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1 **Variability of ash deposits at Piton de la Fournaise (La Reunion Island): insights into fragmentation**  
2 **processes at basaltic shield volcanoes**

3

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20 **Key-words**

21 Ash; basaltic; fragmentation; grain size; Piton de la Fournaise; texture

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24

25 **Abstract**

26 It is commonly accepted that effusive activity emplaces the main emitted magmatic volume in basaltic  
27 shield volcanoes. At Piton de La Fournaise (La Réunion Island, France), eruptive activity occurs mostly within the  
28 non-populated Enclos Fouqué caldera and generally does not pose any risk to the population. However, historical  
29 observations, recent monitoring data and field work on tephra deposits suggest that some eruptions have produced  
30 unusual and unexpected explosive phases. A comprehensive sampling of tephra from major historical and recent  
31 eruptions on this volcano allowed us to perform systematic componentry, grain size, chemical and morphological  
32 analyses in order to characterize the eruptive dynamics involved in these explosive basaltic eruptions. This  
33 integrative approach reveals highly variable characteristics of the studied tephra reflecting different fragmentation  
34 efficiencies and multiple associated mechanisms. Primary ductile and partially brittle fragmentation of various  
35 intensity of juvenile magma emitted during Hawaiian fountaining or mild Strombolian explosions were identified  
36 as the most common fragmentation mechanisms, in particular during the 2014-2018 dominantly effusive eruptive  
37 sequence. In parallel, we distinguished more efficient short-lived fragmentation events related to (i) plug  
38 pressurization and brittle fragmentation enhanced by syn-eruptive crystallization, (ii) magma/water interactions,  
39 (iii) rare phreatic explosions and (iv) secondary fragmentation producing fine ash during caldera collapse. We  
40 conclude that textural, geochemical and morphological analysis make it possible to identify and characterize the  
41 variability in eruptive processes, with the grain size and the componentry of the ash particles being probably the  
42 most important parameters to quantify both the efficiency and the nature of the fragmentation.

43

44 **1. Introduction and objectives**

45 Basaltic shield volcanoes such as Kilauea (Hawai'i Island, USA) and Piton de la Fournaise (La Réunion  
46 Island, France; noted hereafter as PdF), are dominated by effusive activity often associated with low-energy  
47 explosive Hawaiian to Strombolian eruptions (e.g. Houghton et al. 2016; Gurioli et al. 2018; Thivet et al. 2020a,  
48 2020b). However, field evidences and recent syn-eruptive observations at basaltic volcanoes also highlight more  
49 explosive events capable of producing fine-grained tephra, for instance at Etna (e.g. Taddeucci et al. 2002, 2004;  
50 Andronico et al. 2008; Corsaro et al. 2017; Polacci et al. 2019), Stromboli (e.g. Ripepe et al. 2005; Polacci et al.  
51 2009; Lautze et al. 2012), Kilauea (e.g. Dvorak 1992; Mastin 1997; Neal et al. 2019) and Piton de la Fournaise  
52 (e.g. Michon et al. 2007, 2013; Staudacher et al. 2009, 2016; Ort et al. 2016). Ash-forming events, with their  
53 widespread impacts, represent a different hazard from effusive activity. Ash eruptions can impact the areas close

54 to the vents where touristic activity is frequent, as well as the communities living downwind, leading to e.g. adverse  
55 health effects for humans and animals, damage to crops, infrastructure and networks disruption, water  
56 contamination (Horwell and Baxter 2006; Wilson et al. 2015). They also affect the airspace used by aviation (e.g.  
57 Song et al. 2014; Guffanti and Tupper 2015). Ash-forming events are highly unexpected in basaltic settings and  
58 the mechanisms controlling their occurrence are poorly constrained, making their forecasting challenging. To  
59 improve our ability to predict such events and mitigate their impacts, it is thus essential to understand (i) what type  
60 of fragmentation processes can produce ash (e.g. magmatic, phreatomagmatic, phreatic or volcano-tectonic), and  
61 (ii) what are the characteristics of the associated deposits (e.g. componentry, texture, grain size, morphology).

62 Here, we investigate a range of fragmentation mechanisms that can occur at basaltic shield volcanoes,  
63 through the comprehensive study of several tephra deposits composed of lapilli (particle diameters between 64  
64 and 2 mm) and ash (< 2 mm, with fine ash particles < 63  $\mu\text{m}$ , according to the Schmid 1981 nomenclature). These  
65 deposits, originated from prehistorical, historical and recent eruptions at Piton de la Fournaise (PdF), and cover a  
66 wide range of eruption styles. Based on the phenomenology of the studied events as well as the grain size,  
67 componentry, texture, morphology and chemical analysis of the tephra particles, this work brings new insights into  
68 the different eruptive mechanisms that can lead to ash-generating events at basaltic volcanoes. We used grain size  
69 and morphology analyses to constrain the fragmentation mechanisms controlling the studied events, and  
70 componentry analyses to assess the role of juvenile magma and identify the key textural features depicting the  
71 origin of both juvenile and non-juvenile components. Finally, chemical analysis provided us constraints on magma  
72 composition as well as pre- and syn-eruptive conditions.

73

## 74 **2. Case studies and field sampling**

75 PdF is the currently active volcano of the Reunion Island (Fig. 1a), situated in the Indian Ocean,  
76 approximately 750 km to the East of Madagascar. This volcano is one of the most active hotspot shield volcanoes  
77 in the world, with two eruptions per year on average (Peltier et al. 2018). The main and most recent (< 5000 years)  
78 volcano-tectonic structure of PdF is the Enclos Fouqué caldera (Fig. 1b). This horseshoe-shaped depression is 250  
79 m deep, 13 km long in the E-W axis and 6 to 8 km wide in the N-S axis (Michon and Saint-Ange 2008). Since the  
80 settling of permanent population on La Réunion Island (second part of the 17<sup>th</sup> century), the volcanic activity is  
81 mainly concentrated within this structure (e.g. Villeneuve and Bachèlery 2006; Staudacher et al. 2016; Peltier et  
82 al. 2018).

83            Sampling of all the deposits presented in this study was performed during several field campaigns. All  
84 the samples studied here are located in Figure 1b and listed in Figure 1c, in which some of the deposit  
85 characteristics are summarized.

86            Sampling of deposits from recent eruptions (part of routine sampling performed by the Observatoire  
87 Volcanologique du Piton de la Fournaise (OVPF) for each recent eruptive event, represented by green squares in  
88 Fig.1b) is either done during the eruptive activity and close to the source or just after the eruption, to be able to  
89 access strategic and very proximal deposits (Gurioli et al. 2018; Thivet et al. 2020a, 2020b). Other sampling was  
90 performed on older and key deposits representative of explosive events at PdF (orange squares in Fig. 1b). All  
91 eruptions/deposits studied here are classified by style and phenomenology (Figs. 2 and 3), which are inferred from  
92 observational data (recent eruptive events) and reviews of the literature (older events).

### 93 **2.1. Historical and recent Hawaiian fountaining activity**

94            Tephra produced during Hawaiian fountaining activity are the first type of samples described in this study.  
95 We collected (i) deposits emplaced between 2014 and 2018 (fountain heights of 30 to 100 m) and (ii) the Black  
96 Tephra Deposits (1450 to 1630 AD) that were emitted from Piton Chisny, a scoria cone situated outside the Enclos  
97 Fouqué caldera (Fig. 1b), likely during intense Hawaiian fountaining activity (fountain heights of hundreds of  
98 meters; Morandi et al. 2016).

99            (i) During the recent fissure-fed eruptions at PdF, Hawaiian fountaining phases are generally observed at  
100 the beginning of the eruptive activity, when the lava and gas flux are relatively high (Coppola et al. 2017; Thivet  
101 et al. 2020b). Figure 2a shows the sustained Hawaiian fountaining (height of around 50 m) which occurred during  
102 the entire duration of the September 2016 eruption. Two months after the eruption, the whole sequence of this  
103 Hawaiian fountaining was sampled a 100 m from the main active vent. We also added samples from five additional  
104 eruptions that happened during the 2014-2018 period and were collected during the fountaining (with plastic sheets  
105 on the ground for February, May and July 2015 eruptions) or a few months after the eruptions, when the deposits  
106 were finally accessible, such as the June 2014 eruption (Gurioli et al. 2018). All these deposits were sampled  
107 approximately 100 m from the eruptive vents.

108            (ii) The entire stratigraphic sequence (3 m thick) of the last Piton Chisny activity (1450 to 1630 AD)  
109 called the Black Tephra Deposit (Morandi et al. 2016; Principe et al. 2016) was sampled (Fig. S1a). The two  
110 samples shown in this study (Fig. 2b) represent the initial or opening phase (tephra layer named CH1, 5 to 10 cm

111 thick) and a relatively steady phase of long-lived lava fountaining (tephra layer named CH5, 20 cm thick  
112 homogeneous bed of scoria lapilli between two bomb-rich layers).

## 113 **2.2. Recent mild Strombolian and transient explosions**

114 The second type of samples described in this study are tephra emitted during (i) mild Strombolian  
115 eruptions (e.g. July and September 2018 eruptions) and (ii) more intense transient explosions (September 2016  
116 eruption; Thivet et al. 2020a).

117 (i) During the recent fissure-fed eruptions at PdF, mild Strombolian activity (bubble bursting at the  
118 magma free surface) is generally observed a few hours or days after the beginning of the eruptive activity, when  
119 the lava and gas fluxes are progressively decreasing (Thivet et al. 2020b). This mild Strombolian activity usually  
120 occurs as discrete explosions ejecting pyroclasts to heights less than 10 m (Gurioli et al. 2018) and ends with  
121 decreasingly energetic spattering. The samples from the July (Fig. 2c) and September 2018 eruptions, which lasted  
122 around 1 and 47 days respectively, were collected 50 m from the vents, one day after the end of the Strombolian  
123 phases.

124 (ii) The final day of the one week long eruption of September 2016, mostly dominated by sustained lava  
125 fountaining, was characterized by a shift in activity (Thivet et al. 2020a): one of the active vents turned from mild  
126 ejections and slight degassing to relatively more intense, transient bomb- and lapilli-dominated explosions (one  
127 explosion every 10 to 20 seconds producing both incandescent ejections up to 30 m high, yellow circle labeled 1  
128 in Figure 2d) associated with synchronous sporadic emission of dark ash-dominated plumes (up to 300 m high,  
129 the ash particles being transported to proximal areas downwind, yellow circle labeled 2 in Figure 2d). Two months  
130 after the eruption, the whole sequence of deposits from the intense explosions and ash plumes was collected from  
131 a 1 m thick log, approximately 150 m from the explosive vent.

## 132 **2.3. The April 2007 explosions caused by lava flow entry in the sea**

133 During the caldera-forming April 2007 eruption (Piton Tremblet, source of emission only 600 m above  
134 sea level and only 3 km from the coastline; Fig. 1b), relatively high lava flux (maximum of  $200 \text{ m}^3 \text{ s}^{-1}$ , Staudacher  
135 et al. 2009) was associated with intense Hawaiian fountaining activity (lava fountain heights up to 200 m). This  
136 intense activity (from April 2 to May 1) supplied a large lava flow that rapidly reached the ocean (Michon et al.  
137 2007, 2011), forming a new emerged lava platform of around  $5 \times 10^5 \text{ m}^2$  at the end of the eruption. The interaction  
138 between seawater and the hot lava generated multiple explosions, characterized by the emission of steam clouds

139 rich in acidic aerosols and glass shards (laze) which are the third type of samples described in this study (Fig. 3a).  
140 The related tephra deposit (5 mm thick layer emplaced on a clean surface, near the Tremblet village) was collected  
141 on the day of emission (May 5), 1 kilometer from the ocean entry.

#### 142 **2.4. The April 2007 caldera collapse**

143 Tephra can also be produced during caldera (or pit-crater) collapse events that originate from paroxysmal  
144 eruptions. Tephra deposits related to the April 2007 caldera collapse event are studied here. The intense April 2007  
145 eruption drained the shallow magmatic system, situated ~2 km below the summit cone (Di Muro et al. 2014). On  
146 April 5 at 8:48pm UTC, a major collapse occurred which was followed by numerous but weaker collapses until  
147 April 15 (Fontaine et al. 2014), producing a 340 m deep depression of a total volume of  $96 \times 10^6 \text{ m}^3$  (Urai et al.  
148 2007) at the summit. After the main collapse associated with ash emission (Fig. 3b), several lava flows occurred  
149 on the newly formed inner caldera walls (Staudacher et al. 2009). The emitted ash mostly dispersed within the  
150 Enclos Fouqué caldera (Staudacher et al. 2009; Michon et al. 2011, 2013). In this study, we focus on a sample  
151 collected 1.5 km from the source (Fig. 3b) on the western part of the Enclos Fouqué caldera on a clean solar panel  
152 surface of an OVPF station (named “La Dalle”) in order to avoid any contamination. At the time of sampling the  
153 ash thickness was approximately 4 mm.

#### 154 **2.5. The March 1860 violent Strombolian eruption**

155 Two samples representative of the first phase of the March 1860 summit violent Strombolian eruption  
156 were collected within the summit area of PdF, and within the stratigraphic sequence (Fig. S1b) described by  
157 Michon et al. (2013). The drawing shown in Figure 3c was realized by Velain (1878) a few years after the eruption  
158 and represents the Brûlant crater which was situated on the Eastern part of the summit area where the Dolomieu  
159 caldera is situated today. After overflow then draining of an active lava lake located within the Brûlant crater  
160 (Lénat et al. 2016), several eruptive fissures opened on the flank of the volcano in February and March 1860.  
161 Summit explosions producing ash-rich plumes and block ejections were observed on March 19, 1860. The Brûlant  
162 crater was partially eroded and widened by about 300 m. The products of this paroxysmal event were dispersed  
163 over all the PdF edifice (Villeneuve and Bachèlery 2006), but today, the deposits from this eruption are only found  
164 in the summit area of the central cone. Here, we focus on two samples (Fig. 3c) representative of the relatively  
165 fine-grained matrix (lapilli and coarse ash) of the first half of the coarse-grained (bombs, blocks and lapilli) deposit  
166 sequence (Fig. S1b), collected approximately 500 m from the Brûlant crater (REU131112 at the base and PF\_2 at  
167 the top).

## 168 **2.6. Prehistorical ash-rich deposits related to the formation of the Enclos Fouqué caldera**

169 According to Michon et al. (2016) and Ort et al. (2016), the Enclos Fouqué caldera may have formed  
170 during successive mass-sliding events towards the East. These authors have related the caldera forming events  
171 with the Bellecombe Tephra (4900 to 2250 BP), a thick and widespread deposit that can be found all over the PdF  
172 edifice. Ort et al. (2016) divided the stratigraphic sequence of the Bellecombe Tephra into two main units. The  
173 Lower Bellecombe Tephra (around 60 cm thick in a proximal outcrop named “Petite Carrière”) may be related to  
174 the opening of steeply dipping fissures tapping a hydrothermal and/or magmatic system located under the newly  
175 formed Enclos Fouqué caldera. The upper Bellecombe Tephra (around 210 cm thick) is related to a fissure opening  
176 linked to a relative more mature hydrothermal and/or magmatic reservoir, initially emplaced westwards within the  
177 Plaine des Sables area. Two samples were collected within the lower unit and two other ones within the upper unit  
178 (Figs. 3d and S2).

179

## 180 **3. Laboratory methods**

### 181 **3.1. Grain size**

182 Grain size analyses were performed on all the studied deposits. Beforehand, the samples were dried in the  
183 oven at 90 °C during 24 to 48 hours. Manual sieving (grain size bins in wt%) at a  $\frac{1}{2} \Phi$  scale between  $-6 \Phi$  (64 mm,  
184  $\Phi = -\log_2(d \text{ in mm})$ ) and  $5 \Phi$  (32  $\mu\text{m}$ ) was carried out following the procedure described in Gurioli et al. (2018).  
185 When observed, particle clusters were dismantled either by hand or using ultrasonic cleaning baths. Samples rich  
186 in fine ash were analyzed by laser diffraction using a Malvern Mastersizer 3000 (grain size bins in vol%) in an  
187 aqueous solution, in order to quantify the Particle Size Distribution (PSD) of the sample fraction below 63  $\mu\text{m}$   
188 through the application of the Mie theory (Riley et al. 2003), which allow to measure the equivalent particle  
189 diameters, regardless of the real particle shapes. Raw grain size data are available in the supplementary material  
190 (Table S1). In order to combine the two grain size methods (Eycheenne et al. 2012), (i) we assumed that grain  
191 density variations below 63  $\mu\text{m}$  are negligible and that consequently vol% and wt% are equivalent (no porosity  
192 variations were observed, and the components with contrasting densities are evenly distributed though the ash  
193 fraction), and (ii) we normalized the laser data to the mass proportion of sample finer than 63  $\mu\text{m}$ .

### 194 **3.2. Componentry, texture and chemical analysis**



195 For each sample, componentry analyses were performed on one specific grain size fraction, depending  
196 on the sample PSDs (cf. Figs 2 and 3), in order to distinguish juvenile from non-juvenile particles (White and  
197 Houghton 2006) based on their surface texture and morphology (angularity and roughness), porosity (high vs. low  
198 vesicle content), crystal assemblage (presence of magmatic glass and/or microlites and/or phenocrysts) and degree  
199 of alteration (fresh vs. altered and/or eroded). Textural and componentry analyses were performed (i) on the 500-  
200 710 grain size fraction for coarse deposits, and (ii) on finer fractions close to PSD modes for fine deposits. At least  
201 200 particles were counted in each fraction, using a binocular microscope (for lapilli) and Scanning Electron  
202 Microscopy (SEM) (for ash) on epoxy-impregnated polished sections. Internal textures of the samples were  
203 investigated by Backscattered Electron (BSE) imagery using both the JEOL JSM-5910 LV SEM and the Cameca  
204 SxFiveTactis Electron Probe Micro Analysis (EPMA) at LMV, with an acceleration voltage of 15 kV.

205 When applicable, vesicle and crystal contents were estimated based on the analysis of 20 particles on  
206 average. For the September 2016 eruption, detailed textural analysis was previously performed by Thivet et al.  
207 (2020a). Juvenile particles are characterized by different groundmass textures: in the results section, they will be  
208 described as sideromelane (glassy and transparent, with possible presence of microlites) or tachylite (fully  
209 microcrystalline and opaque) particles.

210 Some chemical analyses were also performed on a few samples (Table S2) by in-situ glass analyses with  
211 Cameca SxFiveTactis EPMA (acceleration voltage of 15 kV and a current intensity of 8 nA) following the  
212 procedure described in Gurioli et al. (2018). For some key ash samples, mineral phases were identified by X-ray  
213 diffraction (XRD) analysis with an Empyrean diffractometer, operated at 43 kV and 38 mA, with the use of  $K\alpha$   
214 radiation from a Cu anode. Inorganic Crystal Structure (ICSD) and Crystallography Open (COD) databases were  
215 used to identify the mineral phases. The ash samples were analyzed from 2 to 50 degrees  $2\theta$ , with a step width of  
216 0.02 degrees  $2\theta$  and two second counting time for each step.

### 217 **3.3. Ash morphology**

218 2D ash morphology was quantified using the automated Malvern Morpho-Grainsizer Morphologi G3  
219 following the method developed by Leibbrandt and Le Pennec (2015) and adapted by Thivet et al. (2020a). Before  
220 the analysis, specific grain size fractions were selected within  $\frac{1}{4} \Phi$  bins from  $0.75 \Phi$  (600  $\mu\text{m}$ ) to  $4 \Phi$  (32  $\mu\text{m}$ ),  
221 depending on the deposit grain size. The narrow  $\frac{1}{4} \Phi$  range favors the grain size homogeneity for the  
222 morphological analysis. When possible, we performed the morphology measurements on similar grain size  
223 fractions from one deposit to another to allow data comparison without grain size bias. Several grain size fractions

224 were also analyzed within the same sample to check the effects of the grain size on the morphology. Apparent  
225 Projected Shape of Ash (APASH) was automatically measured by the instrument, via several morphology and  
226 roughness parameters, on a high number of ash particles dispersed on a glass slide. Front lighting with various  
227 magnifications (from 2.5 x to 20 x) was used depending on the particle sizes. In this study we focused on two  
228 roughness parameters, namely the solidity (SLD) and the convexity (CVX), which represent morphological  
229 (particle scale) and textural (smaller scale) roughness of the particles, respectively. The study of Liu et al. (2015)  
230 gives a global overview on how to interpret the different shape parameters, especially SLD and CVX, and how to  
231 link these with magmatic textures. For instance, a perfect round or square particle has a SLD and CVX value of 1.  
232 On the other hand, SLD and CVX decrease as soon as shape irregularities appear. We also focused on one  
233 morphological parameter, the aspect ratio (AR), which represents the elongation of the particles. These three  
234 parameters are defined as follow (Leibrandt and Le Pennec 2015):  $SLD = A/A_{CH}$ ,  $CVX = P_{CH}/P$  and  $AR = W_b/L_b$   
235 where A is the particle area ( $\mu\text{m}^2$ ),  $A_{CH}$  is the area ( $\mu\text{m}^2$ ) of the convex-hull (smallest convex polygon that contains  
236 all the pixels of the particle), P is the particle perimeter ( $\mu\text{m}$ ),  $P_{CH}$  the convex-hull perimeter ( $\mu\text{m}$ ),  $W_b$  the minor  
237 axis and  $L_b$  the major axis of the particle. In specific cases and in order to distinguish between angular and spherical  
238 shapes, we also studied the sphericity (SPH, also known as HS circularity or Shape Factor) which relates the  
239 perimeter to the area of the particles (Riley et al. 2003; Leibrandt and Le Pennec 2015):  $SPH = (4\pi A)/(P^2)$ . SLD,  
240 CVX, AR and SPH are dimensionless parameters varying between 0 and 1. The raw data for each parameter are  
241 shown in Table S3. These data were previously filtered to remove artificial particle clusters form the database  
242 (several particles that touch each other): a manual but rigorous verification was systematically performed on each  
243 automatically identified particle.

244

## 245 **4. Results on the deposits characterization**

### 246 **4.1. Tephra deposits from Hawaiian fountaining activity**

#### 247 **4.1.1. Recent investigated eruptions (2014 to 2018)**

248 The deposits are mostly composed of lapilli with a moderate ash content between 10 and 58 wt%, but  
249 none of them contain detectable fine ash. All these Hawaiian deposits are only composed of juvenile particles,  
250 characterized by vesicle-rich and microlite-poor sideromelane particles (golden pumice and fluidal scoria  
251 pyroclasts). Thivet et al. (2020a) presents detailed textural analysis within the 500-710  $\mu\text{m}$  size fraction on the

252 September 2016 Hawaiian fountain deposits, showing that these ash particles are composed of 42 vol% of vesicles  
253 and only 1 vol% of microlites. Microlites are mainly plagioclase and scarce clinopyroxene. Micro-phenocrysts of  
254 olivine (Fig. 2a), plagioclase or clinopyroxene can also occur depending on the eruptions. Vesicles can be either  
255 round and isolated within the glassy matrix or highly elongated. The PSDs of these samples (Figs. 4a) are unimodal  
256 with modes between 2 and 8 mm depending on the eruptions. The morphology of these particles, measured within  
257 the 250-300 grain size fraction (Fig. 5a), are relatively heterogeneous (CVX between 0.61 and 1.00, SLD between  
258 0.39 and 1.00, and AR values between 0.10 and 1.00), reflecting the systematic presence of round droplets (CVX,  
259 SLD and AR values near 1), fluidal and poly-lobed fragments, as well as elongated particles with sometimes Pele's  
260 hairs (lower CVX, SLD and AR values). Note that the July 2015 sample was measured within a larger grain size  
261 fraction (500 to 600  $\mu\text{m}$ ) but no significant differences are observed in terms of shape except that elongated  
262 particles are slightly more abundant in this fraction (lowest AR median (0.65) measured in this study). Depending  
263 on the samples, SPH medians span a range from 0.49 to 0.65 (Table S3).

#### 264 **4.1.2. The Black Tephra deposits of Piton Chisny (1450 to 1630 AD)**

265 The initial phase of the sampled sequence (labelled CH1 in Fig. 2b), is composed of juvenile lapilli (39  
266 %) and juvenile ash (56 wt% of coarse ash and 5 wt% of fine ash). This ash is either composed of sideromelane  
267 or tachylite particles (Fig. 2b) showing heterogeneous magmatic textures that range from vesicle-rich and crystal-  
268 poor to vesicle-poor and crystal-rich matrixes. The PSD of this sample is multimodal (Fig. 4b) with two main  
269 modes between 11.3 and 16 mm (only composed of sideromelane lapilli, golden pumice and fluidal scoria) as well  
270 as between 180 and 250  $\mu\text{m}$  (mostly composed of tachylite ash with scarce sideromelane ash). Interestingly, the  
271 content of tachylite particles within the deposit increases towards the fine grain sizes, from 6 % (2-2.8 mm), to 10  
272 % (1.4-2 mm), to 11 % (1-1.4 mm) to 19 % (500-710  $\mu\text{m}$ ) to finally 83 % (180-250  $\mu\text{m}$ ). Scarce individual olivine  
273 phenocrysts were also observed (Fig. 2b). Looking at the BSE images it is clear that the tachylite particles show  
274 relatively different shapes with rougher surfaces than sideromelane ones. However, no clear differences are  
275 observed in term of morphology (Fig. 5b) compared to the 2014-2018 Hawaiian ash particles because the tachylite  
276 particles within the CH1 sample are dispersed with the sideromelane particles which have more heterogeneous  
277 shapes.

278 This CH1 layer is overlain by a sequence of about 3 m thick coarser layers (separated by several bomb-  
279 rich layers) which have very similar PSD (Fig. S1a). The labelled sample CH5 (Fig. 2b) is globally representative  
280 of these subsequent deposits which are only composed of juvenile sideromelane particles (golden pumice and

281 fluidal scoria pyroclasts), with only 6 wt% content of ash and with no detectable fine ash. Thus, this deposit is  
282 relatively different from the opening phase. In spite of its significant thickness and large dispersion (Morandi et  
283 al., 2016), the Chisny lapilli deposit is very similar to the recent and weak Hawaiian fountaining deposits described  
284 earlier in this section, with a unimodal PSD, the grain size mode being between 5.6 and 8 mm (Fig. 4b). The ash  
285 morphologies are also quite similar to the recent ash produced during weak Hawaiian fountaining (Fig. 5b).

## 286 **4.2. Tephra deposits from recent mild Strombolian and transient explosions**

### 287 **4.2.1. The July and September 2018 mild Strombolian activities**

288 The deposits collected from mild Strombolian activities during July 2018 and September 2018 (Fig. 2c)  
289 eruptions are mostly composed of lapilli with a very low ash content (5 to 6 wt%) and with no detectable fine ash.  
290 These deposits are only composed of juvenile sideromelane particles. Textural observations performed on the 500-  
291 710  $\mu\text{m}$  grain size fraction highlights an increase in the microlite content (20 to 30 vol%) and a decrease in the  
292 vesicle content (10 to 20 vol%) compared to the Hawaiian particles. Most of the vesicles are round, while some  
293 vesicles are larger and characterized by irregular shapes. The microlite populations are composed by plagioclase,  
294 clinopyroxene and very scarce olivine, in order of abundance. The PSD of these two 2018 samples are unimodal,  
295 relatively similar to the Hawaiian samples (apart for the ash content), with grain size modes between 5.6 and 8  
296 mm (Fig. 3c). Ash morphology measurements were performed on the 250-300  $\mu\text{m}$  grain size fraction and the  
297 particle CVXs, SLDs and ARs are respectively higher than 0.68, 0.58 and 0.19 (Fig. 5c), reflecting fluidal shapes  
298 associated with spiny, rough particles.

### 299 **4.2.2. The specific case of the September 2016 transient explosions**

300 Deposits from the September 2016 transient explosions contain two different type of juvenile particle  
301 components, respectively sideromelane (REU161211\_10, labelled 1 in Fig. 2b) and tachylite ones (REU161211\_6,  
302 labelled 2 in Fig. 2b), as already described in Thivet et al. (2020a). The deposit formed by the sideromelane  
303 component is a typical coarse-grained Strombolian deposit composed of only 8 wt% of ash, while the deposit  
304 related to the tachylite component is composed of 97 wt% of coarse ash. None of these deposits contain detectable  
305 fine ash. The sideromelane ash textures have similar textures than the ones described in the Mild Strombolian  
306 section (labeled 1 in Fig. 2d). Thivet et al. (2020a) already performed precise textural analysis on these samples,  
307 showing a low content of vesicles (13 vol%) and microlites (17 vol%) of plagioclase, clinopyroxene and olivine  
308 (within the 500-710  $\mu\text{m}$  grain size fraction). Vesicles are sometimes deformed. The tachylite ash particles (labeled

309 2 in Fig. 2d) are characterized by a relatively low content of vesicles (10 vol%), and a fully crystallized matrix  
310 (100 vol%) composed of normally zoned microlites of plagioclase and clinopyroxene associated with olivine  
311 microlites and Fe-Ti rich micro-oxides (still within the 500-710  $\mu\text{m}$  grain size fraction). Sideromelane and tachylite  
312 textures are sometimes mingled together (mingled clasts) even at microscopic scale. XRD analyses discard the  
313 presence of any hydrothermal-induced mineral phases on both sideromelane and tachylite deposits. PSD of the  
314 sideromelane deposit shows a similar pattern as the mild Strombolian samples previously described, although with  
315 a coarser grain size mode between 22.6 and 32  $\mu\text{m}$ . In contrast, the PSD of the fine-grained tachylite deposit is  
316 unimodal with a mode between 500 and 710  $\mu\text{m}$  (Fig. 4c). Ash morphology, performed on the 250-300  $\mu\text{m}$  grain  
317 size fraction (Fig. 5c), show that the sideromelane ash has very similar morphology signatures as the mild  
318 Strombolian samples. However, the tachylite ash particles do not have smooth surfaces anymore (CVX and SLD  
319 values  $< 0.95$ ) and are rougher both in terms of SLD (particle scale) and CVX (micron scale), than the sideromelane  
320 ash particles, with no evidence of significant elongation features (AR values  $> 0.40$  and a slightly higher SPH  
321 median of 0.69 compared to the Hawaiian and Strombolian particles).

#### 322 **4.3. Tephra deposits from the explosions caused by the April 2007 lava flow entry in the sea**

323 The hydromagmatic deposit from the April 2007 lava flow entry is only composed of juvenile coarse ash  
324 among with very scarce fine ash (1 wt%). Particles are dense and glassy, and looking at the 500-710  $\mu\text{m}$  grain size  
325 fraction they are characterized by relatively low vesicle content (less than 20 vol%), very low microlite content  
326 (less than 10 vol%) and variable but relatively high olivine phenocryst content (Fig. 3a), in agreement with the  
327 fragmentation of the olivine-rich lava from whom they are originated (Staudacher et al. 2009; Di Muro et al. 2014).  
328 These olivine phenocrysts (both free crystals and composite particles of liquid and phenocrysts) are highly  
329 fractured. Very scarce particles have a dendritic texture (Fig. 3a). The PSD of this deposit is relatively fine-grained  
330 and unimodal, mode being between 500 and 710  $\mu\text{m}$  (Fig. 4d). Morphology of these particles (performed on many  
331 grain size fractions, Fig. 5d) is highly distinctive with respect to Hawaiian and Strombolian deposits mostly  
332 because these particles are relatively elongated with extreme AR values down to 0.03, reflecting the presence of  
333 abundant glassy Limu o Pele shards (following the nomenclature described in Maicher and White 2001). These  
334 kind of shards show elongation, deformation and folded features, with fluidal shapes and fractured surfaces. No  
335 significant morphology differences are observed depending on the different grains size fractions measured.

#### 336 **4.4. Tephra deposits from the April 2007 caldera collapse**

337 The studied sample related to the ash emission during the caldera collapse of April 2007 is only composed  
338 of ash particles with a large amount of fine ash (87 wt%), the largest in our sample set. This ash deposit contains  
339 particle clusters forming irregular-shaped aggregates, composed of both coarse and fine grains and showing  
340 random particle grain size arrangement (Fig. 3b). Most of the particles are considered as non-juvenile (> 99 wt%).  
341 These non-juvenile particles are sometimes altered and systematically characterized by the absence of vesicles, by  
342 fully crystallized gabbroic textures (assemblage of plagioclase, pyroxene, olivine and oxides), with also the  
343 presence of individual and fractured crystal-free particles, which are not coated with magmatic glass. However,  
344 very scarce particles (< 1 wt%) are considered as juvenile. They are either represented by individual glass  
345 fragments with a moderate content in MgO ( $4.9 \pm 0.1$  wt%) and in K<sub>2</sub>O ( $1.3 \pm 0.1$  wt%), or by tachylite particles  
346 with no clear evidence of significant alteration. We also note that a small amount of Pele's hairs is dispersed in  
347 this deposit. These Pele's hairs have a bimodal composition (Table S2) with magnesian (MgO:  $8.1 \pm 0.3$  wt%) and  
348 evolved, low MgO ( $2.9 \pm 0.8$  wt%) melts.

349 XRD analysis performed on the April 2007 caldera collapse deposit confirmed the presence of  
350 plagioclase, clinopyroxene and olivine crystals (observed in the gabbro fragments) and also suggest the presence  
351 of both montmorillonite (clay) and hematite phases. The PSD of this sample is unimodal with a mode between 15  
352 and 8  $\mu$ m (Fig. 4e). Ash morphology performed on this sample and on three different grain sizes (Fig. 5e) show  
353 very different results from the other samples: most of the CVX and SLD values range between 0.90 and 1.00  
354 reflecting the presence of both fine grained, blocky and large, round particles without significant rough surface  
355 features (Fig. 3b). Looking at the relatively high AR values (median between 0.70 and 0.80) they suggest that these  
356 particles are mostly blocky without elongation features. Depending on the grain size, slight differences on the  
357 averaged SPH are observed (Table S3): the coarsest analyzed grain size bins (150-180  $\mu$ m and 63-75  $\mu$ m) show  
358 the highest SPH values measured in this study (0.83 and 0.84 respectively), while the finest analyzed grain size  
359 (32-43  $\mu$ m) show slightly lower values (0.79). These SPH values are in accordance with the SEM observations:  
360 in this sample, the ash particles tend to be more angular towards the finer grain sizes.

#### 361 **4.5. Tephra deposits from the March 1860 violent Strombolian eruption**

362 The sample collected at the base of the deposit (REU131112, Fig. S1b) is composed of lapilli (44 wt%)  
363 and ash (55 wt% of coarse ash and 1 wt% of fine ash). Within the 500-710  $\mu$ m grain size fraction, juvenile particles  
364 represent only 2 % which are characterized by non-altered, more or less vesiculated glassy matrix. The non-  
365 juvenile particles (98 %) are dense, characterized by fully crystallized matrix (Fig. 3c). The size of the crystals of

366 these particles vary from a few tens of microns (microlite-rich and altered or oxidized particles, thus considered  
367 as recycled non-juvenile tachylite particles) to several hundreds of microns (fully crystallized micro-gabbro  
368 texture, thus considered as non-juvenile intrusive fragmented material). Micro-gabbro textures are characterized  
369 by larger crystal in terms of length and width (plagioclase, clinopyroxene and olivine) than in the juvenile or  
370 recycled particles (microlites and micro-phenocrysts sometimes associated with unaltered glass). Scarce olivine  
371 individual crystals are counted within the non-juvenile particles, because they are not wetted with fresh glass. The  
372 PSD of this sample is unimodal with a grain size mode between 1.4 and 2 mm (Fig. 4f), finer than that of Hawaiian  
373 and Strombolian deposits, but coarser than that of hydromagmatic ash from the April 2007 lava flow entry.

374 PF\_2 sample (Fig. S1b) is only composed of ash with the presence of a little amount of fine ash (4 wt%).  
375 Within the 355-500  $\mu\text{m}$  grain size fraction, most of the particles are non-juvenile (89 %). All the particles of this  
376 layer show the same textures as in the REU131112 sample. XRD analysis performed on the PF\_2 samples suggests  
377 the presence of plagioclase, clinopyroxene, olivine (already observed with the SEM) and hematite phases. The  
378 PSD of this sample is unimodal with a grain size mode finer than the previous sample, between 355 and 500  $\mu\text{m}$   
379 (Fig. 4f).

380 Despite a small difference on the measured grain size fractions, the particle morphology of the two  
381 samples representative of this eruption are very similar to each other (Fig. 5f): CVX and SLD medians for both  
382 samples are around 0.95 and AR values are relatively high with medians at 0.78, mostly reflecting gabbroic or  
383 altered particles with rough and blocky shapes. The median values of SPH (Table S3) measured on both samples  
384 are relatively high (0.73 and 0.75) reflecting the presence of blocky non-juvenile particles without any significant  
385 irregularity features, in accordance with the previous observations.

#### 386 **4.6. The Bellecombe Tephra deposits (4900 to 2250 BP)**

387 Samples collected within the first 20 cm fall deposits of the Lower Bellecombe Tephra (BE\_BASE\_1 and  
388 BE\_BASE\_2, Fig. S2) have similar and very high ash content (between 85 and 91 wt%), with however slightly  
389 different fine ash content (32 and 11 wt% respectively). Based on the componentry analysis performed on the 250-  
390 355  $\mu\text{m}$  grain size fraction of the BE\_BASE\_1 sample (Fig. 3d), we suggest that this ash fraction is only composed  
391 of non-juvenile particles either characterized by altered glassy clasts (40%), micro-gabbro (40%), and individual  
392 olivine crystal fragments (20%). We consider that the alteration observed on these particles is representative of the  
393 fragmentation conditions but the alteration may also have occurred after the tephra deposition regarding the  
394 relatively old age of the eruption, and this is the limit of the componentry analysis. Grain size mode of these two

395 samples (Fig. 4g) are skewed toward relatively fine grain size fractions (between 31 and 63  $\mu\text{m}$  for the  
396 BE\_BASE\_1 sample and between 180 and 250  $\mu\text{m}$  for the BE\_BASE\_2 sample).

397 Samples collected within the primary (non-reworked) layers of the Upper Bellecombe Tephra  
398 (BE\_TOP\_1 and BE\_TOP\_2, Fig. S2) have lower ash content than the Lower Bellecombe Tephra samples  
399 (between 47 and 59 wt%) with also lower fine ash content (between 2 and 3 wt%). Similarly to the Lower  
400 Bellecombe Tephra samples, componentry performed on the 250-355  $\mu\text{m}$  grain size fraction of the BE\_TOP\_1  
401 samples suggest that this ash fraction is only composed of non-juvenile particles, consisting in the same types of  
402 fragments of the BE\_BASE\_1 sample (Fig. 3d). Irregular-shaped particle clusters (following the Brown et al. 2012  
403 nomenclature) are much more abundant than in the layers of the Lower Bellecombe Tephra and are characterized  
404 by a random grain size organization. Grain size mode of these two samples are skewed toward coarser grain size  
405 fraction (Fig. 4g) than the Lower Bellecombe Tephra samples (two modes between 8 and 11.3  $\mu\text{m}$  as well as  
406 between 1.4 and 2.8  $\mu\text{m}$  for the BE\_TOP\_1 sample and one mode between 1.4 and 2  $\mu\text{m}$  for the BE\_TOP\_2  
407 sample). In addition to the plagioclase, clinopyroxene and olivine phases, XRD analysis performed on both Lower  
408 and Upper Bellecombe Tephra suggest the presence of montmorillonite and quartz.

409 Ash morphology measurements performed on the four Bellecombe samples show quite similar patterns  
410 (Fig. 5g) reflecting the presence of three different types of non-juvenile particles, namely the glassy altered ones  
411 which have fluidal to spiny shapes (especially for the Lower Tephra samples), as well as the individual crystal-  
412 free and micro-gabbro blocky fragments. Similarly to the March 1860 analyzed deposits, the average values of  
413 SPH (Table S3) measured on the four Bellecombe samples are relatively high (between 0.71 and 0.77) reflecting  
414 the presence of blocky, non-juvenile particles without any significant irregularity features.

415

## 416 **5. Discussion on the fragmentation mechanism variability**

417 In this study, we chose to study a wide range of tephra deposits (in terms of texture, morphology, grain  
418 size, and chemistry) in order to depict the variability of ash-forming events occurring at a basaltic shield volcano.  
419 As a consequence, we could only analyze a limited number of samples for each selected deposit. In order to work  
420 on thick deposits and visualize the entire stratigraphy representing the whole of the eruptive sequences, we  
421 systematically collected samples from proximal locations (50 to 1500 m from source). We are confident that this  
422 sampling strategy is appropriate for pursuing the objectives of this study, i.e. deciphering the processes of ash



423 formation and describing the characteristics of the resulting particles. One sample is representative enough of the  
424 weakest, very short-lived explosions, which are limited in time (very brief) and space (poorly dispersed). The  
425 tephra deposits from older eruptions (March 1860, Piton Chisny, and Bellecombe) are relatively heterogeneous in  
426 terms of lithology and grain size (Figs. S1 and S2), but we identified the best outcrops representing the whole  
427 sequence of events, and carefully selected specific layers representative of the main eruptive phases of each  
428 eruption, based on our observations and the literature when available.

429 All the samples described in this study were affected by transport-dependent processes, which normally  
430 limit the comparison between samples collected at different distances from vent, especially when dealing with  
431 PSD. However, all the samples were collected at similar distances close to their sources (most of them less than  
432 500 meters from source, Fig. 1c), thus effects of transport processes are negligible. Only the two 2007 samples  
433 were collected 1.5 km from source, because we favored clean sampling surfaces to avoid mixing between the  
434 multiple ash-sources observed during the 2007 eruption. More proximal samples were available (300 meters from  
435 source) and show similar PSDs to the more distal samples selected here, which demonstrates that the variations in  
436 PSDs can be confidently interpreted in terms of differences in fragmentation efficiency.

## 437 **5.1. Variability and origin of the eruptive dynamics**

### 438 **5.1.1. Hawaiian fountaining: breakup of vesicle-rich and crystal-poor juvenile magmas**

439 The investigated Hawaiian fountaining events at PdF produced sustained lava fountains with heights  
440 ranging from 30 to 100 m (Fig. 2a). The associated deposits are characterized by lapilli-dominated (Fig. 4a),  
441 vesicle-rich, and crystal-poor juvenile pyroclasts (Fig. 2a). Despite their homogenous textures, the emitted ash  
442 particles show a large range in morphology (Fig. 5a and Table S3). The presence of round droplets and elongated  
443 particles with smooth surfaces, highlight that magma fragmentation occurred at low viscosity and that the final  
444 pyroclast shape was acquired in the air post-fragmentation just before quenching. Round liquid droplets formed  
445 due to surface tension while the high projection velocity shaped the elongated filaments (Shimozuru 1994).  
446 Particles with poly-lobed contours show that magma fragmentation was also controlled by the numerous thin  
447 bubble walls that rapidly quenched and broke up. The low magma viscosity interpretation is in agreement with a  
448 previous estimation ( $10^2$  Pa s) based on a three-phase viscosity model (vesicle and crystal content as well as melt  
449 composition) performed on the magmas from the Hawaiian-style activity of the September 2016 eruption at PdF  
450 (Thivet et al. 2020a). Moreover, most of the studied deposits contain Pele's hairs of different size with various  
451 content in deformed vesicles, representative of low viscosity magmas exhibiting different quenching rates and

452 residence time in the hot lava fountain (Porritt et al. 2012; Mangan et al. 2014; Cannata et al. 2019). The relatively  
453 high vesicle number density ( $N_V$ ) and low microlite number density ( $N_C$ ) measured in the ash, especially during  
454 the September 2016 event (Thivet et al. 2020a), reflect hot magmatic liquids exhibiting high decompression rates  
455 (Toramaru 2006) and low syn-eruptive outgassing and crystallization (Toramaru et al. 2008). This is consistent  
456 with closed system, low-viscosity magma degassing during Hawaiian fountaining that has been related to intense  
457 bubbly flow (Cashman and Scheu 2015) and even annular flow dynamics within the volcanic conduit (e.g. Jaupart  
458 and Vergnolle 1988, 1989; Houghton and Gonnerman 2008). Similar textures are found in lava fountain deposits  
459 from other volcanoes as Etna (e.g. Polacci et al. 2006; Andronico et al. 2008; Andronico and Corsaro 2011) and  
460 Kilauea (e.g. Stovall et al. 2011, 2012; Parcheta et al. 2013; Cannata et al. 2019) but the heights and the intensity  
461 of the lava fountains are generally higher in these latter volcanoes compared to PdF. Moreover, Etna eruptions  
462 stand out from the typical Hawaiian activity for the recurrent formation of ash-rich eruptive columns up to several  
463 kilometers high (e.g. Andronico et al. 2015; Corsaro et al. 2017; Potter et al. 2019).

464         The textural variability of juvenile tephra from a same eruption is well-known in a basaltic context (e.g.  
465 Pioli et al. 2014; Gurioli et al. 2014; Polacci et al. 2019). This is the case of the deposits from the opening phase  
466 of Piton Chisny historical activity (CH1 sample) which is much richer in ash particles. Interestingly, the smaller  
467 grains of these deposits are mostly composed of crystal-rich and vesicle-poor sideromelane as well as tachylite  
468 particles, while the coarser grains mostly show crystal-poor and vesicle-rich sideromelane textures (Fig. 2b). The  
469 texture variation from sideromelane to tachylite can be interpreted as syn-eruptive degassing and crystallization  
470 within the eruptive conduit, as demonstrated at other basaltic systems (e.g. Taddeucci et al. 2002; Sable et al. 2006;  
471 D’Oriano et al. 2014; Polacci et al. 2019) and in experiments (e.g. Simakin et al. 1999; Applegarth et al. 2013;  
472 Arzilli et al. 2015). This shows that the portion of viscous magma exhibits the highest degree of fragmentation.  
473 We thus suggest that syn-eruptive degassing-driven crystallization of the Piton Chisny opening phase drastically  
474 increased the magma viscosity and enhanced magma fragmentation (Arzilli et al. 2019), which was probably more  
475 brittle than the Hawaiian counterpart. The presence of both vesicle-rich (high  $N_V$  reflecting high decompression  
476 rates) and crystal-rich particles (high  $N_C$  reflecting an increase in magma viscosity in the conduit) in the CH1  
477 sample, provides additional evidence of enhanced magma fragmentation (Cimarelli et al. 2010; Cashman and  
478 Scheu 2015). Syn-eruptive crystallization may be facilitated by a long magma residence time during initiation and  
479 propagation of the eruptive conduit from a deep reservoir (Morandi et al. 2016) to the surface. After the opening  
480 phase and the formation of the eruptive conduit, the magma residence time in the conduit decreased resulting in  
481 typical lava fountain activity (CH5 sample).

### 482 **5.1.2. Strombolian explosions: towards brittle fragmentation of relatively degassed juvenile magmas**

483 At PdF, mild Strombolian explosions occur towards the end of dyke eruptions and temporally follow the  
484 fountaining activity, when the lava fluxes are decreasing (Thivet et al. 2020b). Mild Strombolian activity generally  
485 focuses in a single vent/cone producing weak projections, not exceeding heights of a few tens of meters (Fig. 2c).  
486 Bubble explosions at the magma free surface, within generally well-constructed scoria cones, are relatively weak  
487 but temporally close (only a few seconds between each burst, similar to the mild-Strombolian activity described  
488 by Spina et al. 2017 at Etna volcano) compared to the typical regime at Stromboli (e.g. Chouet 2003; Ripepe et al.  
489 2001; Harris and Ripepe 2007). The associated deposits are characterized by bomb and lapilli-dominated (Fig. 4c)  
490 juvenile pyroclasts with crystal-rich sideromelane textures. A decrease in porosity and an increase in micro-  
491 crystallinity (Fig. 2c) are thus observed compared to the magma emitted during the lava fountain activity (golden  
492 and fluidal material). The decrease of  $N_V$  and the increase of  $N_C$  as well as some evidence of vesicle coalescence  
493 (occurrence of a second, irregularly-shaped vesicle population of larger size) within the sideromelane ash particles  
494 suggest that the mild Strombolian dynamics, typically active during slug flow (e.g. Jaupart and Vergnolle 1988,  
495 1989; Houghton and Gonnerman 2008; Chouet et al. 2003; Oppenheimer et al. 2020), is related to lower  
496 decompression rates associated with higher syn-eruptive degassing and crystallization (Thivet et al. 2020b). It also  
497 marks the transition from closed to open system degassing. Sideromelane particle shapes from bubble explosions  
498 (Fig. 5c) differ from the clast shapes produced by the Hawaiian fountaining activity. First, the absence of  
499 significant post-fragmentation features as well as the absence of Pele's hairs and tears, suggest that the relatively  
500 low temperature and high viscosity do not allow particles to deform during and after fragmentation. Moreover, the  
501 rough and spiny shapes of the Strombolian particles may reflect primary brittle fragmentation behavior, mostly  
502 due to lower magmatic temperatures and higher crystal contents (both cooling and degassing of the magma in the  
503 conduit enhanced by a longer residence time compared to the Hawaiian fountaining dynamics), resulting in an  
504 increase in viscosity ( $10^3$  Pa s, calculated value for the sideromelane September 2016 magma, Thivet et al. 2020a).

505 We conclude that the magmas emitted during these mild Strombolian explosions have slightly higher  
506 viscosities than their Hawaiian counterparts. However, vesicles are well coupled with the melt in the Hawaiian  
507 regime, but open degassing system and volatile percolation occur in the Strombolian regime, which drastically  
508 decrease the fragmentation efficiency. This results in the lowest ash content of the investigated dynamics at PdF.

509 Similarly to the textural variations observed between the CH1 and CH5 samples, we observed an  
510 evolution from sideromelane to tachylite particles emitted during the specific case of the September 2016 transient

511 explosions. The texture and associated features of the tachylite ash emitted during these explosions, already  
512 quantified in a previous study, highlight that these juvenile particles are totally crystallized and degassed (0 wt%  
513 of dissolved H<sub>2</sub>O, 10 vol% of vesicles, Thivet et al. 2020a). In-situ chemical and textural analysis, as well as syn-  
514 eruptive observations, suggest that the emitted ash originated from sub-surface degassing and crystallization of the  
515 initial magma (Thivet et al. 2020a). The degassed/cooled magma formed a rheological and impermeable layer,  
516 which played the role of a plug, within one of the two active vents. The ascent of still partially undegassed magma  
517 (sideromelane) fragmented the plug layer composed of tachylite magma (brittle fragmentation by plug  
518 pressurization/decompression). Note that, XRD analysis discard the presence of any hydrothermal-induced  
519 mineral phases that can be related to involvement of hydrothermal fluids or fragmentation of surrounding rocks  
520 (phreatomagmatic mechanisms). Brittle fragmentation is also evidenced by the ash morphologies (Fig. 5c). CVX,  
521 SLD, AR and SPH values are skewed toward higher median values for tachylite ash than sideromelane particles,  
522 reflecting typical tachylite textures with rough surfaces and no significant particle deformation. Moreover, ash  
523 particles with CVX, SLD and AR values of 1.00 are not observed, suggesting that magma viscosity is too high to  
524 produce round droplets like it was the case in the golden ash component.

### 525 **5.1.3. Lava-ocean interactions provoking hydrovolcanic explosions**

526 When a lava flow enters a body of water like the sea at relatively high fluxes ( $> 4 \text{ m}^3 \text{ s}^{-1}$ ), hydrovolcanic  
527 explosions producing laze can occur like it was the case at Kilauea volcano (Mattox and Mangan 1997; Neal et al.  
528 2019), and at PdF during the April 2007 eruption (Fig. 3a), which was fed by a lava peak flux of at least  $200 \text{ m}^3 \text{ s}^{-1}$   
529 (Staudacher et al. 2009). Laze is characterized by the emission of steam clouds highly concentrated in  
530 hydrochloric acid and glass shards (Carlos et al. 2018). According to Mattox and Mangan (1997), the ideal  
531 water/magma ratio to provoke littoral explosions is around 0.15 and an open mixing of lava and seawater (when  
532 the mouth of several lava tubes or lava channels are exposed to sea waves) is the most efficient situation to produce  
533 unconsolidated deposits of glassy, dense lava fragments. The particles formed during the 2007 seawater-provoked  
534 explosions were immediately quenched and show a glassy matrix with rare microlites (Limu o Pele glass shards)  
535 and more abundant phenocrysts of olivine, already present within the lava flow itself, as it was described in Hawaii  
536 (Potuzak et al. 2008). The dendritic texture observed in very scarce particles (Fig. 3a) suggest that some of the  
537 fragmented material experienced not instantaneous but fast quenching. The particles show typical fractured  
538 features on their surfaces and inside the matrix glass. Concentric fractures are often observed around the olivine  
539 phenocrysts (Fig. 3a). We suggest that the intensity of the sudden fragmentation, the almost instantaneous  
540 quenching as well as the thermic contraction are the main cause of these brittle features (Büttner et al. 1999;

541 Zimanowski et al. 2015). Extreme AR values (down to 0.05) reflect the formation of typical elongated glassy  
542 shards particles (Fig. 5d) during these hydrovolcanic explosions.

#### 543 **5.1.4. The April 2007 caldera collapse: phreatic (or phreatomagmatic) explosions associated with secondary** 544 **fault-shear fragmentation**

545 The deposit associated with the ash emission during the April 2007 caldera collapse (Figs. 6a and 6b) is  
546 characterized by the quasi absence of juvenile fragments. Fully crystallized gabbro fragments suggest that the  
547 emitted particles originate from intrusive levels of the caldera wall and from the destroyed, collapsed piston. The  
548 very scarce non-altered juvenile particles may belong to recent and still partially molten magmas stored at shallow  
549 levels within the lava pile that accumulated within the Dolomieu caldera, as molten lava flows within the newly  
550 formed caldera wall were visible just after the main collapse (Fig. 6b). These scarce juvenile particles cannot be  
551 related to the eruptive triggers, but the interstitial and relatively viscous melt within a crystal-rich gabbroic system  
552 might play a significant role in the fragmentation efficiency.

553 The coarser particles ( $> 125 \mu\text{m}$ ) of the deposits generally show round or smooth shapes while the finer  
554 particles ( $< 63 \mu\text{m}$ ) are mainly composed of blocky fragments of gabbroic crystals (Figs. 3b, 5e and 6). This is  
555 clearly evidenced by the SPH values that decrease toward finer grain sizes. SEM images (Fig. 3b and 6) confirm  
556 that the fine particles are significantly more angular than the coarse particles and that the coarse round particles  
557 are often fractured. The ash emissions observed during the April 2007 incremental caldera collapse at PdF have  
558 been firstly interpreted as phreatic driven, based on preliminary petrologic observations (Staudacher et al. 2009).  
559 The quasi absence of juvenile particles, and the presence of particle clusters, and montmorillonite (clay) and  
560 hematite phases, are in accordance with this first interpretation. As shown by analogue experiments (Buckland et  
561 al. 2018), we suggest that the coarse and fine particles are genetically linked, with the fine and blocky particles  
562 originating from the abrasion of the coarse and smoothed particles. We also suggest that wall rock abrasion and  
563 fragmentation was effective during the caldera piston collapse, forming a syn-collapse cataclasite as we observe  
564 very similar brittle features as those found within fault gauge (e.g. Engelder 1974; Marone and Sholtz 1989;  
565 Heilbronner and Keulen 2006), basal shear zones of thick lava flows (Latutrie et al. 2017), dome boundaries  
566 (Cashman et al. 2008) or conduit margins (Pallister et al. 2013). As these finely fractured rocks were transported  
567 in the atmosphere before deposition, we lost some of the features observed in typical fault gauge (such as the  
568 microstructures and faults as well as specific organizations of the particles). But our observations (Figs. 6c and 6d)  
569 support that the ash shapes produced during the April 2007 caldera collapse are very likely the product of fault-

570 shear mechanism. The ubiquity of fine ash particles is also in accordance with this hypothesis, although elutriation  
571 mechanisms may occur within the ring fault system and during the collapse, preferentially injecting the fine ash  
572 particles into the atmosphere.

#### 573 **5.1.5. The March 1860 violent Strombolian eruption: identification of phreatomagmatic interactions**

574 Contemporaneous observations suggest that the March 1860 eruption was triggered by a sudden arrival  
575 of hydrothermal and/or meteoritic water within the newly drained and still hot volcanic conduits and reservoirs  
576 (Michon et al. 2013). This hypothesis is supported by our results. The ubiquity of non-juvenile particles (89 to 98  
577 % of gabbroic particles with rough or blocky shapes as well as altered recycled particles) suggest an origin of the  
578 emitted material from erosion of the conduit walls at different depths (Fig. 3c). The low juvenile content indicates  
579 that the magma involved was not sufficient to provoke such an eruption. Moreover, the scarce juvenile particles  
580 have a very low vesicle content. The contribution of external and/or hydrothermal fluids could have indeed  
581 provoked the fragmentation of the conduit wall rocks still partially hot and partially filled with molten juvenile  
582 magma (2 to 11 % of observed juvenile ash). Small amount of hematite in these deposits may be related to acid  
583 leaching or post-depositional alteration due to the presence of hydrothermal fluids, even if further studies are  
584 needed to confirm this hypothesis. No significant morphology trends are observed in these two samples (Fig. 5f)  
585 due to the wide variety of ash components. However, CVX, SLD and SPH medians are systematically high,  
586 reflecting the presence of blocky particles associated with brittle fragmentation. Slightly higher AR values  
587 compared to the juvenile ash formed by ductile fragmentation, also suggest that no post-fragmentation deformation  
588 as well as no significant ductile mechanisms played a role in these phreatomagmatic eruptions.

#### 589 **5.1.6. The Bellecombe Tephra: identification of phreatic explosions**

590 The investigated samples of the Bellecombe Tephra (BE\_BASE\_1 and BE\_TOP\_1) are only composed  
591 of non-juvenile particles. Alteration of the particles and presence of montmorillonite (clay) and quartz (generally  
592 formed within high temperatures hydrothermal systems) highlight that these particles were fragmented during  
593 phreatic phases that implicated one or several hydrothermal systems. We also suggest that these phreatic (and  
594 possibly phreatomagmatic for some layers which are not studied in this paper, Ort et al. 2016) explosions involve  
595 variably crystallized intrusive rocks, because of the presence of gabbro fragments, similarly to the March 1860  
596 deposits. The presence of particle clusters can also be due to aggregation in the air favored by hydrothermal water,  
597 (Mueller et al. 2017) or post-depositional cementation (clay). Similarly to the phreatomagmatic deposits of the

598 March 1860 eruption, no significant trends are observed in morphology data (Fig. 5g) and the ubiquity of the non-  
599 juvenile blocky particles are revealed by the relatively high SPH measured values.

## 600 **5.2. Magmatic and/or non-magmatic fragmentation efficiencies**

601 The yellow field in Figure 7 show that the investigated Hawaiian deposits are moderately well sorted and  
602 with median grain size values skewed toward relatively coarse grains: the magma breakup (or ductile  
603 fragmentation) within lava fountains has a homogenous and moderate efficiency. Our results highlight that  
604 fragmentation efficiencies attained during Hawaiian activity at PdF is not necessarily correlated with the eruption  
605 intensities. When magma ascends in a conduit following a closed system degassing path and tends to crystallize,  
606 the fragmentation mechanism moves from ductile to brittle because of increasing viscosity. This higher  
607 fragmentation efficiency is recorded in the CH1 sample from the Black Tephra deposits of the Piton Chisny (orange  
608 circle in Fig. 7). The fact that this sample is very poorly sorted reflect the heterogeneity of the magmatic textures  
609 (ranging from crystal-poor and vesicle-rich sideromelane to crystal-rich and vesicle-poor tachylite particles) which  
610 plays a key role on the fragmentation mechanisms (ranging from ductile to brittle).

611 As soon as the lava flux decreases, the magmas have more time to degas, degassing moves from closed  
612 to open system, and the melt crystallizes within the eruptive conduit, increasing the magma viscosity. The deposits  
613 related to these mild Strombolian or transient explosions are well sorted and have relatively coarse grain size  
614 median values (grey field in Fig. 7): the partially brittle fragmentation during the bubble explosions at PdF has the  
615 weakest fragmentation efficiency investigated in this study and it is likely related to the transition from closed to  
616 open system degassing (large bubbles, volatile percolation and gas slugs). The tachylite deposits from the  
617 September 2016 eruption (big black circle in Fig. 7) reflects a better fragmentation efficiency enhanced by the  
618 overpressure caused by the local occurrence of a shallow and highly crystallized magma plug.

619 The deposit associated to the hydrovolcanic explosions caused by the lava entry in the sea observed during  
620 the April 2007 is very well sorted with an intermediate grain size median (blue triangle in Fig. 7), reflecting a quite  
621 efficient fragmentation, which results both from the lower temperature and fully degassed nature of the melt in the  
622 lava flows, producing an increase in melt viscosity with respect to conduit conditions.

623 The two samples representative of the March 1860 summit eruption (pink squares in Fig. 7) are well  
624 sorted and have intermediate median sizes. These results highlight that phreatomagmatic interactions enhance both  
625 magmas and surrounding rocks fragmentation.

626 The Upper Bellecombe samples show lower median grain sizes and slightly better sorting compared to  
627 the Lower Bellecombe ones (green circles in Fig. 7). The wide range of median grain sizes and sorting that span  
628 the phreatomagmatic as well as phreatic deposits reflects the emission of heterogeneous tephra in terms of  
629 components and a potentially high fragmentation efficiency for some explosions.

630 Finally, the deposit related to the April 2007 caldera collapse event is interestingly the finest of this study  
631 (green square in Fig. 7) with an intermediate sorting value. We suggest that the addition of the phreatic explosions  
632 that occurred during the caldera collapse plus the secondary fault-shear fragmentation, may have produced the  
633 extreme fragmentation recorded by this very fine deposit. Scarce residual interstitial melt within the shallow  
634 magmatic system might also contribute to the gabbro fragmentation.

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## 636 **6. Conclusions and risk implications**

637 In our study, we were able to identify a broad range of processes that control and/or enhance the magmatic  
638 and/or non-magmatic particle fragmentation at Piton de la Fournaise shield basaltic volcano. We list them here  
639 with their associated hazard implications.

640 (i) Hawaiian fountains usually produce a moderate amount of coarse ash during their ductile magmatic  
641 fragmentation. The hazard is related mostly to the formation of abundant Pele's hairs that can accumulate within  
642 proximal areas and sometimes in densely inhabited areas, like during the April 2007 eruption (Di Muro et al.  
643 2015), impacting people and livestock due to their fibrous, acicular shape. More recently, the April 2020 eruption  
644 at PdF produced a large amount of Pele's hairs, which were found over a large part of the Reunion Island (Fig. S3,  
645 Table S4, [http://opgc.fr/vobs/so\\_interface.php?so=dynvolc](http://opgc.fr/vobs/so_interface.php?so=dynvolc)). In addition, a previous study showed that Pele's hairs,  
646 can host a large number of smaller particles, which represent a potential surface for heterogeneous nucleation of  
647 salts from the vapor phase (Moune et al. 2007). Ash particles in general can react with the acid gases during their  
648 transport in the volcanic plume and transport pollutants over a wide area (Delmelle et al. 2007).

649 (ii) Mild Strombolian explosions at PdF produce few ash during the bubbles (or slug) explosions at the  
650 magma free surface. However, syn-eruptive degassing-driven crystallization can enhance ash emissions because  
651 of the increase in magma viscosity, triggering brittle fragmentation of still gas-rich magmas. These relatively  
652 sudden changes in activity are unpredictable and can impact people approaching the eruptive sites.



653 (iii) On volcanic islands, more particularly on basaltic shield volcanoes such as PdF or Hawaii, low  
654 viscosity lava flows are likely to reach the ocean, sometimes creating gas and ash emissions. This also represents  
655 a risk to the local population because of the ash and shard fragments but also because of the acid gas emissions  
656 (Carlos et al. 2018). This emphasizes the need to monitor and anticipate the lava flow expansions and directions  
657 depending on the topography, lava flux and viscosity (Harris et al. 2019).

658 (iv) The presence of fluids at shallow levels within the PdF magmatic system has also been identified in  
659 this study as an amplifier of the magmatic and the country rock fragmentation. The interaction between magma  
660 and external fluids is interpreted as playing a potential role in some unusually high explosivity PdF eruptions (i.e.  
661 March 1860 eruption). In some cases, non-juvenile dense particle ubiquity within the investigated samples can be  
662 related to phreatic phases.

663 (v) Finally, secondary fault-shear fragmentation, have been identified as the most efficient mechanisms  
664 to form non-juvenile fine ash particles during the Dolomieu caldera collapse in April 2007. The extremely fine ash  
665 formed during such events, even if localized, can stay longer in suspension and can affect touristic and local  
666 populations.

667 In conclusion, the observations made on the eruptive dynamics associated with the ash characterization  
668 highlights the diversity of the basaltic fragmentation mechanisms at PdF (and this can be extrapolated to other  
669 basaltic volcanoes), which are important to understand in order to anticipate the associated hazards and potential  
670 risks.

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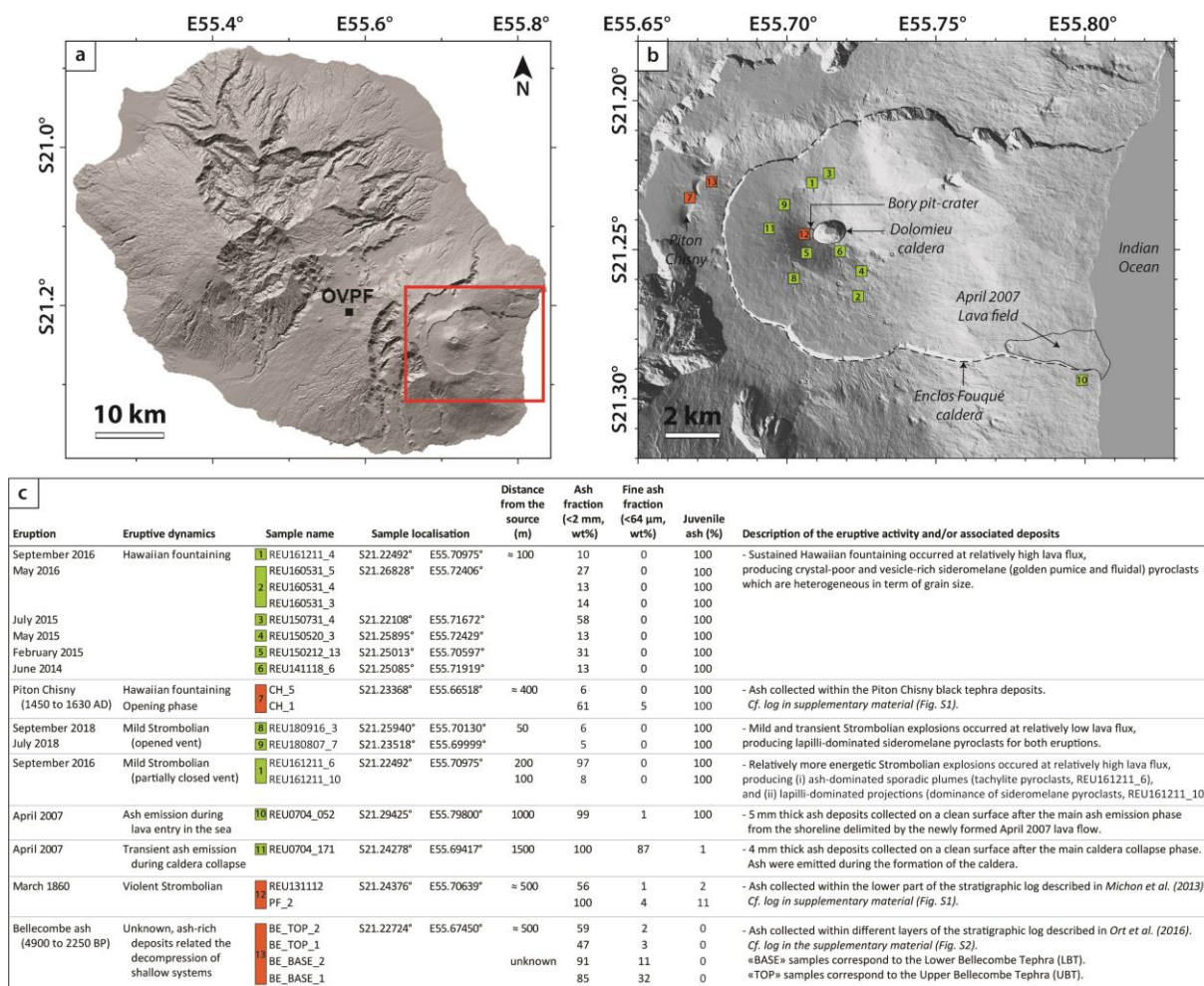
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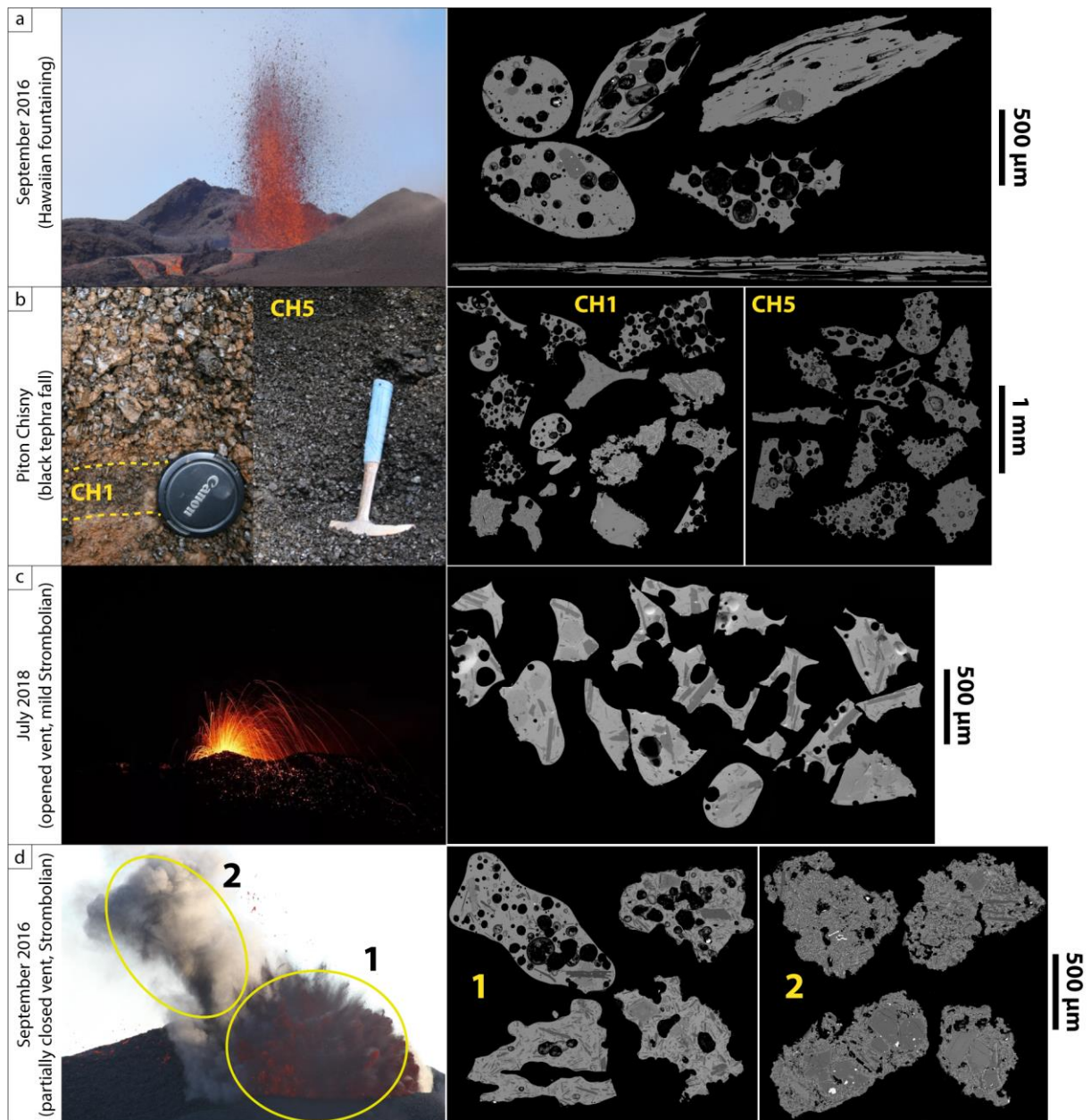
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 681 **Fig. 1** – (a) Digital Elevation Model of La Reunion Island hosting the Piton de la Fournaise (PdF) volcano (red  
 682 square) and the Observatoire Volcanologique du Piton de la Fournaise (OVPF). (b) Map of PdF volcano with the  
 683 studied deposits. The green and orange squares correspond to the deposits sampled during recent activity (2014-  
 684 2018) and historical eruptions, respectively. (c) List and description of all the studied samples. The percentage of  
 685 juvenile ash is measured on specific grain size bins which are described in the results part.

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692 **Fig. 2** – Eruptions and deposits pictures and Backscattered Electron Scanning Microscope (SEM-BSE) images of

693 the respective ash particles, part 1. (a) Hawaiian fountaining activity producing golden pumice and fluidal scoria

694 pyroclasts. (b) Deposits from intense Hawaiian fountaining. CH1 corresponds to the initial phase and CH5

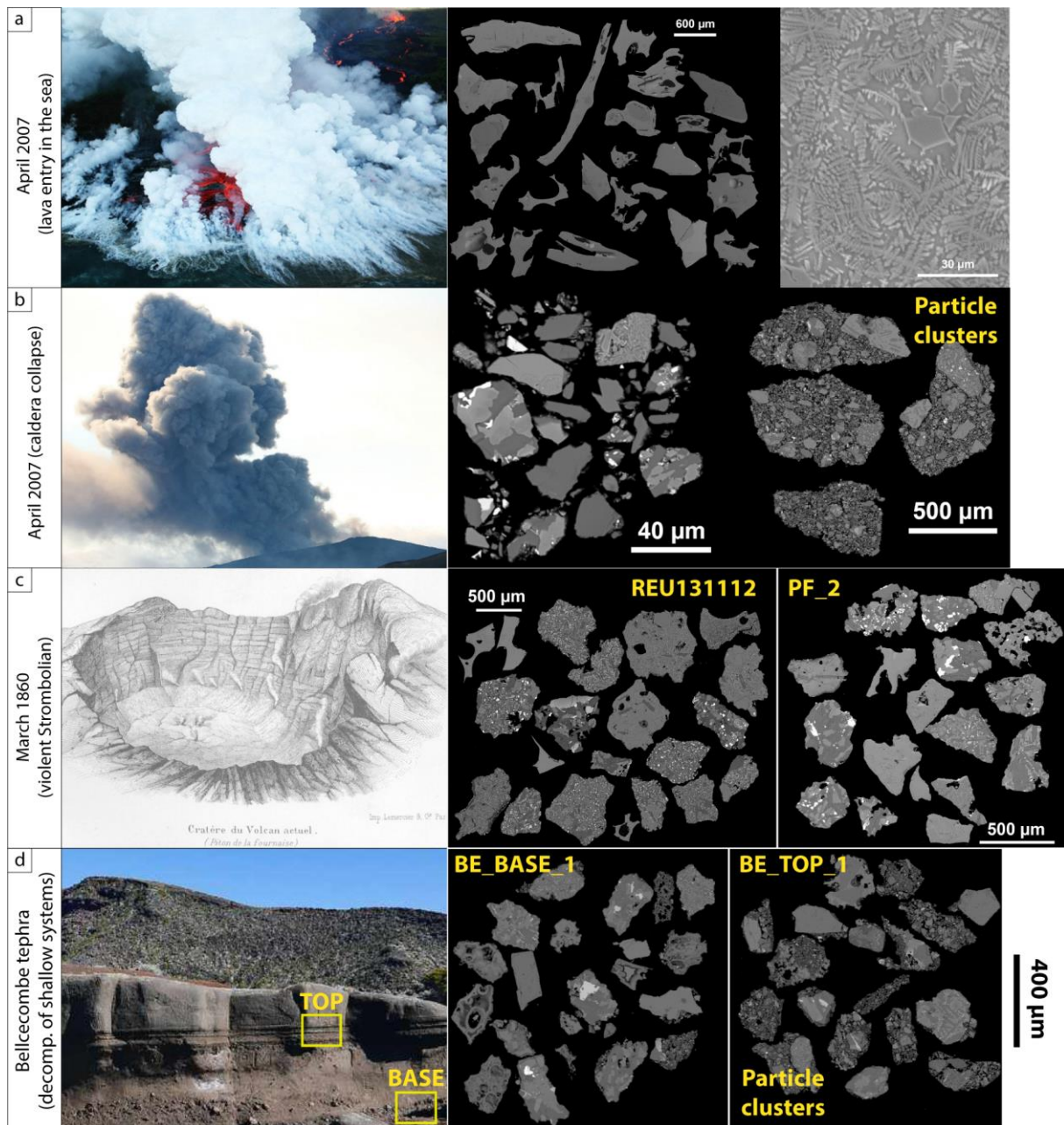
695 corresponds to the later, steady phase. (c) Mild Strombolian activity producing sideromelane pyroclasts. (d)

696 Transient explosions producing incandescent particules (sideromelane pyroclasts, labelled 1) associated with

697 discreet ash plumes (tachylite pyroclasts, labelled 2).

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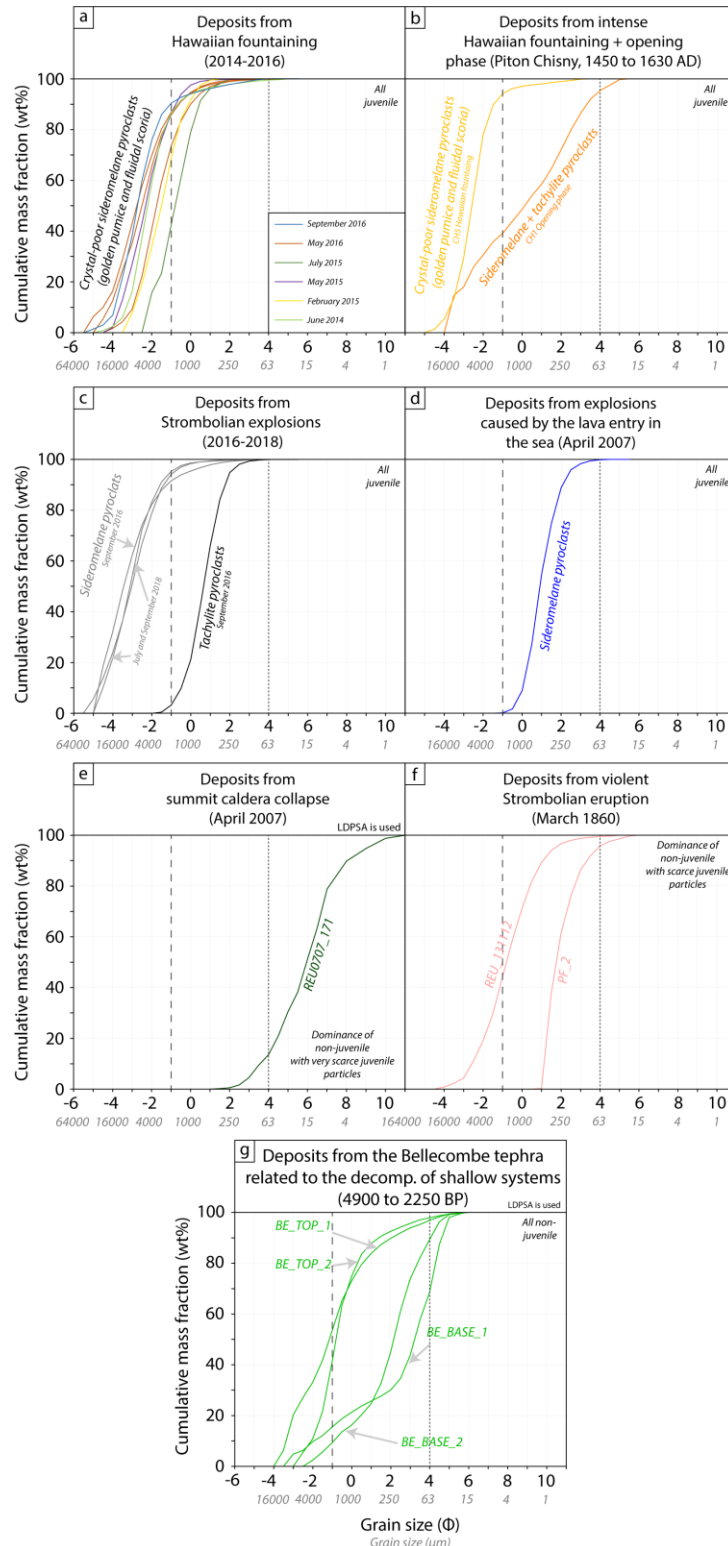
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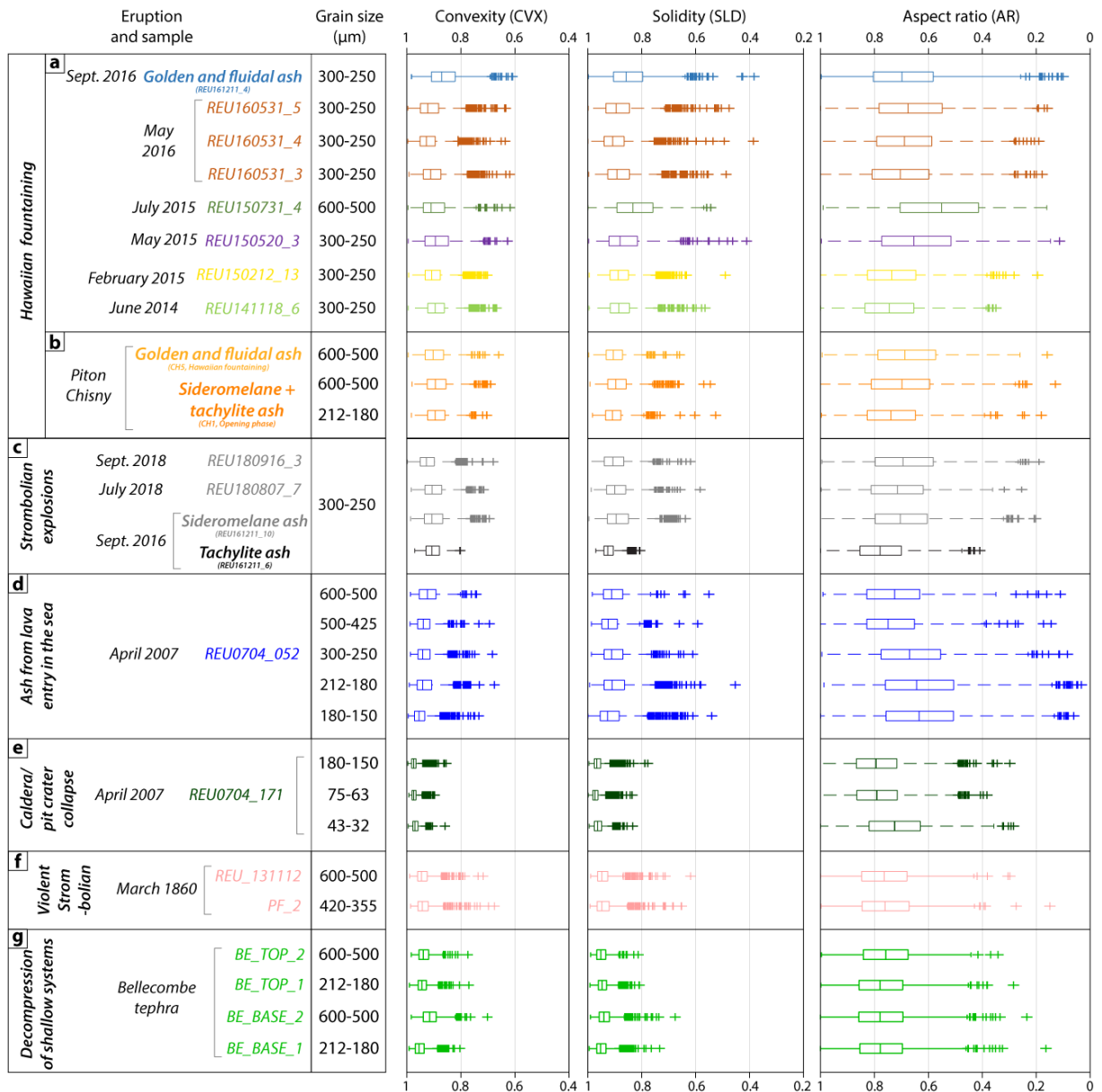
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**Fig. 3** – Eruptions and deposits pictures and SEM-BSE images of the respective ash particles, part 2. (a) Lava entry in the sea producing a laze and ash emission. (b) Ash emission during major caldera collapse at PdF. Particles within the particle clusters are randomly distributed. (c) Ash deposits from the March 1860 summit eruption at PdF. Sketch from Velain (1878) representing the Brûlant crater a few years after the eruption. (d) Bellecombe tephra. Particles within the particle clusters are randomly distributed.



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709 **Fig. 4** – Particle Size Distribution of the different studied deposits. Coarse ash boundary is represented by the  
 710 black dashed line at  $-1 \Phi$  (2 mm). Fine ash boundary is represented by the black dashed line at  $4 \Phi$  (64 mm). The  
 711 samples are represented with a color code reflecting their associated eruptive styles. LDPSA is for Laser  
 712 Diffraction Particle Size Analyzer.



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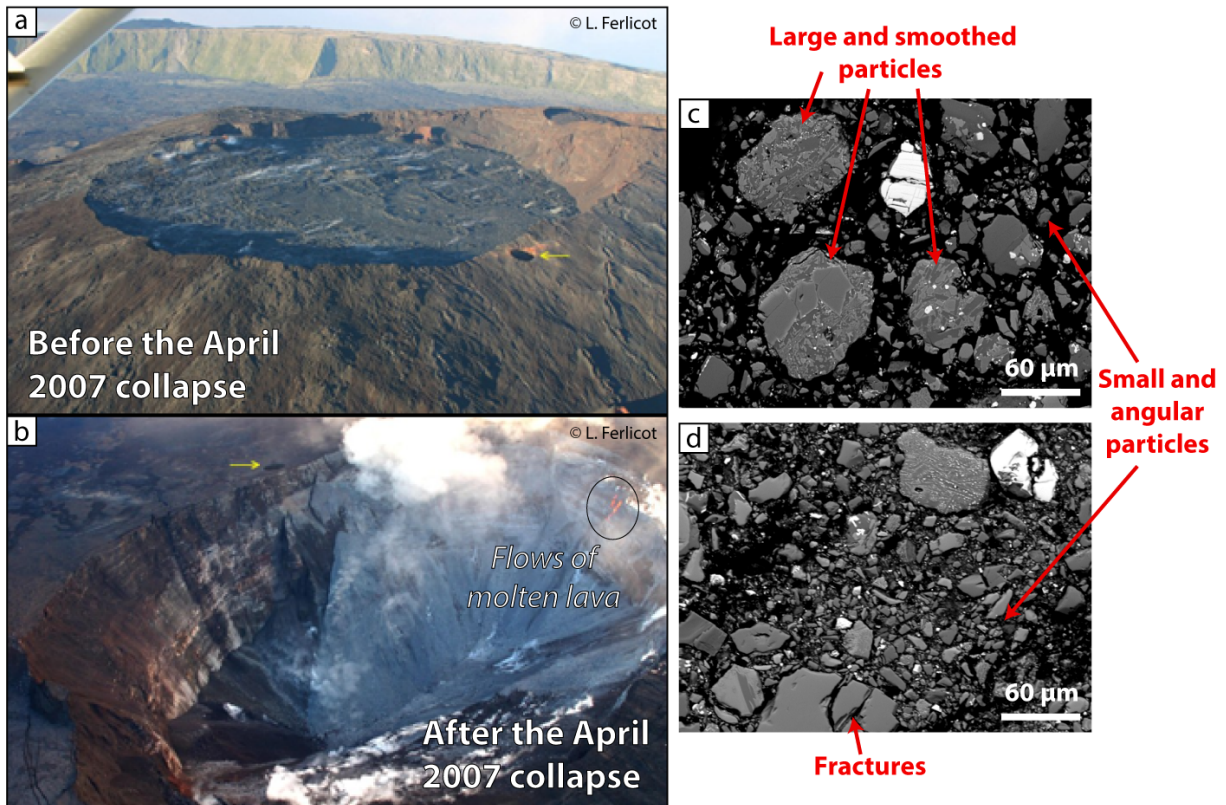
714 **Fig. 5** – Apparent Projected Shape of Ash for each sample, in terms of convexity, solidity and aspect ratio, with  
 715 the grain size used for each measurement. Vertical line within the boxes represents the median and box  
 716 boundaries represent the 25<sup>th</sup> and 75<sup>th</sup> percentiles of each dataset. Error bars represent 2.7σ of each dataset and  
 717 outlier data are represented with crosses. Sample color code as per Figure 4.

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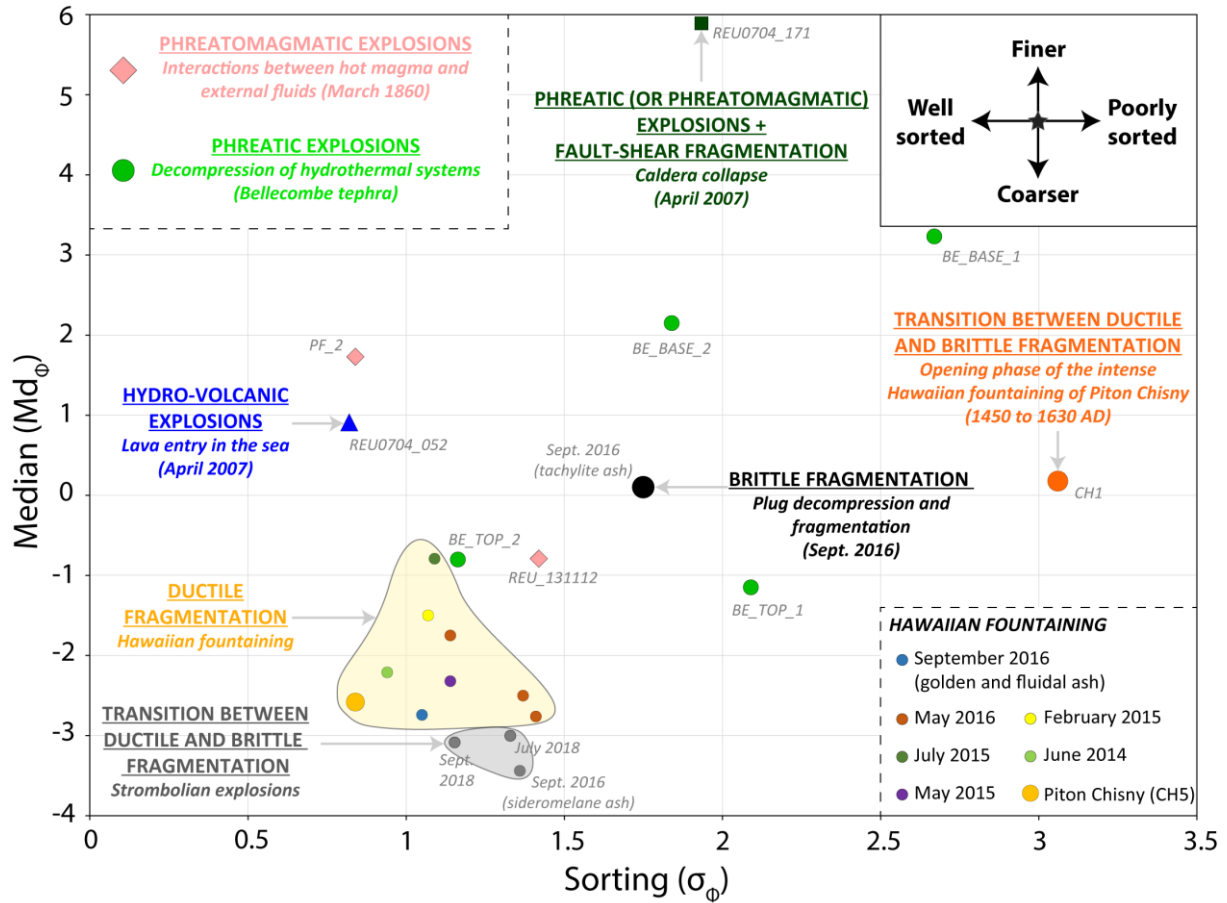
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**Fig. 6** – The Dolomieu caldera of PdF before (a) and after (b) the collapse, with visible lava flows. The yellow arrows mark the La Soufrière crater. (c) and (d) SEM-BSE images of the products emitted during the collapse.



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**Fig. 7** – Median ( $Md_\phi$ ) vs. sorting ( $\sigma_\phi$ ) plot. Sample color code as in Figures 4 and 5. Considering the proximal nature and similar sampling distance of all samples,  $Md_\phi$  is mostly considered as a fragmentation efficiency proxy, and  $\sigma_\phi$  reflects the degree of homogeneity of the deposits in term of texture, of componentry, and thus of fragmentation mechanisms.

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