., 2016 and 539 references therein) defined for PdF magmas. For the June 2014 melt inclusions, the post 540 entrapment crystallization (PEC) ranges from 2.9 to 10.5 wt%. Raw and corrected major and 541 volatile element concentrations of MIs are reported in Table S6.

542 Host olivines span a large compositional range from Fo₈₀ to Fo₈₆. Despite the evolved bulk composition of the magma, most olivines are quite magnesian (Fo₈₃₋₈₅) and are not in 543 equilibrium with the evolved host magma. On the contrary, Mg-poor olivines (Fo₈₀₋₈₁) can be 544 considered as being in equilibrium with the bulk rock composition. The corrected 545 compositions of MIs in phenocrysts from the different samples partly overlap with the 546 547 evolved bulk rocks (MgOwr: 6.1-7.2 wt%) and extend to higher MgO contents of up to 8.8 wt% (Table S6). MIs display a narrow range of transitional basaltic compositions ($K_2O=0.5$ -548 549 0.9 wt%) and show no significant difference between the three types of scoriae. The major element composition of melt inclusions correlates with that of the host olivines. Melt 550 551 inclusions in the high Fo-olivines have the highest MgO, CaO and TiO₂ and lowest K₂O concentrations (Table S6). It is interesting to note that the June 2014 products contain two 552 553 populations of magnesian (Fo_{>83}) olivines hosting melt inclusions with two distinct Ca contents. Most of the magnesian olivines contain MIs with unusually high CaO contents (11.6 554 -12.9 wt%) and high CaO/Al₂O₃ ratios (0.8-0.9), higher than that of the bulk rocks (0.8) (Fig. 555 556 8). The occurrence of olivines with "high Ca" melt inclusions has been observed in all three different types of scoriae. A few magnesian olivines and all Mg-poor olivines (Fo_{80.5-83.6}) host 557

MIs with lower CaO contents (11.4 wt%). This latter composition overlaps with that of the 558 bulk rock (Fig 8). The "high Ca" population of inclusions is also enriched in TiO_2 and Al_2O_3 559 and depleted in MgO, FeO_T and Na₂O for a given olivine Fo content with respect to the "low 560 Ca" population. Both low- and high-Ca populations of melt inclusions have similar K_2O 561 contents and total alkali content increases from 3 wt% at 12.6 wt% CaO, to 3.5 wt% at 10.8 562 wt% CaO. However, we remark that high Ca melt inclusions from the June 2014 activity 563 record a significant scattering in K₂O contents, which range from 0.55 to 0.9 wt%. These 564 565 anomalous compositions potentially track processes of crystal dissolution (e.g. pyroxene 566 dissolution).

MIs in olivines from June 2014 can best be compared with those of other recent small-567 volume and short-lived eruptions which emitted basalts with low phenocryst contents, like 568 those in March 2007 (0.6 x 10^6 m³) and November 2009 (0.1 x 10^6 m³) (Roult et al., 2012). 569 March 2007 aphyric basalt has a bulk homogeneous composition with intermediate MgO 570 content (MgOwr: 7.33 wt%; K₂O: 0.67 wt%). Their olivines (Fo 81) are in equilibrium with 571 572 the bulk rock and their composition is unimodal (Di Muro et al., 2014). November 2009 products are the most magnesian lavas emitted in the 2008-2014 period, slightly zoned 573 (MgOwr: 7.6-8.3 wt%; K₂O: 0.75 – 0.62 wt%) and contain a few percent of normally zoned 574 olivine macrocrysts with bimodal composition (Fo81 and Fo83.5, see Di Muro et al., 2016). 575 June 2014 bulk rocks (MgOwr: 6.7 wt%; K₂O: 0.75 wt%) and melt inclusions in Fo₈₀₋₈₁ 576 olivines are quite evolved. Their composition is close to that of products emitted by summit 577 intracaldera eruptions in 2008, ca. 1.5 years after the large 2007 caldera forming eruption (Di 578 Muro et al., 2015) (Fig. 8). As already reported for 2008 products, many olivine macrocrysts 579 of 2014 are clearly too magnesian to be in equilibrium with the relatively evolved host melts. 580 Overall, MgO content in 2007-2014 melt inclusions tends to decrease with decreasing Fo 581 content of the host olivines. MIs in olivines also exhibit a trend of linear decrease in MgO and 582 increase in FeO from April 2007 to 2009-2014 products (Fig. 9). Melt inclusions in March 583 2007, November 2009 and June 2014 follow the same trend of FeO enrichment (Fig. 9). In the 584 large-volume and olivine-rich April 2007 products, MIs in magnesian olivines with Fo_{>82} have 585 distinctly higher MgO, FeO and lower SiO₂ and Al₂O₃ than MIs in 2009-2014 products. The 586 587 distinctive FeO enrichment of many of the MIs from the April 2007 oceanite has been interpreted by Di Muro et al. (2014) as a result of post-entrapment modification related to new 588 589 magma inputs into long lasting magma storage.

590 Two populations of low- and high-Ca melt inclusions are also found in the November 591 2009 olivines. Low-Ca melt inclusions from the November 2009 and June 2014 eruptions

indicate a single trend of chemical evolution (Fig. 8), consistent with bulk rock compositions. 592 593 June 2014 products have lower MgO and CaO contents than those from November 2009. Significant scattering in K₂O content (0.6-0.9 wt%) is found in low-Ca inclusions from 2009, 594 as observed in high-Ca inclusions from the 2014 eruption, but they share similar K_2O 595 contents. In 2009 and 2014 products, K₂O content of melt inclusions is partly anti-correlated 596 with the olivine Fo content. This observation has been attributed to moderate heterogeneity of 597 primary melts feeding the plumbing system of PdF. Rapid temporal changes of K₂O content in 598 599 PdF basalts have been reported (Boivin and Bachelery, 2009).

600 4.5 Mineral composition and glass – plagioclase equilibrium

All 2014 scoriae (spiny, fluidal, golden) contain the same paragenesis of olivine,
clinopyroxene and plagioclase. The composition of minerals found in golden, fluidal and
spiny scoriae is indistinguishable.

In olivines, average MgO content decreases from macrocrysts (Fo_{84.1}) to mesocrysts 604 605 (Fo_{79.6}) to microcrysts. Olivine microcrysts (Table S5) are normally zoned. Their composition ranges from Fo_{78.0-75.3} in the cores to Fo_{74.3-70.5} in the rims. Overall, olivines in 2014 products 606 607 span the full range of typical Fo contents of recent PdF magmas (Boivin and Bachèlery, 2009; Di Muro et al., 2014; 2015). Clinopyroxene composition (augites) ranges from En₅₃Fs₁₅Wo₃₂ 608 609 to En₄₁Fs₁₄Wo₄₅. Their average composition (En₄₅Fs₁₄Wo₄₁) is consistent with that found in other recent evolved melts like those emitted by the 2008 eruptions (Di Muro et al., 2015) and 610 more generally in recent PdF products (Boivin and Bachèlery, 2009). Clinopyroxenes are 611 unzoned, the composition of cores and rims is very similar and close to that found in 612 microcrysts and mesocrysts. Plagioclase composition ranges from An_{79.5}Ab_{19.9}Or_{0.6} to 613 An_{63.1}Ab_{35.7}Or_{1.2} with a bimodal distribution (An_{76.5-79.5} and An_{63.1-72.9}, Fig. 10a). Similar 614 bimodal distributions were observed in many other products at PdF (Di Muro et al., 2015). 615 616 Mesocrysts (An_{75.5}Ab_{23.8}Or_{0.7} on average) are more calcic with respect to microcrysts (An_{65.7}Ab_{33.1}Or_{1.2} on average). Normal zoning is found from plagioclase cores to rims (Fig. 617 10a). The composition and zonation of 2014 plagioclases clearly contrast with the complex 618 and often reverse zoning patterns and intermediate composition of the 2008 PdF products that 619 were attributed to pre-eruptive magma heating (Di Muro et al., 2015). 620

Plagioclase-melt equilibrium and melt composition in pyroclastic rocks and waterquenched lavas were used to estimate both temperature and water content dissolved within the melt (Fig. 10b and Table S5). Temperature estimates are based on the (dry) equation of Helz and Thornber (1987) recalibrated by Putirka (2008). Dissolved water content was calculated

from the plagioclase hygrometer of Lange et al. (2009) at 50 MPa. This pressure corresponds 625 to the average CO_2 -H₂O saturation pressure (recalculated with Papale et al., 2006) typically 626 recorded in melt inclusions from central products at PdF (e.g. 1931 eruption in Di Muro et al., 627 (2016) and references therein). This pressure roughly corresponds to the sea level depth, 628 which is inferred to be the location of the potential main shallow magmatic reservoir (Peltier 629 et al., 2009; Lengliné et al., 2016; Coppola et al., 2017). The application of the plagioclase 630 hygrometer of Lange et al. (2009) makes it possible to estimate the dissolved water content in 631 the melt with a nominal uncertainty of 0.15 wt% and is only slightly dependent on pressure. 632 Plagioclase compositions not in equilibrium with the melt (glass or bulk rock) are those of 633 mesocryst cores with the highest (An_{>76.5}) anorthite content (Fig. 10a and Table S5). Such 634 compositions are more in equilibrium with CaO-richer magnesian melts than those measured 635 in matrix glasses and bulk rocks of 2014 eruption and likely formed during early stages of 636 637 shallow magma differentiation (Fig. 10a).

In order to determine pre-eruptive conditions, calculations were performed only on 638 639 paired plagioclase rims and matrix glasses in equilibrium, using the plagioclase-melt equilibrium constant of Putirka (2008) calibrated for melts whose temperature exceeds 640 1050° C (Kd_{An-Ab} = 0.27±0.05). Our review of published and unpublished data shows that melt 641 temperature progressively decreases from April 2007 (1188+/-16 °C) to January-October 642 2010 (1147+/-9°C) and positively correlates with K_2O content in melts which increases from 643 0.70 to 0.96 wt% (Fig. 10b). The melts from the June 2014 eruption record the lowest 644 temperatures in post-2007 eruptions (1131 \pm 15 °C) together with the highest K₂O-enrichment 645 (K₂O: 0.90 ± 0.12 wt%). The lowest temperatures are recorded by spiny scoriae, while the 646 temperature of golden scoriae overlaps with that of 2010 products emitted before the 2010-647 2014 phase of quiescence. In spite of the large variability in melt composition and 648 temperature, average pre-eruptive water content dissolved in the melts (0.5 + - 0.2 wt) is 649 quite homogeneous for the whole 2008-2014 period. In 2014, the lowest estimated dissolved 650 water content (down to 0.38 wt%) is for the golden and some fluidal scoriae, while the 651 652 maximum amount (0.68 wt%) is for the spiny opaque scoriae. However, water content estimated from core-bulk rock equilibrium $(0.3\pm0.1 \text{ wt\%})$ is slightly lower than that estimated 653 654 from rim and microlite-matrix glass equilibrium $(0.5\pm0.2 \text{ wt\%})$, but the difference broadly overlaps the nominal uncertainty related to calculations. Dissolved water contents in melts of 655 the pyroclasts are thus intermediate between those measured in 2007 melt inclusions (H_2O : 656 0.8 ± 0.15 wt% and up to 1.1 wt%) and those typically found in degassed matrices of lava 657 658 and Pele's hairs of 2007 (Fig. 10b; 0.2 wt%; see Di Muro et al., 2015; 2016).

659 5 Discussions

660 **5.1 Eruptive dynamics**

The activity fed by the uppermost WF and UF (Fig. 1) was very short-lived, as shown by the 661 presence of only scattered bombs and coarse lapilli (Figs 3d and 3e). The homogeneity of 662 these clasts, their coarse grained nature and the fluidal smooth texture are in agreement with 663 very short-lived fire-fountaining/magma jets. Glassy outer surfaces of clasts have been 664 interpreted as a late-stage product of fusion by hot gases streaming past the ejecta within the 665 jet/fountain (Thordarson et al., 1996; Stovall et al., 2011). However, the occurrence of this 666 process is not supported by the homogeneous glass composition in our fluidal clasts. 667 Therefore, we interpret these features here just as rapid quenching and not re-melting. 668 Vlastélic et al. (2011) documented the mobility of alkalis and other elements on PdF clasts 669 that experienced long exposures to acid gases. In the 2014 eruption pyroclasts, the mobility of 670 elements was prevented by the short duration of the events. 671

At lower altitude and close to the MV (Fig. 1), the 5 cm layer at the base of the fall 672 deposit is fine-grained (Figs. 3b and 3c), rich in fluidal and golden fragments (Fig. 5), with a 673 674 perfect Gaussian grain size curve (Fig. 5), and similar to that reported from the weak 2010 fountaining event (Fig. 3f and Hibert et al., 2015). Therefore, we interpret this deposit as 675 676 being due to weak Hawaiian like fountaining (sustained, but short-lived) activity. We want to remark here that this activity happened during the night and was not observed. The top of the 677 678 same deposit is coarse grained (Figs 3b and 3c), bimodal, has a lower content in coarse ash (Table S2) and is rich in spiny opaque and spiny glass fragments (Fig. 5). The reverse grain 679 680 size likely records the transition from early continuous fountaining to late discrete Strombolian activity (observed and recorded on the 21 of June 2014, Fig. 2). This transition in 681 activity is typical of many eruptions at PdF (Hibert et al., 2015). The reverse grading of the 682 683 whole deposit (Figs. 3b and 3c) is thus not correlated with an increase in energy of the event, but with two different eruptive dynamics and fragmentation processes. The decrease in coarse 684 ash, which correlates with the decrease in energy of the event, highlights the most efficient 685 686 fragmentation process within the Hawaiian fountaining with respect to the slow gas ascent and explosion of the Strombolian activity. These conclusions are consistent with (i) the 687 continuous and progressive decrease in intensity of Real time Seismic Amplitude 688 Measurement recorded by the OVPF seismic network (unpublished data), and (ii) satellite 689 derived TADR which suggest continuous decay of magma output rate after an initial short-690 lived intense phase (Coppola et al., 2017). 691

5.2 Interpretation of the different textural signatures and the meaning of the 4 typologiesof clasts.

694 1) Background on the texture of clasts from Hawaiian and Strombolian activities

The first microtextural analysis of Hawaiian ejecta was performed by Cashman and Mangan 695 696 (1994) and Mangan and Cashman (1996) on pyroclasts from 1984 to 1986 Pu'u 'O'ō fountainings. The authors defined two clast types: 1) 'scoria' consisting of closed-cell foam of 697 \leq 85% vesicularity, with round, undeformed, broadly-sized vesicles, and 2) 'reticulite', an 698 open-cell polyhedral foam with $\sim 1 \mu m$ thick vesicle walls with >95% vesicularity. They stated 699 that the scoria to reticulite transition is a consequence of Ostwald ripening, where larger 700 bubbles grow at the expense of smaller bubbles due to post-fragmentation expansion of clasts 701 within the fountain. According to this model, scoria preserves textures closer to conditions at 702 fragmentation, whereas continued vesiculation and clast expansion in the thermally-insulated 703 core of the fountain results in reticulate. This model was confirmed at lava fountains at Etna 704 (Polacci et al., 2006), Villarrica (Gurioli et al., 2008), Kīlauea Iki, (Stovall et al., 2011 and 705 706 2012), Mauna Ulu (Parcheta et al., 2013) and Al Madinah (Kawabata et al., 2015). These last 707 authors also measured the connected and isolated porosity in the AD1256 Al-Madinah Hawaiian fountaining eruptions. They found that the reticulite-like textures from the central 708 709 part of these very high fountains showed isolated vesicles in agreement with low shear rates and low viscosity melts, where bubbles may grow spherically and remain isolated. In contrast, 710 711 at margins of the fountains, high shear may lead to stretching and mechanical coalescence of bubbles, forming the common, fluidal types of particles seen also in the deposits. They also 712 713 stated that lower vesicularity and greater isolated porosity were found in some tephra interpreted as resulting from violent Strombolian eruptive phases. 714

The data that we found in our study of the typical activity of PdF agree only partially 715 716 with all these interpretations. The reason is that we sampled and measured products of very weak Hawaiian to Strombolian activities. If we plot the approximate durations and masses of 717 these events on the Houghton et al. (2016) diagram, the 2014 activity of PdF falls into the two 718 fields for transient and fountaining activity, but at the base of the diagram. We here show for 719 the first time that short lived and weak fountaining can preserve pyroclast textures that record 720 magma ascent and fragmentation conditions before the explosions and also provide some 721 722 information about the pre-eruptive storage conditions. The occurrence of time-variable ascent 723 conditions is also reflected in the time evolution of eruptive dynamics, with the golden and fluidal scoriae emitted from the low Hawaiian fountaining episodes and the spiny fragmentsfrom the Strombolian-like explosions

726 <u>2) The four typologies of clasts and their distribution in space and in time in the 2014</u> 727 eruption at PdF

728 So, as described in 5.1, longitudinal variation in eruptive style along the fracture system produces a spatial variability in the proportions of the four typologies of clasts. The 729 uppermost fractures (WF and UF, Fig. 1a) are characterized solely by fluidal fragments (Fig. 730 4b); they lack both the spiny and the golden components. In addition, these fluidal clasts are 731 the ones showing the smoothest surfaces (indicative of rapid quenching in a very hot 732 environment), low porosity values (between 50 to 77 vol. %, Fig. 6b), the highest content in 733 isolated vesicles (~ 30 vol. % Fig. 4c), and low vesicle numbers (3 to 5 x 10^6 , Fig. 4b), 734 comparable to the spiny fragments. They have scarce mesocrysts (1-2 vol. % Table S3) and 735 very low amount of microcrysts of plagioclase and clinopyroxene (3 to 11 vol. %, Table S3). 736 These fluidal scoria fragments were emitted by short lived jets of magma, therefore they 737 underwent rapid quenching in a very hot environment that prevented any expansion or further 738 739 vesiculation and preserved a very high number of isolated vesicles (Fig. 6d). Syn-eruptive crystallization was hindered by high ascent velocities in the dyke, due to the sudden release of 740 741 over-pressure in the shallow magma reservoir.

The four typology of clasts, golden pumice, fluidal scoria and the spiny fragments 742 743 (Fig. 4) were found associated only at the MV. The relative proportions of these four typologies of clasts correlate with the eruptive dynamics. The golden lapilli and fluidal clasts 744 745 were in fact dominant in the Hawaiian, more energetic activity at the beginning of the eruption (during the night between the 20 and the 21 of June 2014). In contrast, the spiny 746 747 fragments were dominant during the Strombolian activity, coinciding with the decreasing in 748 Mass Discharge Rate (MDR, early in the morning of the 21, Fig. 2 and Coppola et al., 2017). The golden and fluidal fragments from the MV show the highest porosity (86 %, Fig. 6a), 749 750 variable proportions of isolated vesicles (Fig. 6c) and high, but variable, N_V numbers (Figs. 4a). They are also characterized by a uniform vesicle size population with clear evidence of 751 752 incipient expansion, especially in the fluidal fragments (Figs. 4a and 4b). From the connectivity graph, there is a clear decrease in isolated vesicles with the increase in 753 vesicularity (Fig. 6c). The content in crystal, mostly formed by microcrysts of sodic 754 755 plagioclase (Fig. 10a) due to magma degassing during its ascent and decompression in the conduit (Di Muro et al., 2015), is very low, especially in the golden pumice (up to 15 vol. %), 756

and slightly higher for the fluidal clasts (up to 23 vol. %). We interpret the golden fragments, 757 758 at the MV, to be the fastest (low amount of microcrysts) and less degassed magma (high vesicularity coupled with high N_V), which experienced only a very short residence time in the 759 magma transport system (dyke+vent), followed by the fluidal fragments. In contrast the spiny 760 fragments, characterized by higher percentage of microcrysts and mesocrysts, by the lack of 761 isolated vesicles, by the presence of coalescence signature and low N_v values (Figs. 4c and 762 4d), are indicative of an extensively degassed and cooled magma. The presence of the 763 764 mesocrysts (that formed in the shallow reservoir) in the spiny fragments, and their slightly cooler temperature (Fig. 10b), strongly support this interpretation. The spiny fragments likely 765 766 record the slowest ascent velocity and the longest residence time in the reservoir+dyke+vent 767 system compared to the golden/fluidal counterpart. Therefore these fragments are associated with Strombolian events, and decreasing MDR, in agreement with their slower ascent that 768 769 allows extensive syneruptive crystallization.

Among spiny fragments, the opaque ones are the densest, they lack a uniform glassy surface, and they are characterized by i) very high microcrysts content, ii) strong coalescence signature (Fig. 4d), iii) heterogeneous glass chemistry, and iv) mingling with hotter magma at the clast edges (Fig. 8a). All these features reveal the composite nature of these clasts. We interpret the spiny opaque as spiny glass fragments recycled inside the eruptive vent during the explosions, being the densest portion of the magma prone to fall back in the vent/fracture (Fig. 2b).

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778 3) <u>Degassing-driven versus cooling-driven crystallization</u>

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Syn-eruptive degassing is favoured by bubble connectivity/permeability (Figs. 6c and 6d) in 780 the ascending magma, enhanced by syn-eruptive crystallisation in the conduit (especially 781 microcrysts of plagioclase, Fig. 10a), even for magmas at low vesicularity. However, our 782 dataset also supports the occurrence of magma stratification in the reservoir. Textural and 783 784 petrological data demonstrate that the initial activity emitted a small volume of melt (represented by golden and large part of the fluidal fragments) with very scarce crystals. This 785 786 crystal-poor melt was followed in time by the main volume of magma that contains a larger amount of mesocrysts (spiny clasts and lava). Lava flows represent the main volume emitted 787 788 in the 2014 eruption. Mesocrysts are absent in the golden, scarce in the fluidal and more abundant in the spiny (Figs 4b, 4c and 4d) and lava (Fig. 4e) fragments and consist in an equal 789 790 percentage of plagioclase and clinopyroxene and minor olivine. Their composition indicates

that they formed in the reservoir, as shown by their different composition in respect to the 791 microcrysts counterparts that formed during melt degassing in the conduit (Fig. 10a). Most 792 important, a large amount of microcrysts in lava formed in the reservoir as well during 793 magma cooling (Figure 10a). So, we have a range of crystallization conditions. The fact that 794 the lighter plagioclase are not concentrated in the upper and early erupted portion of the 795 reservoir can be due either to the fact that often they are locked in clusters with the 796 clinopyroxene or that this melt was expelled from the crystal-rich portion of the reservoir (see 797 Figure 10b). Water exsolution from the melt can result from its extensive crystallization, 798 799 which induces an increase in dissolved volatile content, up to saturation (second boiling) and can drive melt-crystal separation. 800

801 In conclusion, the crystals in the 2014 fragments do reflect the shallow reservoir 802 conditions and the ascent degassing processes.

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804 <u>4) Textural syn-eruptive versus post fragmentation modifications</u>

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To prove that the 2014 vesiculation of the clasts have been not modified by post 806 fragmentation expansion process, following Stovall et al. (2011), we use a plot of vesicle-to-807 melt ratio (V_G/V_L, after Gardner et al., 1996) and vesicle number density (N_V, Fig. 11). As 808 demonstrated by Stovall et al. (2011), addition of small bubbles leads to an increase in N_V and 809 only a slight increase in V_G/V_L. Bubble growth by some combination of diffusion and 810 decompression leads to an increase in V_G/V_L at constant N_V. N_V decreases while V_G/V_L 811 increases during bubble coalescence, whereas loss of bubbles via collapse or buoyant rise 812 leads to a reduction in both parameters. Intermediate trends on the diagram reflect 813 combinations of more than one of these processes. The pumice and the scoria from the MV of 814 PdF show the highest V_G/V_L, but also the highest Nv, suggesting preservation of small 815 vesicles and growth by some combination of diffusion and decompression. The presence of 816 the small vesicles and the lack of a strong coalescence/expansion signature confirm that the 817 818 weak PdF activity leads to only limited post-fragmentation expansion inside the hot portions of the short-lived fountains. These data contrast with the data from the more energetic 819 820 fountaining events observed at Kilauea or elsewhere, where pre-eruptive information is 821 basically erased because pumice textures are dominated by expansion effects due to their 822 longer residence within the long-lived energetic fountaining. In contrast, the densest, spiny scoriae and the scoria from the Fractures activity show the lowest values of Nv and V_G/V_L , 823 824 due to incipient coalescence and/or loose/lack of small bubbles.

According to previous works (listed above), the golden pumice of PdF should be 825 826 derived from the central part of the fountains, but they do not show the strong post expansion signatures reported by other samples collected from more energetic Hawaiian fountainings 827 (Fig. 11). It is interesting to note that the fluidal fragments at the MV are less smooth (Fig. 4), 828 more vesiculated, and have a lower content of isolated vesicles than the fluidal scoria from the 829 uppermost Fractures (Fig. 6). Therefore fluidal fragments at the 2014 MV could indeed 830 represent clasts that have been partly modified during their residence in the external part of 831 the fountains, while the golden samples could come from the central part (Stovall et al., 2011 832 833 and 2012). However, the slight differences in crystallinity and glass chemistry between the fluidal and golden fragments support the idea that each of these fragments has an imprint from 834 835 the pre-fragmentation setting. In contrast, the spiny fragments from the MV and the fluidal fragments from the Fractures show low N_V and low V_G/V_L in agreement with loss of vesicles 836 837 and coalescence. However, the presence of large numbers of isolated vesicles within the fluidal scoria from the Fractures agrees with their provenance from a fast hot ejection of 838 839 relatively degassed magma (low N_V). In contrast the spiny fragments, especially because of the presence of abundant mesocrysts and increase in syneruptive microcrysts, are indicative of 840 the slowest ascent velocity and extensively degassing and cooled magma. The spiny 841 fragments are the most degassed, densest and the most crystal rich magma that was emitted 842 during low-energy activity by Strombolian explosion, where recycling phenomena were also 843 very frequent (Fig. 2f). 844

Our vesicle connectivity results are in full agreement with the recent review of 845 Colombier et al. (2017b). According to these authors, connectivity values can be used as a 846 useful tool to discriminate between the basaltic scoria from Hawaiian (fire fountaining) and 847 Strombolian activity. The broad range in connectivity for pumice and scoria from fire 848 fountaining is interpreted simply as being due to variations in the time available before 849 quenching due to differences in location and residence time inside the fountain. The fluidal 850 fragments from the WF are the richest in isolated vesicles because they are transported by 851 852 very short lived hot lava jets. In contrast, the higher connectivity observed in scoria from Strombolian activity is probably related to their higher average crystallinity, and more 853 854 extensive degassing prior to the eruption (Colombier et al., 2017b). The spiny surface of these 855 Strombolian fragments is due to the fact that these weak explosions emit only a small solid mass fraction and the partially quenched dense clasts land quickly after a short cooling path 856 through the surrounding atmosphere (e.g. Bombrun et al., 2015). 857

All the clast, from golden to spiny, are very permeable, independent on their 858 859 vesicularity, crystal content and/or of the presence of isolated vesicles. This is in agreement with our interpretation that magma degasses during its ascent in the conduit and that promotes 860 microlite nucleation (see the sodic plagioclase, Fig. 10a) before magma fragmentation (see 861 also Di Muro et al. 2015 with the Pele's hairs and tears samples for the three 2008 eruptions). 862 Moreover, we always find that some of the spiny clasts (especially the opaque ones) are 863 slightly less permeable than the golden and fluidal ones, but not with a low permeability as we 864 865 would expect by their low vesicularity.

866 In conclusion, we can state that i) the crystals lower the percolation threshold and 867 stabilize permeable pathways and ii) this is true for the syn-eruptive sodic plagioclase that 868 favor an efficient degassing in the relatively crystal-rich magma, because of their low wet angles that favor degassing against nucleation (Shea, 2017) and their aspect ratio (e.g. Spina 869 870 et al. 2016) iii) therefore permeability develops during vesiculation through bubble coalescence, which allows efficient volatile transport through connected pathways and 871 872 relieves overpressure (Lindoo et al., 2017). Pervasive crystal networks also deform bubbles and therefore enhance outgassing (Oppenheimer et al., 2015). Based on Saar et al. (2001) 873 crystals should start to affect the behavior of the exsolved volatile phase when they approach 874 20 vol. % (Lindoo et al., 2017). In our dataset, apart from the golden and part of fluidal, all 875 the other clasts do have microcrysts >20%. Our data completely support that slow 876 decompression rate allows more time for degassing-induced crystallization, which lowers the 877 vesicularity threshold at which bubbles start to connect. 878

Rapid re-annealing of pore throats between connected bubbles can happen due to short 879 melt relaxation times (Lindoo et al; 2016). This phenomenology could explain the high 880 amount of isolated vesicles in the fountaining samples. However, vesicle distributions of the 881 882 golden and fluidal fragments are almost perfect Gaussian curves, so it seems that if the relaxation process happens it just merged perfectly with the expected vesicle distribution. In 883 contrast, coalescence and/or expansion (as we observe in the spiny fragments) do not fit the 884 885 curves (Fig. 4). In addition, we should expect that in crystal-poor fragments, due to melt relaxing and pathway closure, the clasts became almost impermeable after quenching, as 886 887 revealed by some petrological experiments performed on crystal-poor basaltic magma (Lindoo et al., 2016). In contrast, in high crystalline magmas, the presence of micro-crystals 888 889 increases viscosity thus preserving the coalesced textures (see Moitra et al., 2013). The isolated vesicle-rich fragments of the 2014 PdF eruption are highly permeable, and are 890 891 characterized by variable ranges of porosity and numbers of vesicles (Fig.4 and Fig. 6d) that seem more related to the pre-eruptive conditions than to the post relaxation of low-viscosity melts. In the 2014 crystal-poor samples, the permeability increases rapidly once the percolation threshold has been reached, and efficient degassing prevents bubble volumes from expanding past the percolation threshold (Rust and Cashman 2011).

In conclusion, also the vesicles in the 2014 fragments do partly reflect the shallowreservoir conditions and mostly the ascent degassing processes.

5.4 Integration between the physical and textural characteristics of the products and their geochemical signature: insight into the feeding system

900 According to Peltier et al. (2016), the June 2014 eruption emitted magma from a shallow pressurized source located only 1.4-1.7 km below the volcano summit. Coppola et al. (2017) 901 suggest that the 2014 event was fed by a single shallow and small volume magma pocket 902 stored in the uppermost part of the PdF central plumbing system. All 2014 clasts show 903 904 homogeneous and evolved bulk compositions, irrespective of their textural features. June 2014 products are among the most evolved products erupted since at least 1998 and are 905 moderately evolved with respect to those emitted in 2010, just before the 2010-2014 906 907 quiescence. Bulk rock and melt inclusion data suggest that the 2014 evolved magma can be produced by crystal fractionation during the long lasting (4.6 years) storage and cooling of the 908 909 magma injected and partly erupted in November 2009. The different types of scoria and pumice emitted in 2014 show significant variations in glass composition (Fig. 8b) due to 910 911 variable degrees of micro-crystallization. In theory, microcrysts can reflect late stage (during magma ascent and post-fragmentation) crystallization. In this case, their variable amount 912 913 within, for instance, the glassy and opaque parts of the spiny scoria might reflect slower ascent velocity or longer residence time in the system (e.g. Hammer et al., 1999, Stovall et al., 914 2012; Gurioli et al., 2014) in agreement also with the vesicle signature. However, the four 915 916 typologies of clasts differ also in terms of mesocryst content (from rare to 5 vol. % for the golden and fluidal and 14-23 vol. % for the glassy spiny and spiny opaque, respectively). 917 Equilibrium plagioclase-melt pairs record an almost constant and moderate dissolved water 918 919 content, intermediate between that expected for melts sitting in the main shallow reservoir (located close to sea level) and the degassed matrix of lavas. Dissolved water contents are 920 921 thus consistent with pre-eruptive magma water degassing during its storage at shallow level, as suggested by geophysical data, and suggest that the plagioclase mesocrysts and some of the 922 microcrysts in the spiny scoria and in the lava grew during magma storage (Fig. 10a). Melt 923

924 composition records a potential pre-eruptive thermal gradient of ~30 °C between the hotter
925 (pumice and fluidal) and the cooler (spiny) magma (Fig. 10b).

Tait et al. (1989) suggest that magma evolution can lead to oversaturation of volatile 926 species within a shallow reservoir and trigger a volcanic eruption. At PdF, the golden and the 927 fluidal clasts might represent the portion of magma located at the top of the shallow reservoir 928 and enriched in bubbles of water rich fluids, released by the cooler, more crystallized and 929 more degassed "spiny-lava" magma (Fig. 10b). The small volume of magma, its constant bulk 930 931 composition and the very small inflation recorded prior to the eruption (Fig. 1d) could be 932 consistent with an internal source of over-pressure related to volatile exsolution. Larger 933 inflation rates over a broader area are expected when shallow reservoir pressurization is 934 related to a new magma input from a deeper source. Slight baseline extensions both on distal and proximal sites suggest that magma transfer towards shallower crustal levels started short 935 936 before (11 days) the final magma eruption. Geochemical data do not support the occurrence of a new magma input in the degassed and cooled 2014 reservoir. We can thus speculate that 937 938 stress field change related to progressive deep magma transfer has promoted volatile exsolution, melt-crystal separation and melt expansion in the shallow reservoir. Textural 939 940 heterogeneity of the 2014 products partly reflects a pre-eruptive physical gradient recorded by the variability in crystal and bubble contents in the shallow reservoir feeding this eruption. 941 The golden and fluidal fragments are the bubble richer and hotter portion of the melt. The 942 spiny fragments are the degassed and cooler portion of the reservoir, whose progressive 943 tapping led to a decrease in explosive intensity (from fountaining to Strombolian activity). 944 Our results are also consistent with processes of mechanical reservoirs/dyke stratification, as 945 observed by Menand and Phillips (2007). As explained earlier, magma ascent promoted 946 947 syneruptive degassing induced crystallization. The spiny opaque clasts can be considered as being recycled material that fell back into the system. Accumulation of olivine crystals out of 948 equilibrium with the host magma produces minor variations in mesocryst contents as 949 observed within the same type of clasts sampled at different times/locations during the 950 951 eruption, with the scoria from the WF and early erupted lava being the ones with the lowest amount of olivine (Table S4 and Fig. 7b). Again, this temporal variation supports an increase 952 953 in large heavy crystals within the most degassed magma emitted toward the end of activity, 954 further suggesting that it corresponds to the lower part of the reservoir.

Our dataset permits us to propose that the 2014 eruption was fed by a physically zoned magma reservoir. The low-density, crystal-poor, bubble-rich magma located in the upper part of the storage system, ascended first, rapidly and fed the early, more energetic phase, the

Hawaiian fountaining. This low-density magma is not more evolved than the spiny one (same 958 959 bulk compositions) and it is not necessarily richer in dissolved volatile amounts; it is just poorer in crystal and richer in bubbles. Second boiling, possibly triggered a few days before 960 the eruption by stress field change, is responsible of the extraction of bubble rich melt from a 961 crystal-rich network. This last one will represent the main volume of the erupted lava. Fast 962 ascent of the foam hinders its crystallization and preserves high number of vesicles, high 963 vesicularity and it is only little modified by post-fragmentation expansion. Decrease in initial 964 965 overpressure translates in a progressive decrease in magma ascent rate and output rate (e.g. 966 Coppola et al., 2017 and references therein). Nucleation of microcrysts is enhanced in melt 967 ascending with lower speed and is mostly related to syneruptive degassing (for the spiny). The 968 larger volume (dense lava) corresponds to crystallized and less vesiculated magma which experiences a slow ascent in the dyke and even further micro-crystallisation during its 969 970 subaerial emplacement.

971 Melt inclusion results allow us to confirm the involvement of a single and only slightly 972 heterogeneous magma source in 2014, related to cooling and fractional crystallisation of an 973 older magma batch (November 2009). Interestingly, this latter short lived summit eruption 974 was also characterized by the same large textural range of pyroclastic products found in 2014 975 in spite of its more mafic composition.

This suggests that bubble accumulation and source pressurisation is highly dependent on the shallow storage depth, which facilitates rapid water exsolution (Di Muro et al., 2016), and it is not necessarily the outcome of slow magma cooling and differentiation (Tait et al., 1989).

980 6. Proposed model for the 2014 eruption and conclusions

In this paper we show that textural and petro-chemical study of the eruptive products can be used to characterize the on-going activity at PdF and to constrain both the trigger and the evolution of short-lived and small-volume eruptions. This approach is extremely valuable in i) understanding processes that lead to an eruption which was preceded by short-lived and elusive precursors, and ii) in reconstructing the time evolution of eruptive dynamics in an eruption with poor direct observations.

Following the sketch in Figure 12, we infer that residual magma from the 2009 eruption ponding at shallow levels experienced long-lasting cooling and crystallization (Fig. 12a). Between 2010 and 2014 the volcano progressively deflated (Fig. 12b) possibly because of magma degassing and cooling, facilitated by the shallow depth of the reservoir. During thisphase mesocrysts and some microcrysts formed (Figs. 4e and 10a).

The occurrence of deep (>10 km bsl) lateral magma transfer since March-April 2014 992 has been inferred by Boudoire et al., (2017) on the basis of deep (mantle level) seismic 993 swarms and increase in soil CO₂ emissions on the distal western volcano flank. The incipit of 994 magma transfer towards shallower crustal levels is potentially recorded by subtle volcano 995 inflation about 11 days before the June 2014 eruptions (Figs. 1d and 12c). We suspect that 996 997 these deep processes can have progressively modified the shallow crustal stress field and favoured magma vesiculation and melt-crystal separation. Second boiling could thus have 998 999 over-pressured the shallow seated reservoir and triggered magma ascent (Fig. 12c).

1000 Without this deep magma transfers we believe that the small reservoir activated in 2014 would have cooled down completely to form an intrusion (as suggested by the pervasive 1001 1002 crystallization of the lava, one of the densest emitted from 2014 to 2017, Harris et al. 2017). 1003 The 2014 event represented instead the first of a long series of eruptions, whose magmas 1004 became progressively less evolved in time (Coppola et al., 2017). In this scenario the trigger 1005 mechanisms of 2014 activity are both internal and external in the sense that the small shallow 1006 reservoir hosting cooled magma permitted to create the conditions favourable to a second 1007 boiling (Fig. 12c, and Tait et al., 1989). The second boiling was likely triggered by an almost undetectable stress field change, and was favoured by the shallow storage pressure of the 1008 magma (Fig. 12c) that promoted fast water exsolution and rapid magma response to external 1009 triggers. The second boiling possibly contributed to the inflation registered 11 days before the 1010 eruption at 1.4-1.7 km (Fig. 12c) caused both by magma expansion and transfer of hot fluids 1011 1012 to the hydrothermal system (Lénat et al., 2011).

1013 Our data permit to exclude (i) new magma input and/or fluid inputs (CO2-rich fluids) 1014 from deep magmatic levels to trigger the June 2014 eruption. We also exclude (ii) heating and 1015 enhanced convection of the shallow magma reservoir (due to heat diffusion without fluid or mass transfer), because this process is very slow. Furthermore, the 2014 minerals do not 1016 1017 record evidences of magma heating. We can exclude equally (iii) deformation of the volcanic edifice and decompression of the magma reservoir and/or hydrothermal system due to flank 1018 1019 sliding because geodetic data show no evidence of flank sliding able to produce stress change 1020 in the hydrothermal and magmatic system. Geophysical and geochemical data have permitted 1021 to track vertical magma and fluid transfer below the volcano summit in April 2015, that is about one year after the early deep lateral magma transfer (Peltier et al., 2016). Deep 1022 1023 processes are difficult to detect for any monitoring network.

We conclude that the overpressure, caused by the second boiling, triggered the 1024 1025 eruption. The occurrence of a hydrous almost pure melt at shallow depth permitted its fast 1026 vesiculation upon ascent towards the surface. In turn, fast ascent of the foam (Fig. 12d) hindered its crystallization and preserved high number of vesicles. Decrease in initial 1027 1028 overpressure translated in a progressive decrease in magma ascent rate and output rate (e.g. 1029 Coppola et al., 2017 and references therein) and a temporal transition from Hawaiian activity to Strombolian activity (Fig. 12 d). Nucleation of microcrysts was enhanced in melt ascending 1030 1031 with lower speed and in turn this syn-eruptive crystallization favoured bubble 1032 connectivity/permeability in the ascending magma, even for magma at low vesicularity. The 1033 largest volume (dense lava) corresponds to highly-crystallized and degassed magma already 1034 in the reservoir, that experienced a slower ascent in the dyke and even further micro-1035 crystallisation during its subaerial emplacement.

1036 The texture of the products allowed us to follow the dynamic evolution of the system 1037 in space, from smooth fluidal scoria emitted from rapid jet of lava at the fractures, to a more 1038 stable activity at the MV, and in time. At the MV, in fact, we observed the transition from the 1039 golden and fluidal fragments emitted from Hawaiian fountaining, at the peak of the intensity 1040 of the eruption, to the spiny fragments, emitted from a declining Strombolian activity at the 1041 end of the eruption.

1042 Therefore we here show for the first time that short lived and weak Hawaiian 1043 fountaining and Strombolian events can preserve pyroclast textures that can be considered a 1044 valid approximation to shallow reservoir conditions and ascent degassing processes before the 1045 explosions and correlate to the eruptive dynamics as well.

1046 To conclude, these results highlight the importance of petrological monitoring, which 1047 can provide complementary information regarding the ongoing volcanic activity to other 1048 geophysical and geochemical monitoring tools commonly used on volcanoes.

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Figure 1 a) Digital elevation model of the summit crater area at Piton de la Fournaise, La Réunion, France; orange = fractures generated by pre-2014 eruptions (reported are the dates of their activities); b) red = fractures active during the 2014 eruption: WF (Western Fracture), UF (Upper Fracture), LF (Lower Fracture), MV (Main Vent). Black= outline of the 2014 lava field; c) locations of sample collection points. The coordinates are in UTM, zone 40 south. (d) Distance change (baseline) in centimetres between two GNSS summit stations: DSRG and SNEG (see location in the inset). Increase and decrease of the signal mean a summit inflation and deflation, respectively. The yellow areas represent eruptive and intrusive periods. In Figure 1d, the rapid and strong variations linked to dike injections preceding intrusions and eruptions by a few tens of minutes have been removed; (e) Digital Elevation Model of La Réunion island.

1409

June 2014 eruption at PdF

Early morning, June 21



June 21 ~ 7h00



June 21, 7h38

June 21, 13h35





June 21,17h00





1412	Figure 2 Photos collection from the 2014 eruption at the MV, highlighted with a white cross
1413	(see location in Fig. 1). From a to g: evolution of the Strombolian activity from early morning
1414	to evening, June 21 that shows a decline in the activity with time. Unfortunately, the relatively
1415	more energetic Hawaiian fountaining events that happened during the night were not
1416	documented. a) Strombolian activity at the MV and associated lava flow; b) zoom view of the
1417	Strombolian activity at the MV. The images in a, b and the inset in b are from Laurent Perrier;
1418	c) aerial view of the SE flank of the PdF, taken by the OVPF team from the helicopter of the
1419	gendarmerie of La Réunion; d) Eastern front of the lava where the OVPF team collected a
1420	quenched lava sample; e) low Strombolian activity at the MV and the associated lava flow,
1421	photo from: <u>http://www.ipreunion.com/volcan/reportage/2014/06/21/eruption-du-piton-de-la-</u>
1422	fournaise-actualise-a-17h-la-lave-coule-sur-1-5-kilometre,26023.html; f) and g) decline of the
1423	Strombolian activity at the MV, the photo in e) is from <u>http://www.zinfos974.com/L-</u>
1424	eruption-du-Piton-de-la-Fournaise-Le-point-de 17h_a72981.html; and the photo if f) is from:
1425	f) <u>http://nancyroc.com/eruption-a-la-reunion</u>



Figure 3 a) Continuous blanket of scoria fall out deposit emitted from the MV (Fig. 1 for location) during June 2014 eruption at PdF. The black cross locates the position of the MV (see Fig. 1 for the location); b) schematic stratigraphic log of the scoria fall out deposit emplaced during June 2014 eruption at the MV. c) grain size histograms of the base and the top of the deposit of the MV, the particle diameters are at half phi; d) scattered scoria (outlined in yellow) from the WF (see Fig. 1 for the location); e) grain size histogram of the scoria deposit at the WF, the particle diameters are at half phi; f) comparison between the grain size histograms for the 2010 Hawaiian fountaining and the 2014 MV activity, both the particle axes are reported in full phi for comparison.

Туре	Clast	Thin section	Microscope	SEM (25X)	VSD	Crystal vol %	N _v
Golden Pumice (a)			0.02 mm		25 20 n = 2 15 10 5 0 0 0 0 0 0 0 0 0 0 0 0 0	Tot = 8-15 Mplg= rare μ plg = 6-11 Mcpx = rare μ cpx =(1-3)	2x10 ⁷ 9x10 ⁶
Fluidal Scoria (b)	WF MV		0.02 mm		n = 3	Tot = 4-23 Mplg = 0.4-1 μplg = 2-19 Mcpx = 0-1 μcpx = 1-4	2x10 ⁷ 5x10 ⁶ 3x10 ⁶
Spiny glassy scoria (c)			0.02 mm		¹⁵ 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	Tot = 51 Mplg = 11 µplg = 23 Mcpx =15 µcpx = 2	6x10 ⁶
Spiny opaque scoria (d)	T cm	(Marine)			¹⁵ ¹⁰ ¹⁰ ¹⁰ ¹⁰ ¹⁰ ¹⁰ ¹⁰ ¹⁰	Tot = 55 Mplg = 11 µplg = 25 Mcpx = 10 µcpx = 9	4x10 ⁶
Lava (e)	The second secon	<u>а</u>	0.02 mm	l mm	n = 1 n = 1 0 0 0 0 0 0 0 0 0 0 0 0 0	Tot = 100 Mplg = 2 µplg = 64 Mcpx = 3 µcpx = 31	2x10 ⁴

Figure 4 Textural features of June 2014 pyroclasts and lava. Clast = photo of the different types of juvenile pyroclasts and lava channel. The 1456 photo of the lava channel is from Laurent Perrier. WF = Western Fracture (smooth fluidal scoria), MV = Main Vent (fluidal scoria, less smooth 1457 than the ones at the WF). Thin section = thin section imaged with a desktop scanner. Microscope = picture taken with an optical microscope 1458 using natural light; SEM (25X) = image captured using a scanning electron microscopy (SEM), in BSE mode at 25x magnification: black are 1459 vesicles, white is glass, grey are crystals. VSD = vesicle size distribution histograms, where the diameter, in mm, is plotted versus the volume 1460 percentage, n = number of measured clasts; Crystal vol. % : Tot = total percentage of crystals corrected for the vesicularity; Mplg = percentage 1461 of mesocrysts of plagioclase; $\mu plg = percentage$ of microcrysts of plagioclase; Mcpx = percentage of mesocrysts of pyroxene; $\mu cpx = percentage$ 1462 of microcrysts of pyroxene; Nv = number density corrected for the vesicularity. 1463

1464



Figure 5 Proportion of each type of clast measured from the base to the top of the 10 cm thick deposit emplaced during the eruption, at the MV site. The deposit is dominated by Hawaiianlike lapilli fragments at the base (golden pumice and fluidal scoria) and Strombolian-like bombs and lapilli at the top (spiny scoria): (a) componentry within the different grain size classes; b) normalized componentry composition from the base to the top of the deposit.





1482

Figure 6 Density, connectivity and permeability data of June 2014 pyroclast and lava 1483 1484 fragments: a) density distribution histogram for all the pyroclast fragments measured at the MV + two lava fragments collected from the Eastern front of the lava flow (see Fig. 1 for 1485 location). n = number of measured clasts; b) density distribution histogram for the pyroclasts 1486 sampled at the WF and the bomb sampled at the UF. The bomb broke in five fragments (2 1487 fragments from the core, the least dense, and three fragments from the quenched edges, the 1488 1489 densest). In both the density histograms the stars represent the density intervals from which we picked the clasts for the textural measurements; c) graph of the connected vesicularity 1490 versus total vesicularity. The diagonal line represents equality between the connectivity and 1491 vesicularity, beneath this line the samples have isolated vesicles and the straight lines 1492 represent lines of equal fraction of isolated vesicles. To note that the bomb from the UF has 1493 1494 the high vesicular core with less than 5% of isolated vesicles, while the three low vesicular fragments from the quenched edge have more than 25% of isolated vesicles (see pink spots); 1495 d) Darcian viscous permeability (k_1) versus vesicularity fraction for the four typologies of 1496 clasts collected at the MV. For comparison, two fluidal fragments and one spiny opaque 1497 fragments from February 2015 eruption are reported. 1498



Figure 7 Ni-Cr concentration plot. (a) Ni-Cr signature of the June 2014 lavas compared to 1501 that of recent eruptions (Di Muro et al. (2015) and unpublished data). Whole-rock (circles) 1502 1503 and glass (triangles) compositions are shown for the June 2014 eruption. Olivine controlled lines are indicated for olivine hosting 1.2 and 0.6 wt.% Cr-spinel. Compositions used for 1504 1505 olivine (Ni=1900 ppm, Cr=300ppm), clinopyroxene (Ni=970 ppm, Cr=4800 ppm), and Cr spinel (Ni=1500 ppm, Cr=25%) are inferred from Welsch et al. (2009), Salaün et al. (2010), 1506 1507 and Di Muro et al. (2015). (b) Zoom of the Ni-Cr relationship between glass (triangles) and whole-rock (circles) samples from the June 2014 eruption. Fracture I = Western Fracture, 1508 1509 Fracture II = Upper Fracture. Careful sample selection has permitted to obtain a set of virtually olivine-cpx free crystals. Any addition of mafic crystals translates into enrichment in 1510 Ni-Cr; those samples that contain a few % of crystals (consistent with textural and 1511 1512 petrological observation) are slightly enriched in compatible elements.

1513



Fig. 8 (a) Evolution of CaO/Al₂O₃ ratio in the matrix glasses of recent eruptions at Piton de la Fournaise as a function of MgO content (directly proportional to melt temperature). MI = Melt inclusions (grey area for the 2014 samples). (b) CaO versus MgO content for Piton de la Fournaise products. WR = whole rock, GM = ground mass; MI = melt inclusion, EM = embayment glass



1523 Figure 9 FeO_T in melt inclusions as function of Fo content of the olivine host for recent
1524 eruptions at Piton de la Fournaise
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Figure 10 a) Anorthite versus Albite compositions for the plagioclase crystals measured for June 2014 eruption of PdF; b) Temperature, composition (K_2O) and dissolved water content (H_2O) for the evolution of 2007-2014 melts from glasses. The data have been obtained by

studying the glass-plagioclase equilibrium or on the basis of matrix glass analyses. Temperature estimation based on the MgO-thermometer of Helz and Thornber (1987) modified by Putirka (2008). Water content is from the plagioclase hygrometer of Lange et al. (2009). Only plagioclases in equilibrium with melts are considered, following the procedure described by Putirka (2008) for >1050°C melts (Kd = 0.27 ± 0.05). Error bars reported in Figure 10b correspond to the standard deviation of the plagioclase dataset, whose range is larger than error of the method. We stress that the reported temperatures are obtained using Helz dry model. Further uncertainty arises from the dependence of the method on dissolved water content as shown recently by Putirka (2008). In order to minimize the number of assumptions and perform a comparison between distinct eruptions, we preferred to adopt the dry model.



1573 Figure 11 Volumetric ratio of vesicles to melt (V_G/V_L) versus vesicle number density



Figure 12 Schematic model of the evolution of PdF volcanic system from the new deep magmatic input of November 2009 up to June 2014
eruption. See explanation in the text