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# Magmatic and phreatomagmatic contributions on the ash-dominated basaltic eruptions: Insights from the April and November–December 2005 paroxysmal events at Karthala volcano, Comoros

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## ► To cite this version:

Simon Thivet, Jean Carlier, Lucia Gurioli, Andrea Di Muro, Pascale Besson, et al.. Magmatic and phreatomagmatic contributions on the ash-dominated basaltic eruptions: Insights from the April and November–December 2005 paroxysmal events at Karthala volcano, Comoros. 2022. hal-03578724

**HAL Id: hal-03578724**

**<https://uca.hal.science/hal-03578724v1>**

Preprint submitted on 17 Feb 2022

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1 **Magmatic and phreatomagmatic contributions on the ash-dominated basaltic eruptions: insights**  
2 **from the April and November-December 2005 paroxysmal events at Karthala volcano, Comoros**

3

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19

20 **Abstract**

21 Basaltic eruptions are commonly associated with lava emissions and relatively weak  
22 explosive activities, but they can sometimes produce strong explosive eruptive phases. In April and  
23 November 2005, two paroxysmal eruptive events occurred within the summit crater of Karthala  
24 basaltic shield volcano (Grande Comore Island, Comoros), which hosted a water lake before each of  
25 these events. Both 2005 ash plumes spread across the Comoros Archipelago and heavily impacted  
26 the whole Grande Comore Island. Associated deposits on the volcano summit are extremely fine-  
27 grained (up to 50 wt% of fine ash < 63 µm for some analyzed layers) and rich in millimeter-sized  
28 rounded accretionary lapilli aggregates. Field observations, as well as textural and chemical analyses  
29 performed on both coarse- and fine-grained pyroclasts permit to identify juvenile and non-juvenile

30 components and quantify their peculiar characteristics. Coarse ash (710-1000  $\mu\text{m}$ ) mainly consists of  
31 juvenile pumice particles (vesicle number density  $N_v = 4.5 \cdot 10^4 \text{ mm}^{-3}$  and gas to melt ratio  $V_G/V_L = 1.5$ ,  
32 on average), characterized by free groundmasses and representative of magma portions ascending  
33 quickly within the eruptive conduits (up to  $10 \text{ m s}^{-1}$ ). A relatively low amount of juvenile scoria  
34 particles are also observed in the coarse ash fractions, which are characterized by magma degassing  
35 ( $N_v = 4.7 \cdot 10^4 \text{ mm}^{-3}$  and  $V_G/V_L = 0.5$  on average) and associated crystallization (occurrence of dendritic  
36 microlites). Non-juvenile fragments (from blocks to coarse ash) are dense lava or intrusive fragments.  
37 Their amount decreases exponentially toward the fine ash fractions, which are mainly composed of  
38 juvenile, blocky, dense and glassy particles that are characterized by unambiguous textural signs of  
39 brittle fragmentation (hackle lines, stepped features and cracks). We support that Molten Fuel-  
40 Coolant Interactions between highly porous fast ascending basaltic magmas and external waters  
41 occurred during the paroxysmal phases of the studied eruptions, leading to a brittle-dominant and  
42 efficient regime of magma fragmentation. Variable but large amount of fine ash grains through the  
43 stratigraphic depth of the deposits can be ascribed to the brittle failure of the vesicle walls of the  
44 initial porous magma. Concurrently, thermohydraulic explosions caused the host rock fragmentation  
45 at shallow level, generating the relatively coarse non-juvenile particles. A short-lived episode of  
46 intense lava fountaining associated with steam explosions eventually occurred at the end of the  
47 November 2005 paroxysm, forming the last and relatively coarse tephra layer at the top of the  
48 studied eruptive sequence. Each paroxysmal phase lasted about a day as each associated water lake  
49 and shallow water table progressively vaporized and dried away. Both eruptions ended with lava  
50 pond and weak lava fountaining activities confined within the summit crater. We conclude that the  
51 contributions of both magmatic processes and phreatomagmatic interaction mechanisms ultimately  
52 generated the grain size, grain component and grain texture variabilities observed within the  
53 paroxysmal deposits. This work contributes to a better understanding of the generation of unusual  
54 fine ash from basaltic explosions as well as their eruptive dynamics and associated mechanisms, from  
55 magma ascent in the conduit to the fragmentation level and the interaction with intra-crateric lake  
56 waters.

57

## 58 **Keywords**

59 Ash; basaltic; Karthala; Comoros; phreatomagmatism; texture

60

## 61 **Highlights**

- 62 • Karthala basaltic volcano produced two paroxysmal events in 2005
- 63 • Associated fallout deposits were highly spread out and rich in fine ash
- 64 • Tephra deposits record the signatures of shallow water-magma interactions
- 65 • Phreatomagmatism enhanced host rock and magma fragmentation efficiencies

66

## 67 **1. Introduction**

68 Volcanic activity represents an important part of natural hazards and risks on Earth (Small  
69 and Naumann, 2001). Active basaltic volcanism is widespread, present in all tectonic contexts, and is  
70 usually associated with effusive as well as weak explosive events from Hawaiian-like to Strombolian-  
71 like activities (Siebert et al., 2015). Low magma viscosities and low magmatic volatile contents  
72 (Taddeucci et al., 2015) ultimately lead to low efficiency fragmentation mechanisms when the  
73 magma is emitted at the surface, typically forming lava flows and relatively coarse-grained tephra  
74 (Cashman and Scheu, 2015). Many studies on active basaltic volcanoes highlight this common  
75 volcanic behavior, for instance at Piton de la Fournaise (e.g., Gurioli et al., 2018; Edwards et al., 2020;  
76 Thivet et al., 2020b), Stromboli (e.g., Gurioli et al., 2014; Gaudin et al., 2017; Thivet et al. 2021), Etna  
77 (e.g., Polacci et al., 2006; Andronico and Corsaro, 2011; Vergnolle and Gaudemer, 2012), as well as  
78 for Hawaiian (e.g., Head and Wilson, 1987; Stovall et al., 2011; Parcheta et al., 2013) and Icelandic  
79 volcanoes (e.g., Thordarson and Larsen, 2007; Pedersen et al., 2017; Bonny et al., 2018).

80 However, eruptive intensity can fluctuate, even during a single eruption, paroxysmal  
81 phases/events corresponding to the moments of greatest intensity. Basaltic volcanism can indeed  
82 shift toward more intense activities from Strombolian (Pioli et al., 2008) to sub-Plinian (Jordan et al.,  
83 2016) and Plinian-like (Costantini et al., 2010).

84 Basaltic paroxysmal events can be due to specific magmatic processes such as fast ascent of  
85 deep volatile-rich primitive magma (Métrich et al., 2010), gas transfer from deep to shallow levels  
86 resulting in destabilization of the upper magma column (Aiuppa et al., 2010), plug pressurization  
87 resulting in intermittent decompressions of the upper system (Thivet et al., 2020a), syn-eruptive  
88 magma degassing and crystallization resulting in the drastic increase of the magma viscosity in the  
89 volcanic conduit (Giordano and Dingwell, 2003) and sudden tectonic events such as caldera collapse  
90 resulting in secondary tephra fragmentation (Thivet et al., 2020c).

91 Moreover, magmas or lavas can also interact with external fluids at different levels (e.g.  
92 intra-crateric lake waters, fluids from hydrothermal systems, ocean) resulting in the so-called  
93 hydrovolcanism, which can lead to relatively explosive behaviors, as during phreatic or

94 phreatomagmatic activities (e.g., Mastin and Witter, 2000; Belousov and Belousova, 2001;  
95 Graettinger et al., 2013; Houghton et al., 2015; White et al., 2015; Zimanowski et al., 2015; Németh  
96 and Kósik, 2020). This kind of activities is reported for several basaltic systems, such as Piton de la  
97 Fournaise (e.g., Staudacher et al., 2009; Michon et al., 2013; Thivet et al., 2020c), Hawaii (e.g.,  
98 McPhie et al., 1990; Dvorak, 1992; Dzurisin et al., 1995) and Iceland (e.g., Dellino et al., 2012;  
99 Gudmundsson et al., 2012; Magnússon et al., 2012).

100           The large range of possible scenarios associated with basaltic explosive activity makes the  
101 related hazards and associated risks difficult to anticipate. With respect to weak intensity basaltic  
102 volcanism (e.g. lava flows, Hawaiian fountaining and mild Strombolian explosions; Thivet et al.  
103 2020b), paroxysmal basaltic events are more impactful mainly because of the emission of large  
104 amounts of fine-grained tephra, which can disperse over large areas (Jenkins et al., 2015). This is a  
105 critical point for awareness and anticipation of such volcanic crises as well as for hazard assessment  
106 (Nassor, 2001; Morin et al., 2016).

107           In the present study, we focus on two distinct paroxysmal basaltic phases of the Karthala  
108 shield volcano (Grande Comore Island, Comoros, Fig. 1) producing large plumes and regionally  
109 dispersed ash-rich deposits, in April and November 2005. Hazards associated with both paroxysms  
110 impacted most of the Grande Comore Island (Bachèlery et al., 2016): projections of ballistics were  
111 reported within the summit uninhabited area of the volcano, while significant ash fallouts and  
112 associated mudflows spread downslope within inhabited areas, including in Moroni, the capital of  
113 Comoros. Here, we summarize the morphological and eruptive history of the volcano in the last 30  
114 years and present the results of a detailed textural study of the ash-rich fallout deposits the both  
115 2005 paroxysms. These deposits are also compared with more typical Hawaiian-like deposits, in  
116 particular that from the 1977 eccentric eruption that occurred at low altitude on the southwestern  
117 flank of Karthala volcano. A chemical overview is also addressed in order to contextualize the  
118 compositions of the emitted magmas during the recent activity of the volcano. All these insights are  
119 discussed in order to constrain the syn-eruptive dynamics of the studied eruptions, in terms of  
120 reservoir, conduit processes and fragmentation mechanisms as derived from pre-, syn- and post-  
121 eruptive observations, as well as from features of the investigated deposits.

122

## 123 **2. Geological context**

### 124 **2.1. Comoros archipelago**

125 Karthala volcano is located on the Grande Comore (or Ngazidja) Island, which is the  
126 westernmost island of the Comoros archipelago that is formed by four main volcanic islands (from  
127 West to East: Grande Comore, Mohéli, Anjouan and Mayotte) in the northern part of the  
128 Mozambique Channel (Fig. 1a). The tectonic setting and the origin of the Comoros archipelago is the  
129 subject of controversy (e.g., Flower and Strong, 1969; Emerick and Ducan, 1982; Nougier et al., 1986)  
130 but recent tectonic and geomorphological measurements rule out the presence of a hotspot and  
131 interpret the Comoros archipelago as the boundary between two tectonic plates, favoring upwelling  
132 of mantellic magmas (Famin et al., 2020; Tzevahirtzian et al., 2021). Magnetic data support the  
133 presence of oceanic crust in this area (Coffin and Rabinowitz, 1987), which is also compatible with  
134 sedimentary and tectonic arguments to explain the presence of detrital crustal rocks in the area as  
135 well as abundant sandstone enclaves within the Comorian lavas. The Comorian volcanism started in  
136 Mayotte around 20 Ma, emitting a wide range of alkaline and variably undersaturated (from  
137 basanite-tephrite up to phonolite) magma compositions and forming a wide range of eruption styles,  
138 as evidenced by the variability of deposits and edifice morphologies in the archipelago (Bachèlery et  
139 al., 2016; Michon, 2016; Tzevahirtzian et al., 2021).

## 140 **2.2. Grande Comore Island and Karthala volcano**

141 Karthala basaltic shield volcano is the largest emerged active volcano of the area (Bachèlery  
142 et al., 2016). It represents one of the two recently active Comorian volcanic systems, with the newly  
143 formed submarine volcano of Mayotte (e.g., Cesca et al., 2020; Lemoine et al., 2020; Feuillet et al.,  
144 2021). Karthala (summit at 2361 m above sea level) forms the southern two-thirds of the Grande  
145 Comore Island, while La Grille volcano (summit at 1087 m above sea level) forms the northern part of  
146 it (Fig. 1b). A third and old massif, named M'Badjini, has also been identified in the southernmost  
147 part of the Island (Bachèlery and Coudray, 1993; Bachèlery et al., 2016). Karthala eruptions are  
148 mostly effusive and associated with weak explosive activities (lava fountaining and mild Strombolian  
149 explosions), although explosive events have usually occurred as shown by the alternation of lavas  
150 and pyroclastic deposits on the summit crater walls. Products are mainly alkaline basalts (Bachèlery  
151 and Hémond, 2016) that usually contain olivine, clinopyroxene and more rarely plagioclase  
152 phenocrysts.

153 The summit area of Karthala volcano corresponds to the junction of two well-defined rift-  
154 zones and hosts a remarkable caldera complex (Strong and Jacquot, 1970; Poppe, 2012) which is 3.5  
155 km long and 2.8 km wide (Fig. 1c). The presence of several collapse structures, scattered craters and  
156 pit craters of different sizes within the summit area, suggests that several shallow magmatic  
157 reservoirs feed the volcano summit activity. Geophysical investigations (Lénat et al., 1998; Savin et

158 al., 2001, 2005; Bernabeu et al., 2018) have permitted to define the structure of the summit area,  
159 which can be schematically divided in three main units: (i) an upper 200-400 m thick sequence of dry  
160 basaltic rocks, (ii) an intermediate 300 to 1200 m thick water-saturated unit and (iii) a deep  
161 conductive unit hosting the hydrothermal system. The second unit forms the ground water body  
162 whose surface expression was the Choungou-Chahalé intra-crateric lake (visible each of the 2005  
163 eruptions), while the deeper hydrothermal system is better developed below the northern caldera  
164 lobe where is located the Changou-Chagnoumeni pit-crater.

### 165 **2.3. Recent eruptive activity and summit morphology evolution of Karthala volcano**

166 According to the observations of the inhabitants of the Grande Comore Island, Karthala  
167 erupted at least 15 times during the 20th century (Bachèlery et al., 2016). In April 1977, an eccentric  
168 magmatic eruption occurred at low altitude (around 390 m above sea level) on the southwest flank  
169 of the volcano, producing lava fountains and lava flows, which invaded the adjacent villages of  
170 Singani and Hetsa, and which attained the coastline (Krafft, 1982). Then, and since the creation of the  
171 Karthala Volcanological Observatory (OVK) in 1988, five summit eruptions occurred in July 1991, April  
172 2005, November-December 2005, May-June 2006 and January 2007, which all took place within the  
173 Choungou-Chahalé crater, except the 2007 one, which took place within the Choungou-  
174 Chagnoumeni pit-crater (Fig. 1c). Mainly due to these volcanic events, the morphology of the summit  
175 area has frequently changed (Fig. 2). Bachèlery et al. (2016) detailed the pre- and syn-eruptive  
176 phases of each of these recent eruptions and the following paragraph summarizes the most  
177 important points derived from that study.

178 After the one-day, July 11, 1991 phreatic eruption (Bachèlery et al., 1995; Savin et al., 2005),  
179 a new crater formed (around 280 m in diameter and 40 m in depth) and was rapidly filled by an intra-  
180 crateric water lake that regularly changed in terms of water level and color, probably indicating  
181 acidity changes (Fig. 2a). The first precursors of the April 2005 eruption were recorded by mid-2003,  
182 characterized by a progressive increase in the seismic activity. The eruption (Fig. 2b) began on April  
183 16, on the same location of the lake, after a seismic crisis of three hours (drastic increase of the  
184 volcano-tectonic earthquakes frequency). The initial paroxysmal phase of the eruption was  
185 characterized by the emission of a gas-rich and ash-rich plumes associated with the projections of  
186 ballistics. During this phase, which lasted until the night between April 17 and April 18, rumbles,  
187 volcanic lightning, sulphur smell and mudflows were reported by several coastal villages (Morin et al.,  
188 2009). Most importantly, the two thirds of the island were covered by ash due to heavy ash fallouts.  
189 From April 18, the main explosive activity stopped, revealing that the lake disappeared and was  
190 replaced by an active lava pond associated with weak lava fountaining, which progressively in

191 intensity until the end of the eruption on April 19. A water lake formed again in the Choungou-  
192 Chahalé crater around May 8. Between May and November 2005, seismicity rates were oscillating,  
193 and a new eruption began on November 24 within the Choungou-Chahalé crater (Fig. 2c). As during  
194 the April eruption, the initial phase was characterized by the emission of a large plume, which  
195 reached the altitude of 12 km above sea level, with a lateral East-West extension over the whole  
196 Comoros archipelago. During the first day of the eruption, heavy ash falls obscured sunlight over half  
197 part of Grande Comore Island. The paroxysmal phase ended on November 25, again revealing that  
198 the lake disappeared and was replaced by an active lava pond at the bottom of the crater associated  
199 with weak lava fountaining, which progressively decreased in intensity until the end of the eruption  
200 on December 8. In total, these two similar eruptions affected around 250,000 people, mainly  
201 because of syn-eruptive ash fallouts and significant SO<sub>2</sub> emissions (Prata and Kerkmann, 2007) that  
202 affected aviation safety, air quality and drinking water resources (Morin et al., 2009; 2016). Syn- and  
203 post-eruptive mudflows caused a lot of disruption as well (Dille et al., 2020). A few centimeters of  
204 ash accumulated on the coast, while the summit area was covered by several meters of pyroclastic  
205 deposits (Smietana, 2007) made of blocks, bombs and ash (Fig. S1). After the November-December  
206 2005 eruption, the intra-crateric water lake has not returned. In May 2006, an increase in seismicity  
207 was recorded again. On May 28, approximately after two hours of seismic crisis, a new magmatic  
208 eruption started within the Choungou-Chahalé crater (Fig. 2d). The eruption was characterized by  
209 lava fountains, reaching 50 meters in height and forming a lava pond inside the crater, which caused  
210 no major threat to the population. The lava pond progressively cooled down until the end of the  
211 eruption on June 1 and raised up the crater floor by approximately a hundred meters. Finally, a one-  
212 day magmatic eruption occurred on January 13, 2007, within the Choungou-Chagnoumeni pit-crater  
213 (Fig. 1c), which was entirely filled by lava at the end of the eruptive activity (Bernabeu et al., 2018).  
214 This latter eruption ended a three-year cycle of notable activity. The current morphology of the  
215 summit area of Karthala volcano results from these recent eruptive events as well as constant  
216 erosion (Fig. 2e).

217

### 218 **3. Methods**

#### 219 **3.1. Field work and sampling**

220 Samples from 1977, April 2005, November-December 2005, May-June 2006 and January  
221 2007 eruptions were collected during several field expeditions performed in November 2006, April  
222 2007 and October 2018 (Table S1).



223           Concerning both 2005 eruptions, several stratigraphic logs were excavated and described  
224 within or near the summit caldera complex, 500 to 1500 meters from the center of the Choungou-  
225 Chahalé crater (Fig. 1c). Six logs were described in detail in November 2006 (annotated KAR\_5,  
226 KAR\_9, KAR\_10, KAR\_15, KAR\_17 and KAR\_18) and two others were added in October 2018  
227 (annotated KAR\_2005a and KAR\_2005b). In addition, 185 measurements of deposit thickness were  
228 realized within 10 km<sup>2</sup> in the summit area of the volcano to constrain ash dispersal and volumes and  
229 to characterize the lateral facies evolution of both 2005 eruptions (Table 1, Fig. S1). KAR\_10 log (Figs.  
230 3a and 3b), of 130 cm in thickness, represents the best sequence of the whole fallout deposits  
231 representative of both April and November 2005 paroxysms. Within this log, each identifiable layer  
232 (identified thanks to grain size and color changes, annotated with letters from KAR\_10\_a to  
233 KAR\_10\_u) was carefully sampled and analyzed. In addition, the very last identifiable deposit of the  
234 November 2005 paroxysm was also collected from the more proximal outcrop (KAR\_2005b), for a  
235 total of 22 collected samples. The other logs were described and used to (i) better distinguish and  
236 extrapolate the limit between the April 2005 and the November 2005 deposits, (KAR\_5, Fig. S2), (ii)  
237 observe the distal, medial and proximal deposit facies and measure thickness variations (KAR\_9,  
238 KAR\_15 and KAR\_17 respectively, Fig. S3), (iii) identify surge deposits that were also locally emplaced  
239 during the eruptions (KAR\_18, Fig. S4). , (iv) represent the heterogeneity and main features of the  
240 medial to proximal fallout deposit facies (KAR\_2005a, Fig. S5) and (v) complete the sampling for  
241 chemical analysis. Eruptive products from the lava pond and associated weak explosive lava fountain  
242 activities of both April and November-December 2005 eruptions were unfortunately inaccessible due  
243 to their low dispersion (mainly falling and recycling into the active lava pond).

244           Proximal bombs and lapilli from the magmatic May-June 2006 and January 2007 eruptions  
245 were collected near their respective vents (KAR\_2006 and KAR\_2007 respectively, Fig. 1c), in  
246 November 2006 and April 2007 respectively. In October 2018, the purely magmatic lava fountain  
247 deposits of the 1977 eccentric Singani eruption (KAR\_1977\_a and KAR\_1977\_b) were collected 100  
248 m far from the corresponding eruptive vent (Figs. 1b and S6), in order to be compared with the 2005  
249 ash-rich paroxysmal deposits of the summit activity. All the samples were systematically dried in the  
250 oven at 70°C for 48 hours. This low temperature drying step preserved any potential phases present  
251 on the samples, eased the manual sieving and allowed to perform accurate density measurements  
252 (Ross et al., 2022).

### 253 **3.2. Grain size analysis**

254           Grain size analysis was performed on the two pyroclastic samples of the 1977 eruption  
255 (KAR\_1977\_a and KAR\_1977\_b samples) as well as on all pyroclastic samples of the both 2005

256 paroxysms (samples that belong to the KAR\_10 log and KAR\_2005b sample), using both manual  
257 sieving (above 22  $\mu\text{m}$  in intermediate diameter using sieves each  $\frac{1}{2} \phi$ ) and laser diffraction  
258 measurements (below 45  $\mu\text{m}$ , with a detection limit of 0.1  $\mu\text{m}$  in equivalent diameter) for fine ash-  
259 rich samples, following the Eychenne et al. (2012) and Thivet et al. (2020c) procedures. Accretionary  
260 lapilli (when occurring) were gently dismantled beforehand. The overlap between 22 and 45  $\mu\text{m}$  for  
261 the two techniques was used to fit both results, in order to quantify representative particle size  
262 distributions for the whole eruptive sequences (Table S2). Traditional millimeter and phi scales were  
263 used to describe the deposit grain sizes, both being linked by this equation:  $\phi = -\log_2(d)$ , with  $d$  the  
264 particle grain size in mm (Taner, 1969).

### 265 **3.3. Tephra component and ash morphology analyses**

266 Pyroclast components were identified and quantified in the same samples as the grain size  
267 analysis and for different grain size fractions (Table S3), in order to distinguish juvenile (material  
268 representative of the magma feeding the eruptions) and non-juvenile (material representative of  
269 pre-existing fully solidified lithic rocks that are expelled/fragmented during the eruptions) particles  
270 (White and Houghton, 2006). This distinction was based on the color, surface texture, morphology,  
271 porosity, crystal content and assemblage of the particles. Particle counting was performed on a  
272 selection (based on sample quantity and quality) of  $\frac{1}{2} \phi$  grain size fractions previously isolated by  
273 sieving. Lapilli (when occurring) were systematically counted for each layer by hand and using a  
274 binocular microscope. Selected ash fractions between 500  $\mu\text{m}$  and 2 mm were counted using a  
275 binocular microscope. Selected ash fractions below 500  $\mu\text{m}$  were embedded in epoxy resin and then  
276 polished. These ash particles were then counted using a JEOL JSM-5910 LV scanning electron  
277 microscope (SEM), acquiring back-scattered electron (BSE) images with an acceleration voltage of 15  
278 kV. For all analyzed layers and analyzed grain size fractions, at least 500 particles were counted  
279 (when enough were available), in order to perform a statistically representative counting following  
280 the Van der Plas and Tobi (1965) statistic rules.

281 Ash morphology measurements, which correspond to an apparent projected shape of ash  
282 (APASH), were performed using the automated Malvern Morpho-Grainsizer Morphologi G3 following  
283 the method developed by Leibrandt and Le Penec (2015) and adapted by Thivet et al. (2020c). In  
284 total, 20979 particles were analyzed (Table S3) with different magnification (from x2.5 to x20  
285 depending on the analyzed grain sizes), allowing the representative quantification of several particle  
286 shape parameters: solidity (SLD), convexity (CVX), sphericity (SPH) and aspect ratio (AR) parameters  
287 are specifically discussed in this study, respectively representing the morphological roughness  
288 (mostly based on the particle area), the textural roughness (mostly based on the particle perimeter),

289 the angularity (based on both particle area and perimeter) and the elongation (ratio between the  
290 particle width and length) of the measured particles (specific definitions and formulas can be found  
291 in the previously cited studies). Ash morphology measurements were performed on different grain  
292 fractions, almost covering the whole range of ash sizes from 63 to 1400  $\mu\text{m}$  (loss of analytical  
293 precision below and above this range), for some 2005 paroxysmal layers (KAR\_10\_a, b, c, j, k, l, n and  
294 o; Carrier, 2019) as well as a layer from the 1977 eruption (KAR\_1977\_a).

### 295 **3.4. Textural analyses**

296 Textural analyses of tephra particles were then performed using several techniques. The  
297 different components identified were analyzed distinctly. For each component, several tephra layers  
298 have been selected based on sample quality and quantity. First, 3D images were acquired on coarse  
299 (710-1000  $\mu\text{m}$ ) and fine (< 63  $\mu\text{m}$ ) grain size fractions, using a Zeiss-Supra 55VP Gemini Column field  
300 emission scanning electron microscope (FE-SEM) and acquiring secondary electron (SE) images with  
301 an acceleration voltage of 3 kV. All the acquired 3D images are shown in Fig. S7 (98 images in total,  
302 performed on KAR\_10\_c, k, l, n, o, p, t, KAR\_2005b and KAR\_1977\_a samples). These images allowed  
303 to identify specific textural features on the particle surfaces for the different identified components.

304 Then, 2D images were also acquired using the JEOL JSM-5910 LV SEM, in BSE mode with an  
305 acceleration voltage of 15 kV, in order to investigate the internal textures of the ash particles. For  
306 each juvenile sample, the vesicle density number ( $N_V$ ), the vesicle to melt ratio ( $V_G/V_L$ ), the vesicle  
307 size distribution (VSD) and the total porosity fraction were reconstructed in 3D and quantified (Table  
308 S4) following the procedure adapted by Thivet et al. (2020b) and measuring about a thousand  
309 vesicles (equivalent to around seven ash particles on the 710-1000  $\mu\text{m}$  grain size fraction), in order to  
310 perform a statistically representative vesicle count (Van der Plas and Tobi, 1965). The acquired 2D  
311 images as well as their corresponding binarized counterparts are shown in Fig. S8 (68 images in total  
312 performed on KAR\_10\_a, b, c, l, n, o, KAR\_2005b and KAR\_1977\_a samples). Crystal contents are not  
313 quantified (due to their scarcity and/or relatively small sizes in some of the components) but  
314 qualitative observations on crystallinity is nevertheless shown, compared and discussed in this study.

315 Density and derived porosity, as well as connected porosity (vesicle connectivity)  
316 measurements (Table S4) were performed on 31 lapilli fragments from the 1977 eruption  
317 (KAR\_1977\_a and b samples) and both 2005 paroxysms (KAR\_10\_b, c, g, i, s and KAR\_2005b  
318 samples), following the procedure adapted by Thivet et al. (2020b). (i) Clast density measurements  
319 were performed using a Micromeritics Geopyc 1360 envelope density analyzer, (ii) porosity was  
320 derived using a vesicle-free rock density of 2874  $\text{kg m}^{-3}$ , which was determined by powdering four  
321 lapilli fragments (from KAR\_10\_a and KAR\_2005b samples) and by measuring their masses and

322 volumes using the Micromeritics Accupyc 1340 helium pycnometer. (iii) This pycnometer was also  
323 used to determine the skeletal volumes of the samples in order to calculate the connected porosity  
324 for each corresponding clast.

### 325 **3.5. Chemical analyses**

326 Analyses of bulk rock composition as well as in situ matrix and mineral chemistry (Table S5)  
327 were performed on selected clasts from the 2005, 2006 and 2007 eruptions (Smietana, 2007).

328 For bulk rock analyses, each selected sample (bombs or lapilli from KAR\_10\_c, l, s, KAR\_17,  
329 KAR\_2006 and KAR\_2007 samples) was powdered and dissolved in acid solutions. The final solutions  
330 were analyzed using a Horiba Jobin-Yvon Ultima C inductively coupled plasma atomic emission  
331 spectrometer (ICP-AES) for major elements analysis and using an Agilent 7500 quadrupole inductively  
332 coupled plasma mass spectrometer (ICP-MS) for rare earth element (REE) analysis. One additional  
333 bulk lava composition from the 1977 eruption has also been added from Desgrolard (1996) for  
334 comparison.

335 In situ measurements of matrix (glass) and mineral chemistry were performed on  
336 representative samples (KAR\_10\_c, s, KAR\_2006 and KAR\_2007) using a Cameca SX100 electron  
337 probe micro-analyzer (EPMA), with an acceleration voltage of 15 kV and a current intensity of 10 nA.  
338 A 10  $\mu\text{m}$  beam and a focused beam were used for the glass and crystal analyses, respectively.

339 In addition to these analyses, mineral phases on the 1977 (KAR\_1977\_a) and both 2005  
340 paroxysmal (KAR\_10\_f, t and KAR\_2005b) ash deposits were identified by X-ray diffraction (XRD)  
341 analysis (Table S5), using an Empyrean diffractometer, operated at 43 kV and 38 mA, following a  
342 method described in Thivet et al. (2020c). Qualitative Energy-dispersive X-ray spectroscopy (EDXS)  
343 spectrums were also acquired on some ash surfaces from both 2005 eruptions (KAR\_10\_c, p and t),  
344 in order to identify potential secondary phases from the primary ash surface backgrounds, using the  
345 Zeiss-Supra 55VP Gemini Column FE-SEM, with an acceleration voltage of 15 kV (Table S5).

346

## 347 **4. Results**

### 348 **4.1. Fallout deposit features and grain size characteristics**

349 Despite heavy rain falls and strong winds that caused constant soil erosion, the April and  
350 November 2005 deposits were still well preserved during the 2006 and 2018 field campaigns, both  
351 inside the summit Karthala caldera complex and on its outer proximal slopes. Their thickness and  
352 grain size drastically increase when moving towards the eruptive vent. Juvenile bombs and non-

353 juvenile blocks (from a few cm to a meter in equivalent diameter) are also very abundant in the  
354 proximal areas (Figs. 2e, S1 and S3c). Eruptive deposits sampled within the KAR\_10 log, which is the  
355 most complete stratigraphic log of the April and November 2005 fallout deposits (Figs. 3a and 3b),  
356 are located towards the northwest and 500 m of distance from the Choungou-Chahalé crater rim,  
357 which corresponds to a medial area (intermediate deposit thickness, absence of bombs and blocks  
358 and absence of cross-stratified surge deposits that are mainly visible in the northwestern part of the  
359 caldera, cf. Fig. S4). At this location, the April and November deposits are respectively 70 and 60 cm  
360 thick, with 12 (from KAR\_10\_a to KAR\_10\_l) and 10 (from KAR\_10\_m to KAR\_10\_u with also  
361 KAR\_2005b) identifiable and sampled layers, respectively. Each layer has a constant lateral thickness  
362 ranging from a minimum of 1 (KAR\_10\_e) up to 17 cm (KAR\_10\_f) within the April 2005 deposits,  
363 whereas in the November 2005 deposits the variation ranges from 3 (KAR\_10\_m) to 12 cm  
364 (KAR\_10\_t). Most of the layers, especially the fine-grained ones (e.g., KAR\_10\_d, f, h, p, r, and t), are  
365 enriched in millimeter-sized rounded accretionary lapilli.

366 Grain size parameters of each layer are represented in Fig. 3c, which reveal the dominantly  
367 fine-grained characteristics of the 2005 paroxysmal deposits, whose sorting values range between 1  
368 and 3  $\phi$  and whose grain size mean range between 32 and 500  $\mu\text{m}$ , except for the last emitted  
369 products of November (KAR\_2005b) that represent the coarsest tephra layer (grain size mean of 4  
370 mm) of all 2005 beds. In the whole sequence, 16 layers show unimodal grain size distributions (main  
371 modes ranging from 31 to 1000  $\mu\text{m}$ ), whereas 6 are bimodal (finer modes ranging from 31 to 90  $\mu\text{m}$   
372 and coarser modes ranging from 250 to 1400  $\mu\text{m}$ ). The coarsest grains are 1.6 cm in diameter (except  
373 for the KAR\_2005b sample, which contains clasts up to 3.2 cm in diameter) and most importantly, 15  
374 layers contained fine ash particles as fine as 1  $\mu\text{m}$ .

375 April and November 2005 paroxysmal deposits are much finer-grained than the 1977 typical  
376 magmatic deposit (Table S2), which is characterized by unimodal grain size distributions (modes  
377 between 4 and 8 mm), the coarser grains being 22.6 cm in diameter and lacking particles finer than  
378 45  $\mu\text{m}$  (grain size mean values at 5.66 mm).

#### 379 **4.2. Tephra grain components and associated characteristics**

380 Four main tephra components are identified in each sampled layer of the April and  
381 November 2005 deposits: (i) coarse juvenile pumice (Fig. 4), (ii) juvenile scoria (Fig. 5), (iii) non-  
382 juvenile (Fig. 6) and fine juvenile dense glassy (Fig. 7) grains. They are described hereafter based on  
383 the observations made on coarse (lapilli and coarse ash in the 710-1000  $\mu\text{m}$  grain size fraction) and  
384 fine ash (< 63  $\mu\text{m}$ ) grain size fractions. Juvenile vs. non-juvenile component distributions as a function  
385 of grain size (Fig. 8) as well as ash morphological features (Fig. 9) are also described.

#### 386 **4.2.1. Juvenile pumice grains**

387 Juvenile pumice grains are characterized by a yellow and shiny color, well-visible on the  
388 lapilli-sized clasts, whose surfaces are often characterized by bread-crusting cracks (Figs. 4a and 4b).  
389 They have also characterized by a pumiceous matrix. Scarce (< 5 vol%) micro-phenocrysts (up to 200  
390  $\mu\text{m}$  in length) of olivine, clinopyroxene and plagioclase are observed in some lapilli clasts and rare  
391 loose crystals are present in the ash fraction. SEM images of the coarse ash fraction reveal that  
392 pumice grains can either have sub-rounded shapes with no broken angular surfaces (Fig. 4c) or can  
393 have some broken angular surfaces characterized by the occurrence of microscopic hackle lines,  
394 stepped features and branching quench cracks (Fig. 4d). Pumice ash particles are also characterized  
395 by the scarcity of both micro-phenocrysts and microlites, hence are mainly composed of glass and  
396 vesicles, which can be rounded (spongy texture, Figs. 4c and 4e), elongated (tube pumice texture,  
397 Fig. 4d), or highly deformed (shrinkage texture, Fig. 4f) with often thin vesicle walls (around 1  $\mu\text{m}$ )  
398 that can be sometimes broken (Fig. 4g). Highly fluidal, Pele's hairs and tears are not observed in the  
399 2005 deposits.

400 As a comparison, the 1977 magmatic deposit (Fig. S6) is composed of fluidal (with Pele's hairs  
401 and tears) particles, characterized by a slightly lower content of vesicles and slightly higher content of  
402 microlites than the 2005 juvenile pumice grains. The 1977 ash grains do not show any significant  
403 angular faces, with no cracks and stepped features (Fig. S7). Moreover, these particles do not show  
404 any significant deformation features such as vesicle shrinkage or breakage, contrary to some of the  
405 2005 ash particles.

#### 406 **4.2.2. Juvenile scoria grains**

407 Juvenile scoria lapilli grains are characterized by spiny and rough surfaces without any  
408 significant cracks and bread-crusting features, as well as by dark/mat colors and by less abundant  
409 vesicles than their pumiceous counterparts (Figs. 5a and 5b). Their low content in micro-phenocrysts  
410 (< 5 vol%) is similar to those of pumice grains. SEM images of the coarse ash fraction highlight that  
411 scoria grains have irregular and rough shapes (Fig. 5c). These scoria grains are also characterized by a  
412 variable content in microlites (from around 5 vol% to a fully crystallized matrix, representing  
413 sideromelane to tachylite textures) accompanied with a decrease in vesicle abundance compared to  
414 the pumice grains. Microlites (up to 20  $\mu\text{m}$  in length) are sometimes protruding from the grain  
415 surfaces (Fig. 5d) suggesting tachylitic textures. Microlites are made of plagioclase, clinopyroxene,  
416 olivine and Fe-Ti oxides (in order of abundance) and have dendritic textures. The higher the microlite  
417 content, the more coalesced the vesicles (Figs. 5e, 5f and 5g).

#### 418 **4.2.3. Non-juvenile grains**

419 Non-juvenile grains are dense and characterized by blocky shapes (Figs. 6a, 6b and 6c). These  
420 grains are sometimes partially embedded in vesicular juvenile melt (Figs. 6d and 6e) and show  
421 variable crystallized textures, from tachylite (rapid crystallization textures that could correspond to  
422 ancient lavas, Fig. 6f) to micro-gabbro (relatively slow crystallization texture that could correspond to  
423 plutonic rocks, Figs. 6g and 6h), with the occurrence of plagioclase, clinopyroxene and olivine crystals  
424 as well as oxides from 10 to 200  $\mu\text{m}$  in length (Figs. 6e, 6f, 6g and 6h). Note that rare micro-gabbroic  
425 enclaves containing interstitial melt in between their own crystals also occur inside coarse juvenile  
426 fragments (Fig. 6i). Hence, we suggest that these enclaves represent crystallized parts of the involved  
427 magmatic chamber and are thus considered as juvenile material.

#### 428 **4.2.4. Fine ash characteristics**

429 The fine ash ( $< 63 \mu\text{m}$ ) grains include the three components previously described except that  
430 the coarse juvenile pumice component is replaced by the fine juvenile dense glassy one (Fig. 7a).  
431 Accretionary lapilli aggregates are omnipresent in the fine-grained layers of both April and November  
432 2005 deposits and are mainly composed of fine ash particles, with some scarce coarse ash grains, the  
433 coarser ones being in the inner parts and the finer ones being on the outer rims (Fig. 7b). Contrary to  
434 the relatively coarse-grained juvenile pumice grains that are glassy and highly vesicular (Fig. 4), the  
435 corresponding fine-grained particles are glassy but dense, with scarce  $< 50 \mu\text{m}$  vesicles (Figs. 7a and  
436 7b) and are characterized by ubiquitous microscopic hackle lines and stepped features with scarce  
437 branching quench cracks on their surfaces (Figs. 7c, 7d, 7e, 7f, 7g, 7h and S7). Hackles lines are  
438 characterized by repeated irregularities that are parallel to sub-radial on the broken faces of the  
439 particles. The length of these irregularities does not exceed  $40 \mu\text{m}$  and are repeated every 1 to 10  
440  $\mu\text{m}$ . The stepped features yield irregular particles with an uneven surface made up of three-  
441 dimensional polyhedral elements. The branching quench cracks are fracture networks within fresh  
442 glass. SEM images also highlight the occurrence of many very fine particles down to  $0.1 \mu\text{m}$  on the  
443 surface of the fine ash particles (these very fine particles are not visible on the grain size  
444 measurements because of their relatively low masses and volumes). Some particles are also  
445 characterized by secondary thin mineral depositions on their surface ( $< 1 \mu\text{m}$  thin), which could not  
446 be clearly identified by EDXS analysis in term of chemical composition (Table S5). Most importantly,  
447 the juvenile dense glassy grains undoubtedly dominate the fine ash grain size fractions compared to  
448 both non-juvenile (corresponding to loose crystal fragments) and juvenile scoria grains.

449 XRD measurements performed on the fine ash fractions ( $< 45 \mu\text{m}$ ) of KAR\_10\_f and KAR\_10\_t  
450 samples show similar results as the measurements performed on coarser ash fractions (250 to 355  
451  $\mu\text{m}$ ) of KAR\_10\_f (April 2005 paroxysm), KAR\_10\_t and KAR\_2005b (November 2005 paroxysm), as

452 well as KAR\_1977a (1977 eruption) samples: only typical magmatic phases are detected among with  
453 plagioclase, clinopyroxene, rhönite, olivine, and Fe-Ti oxide in order of average abundance (Table S5),  
454 with the notable absence of hydrothermal phases. The analyzed ash sample from the 1977 eruption  
455 (KAR\_1977a, 250 to 355  $\mu\text{m}$ ) show a higher content of olivine than in the 2005 products, in  
456 accordance with the optical observations.

#### 457 **4.2.5. Ash components and morphologies**

458 Juvenile vs. non-juvenile component proportions have been determined within all April and  
459 November 2005 layers and within different grain size fractions. No significant differences are  
460 observed between the different analyzed layers (Table S3). However, the component proportions  
461 significantly vary with the grain size (Fig. 8) for all the layers. Non-juvenile grains occur mostly within  
462 the coarser grain size fractions, with a juvenile grains/non-juvenile grains ratio of 1 within the 22.6-  
463 32 mm grain size fraction, on average. The ratio is then oscillating at low values (between 1 and 7, on  
464 average) for grain size fractions between 32 mm and 500  $\mu\text{m}$ . From 500  $\mu\text{m}$  and towards fine-grained  
465 fractions, the ratio is drastically increasing, reaching an average value of 37 for the 31-45  $\mu\text{m}$  grain  
466 size fraction. Note that standard deviations considering all analyzed layers for each grain size bin  
467 (represented by the error bars in Fig. 8) are decreasing from coarse-grained to fine-grained fractions,  
468 which suggests that component proportion variability from a layer to another is decreasing towards  
469 fine-grained fractions. Note that no distinctive quantification has been made between the different  
470 juvenile components (complexity of the observed textures would often lead to significant and  
471 unquantifiable errors). However, preliminary observations made through different grain size  
472 fractions and layers, suggest that juvenile pumice and dense glassy are dominant compared to the  
473 juvenile scoria within the coarse-grained (lapilli and coarse ash) and fine-grained (fine ash) fractions  
474 respectively. Juvenile scoria grains are almost absent in the fine-grained fractions (Fig. 7).

475 As shown by ash morphology measurements, all 2005 analyzed layers (KAR\_10\_a, b, c, j, k, l,  
476 n and o) show similar morphological signatures and ranges, looking at similar grain size fractions  
477 (Table S3). However, focusing on a representative layer of both 2005 paroxysms (KAR\_10\_a), ash  
478 morphology quantifications (Table S3) show differences in function of the observed grain sizes: SLD  
479 (Fig. 9a), CVX (Fig. 9b) and SPH (Fig. 9c) ranges progressively evolve toward higher values with  
480 decreasing grain sizes, reflecting the occurrence of relatively irregular-shaped and/or smoothed  
481 particles within coarse grain size fractions (e.g., 500-1000  $\mu\text{m}$ ), whereas blocky-shaped and/or  
482 angular particles dominate within finer grain size fractions (e.g., 63-125  $\mu\text{m}$ ). AR parameter exhibits  
483 the higher ranges of values (Fig. 9d) for deposits from the 1977 eruption (KAR\_1977\_a) and both  
484 2005 paroxysms. This variability does not change with grain size and reflects that elongated or blocky



485 equidimensional grains coexist within the deposits. On the whole, 2005 ash particles have a  
486 significantly different morphological signature than to those of the 1977 eruption (Fig. 9). This shift  
487 reflects the presence of both well rounded and highly elongated particles within the 1977 magmatic  
488 sample, leading to a wider range of fluidal morphologies in terms of SLD, CVX, SPH and AR, compared  
489 2005 samples. Note that the significant data overlap between the 1977 and 2005 samples is  
490 reinforced by the occurrence of abundant olivine loose crystals within the crystal-rich 1977 sample,  
491 which have similar morphological properties as the blocky-shapes and angular 2005 particles.

#### 492 **4.3. Textural quantifications on porosity**

493 VSD analysis performed on the coarse juvenile ash particles (700-1000  $\mu\text{m}$ ) shows distinct  
494 results for the pumice grains (Fig. 10a) and for the scoria grains of the April and November 2005  
495 paroxysmal deposits (Fig. 10b). VSDs of pumice grains show, on average, a symmetrical and unimodal  
496 distribution, vesicle diameters ranging from 5 to 400  $\mu\text{m}$  with a main mode around 90  $\mu\text{m}$ . Note that  
497 KAR\_1\_c and KAR\_1\_n samples have a second but minor VSD mode between 200 and 400  $\mu\text{m}$ . On all  
498 these pumice grains, the measured vesicle content is 61 vol% on average. In contrast, VSDs of scoria  
499 grains show, on average, a significantly different distribution, vesicle diameters still ranging from 5 to  
500 400  $\mu\text{m}$  but with an asymmetrical distribution, main modes being shifted towards larger vesicle  
501 diameters (between 100 and 200  $\mu\text{m}$ ) and with second minor modes up to 400  $\mu\text{m}$ . Moreover, the  
502 measured vesicle content is half (31 vol%) that of the pumice grains. The 1977 ash show a slightly  
503 different pattern from the two 2005 components described before (Fig. 10b): a unimodal distribution  
504 is observed, with vesicle diameters ranging from 10 to 400  $\mu\text{m}$  and with a mode at around 120  $\mu\text{m}$   
505 and a total vesicle content of 54 vol%.

506 Textural distinctions between these different components are also visible focusing on  $N_V$  and  
507  $V_G/V_L$  values (Fig. 10c).  $N_V$  values for both pumice and scoria grains range in the same order of  
508 magnitude (from  $2 \cdot 10^4$  and  $8 \cdot 10^4 \text{ mm}^{-3}$ ) and have similar average values ( $4.5 \cdot 10^4$  and  $4.7 \cdot 10^4 \text{ mm}^{-3}$  for  
509 pumice and scoria respectively). On the other hand,  $V_G/V_L$  ratios are significantly lower in the scoria  
510 than in the pumice grains (1.46 and 0.53 respectively). Note that the 1977 sample show the lowest  
511  $N_V$  ( $2.4 \cdot 10^4 \text{ mm}^{-3}$ ) with an intermediate  $V_G/V_L$  ratio (1.17).

512 Finally, porosity and vesicle connectivity measurements (Fig. 10d) highlight that the juvenile  
513 lapilli fragments (both pumice and scoria clasts) from both 2005 eruptions have relatively high  
514 porosities (from 61 to 90 vol%, 82 vol% on average) compared to the juvenile lapilli fragments from  
515 the 1977 lava fountaining eruption (from 45 to 72 vol%, 60 vol% on average). In parallel, the  
516 connected porosity measured within the 2005 lapilli clasts (from 71 to 95 vol%, 84 vol% on average)

517 are slightly lower than that measured on the 1977 lapilli clasts (from 66 to 99 vol%, 91 vol% on  
518 average).

519 All the detailed data described in this section can be found on Table S4.

#### 520 **4.4. Magma chemical composition evolution from 1977 to 2007**

521 Magma compositions, from the 1977 eruption until the 2007 one, show a complex  
522 evolutionary trend (Fig. 11). The 1977 bulk rock composition shows a relatively high MgO content of  
523 13.7 wt%, reflecting the olivine-rich nature of the emitted magma, with also a relatively high  
524 CaO/Al<sub>2</sub>O<sub>3</sub> ratio of 0.90 reflecting the presence of clinopyroxene (in accordance with the XRD  
525 measurements and the optical observations). The next emitted magmas were aphyric, with, in April  
526 2005, the lowest MgO content (6.4 wt%) and the lowest CaO/Al<sub>2</sub>O<sub>3</sub> ratio (0.79) recorded on the 1977-  
527 2007 period. Then, the November 2005 (6.6 wt% of MgO, CaO/Al<sub>2</sub>O<sub>3</sub> ratio of 0.81), May-June 2006  
528 (6.6 wt% of MgO, CaO/Al<sub>2</sub>O<sub>3</sub> ratio of 0.81) and January 2007 (6.7 wt% of MgO, CaO/Al<sub>2</sub>O<sub>3</sub> ratio of  
529 0.83) magmas progressively show a slight increase in these values.

530 Matrix glass compositions (Fig. 11) show similar or lower MgO and CaO/Al<sub>2</sub>O<sub>3</sub> ratio values  
531 from each corresponding bulk rock composition. Glass compositions closed to bulk rocks correspond  
532 to uncrystallized pumiceous and dense glassy grains, while glass compositions that are progressively  
533 depleted in MgO content with decreasing CaO/Al<sub>2</sub>O<sub>3</sub> ratios correspond to partially crystallized scoria  
534 grains.

535 Crystals of plagioclase, clinopyroxene and olivine, as well as Fe-Ti oxides were analyzed for  
536 the 2005, 2006 and 2007 eruptions. Only one euhedral micro-phenocryst of plagioclase was  
537 measured at An<sub>78</sub> (November-December 2005 eruption, in equilibrium with the bulk rock  
538 composition) whereas 27 microlites (dendritic texture in scoria grains) of plagioclase were measured  
539 ranging from An<sub>55</sub> to An<sub>66</sub> for the 2005 and 2007 eruptions (in equilibrium with their surrounding  
540 depleted matrices). 46 clinopyroxenes were analyzed, for the 2005, 2006 and 2007 eruptions,  
541 showing compositions ranging between salite and augite. 60 analyses were realized on olivine  
542 crystals for the 2005, 2006 and 2007 eruptions, ranging from Fo<sub>39</sub> to Fo<sub>86</sub>: Fo<sub>81-86</sub> olivines are euhedral  
543 antecrysts that were measured in the May-June 2006 products, Fo<sub>76-81</sub> olivines are euhedral or  
544 skeletal micro-phenocrysts that were measured in the 2005, 2006 and 2007 eruptions and in  
545 equilibrium with their corresponding bulk rock compositions, and Fo<sub>39-76</sub> olivines are dendritic  
546 microlites in equilibrium with their surrounding depleted matrices that were mainly analyzed in the  
547 2005 scoria products.

548 All the detailed data described in this section, as well as crystal compositions and REE  
549 concentrations can be found on Table S5.

550

## 551 **5. Discussion**

552 The distinction between magmatic and phreatomagmatic fragmentation regimes, especially  
553 concerning mafic magmas, is widely discussed in volcanology (e.g., Houghton et al., 1996; Ort and  
554 Carrasco-Núñez, 2009; Graettinger et al., 2013; Colombier et al. 2019a, Latutrie and Ross, 2020;  
555 Thivet et al., 2020c), but in some cases, is unattainable looking only at deposit features of a given  
556 eruptive event (White and Valentine, 2016). In our study, pre-, syn and post-eruptive visual  
557 observations of the active crater are available, providing a reliable support for interpreting on the  
558 eruption dynamics (Fig. 2). We also had the opportunity to study a complete and well-preserved  
559 deposit sequence (Fig. 3), representative of the two targeted explosive events. In this section, both  
560 magmatic and phreatomagmatic processes are addressed in order to explain the physical  
561 characteristics of the related deposits and products, such as grain size (Fig. 3), texture (Figs. 4, 5, 6, 7  
562 and 10) and shape variations (Fig. 9), as well as grain components distributions (Fig. 8) and chemical  
563 compositions (Fig. 11).

### 564 **5.1. The magmatic contribution on the conduit conditions: magma degassing and ascent rates** 565 **controlling the initial eruptive dynamics**

566 Pre-eruptive seismic activity recorded since mid-2003 suggested a deep magmatic input from  
567 20 km below sea level under the summit caldera (Bachèlery et al., 2016). Deep volcano-tectonic  
568 events were recorded both before and during the 2005 eruptive events by the seismic network of the  
569 Karthala Volcano Observatory (OVK). This seismic unrest anticipated a new phase of intense eruptive  
570 activity, the highest since the phreatic eruption of 1991 and heralded the 2005-2007 sequence of  
571 eruptions (Bachèlery et al., 2016). The decrease of both CaO/Al<sub>2</sub>O<sub>3</sub> ratio and MgO content starting  
572 from April 1977 (composition corrected for the cumulative olivine content, step 1 in Fig. 11) to April  
573 2005 bulk rocks (step 2 in Fig. 11), reflects a differentiation trend within feeding magmatic system  
574 during this period. We can speculate that volatiles concentration increased in the cooling and  
575 evolved melts during the 1977-2005 time period. On the other hand, the progressive increase of  
576 MgO and CaO/Al<sub>2</sub>O<sub>3</sub> values from April 2005 and January 2007 eruptions reflects progressively less  
577 evolved compositions during this period and highlights that a new magma input is progressively  
578 contributing to the April 2005, November-December 2005, May-June 2006 and January 2007  
579 eruptions (step 3 in Fig. 11). The eruption of these magmas is likely the result of the mixing of this  
580 evolved and this more primitive magmatic components.

### 581 **5.1.1. Insights from the juvenile pumice component**

582 Because of their small size and their textural signatures suggesting their interaction with  
583 external water, we are confident that the analyzed ash particles are rapidly quenched when formed  
584 (Xu and Zhang, 2002), hence representative of the magma texture and composition at the  
585 fragmentation level.

586 The juvenile pumice component (Fig. 4), which is predominant within the 2005 fallout  
587 deposits (Fig. 8), represents microlite-free and highly porous magma portions. Pumice ash grains are  
588 characterized by high  $N_v$  values and high  $V_G/V_L$  ratios (Fig 10c), compared for instance to the recent  
589 and typically weakly explosive basaltic eruptions of Piton de la Fournaise volcano (Thivet et al.,  
590 2020a; 2020b). These data, together with mainly unimodal VSDs (Fig. 10a), suggest that the  
591 corresponding magma portion exhibit one significant episode of degassing, resulting in bubble  
592 nucleation and growth, during the relatively high magma decompression in the volcanic conduit  
593 (Shea et al., 2010, 2017). From these insights, we can speculate the occurrence of a closed-degassing  
594 regime for the magmas emitted during both paroxysms (Figs. 12a and 12b).

595 Pre-eruptive magma temperatures are estimated using the MgO-dependent thermometer  
596 for basaltic melts (using bulk rocks) at atmospheric pressure, from Thivet et al. (2020a): 1121 and  
597 1131 °C are the average estimated magma temperatures for the April and November-December  
598 eruptions, respectively. In parallel, Lange et al. (2009) model, based on plagioclase-melt equilibrium,  
599 is used to estimate residual dissolved H<sub>2</sub>O content in the magmatic melts, giving relatively low  
600 dissolved H<sub>2</sub>O contents, typically < 1 wt%, for both eruptions. These estimations together with the  
601 textural  $N_v$  parameter, are also used to quantify magma decompression rates using the  $N_v$ -  
602 dependent decompression rate meter of Toramaru (2006). Relatively high  $N_v$  values measured within  
603 several distinct deposit layers of both 2005 paroxysms lead to variable but relatively high magma  
604 decompression rates between 0.16 and 0.31 MPa s<sup>-1</sup> compared to relatively lower magma  
605 decompression rates (< 0.15 MPa s<sup>-1</sup>) estimated for recent and typically weak explosive basaltic  
606 eruptions of Piton de la Fournaise (Thivet et al., 2020a, 2020b). Magma decompression rates are  
607 typically linked with ascent and mass discharge rates (Gonnermann and Manga, 2003) as well as  
608 eruptive dynamic regimes and fragmentation efficiencies (Cashman and Scheu, 2015). Interestingly,  
609 the higher calculated decompression rate is from the KAR\_2005b layer (0.31 MPa s<sup>-1</sup>, corresponding  
610 to an ascent rate of about 10 m s<sup>-1</sup>), which corresponds to the last identifiable deposits of the  
611 November 2005 paroxysm, which is also the coarsest tephra layer of the 2005 paroxysms sequence  
612 (Fig. 3c). We suggest that this high decompression and ascent rate was the main contributor to the  
613 formation of a short-lived but intense lava fountaining at the origin of the deposition of the

614 KAR\_2005b layer that is part of a relatively large bomb field area (Fig. S1c). Vesicle connectivity  
615 measurements performed on lapilli clasts fit with this interpretation as these measurements fall in  
616 the range characterized by sustained lava fountaining (Fig. 10d). As a comparison, the measured  
617 clasts from the 1977 eruption show relatively lower porosities and lower vesicle connectivity values,  
618 reflecting much less intense magmatic activities ranging between Hawaiian-style and mild  
619 Strombolian-style dynamics. The absence of fine ash particles in the KAR\_2005b layer (Fig. 3c)  
620 suggests the predominance of purely magmatic dynamics that can be explained by the progressive  
621 end of phreatomagmatic dynamics as the intra-crateric water lake and the water table system dried  
622 up. The short-term increase in magma ascent rate may also have contributed to the cessation of  
623 water-magma interactions (Fig. 12b). Note that non-juvenile blocks and coarse ash present in the  
624 KAR\_2005b layer are either attributed to remnant steam explosions or to purely magmatic processes  
625 that can produce such deposits, as supposed for instance at Stromboli (Métrich et al. 2005; Calvari et  
626 al. 2006).

#### 627 **5.1.2. Insights from the juvenile scoria component**

628 Besides the juvenile pumice component, variably crystallized magmatic portions are  
629 associated with the presence of the juvenile scoria component (Fig. 5), which occurred in all the  
630 studied layers but in relatively small proportions (as also shown in Carlier, 2019). We associate this  
631 scoria component with a process of crystallization near the conduit margins, induced by degassing  
632 during the magma decompression that is a common process in basaltic melts (e.g., Lipman et al.,  
633 1985; Applegarth et al., 2013; Thivet et al., 2020b). More than a crystallization process, it can be  
634 referred as a zoning of the textural features of the ascending magma column (Fig. 12). The  
635 occurrence of syn-eruptive crystallization during both 2005 Karthala paroxysms, and also during the  
636 2006 and 2007 relatively weak explosive eruptions, is evidenced by differentiated glass compositions  
637 (microlites of olivine deplete the MgO content of the glass and microlites of clinopyroxene and  
638 plagioclase decrease or increase the CaO/Al<sub>2</sub>O<sub>3</sub> ratio, respectively) with respect to their bulk rock  
639 counterparts (step 4 in Fig. 11). Crystal compositions and textures can also be used as an indicator of  
640 the presence of this degassed magma: degassing-driven crystallization usually forms dendritic  
641 textures with no preferential orientations and most likely occurred during the final stages of the  
642 magma ascent before the fragmentation (Applegarth et al. 2013). This explains why the analyzed  
643 microlites are not in equilibrium with the bulk rocks but rather with their surrounding depleted  
644 matrices.

645 The occurrence of microlites in the scoria component is also associated with vesicle  
646 coalescence and gas loss processes, clearly visible on VSDs and  $N_v$  vs.  $V_G/V_L$  patterns (Figs. 10b and

647 10c). This textural evolution reflects that a large proportion of the magma in the conduit is bubbly  
648 and non-crystallized (corresponding the juvenile pumice component) while small proportion is more  
649 degassed and crystallized (corresponding to the juvenile scoria component). This leads to a drastic  
650 increase in magma viscosity from  $10^2$  (juvenile pumice) to  $10^9$  Pa s (fully crystallized scoria portions)  
651 using the three-phase viscosity model, which integrates Maron and Pierce (1956), Llewellyn and  
652 Manga (2005), Giordano et al. (2008), Mader et al. (2013) and Truby et al. (2015) models. In some  
653 cases, this increase of magma viscosity can drastically change eruptive dynamics and increase the  
654 magma fragmentation efficiency (e.g., Sable et al., 2006, 2009; Thivet et al., 2020a, 2020c). However,  
655 the viscous portion identified in all analyzed layers from both 2005 Karthala paroxysms, was not  
656 dominant enough, especially within the fine tephra fractions, to be considered as the main process  
657 leading to increasing fragmentation. Moreover, the occurrence of differentiated melts within the  
658 2006 and 2007 weak explosive eruptions (Fig. 11), with a composition close to that of the scoria  
659 components, confirm that this syn-eruptive crystallization process cannot be at the origin of the main  
660 fragmentation mechanism observed during the 2005 paroxysmal phases.

## 661 **5.2. The phreatomagmatic contribution on the fragmentation efficiencies: water-magma** 662 **interactions controlling the formation of a wide range of tephra in terms of component and size**

### 663 **5.2.1. Insights from the non-juvenile component**

664 The occurrence of non-juvenile particles within the 2005 paroxysmal deposits (Fig. 6) is  
665 interpreted as a typical consequence of water-magma interactions (e.g., Self et al., 1980; Dvorak,  
666 1992; Dzurisin et al., 1995; Houghton et al., 1996; Doubik and Hill, 1999). The presence of large (up to  
667 meters) blocks of old and dense lavas within proximal fallout areas (Figs. 2e, S1c and S3c), suggests  
668 the occurrence of thermohydraulic explosions (Thiéry and Mercury, 2009; Montanaro et al., 2021)  
669 and associated shock waves (Büttner et al., 2005) able to fragment and project these ballistics (Fig.  
670 12), originating from dense, crystallized and sometimes oxidized lava and micro-gabbro units that are  
671 visible on the crater walls (Fig. 2). The absence of extended hydrothermal alteration on these non-  
672 juvenile particles suggests that they are fragmented within the upper 1 km of the volcano  
673 (corresponding to the thick water-saturated rock unit), as the hydrothermal system is located deeper  
674 and represent a weak activity in the Choungou-Chahalé crater area (Lénat et al., 1998; Savin et al.,  
675 2001, 2005; Bernabeu et al., 2018; Liuzzo et al., 2021).

676 Within medial and distal fallout areas, block occurrence rapidly decreases (Fig. S1c) as they  
677 travelled in ballistic paths. In parallel, non-juvenile particles are mainly observed within relatively  
678 coarse grain size fractions of fallout deposits (Fig. 8). This distribution pattern suggests that

679 phreatomagmatic-induced host rock fragmentation mainly contributes to the formation of coarse  
680 tephra grains but do not contributes to the significant formation of fine ash particles.

### 681 **5.2.2. Insights from the juvenile dense glassy component**

682 Active or interactive particles are tephra grains that exhibit Molten Fuel-Coolant Interactions  
683 (MFCIs), which in the case of phreatomagmatic eruptions, are related to the interaction between  
684 magmatic melts and external fluids, especially water (e.g., Büttner et al., 1999; 2002; Fitch and  
685 Fagents, 2020). As shown by natural and experimental particles (Dürig et al., 2012a), they are  
686 typically characterized by visible branching quench cracks and stepped features on their surface as  
687 well as blocky shapes, reflecting the occurrence of crack bifurcation and shock wave propagations  
688 induced by MFCIs and a dominant brittle fragmentation mechanism (Wohletz et al., 2013).

689 Within the 2005 paroxysmal deposits, these active particles (with stepped features and  
690 cracks) mainly occur in the fine-grained juvenile dense glassy component (Fig. 7), suggesting that  
691 brittle fragmentation induced by phreatomagmatism is the main contributor of the formation of fine  
692 ash particles, which are not considerably formed during purely magmatic and weak explosive  
693 activities (e.g., Parfitt, 1998; Thivet et al., 2020a; 2020c). Note that stepped features are not to be  
694 confused with the ubiquitous hackle lines, which are not a direct diagnostic of phreatomagmatism  
695 but only of brittle fracture of glass. Nevertheless, many examples of interactive grains in the literature  
696 include both stepped features and hackle lines (e.g., Heiken and Wohletz, 1987; Buttner et al. 1999;  
697 Austin-Erickson et al., 2008).

698 The relatively high porosity of the initial magma, as shown by the coarse pumice fragments  
699 (Fig. 4), also facilitated the fragmentation during the water-magma interactions, as relatively low  
700 deformation energy is required to break highly vesicular material (Zhang 1999; Zimanowski et al.,  
701 2015). This efficient brittle fragmentation led to the emission of fine dense ash fragments, made of  
702 glassy vesicle wall fragments or glassy rinds (Mastin, 2007), which are inherited from the  
703 fragmentation of the initial vesicular magma. The genetic link made between the coarse-grained  
704 pumice and fine-grained dense glassy particles is also supported by the VSDs measured on coarse ash  
705 particles, which show that the main porosity is represented by vesicles larger than 50  $\mu\text{m}$  in diameter  
706 (Fig. 10a), thus the main porosity cannot be recorded in particles finer than this size (Fig. 7). This  
707 highlights the role of bubbles (and associated characteristics, especially high  $N_v$ ) in generating fine  
708 ash during hydromagmatic eruptions (Liu et al., 2015). The rapid stress and deformation processes,  
709 as well as volatile degassing from the melt, occurred shortly before and at fragmentation (e.g.,  
710 Dingwell, 1996; Papale, 1999, Dürig et al., 2012b). These fast processes probably caused a rapid  
711 transition from a viscous regime (above the glass transition temperature) to a brittle regime (below

712 the glass transition temperature), whose textural footprints might be identifiable in some ash grains,  
713 in which vesicle walls are highly deformed and/or broken (Figs. 4f and 4g), also suggesting the  
714 occurrence of in situ particle granulation (Colombier et al., 2019a).

715 The fact that no significant variations were observed in ash morphology for the investigated  
716 2005 layers and for a same grain size fraction (also shown in Carlier, 2019), suggests that similar  
717 fragmentation mechanisms occurred during both 2005 paroxysms. The comparison made between a  
718 1977 and a 2005 ash sample has been performed (Fig. 9) in order to focus on ash morphology  
719 variations in function of the nature and origin of the deposits (weak magmatic vs. strong  
720 phreatomagmatic activities respectively) as well as of the grain size fractions. The 1977 deposit,  
721 contain smooth, rounded as well as Pele's hair and tear particles (Fig. 9) that are typically absent in  
722 the 2005 ash deposits but present in other deposits associated with Hawaiian-like activities (e.g.,  
723 Cannata et al., 2019; Thivet et al., 2020a; 2020c). Brittle and efficient fragmentation is better  
724 recorded in the 2005 fine ash fraction, as solidity, convexity and sphericity shape parameters tend to  
725 higher values, typically reflecting blocky shapes (Figs. 9a, 9b and 9c).

726 The occurrence of accretionary lapilli within the 2005 deposits (Fig. S5), gradually composed  
727 of relatively coarse particles in their cores to relatively fine particles in their rims (Fig. 7b), support  
728 the idea that these ash particles were produced by phreatomagmatism and transported within wet  
729 plumes (Houghton et al., 2015), in which accretionary lapilli can develop from a wet nucleus thanks  
730 to surface tension and electrostatic attraction (Colombier et al., 2019b).

### 731 **5.3. Insights from observational data and knowledge of the summit area of Karthala volcano**

732 Lénat et al. (1998) and Savin et al. (2001) highlight that the Choungou-Chahalé crater,  
733 periodically hosting a water lake, is underlain by a maximum of 1 km of water-saturated rocks. Both  
734 April and November-December 2005 eruptions, which began with paroxysmal phases producing high  
735 ash-rich plumes, occurred when water lakes were visible at the bottom of the Choungou-Chahalé  
736 crater. Water-magma interactions can thus have occurred both during dyke propagation through the  
737 water-saturated rocks and at shallow level at the surface water lake. Each paroxysmal phase was  
738 then followed by a lava pond and weak explosive (lava fountaining) activity, from the same vent area  
739 feeding the paroxysmal activity and after the disappearance of the intra-crateric water lake. This  
740 evolution highlights that the lakes were rapidly heated and vaporized in contact with the hot magma  
741 rising to the surface during the initial paroxysmal phases. We can thus infer that the water/magma  
742 ratio evolved from relatively high values at the beginning of the eruption and tended to zero over  
743 time, within a short duration of around 24 hours for each 2005 paroxysm. These contrasting water-  
744 magma interactions (Fig. 12) resulted in different degree of released energy for the magma/rock



745 fragmentation (e.g., Frazzetta et al., 1983; Sheridan and Wohletz, 1983; Wohletz and McQueen,  
746 1984; Wohletz et al., 2013; Houghton et al., 2015; Zimanowski et al., 2015).

747 Also, lava emissions observed during both 2005 eruptions after the paroxysmal phases,  
748 resulted in the elevation of the crater bottom, which caused the absence of visible water at the  
749 surface after the November-December 2005 eruption. The notable absence of water lake before,  
750 during and after the May-June 2006 eruption can thus be correlated with its effusive and weak  
751 explosive behavior (Fig. 2d). Furthermore, these newly emitted lavas represent a new thermo-  
752 lithological boundary within the crater floor, which can significantly drag hydrothermal fluids from  
753 the crater center towards peripheral areas (Bernabeu et al., 2018). Interestingly, the northern  
754 caldera lobe and the northern rift area hosts the strongest hydrothermal activity (fumaroles and soil  
755 degassing) of the volcanic system (Liuzzo et al., 2021) and produce dominantly effusive activities  
756 (e.g., 1965; 1972; 2007), whereas the hydrothermal activity is much weaker in the area of the central  
757 Choungou-Chahalé crater that feeds the most violent explosive activity (e.g., 1918; 1991; 2005). This  
758 geographical pattern suggests that, in the whole 2005-2007 Karthala activity, hydrothermal activity  
759 did not contribute to modulate the degree of explosivity. On the other hand, we can infer that a link  
760 exists between the occurrence of shallow water (i.e. the water lake and associated water-saturated  
761 rocks) and the degree of explosivity.

762

## 763 **6. Conclusions**

764 Field observations and laboratory analyses performed on the 2005 eruptive deposits of  
765 Karthala volcano permit to identify and quantify distinct eruptive processes.

766 1 – Initial syn-eruptive magmatic processes, especially degassing (dominance of highly porous  
767 magma portions) and in a less important proportion crystallization (occurrence of microlite-rich  
768 portions) within the volcanic conduits were relatively significant during the two 2005 paroxysmal  
769 phases compared to the weak explosive basaltic eruptions forming lava fountains (i.e., 1977's  
770 Karthala eruption). However, these processes alone could not lead to the observed characteristics of  
771 the deposits (i.e., occurrence of non-juvenile block to coarse ash particles, as well as abundant  
772 accretionary lapilli and fine ash particles).

773 2 – MFCI interactions between highly porous magmatic melts and external water occurred  
774 during the initial paroxysmal phases, leading to brittle and efficient fragmentation mechanism and  
775 forming a large amount of fine ash from the initial porous magma. Thermohydraulic explosions

776 caused by the water-magma interaction resulted in the host rock fragmentation, forming relatively  
777 coarse (from blocks to coarse ash) non-juvenile particles.

778         3 – Fluctuating host rock and magma fragmentation efficiencies can be controlled by the  
779 variability of water/magmatic melt ratios through the eruptions. In natural systems, access of the  
780 magma to water includes many parameters difficult to assess (e.g. variations of magma ascent rate,  
781 surface area to volume ratios, vent shape, host rock permeabilities, amount and variations of syn-  
782 eruptive debris within the eruptive vent, Houghton et al., 2015). This makes phreatomagmatic  
783 interactions more complex than theoretical and experimental models. However, in the present study  
784 we argued and conclude that the observed deposit variabilities in term of grain size, grain  
785 componentry and layer thickness might be linked by variable phreatomagmatic interactions, which  
786 released variable energy, resulting in variable fragmentation efficiencies. (i) Fine and unimodal (grain  
787 size modes < 90  $\mu\text{m}$ ) tephra layers (KAR\_10\_b, d, f, h, l, m, n, p, r, t and u) result from a relatively  
788 homogenous and efficient phreatomagmatic fragmentation (we can speculate relatively high  
789 water/magma ratios, Fig. 12a). This fragmentation regime can be short-lived or relatively steady as  
790 suggested by highly variable tephra layer thicknesses (from < 5 to 20 cm). (ii) Intermediate-sized and  
791 unimodal (grain size modes between 250 and 710  $\mu\text{m}$ ) tephra layers (KAR\_10\_a, j, k and q) result  
792 from homogeneous but relatively weak phreatomagmatic fragmentation mechanism (we can  
793 speculate intermediate water/magma ratios, Fig. 12a). (iii) Bimodal (the coarser modes being  
794 between 250 and 1400  $\mu\text{m}$  and the finer modes being between 31 and 90  $\mu\text{m}$ ) tephra layers  
795 (KAR\_10\_c, e, g, i, o and s), originated from several combined mechanisms. The finest modes of these  
796 layers are mainly composed of juvenile material which undergone intense and efficient  
797 fragmentation. In contrast the coarser modes are composed of both inefficiently fragmented juvenile  
798 material (residual juvenile part of the phreatomagmatic fragmentation mechanism) and  
799 synchronously emitted non-juvenile coarse grains (from thermohydraulic explosions). (iv) The  
800 coarsest and unimodal (grain size mode between 5,66 and 8 mm) tephra layer (KAR\_2005b), forming  
801 the uppermost part of the 2005 tephra sequence, mainly result from magmatic processes (Fig. 12b),  
802 forming a short-lived (tephra layer < 5 cm in thickness) but sustained lava fountaining episode at the  
803 end of the November 2005 paroxysm (which might be associated with remnant steam explosions  
804 forming non-juvenile blocks and coarse ash grains).

805         4 – Available external water volumes rapidly decreased during both 2005 paroxysms, which  
806 lasted only one day each, as the water vaporized and wet rocks dried in contact with relatively hot  
807 magmas and/or lavas and/or tephra. As the shallow system dried up, phreatomagmatic regimes  
808 progressively stopped for both 2005 eruptions, leading to lava pond and weak fountaining activities  
809 until the end of both 2005 eruptions.

810 In conclusion, and beyond field observations that were made at the time of the eruptions,  
811 the present study highlights the importance of in-depth analysis for such deposits in order to gain  
812 insights on shallow magmatic systems and associated processes potentially leading to relatively  
813 important and uncommon volcanic hazards.

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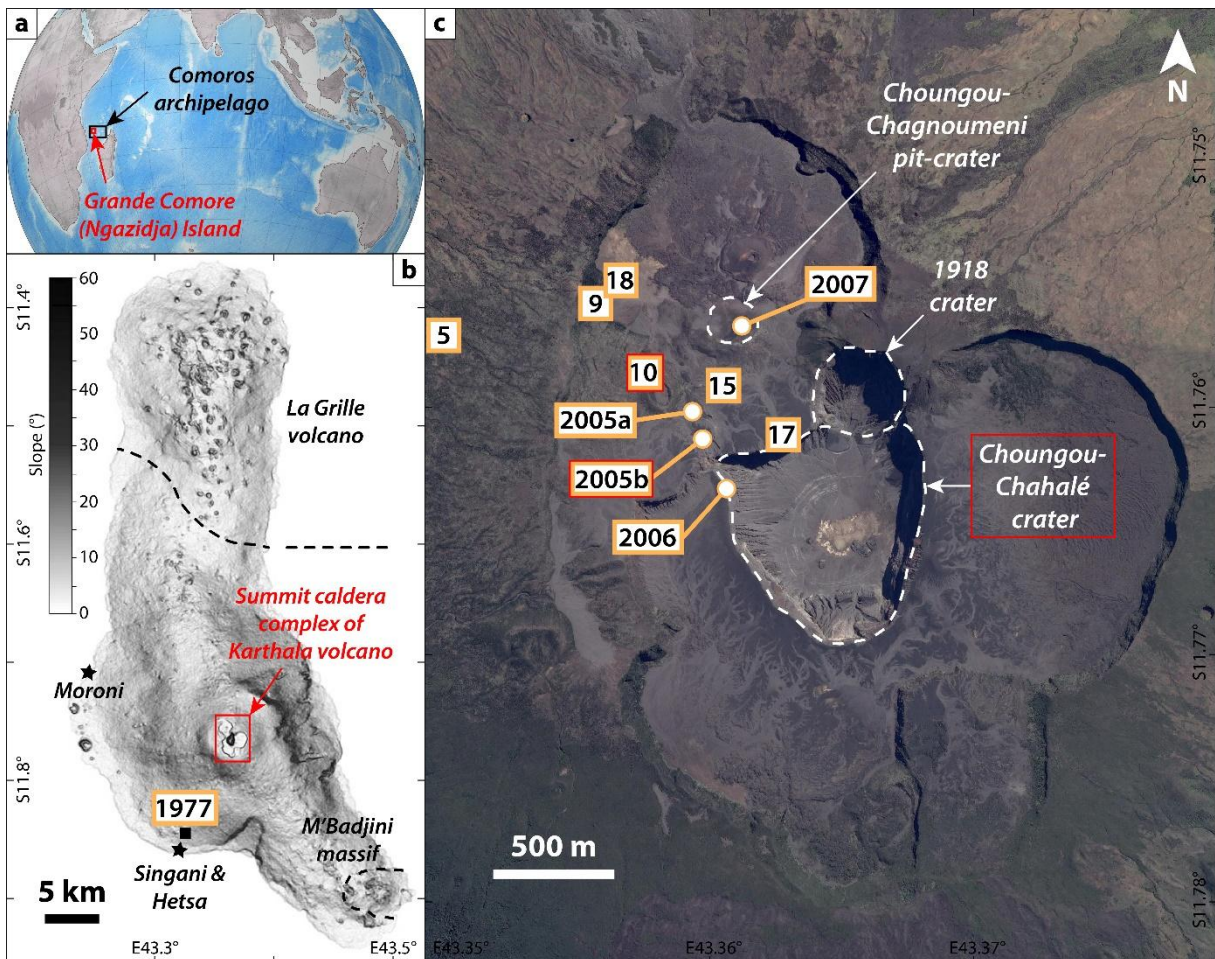
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822 **Figures and tables**



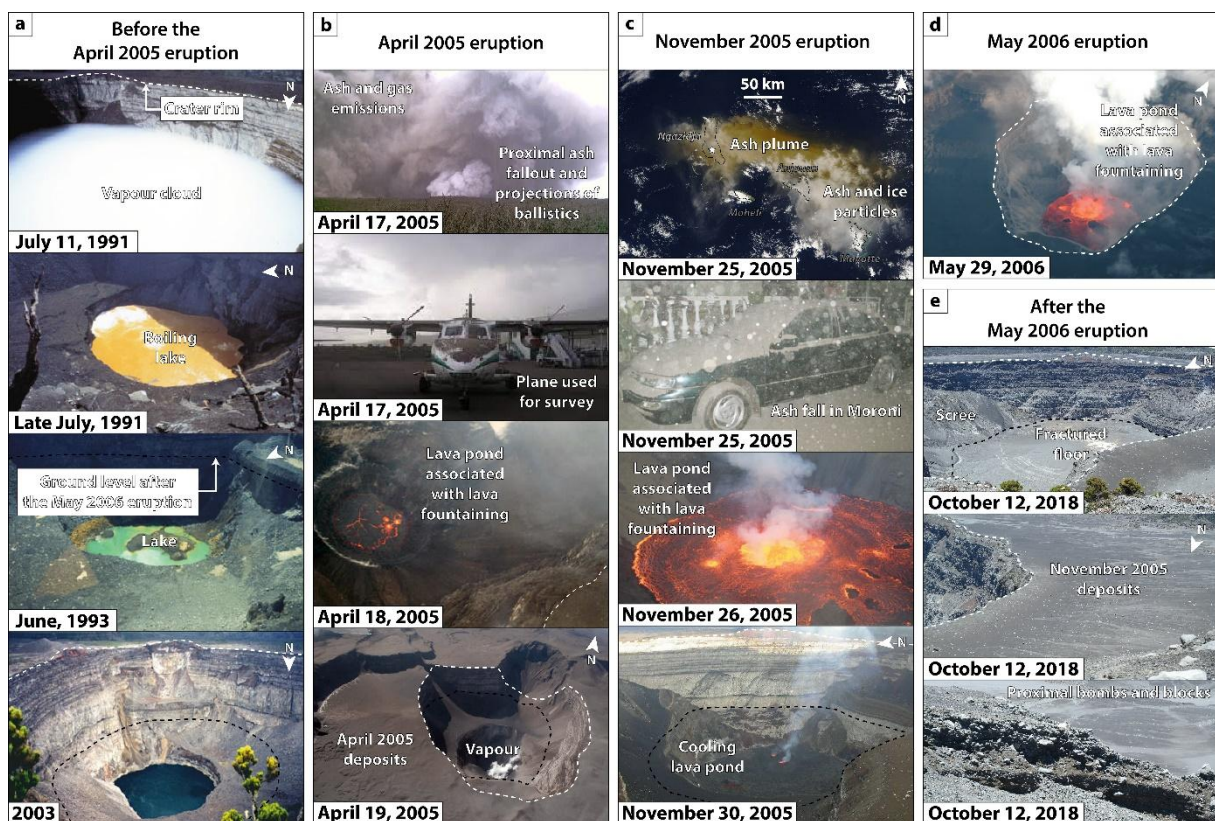
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824 **Figure 1** – (a) Location of the Comoros archipelago and the Grande Comore (Ngazidja) Island within  
 825 the Mozambique Channel. (b) SRTM-derived slope map of the Grande Comore (Ngazidja) Island  
 826 (modified from Bachèlery et al., 2016). The black square shows the location of the 1977 eccentric  
 827 eruption and its associated sampling location (labelled 1977). Black stars show the capital city of  
 828 Moroni and the adjacent villages of Singani and Hetsa. (c) Google Earth satellite image (CNES/Airbus,  
 829 July 31, 2017) of the summit caldera complex of Karthala volcano. April 2005, November-December  
 830 2005 and May-June 2006 eruptive vents opened in the floor of the Choungou-Chahalé crater. The  
 831 January 2007 eruption occurred in the Choungou-Chagnoumeni pit-crater. Numbered squares show  
 832 the studied or sampling sites of both 2005 paroxysmal deposits (labelled 5, 9, 10, 15, 17, 18, 2005a  
 833 and 2005b), as well as May-June 2006 (labelled 2006) and January 2007 (labelled 2007) eruptive  
 834 products. Detailed location and description of each sample are shown in Table S1.

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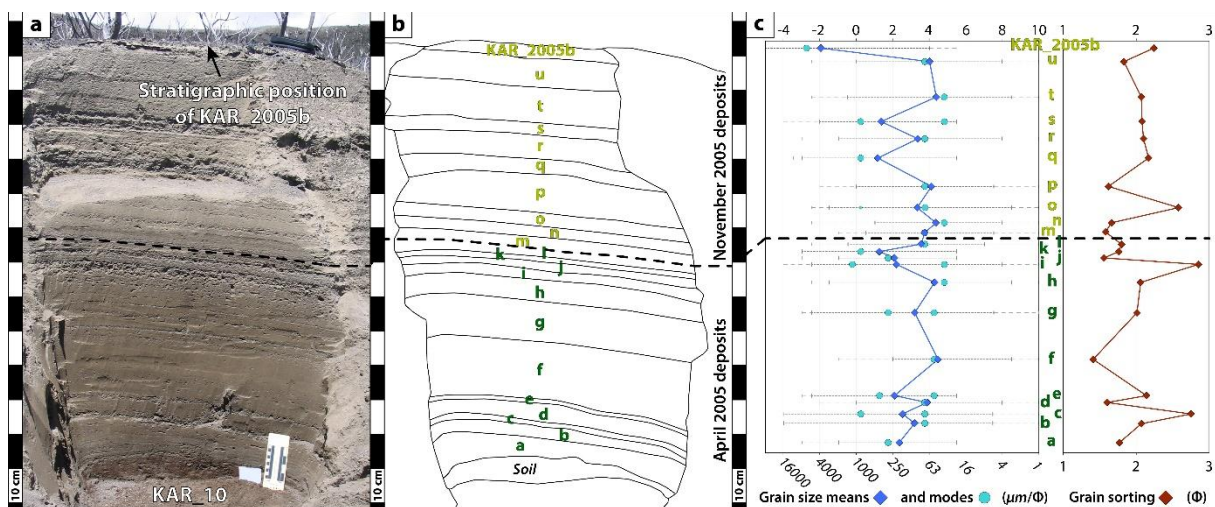
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839 **Figure 2** – Ground-, airplane- and satellite-based photographs showing the morphological evolution  
 840 and eruptive activities that occurred within the Choungou-Chahalé crater, between 1991 and 2018.  
 841 Dashed white lines show the Choungou-Chahalé crater rim and dashed black lines represent the

842 ground level of the Choungou-Chahalé crater floor after the May 2006 eruption. (a) Morphology of  
 843 the Choungou-Chahalé crater and evolution of the water lake before the April 2005 eruption.  
 844 Pictures taken by Patrick Bachèlery (modified from Bachèlery et al., 2016). (b) Chronology and  
 845 impacts of the April 2005 eruption. From top to bottom, pictures taken by (i) Daniel Hoffschir, (ii) and  
 846 (iii) Hamidi Soulé (modified from Bachèlery et al., 2016) and (iv) Nicolas Villeneuve. (c) Chronology  
 847 and impacts of the November 2005 eruption. From top to bottom, (i) satellite image from NASA  
 848 (Terra MODIS) white star representing the eruptive vent, (ii) pictures taken by Hamidi Soulé, (iii) Julie  
 849 Morin (modified from Bachèlery et al., 2016) and (iv) François Sauvestre. (d) May-June 2006  
 850 eruption. Picture taken by Julie Morin (modified from Bachèlery et al., 2016). (e) Morphology of the  
 851 Choungou-Chahalé crater after the May-June 2006 eruption. Pictures taken by Simon Thivet.

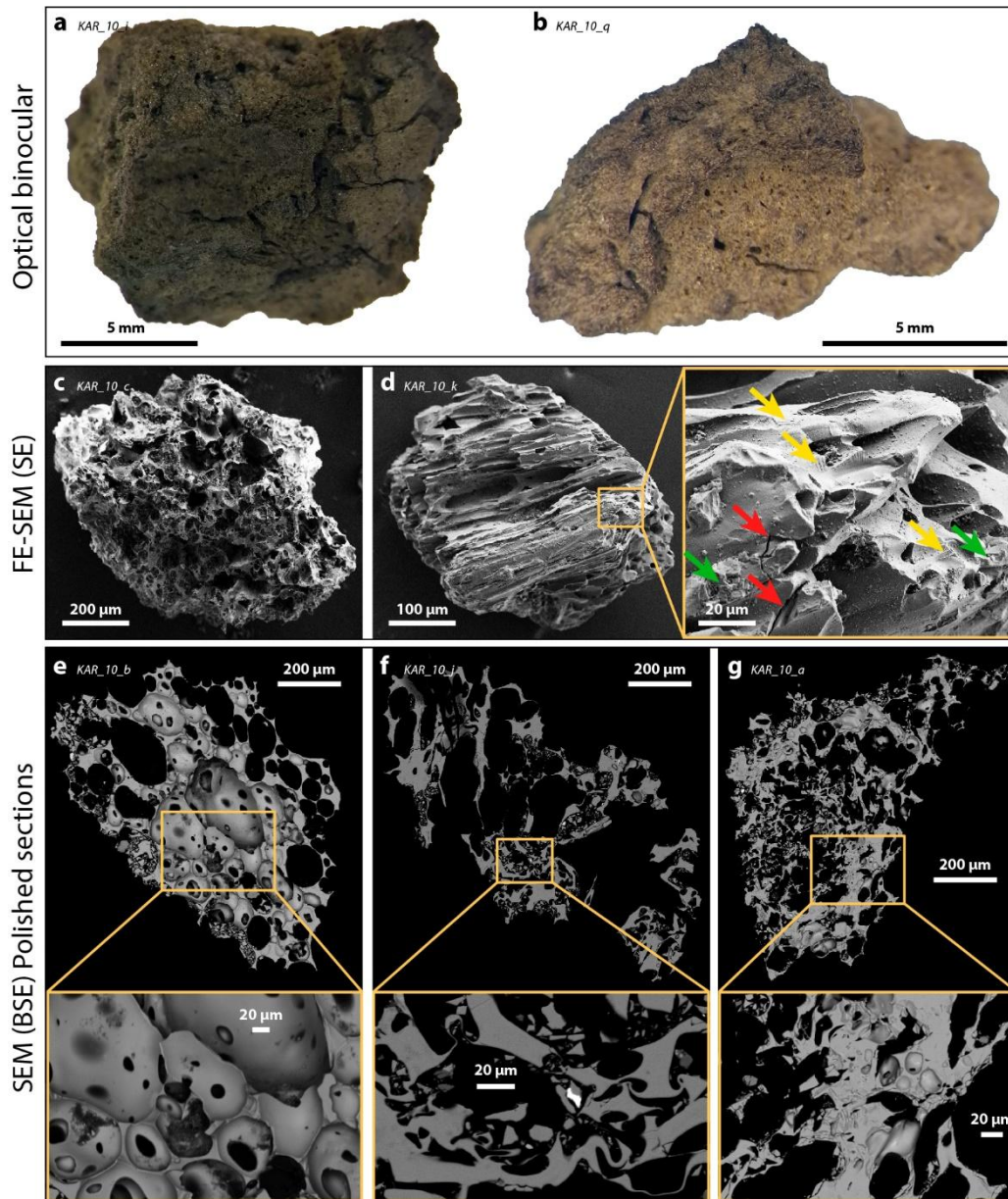
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857 **Figure 3** – (a) Picture of the KAR\_10 stratigraphic log (cf. Fig. 1 for location), representative of the ash  
 858 fallout deposits of both April and November 2005 paroxysmal phases. The thick dashed black line  
 859 delimits the deposits from the two paroxysms. (b) Schematic of the same stratigraphic log with the  
 860 different identified and sampled layers. Note that the very last products emitted by the November  
 861 2005 paroxysm are represented at the top of the log (KAR\_2005b sample, cf. Fig. 1 for location).  
 862 Layer names (from KAR\_10\_a to u, as well as KAR\_2005b) are labelled in dark (April 2005) and light  
 863 (November 2005) green. (c) Grain size mean, mode and sorting values of the sampled layers. Thin  
 864 dashed lines represent the total grain size range of each sample and thick dashed lines only include

865 grain size fractions with > 1 wt% of the total mass of each sample. Detailed grain size analyses are  
866 shown in Table S2.



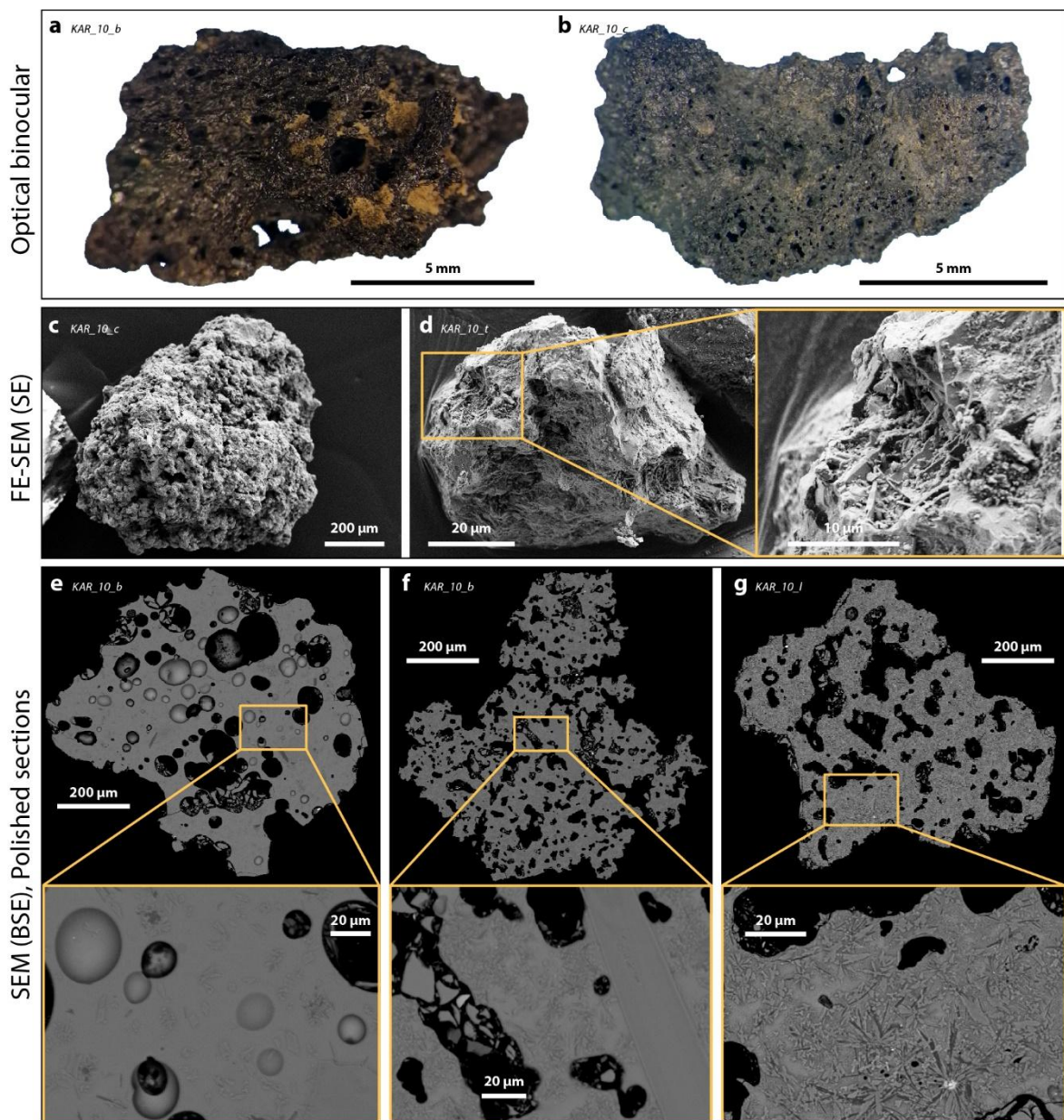
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868 **Figure 4** – Juvenile pumice clasts from both April and November 2005 paroxysmal fallout deposits. (a)  
869 and (b) Optical images of light brown lapilli fragments showing non-fluidal shapes and bread-crust  
870 cracks on their surfaces. (c) and (d) 3D images of coarse ash (710-1000 μm) particles. Vesicles are  
871 either (c) rounded or (d) elongated. (d) Micron-scaled hackle lines (yellow arrows), stepped features  
872 (green arrows) and branching quench cracks (red arrows) are visible on the particle broken surfaces.  
873 (e), (f) and (g) Cross-section images of coarse ash (710-1000 μm) particles showing different  
874 deformation features from (e) rounded to (f) highly deformed and (g) broken vesicle walls.

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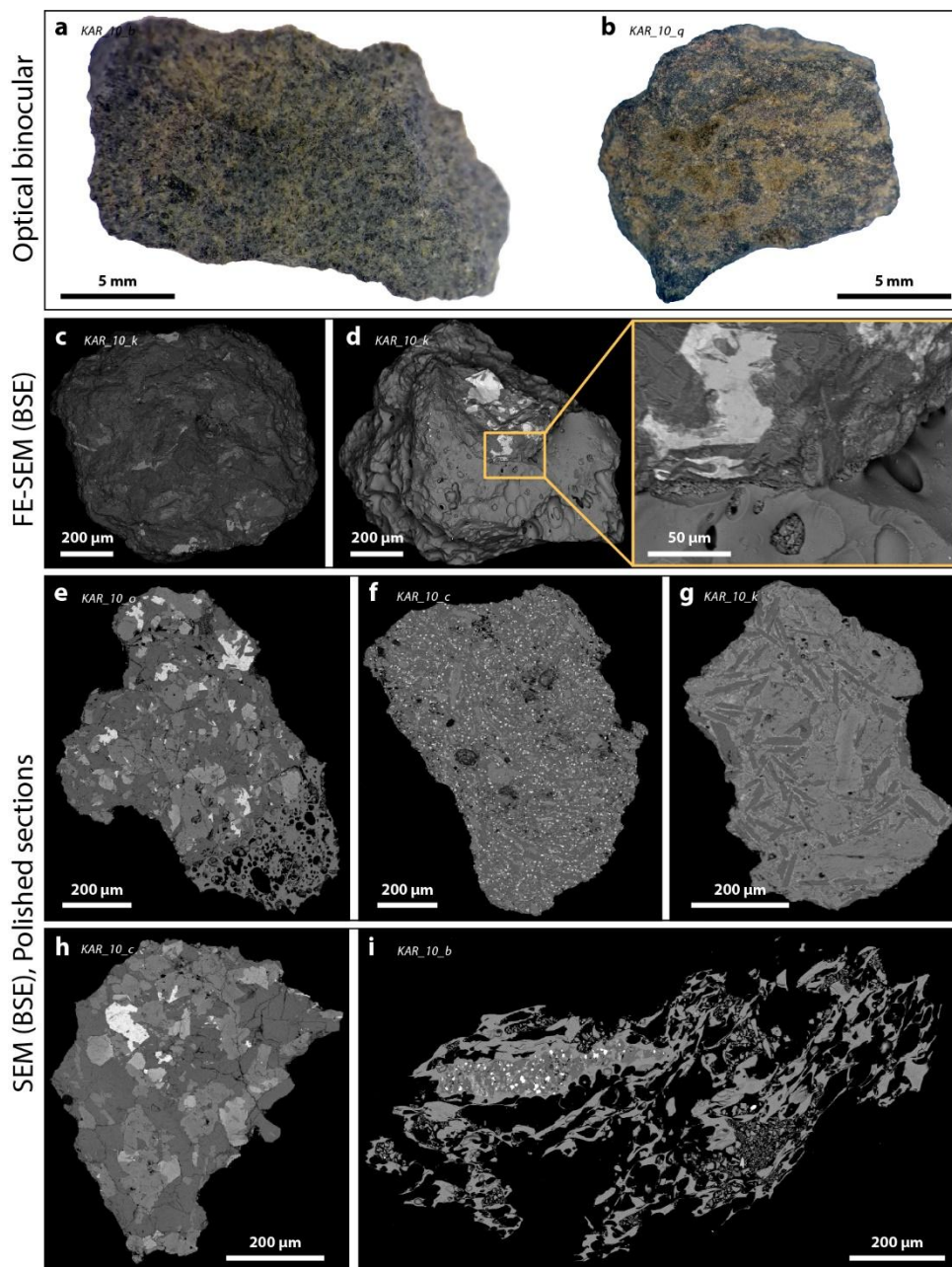
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879 **Figure 5** – Juvenile scoria clasts from both April and November 2005 paroxysmal fallout deposits. (a)  
880 and (b) Optical images of black lapilli fragments showing non-fluidal shapes. (c) and (d) 3D images of  
881 coarse ash (710-1000 μm) particles, characterized by rough or blocky surfaces. (d) Microlites are  
882 sometimes visible on their surface reflecting matrices with a high microlite contents. (e), (f) and (g)  
883 Cross-section images of coarse ash (710-1000 μm) particles showing a high range of crystallinity and  
884 porosity.

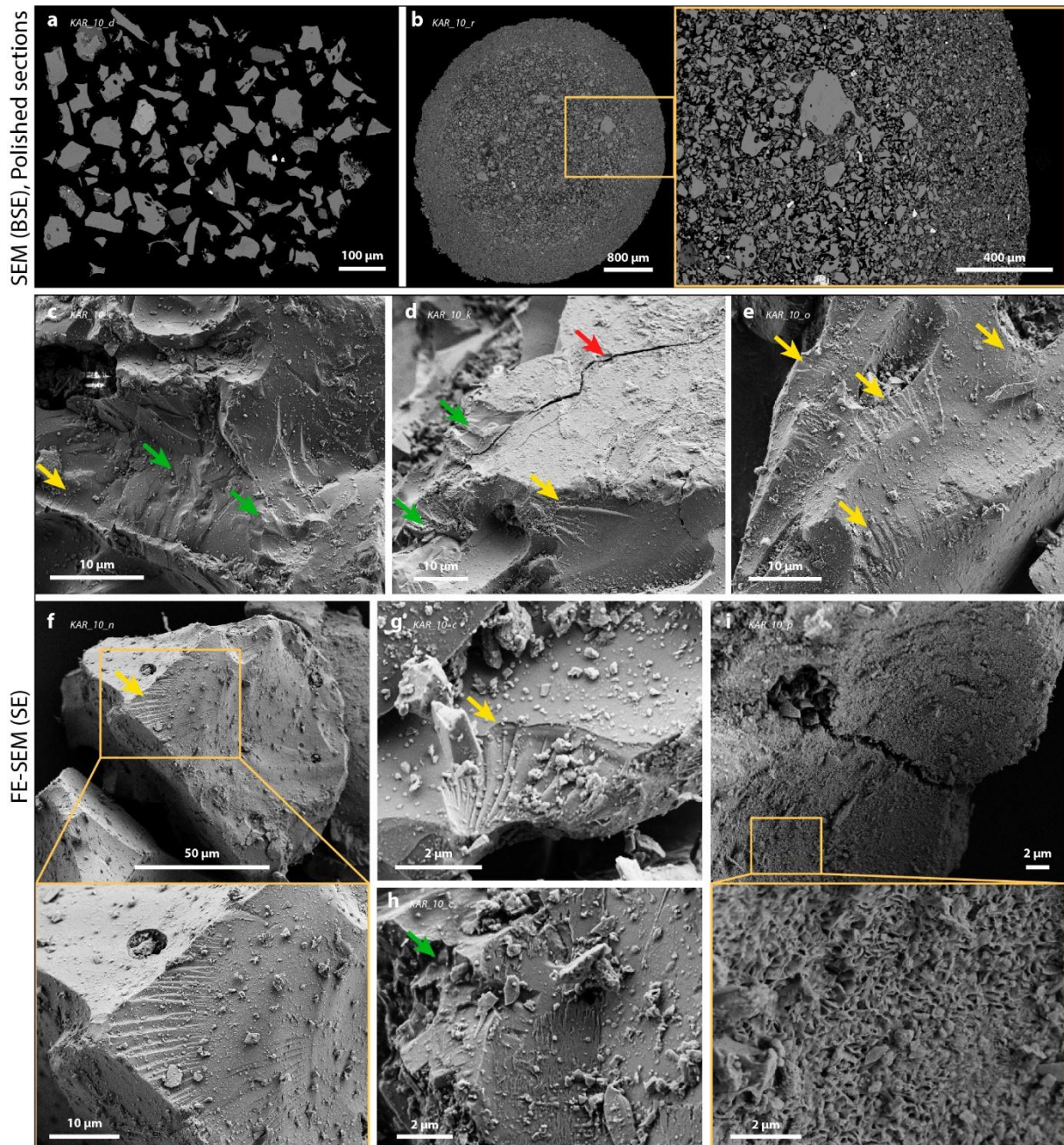
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889 **Figure 6** – Non-juvenile clasts from both April and November 2005 paroxysmal fallout deposits. (a)  
 890 and (b) Optical images of dense lapilli fragments, with visible phenocrysts of plagioclase, pyroxene,  
 891 olivine and oxides. (c) and (d) 3D images of coarse (710-1000  $\mu\text{m}$ ) and dense micro-gabbroic ash  
 892 particles, characterized by blocky surfaces. (d) and (e) These non-juvenile fragments are sometimes  
 893 coated by the juvenile magma. (e), (f), (g) and (h) Cross-section images of coarse (710-1000  $\mu\text{m}$ ) ash  
 894 particles showing different textures in term of crystal size distribution, from tachylite-like to micro-  
 895 gabbro textures. Note the total absence of interstitial melts in these lithic fragments. (i) Cross-section

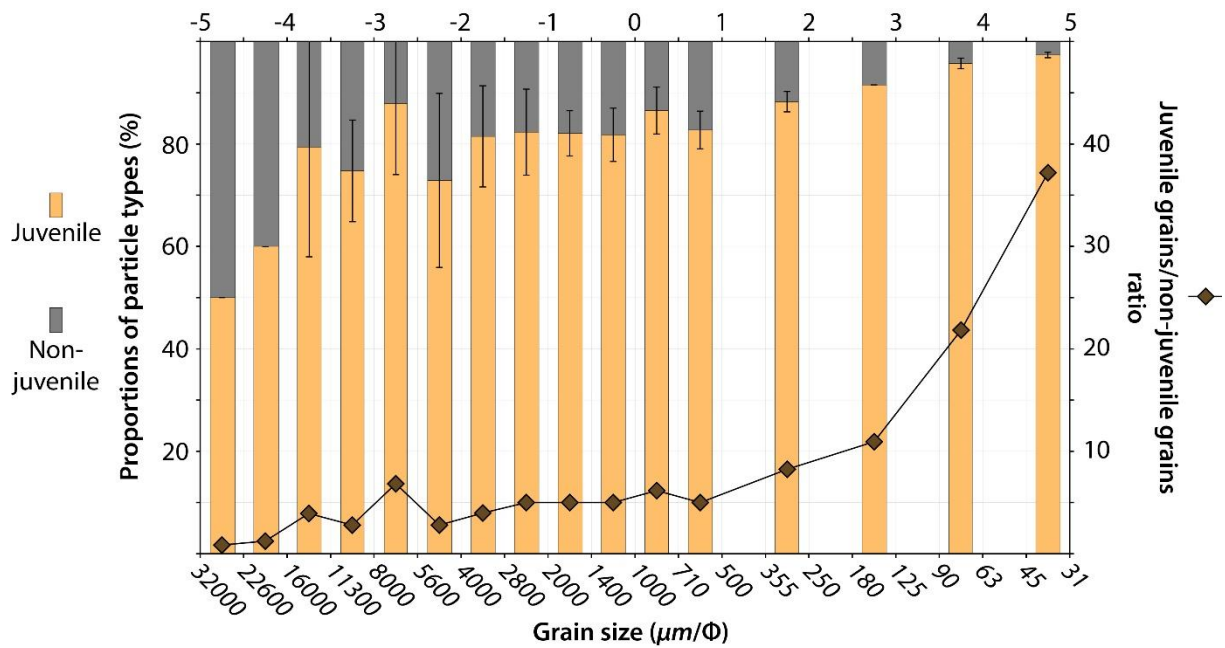


896 image of a micro-gabbroic fragment included within a juvenile pumice ash particle. In this case, this  
 897 micro-gabbroic fragment is considered as juvenile as it is part of the erupted magma, and it also  
 898 contains interstitial melt.



899  
 900 **Figure 7** – Fine ash particles from both April and November 2005 paroxysmal fallout deposits. (a)  
 901 Cross-section image of a sample showing the internal textures of the fine ash (< 63μm) particles. (b)  
 902 Cross-section image of an accretionary lapilli. (c), (d), (e), (f), (g), (h) and (i) 3D images of fine ash (<  
 903 63 μm) particles. The fine-grained dense glassy particles show micron-scaled hackle lines (yellow  
 904 arrows), stepped features (green arrows) and branching quench cracks (red arrows) on their  
 905 surfaces. (h) and (i) Secondary deposition phases are also observed on some of the particle surfaces.

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909 **Figure 8** – Particle componentry analysis (juvenile vs. non-juvenile) of both April and November 2005  
910 paroxysmal fallout deposits, in function of particle grain size. Each bar represents the average value  
911 (number %) for all the analyzed samples. Error bars represent the standard deviation to this averaged  
912 value, hence are representative of some local variations for some tephra layers. Detailed  
913 componentry analyses are shown in Table S3.

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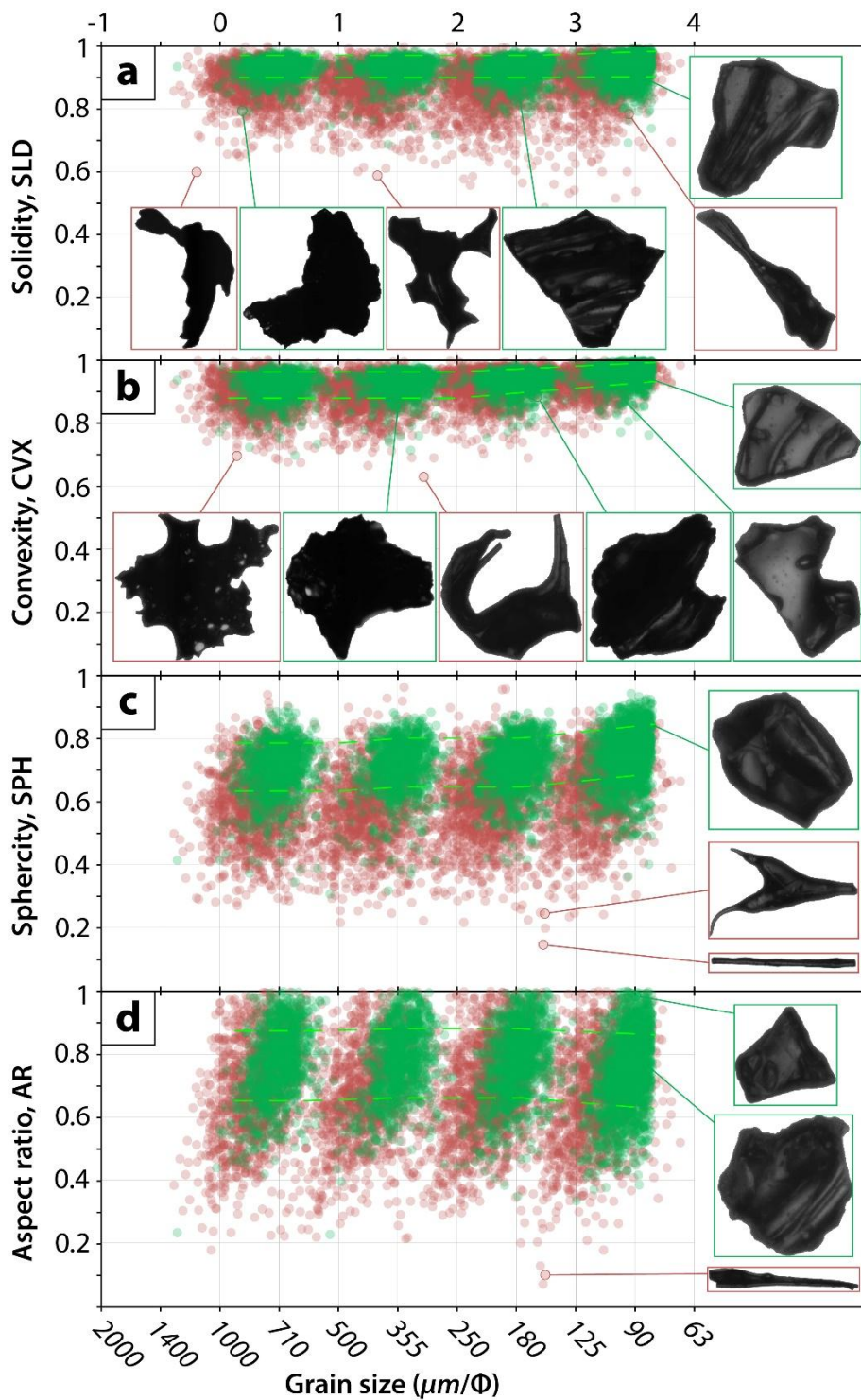
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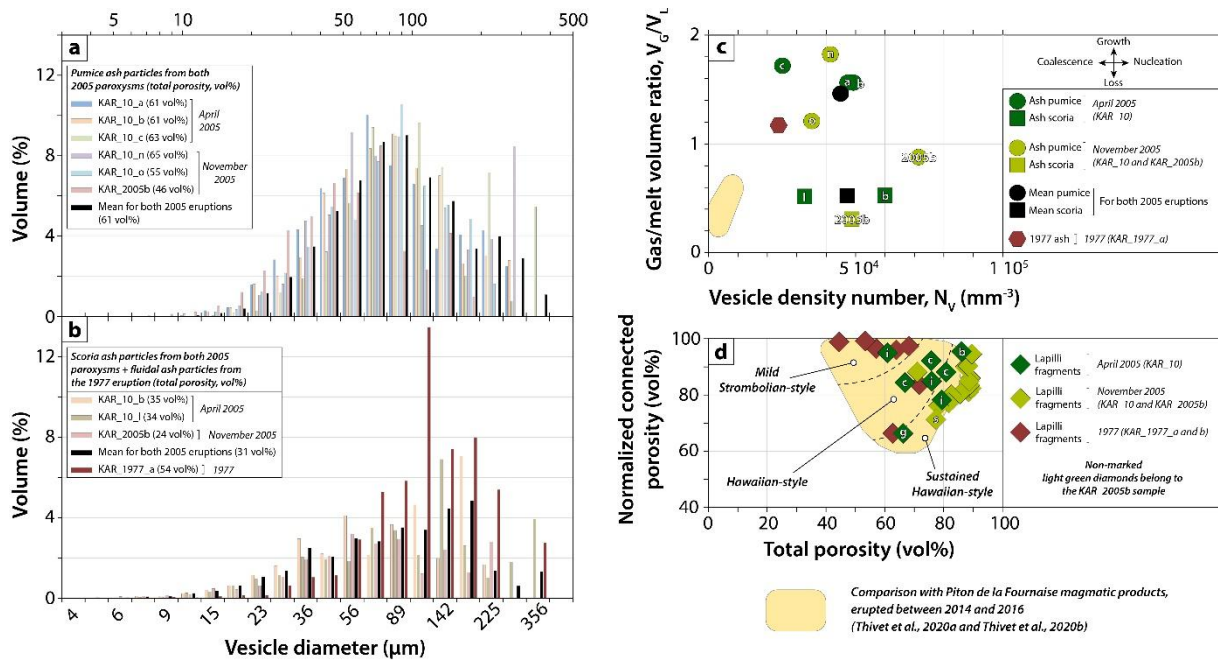


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927 **Figure 9** – Ash particle morphological analysis. (a) Solidity, (b) convexity, (c) sphericity and (d) aspect  
928 ratio values in function of particle grain size. Green dots represent the KAR\_10\_a sample from the  
929 April 2005 paroxysm, whereas the red dots represent the KAR\_1977\_a sample from the 1977

930 magmatic eruption that produced lava fountains. Green dashed lines represent the 2005 ash  
 931 population within the standard deviation. Morphological raw data is presented in Table S3.

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933

934 **Figure 10** – (a), (b) and (c) Micro-texture and (d) bulk-texture analyses of both April and November  
 935 fallout deposits as well as 1977 products. Vesicle size distributions of (a) pumice and (b) scoria (and  
 936 1977 lava fountains) ash particles. (c) Gas to melt ratios in function of vesicle density numbers for the  
 937 same samples presented in (a) and (b). (d) Normalized connected porosity in function of total  
 938 porosity for a selection of lapilli fragments from the 1977 eruption, as well as both April 2005 and  
 939 November 2005 paroxysms. Detailed textural analyses are reported in Table S4.

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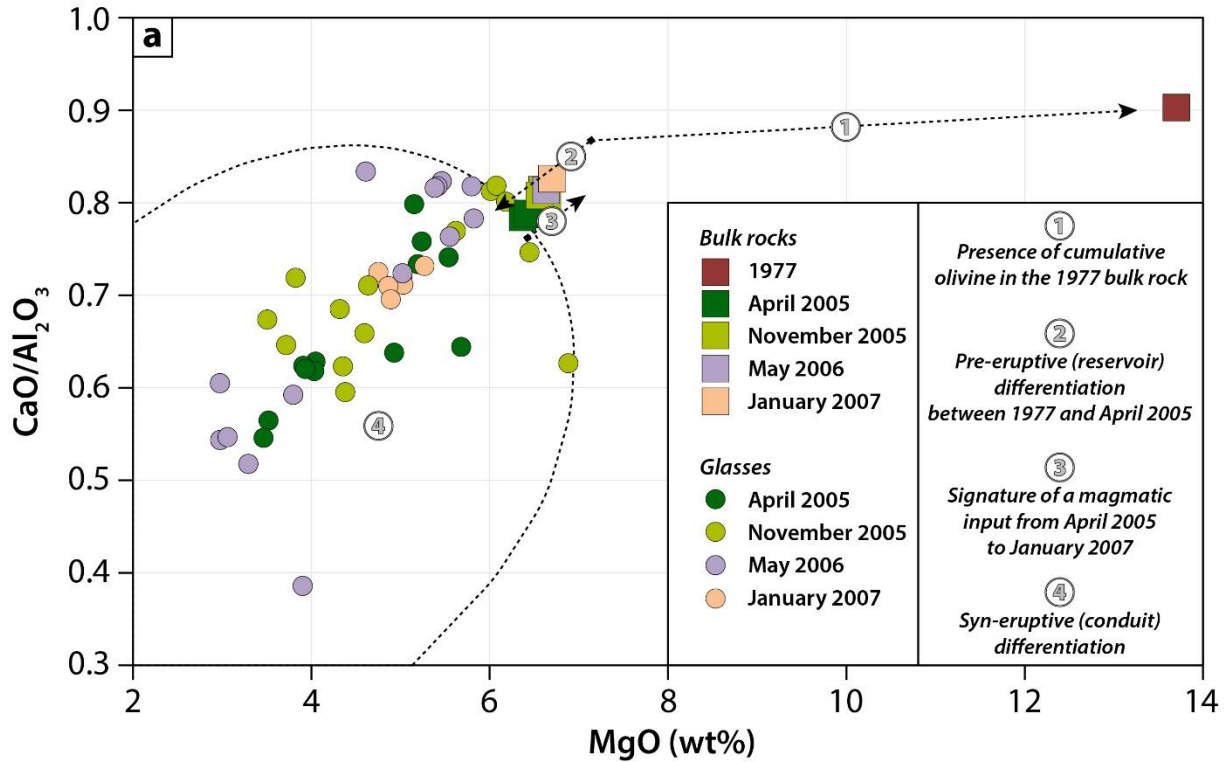
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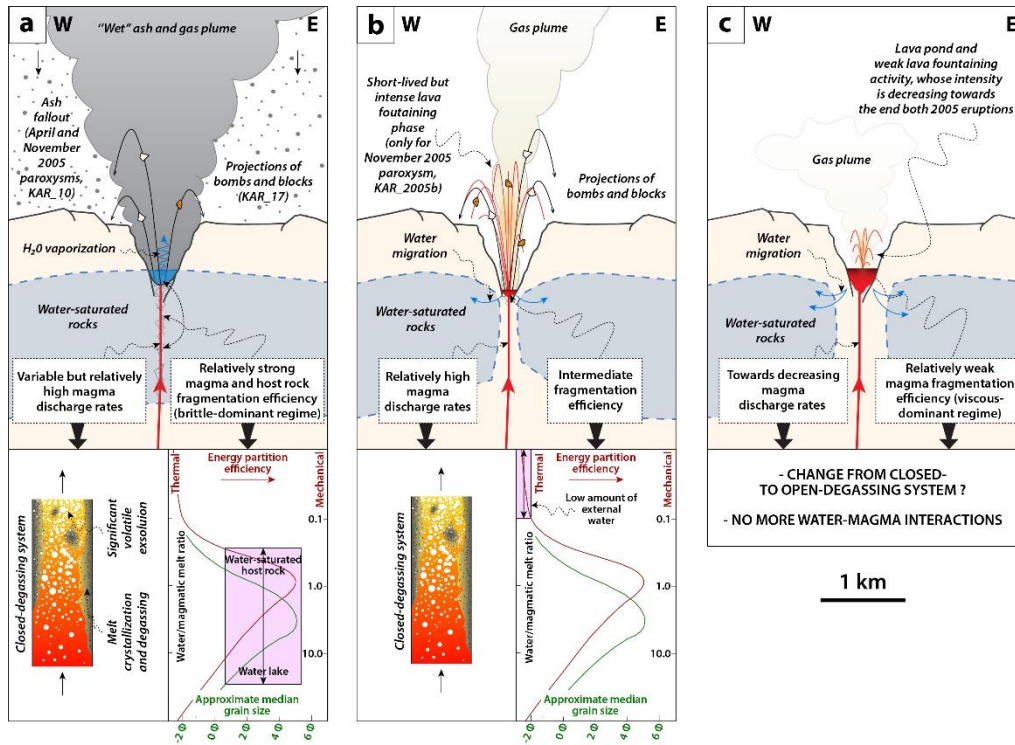
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**Figure 11** – (a) Bulk rock and glass compositions (CaO to Al<sub>2</sub>O<sub>3</sub> ratio in function of MgO) of the magmas emitted between 1977 and 2007 at Karthala volcano, showing a complex magmatic evolution represented by the numbered steps. Error bars are included in the symbols.. Detailed bulk rock, glass and mineral analyses are reported in Table S5.

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968 **Figure 12** – Schematic sections of the shallow magmatic system of Karthala volcano, (a) during both  
 969 2005's paroxysms, (b) during the last phase of the November 2005 paroxysm and (c) during both  
 970 2005's lava pond activities. Shallow magmatic processes are represented on the bottom left hand  
 971 side diagrams and water-magma interactions are illustrated in the bottom right hand side diagrams  
 972 adapted from Wohletz et al. (2013).

973

974 **Acknowledgments**

975 We thank J-L. Devidal and E. Voyer for their invaluable help with the EPMA and SEM at the  
 976 LMV and we thank A. Dalle for its help with the FE-SEM performed at 2MAtech. We are grateful to all  
 977 members of the Observatoire Volcanologique du Karthala (OVK) for their support during the 2018  
 978 field work. We also thank the two reviewers for their highly constructive reviews. We acknowledge  
 979 the support of the Interreg Hatari project. This research was funded by ClerVolc, the French  
 980 Government Laboratory of Excellence initiative n°XXXX, the French government IDEX-ISITE initiative  
 981 16-IDEX-XXXX (CAP20-25), the Action Incitative of the Observatoire de Physique du Globe de  
 982 Clermont-Ferrand (OPGC) and the Alexander Von Humboldt foundation.

983

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