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# **Characterization of an exhumed high-temperature hydrothermal system and its application for deep geothermal exploration: An example from Terre-de-Haut Island (Guadeloupe archipelago, Lesser Antilles volcanic arc)**

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1 **Characterization of an exhumed high-temperature**  
2 **hydrothermal system and its application for deep**  
3 **geothermal exploration: an example from Terre-de-Haut**  
4 **Island (Guadeloupe archipelago, Lesser Antilles volcanic**  
5 **arc)**

6  
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23

24

25

26 **Abstract**

27 We present integrated structural, petrological, mineralogical and geochemical investigations  
28 conducted on an exhumed hydrothermal system identified in Terre-de-Haut Island  
29 (Guadeloupe archipelago). This work demonstrates, for the first time, the occurrence of  
30 ductile shear zones, marked by the development of spaced schistosity planes generated by  
31 pressure-solution processes. We unravel first that the development of spaced schistosity is  
32 coeval with high-temperature hydrothermalism (approximately 350°C) and second that this  
33 hydrothermal alteration occurred at a depth of less than 2 kilometres. We highlight, at  
34 shallow crustal depths, the record of both vertical and lateral fluid pathways in the studied  
35 volcanic arc. Finally, combining this new dataset with the large number of published surface  
36 data obtained in the active geothermal system of Bouillante, we propose a conceptual  
37 scheme for the studied fossil reservoir to document the roots of magmatic arc-related active  
38 geothermal systems and to introduce new perspectives for deep geothermal exploration.

39 *Keywords: exhumed hydrothermal system; hydrothermal fluids pathways; high temperature*  
40 *geothermal reservoir; geothermal exploration; Lesser Antilles arc.*

41

## 42 **1- Introduction**

43 For many geothermal systems, a significant amount of geological and geophysical surface  
44 data is currently available to characterize geothermal resources (Ledru and Guillou-Frottier,  
45 2010; Moeck, 2014). However, deciphering the geological properties of the lowermost part of  
46 geothermal reservoirs, i.e., below commonly drilled depths (1.5 kilometres in the case of the  
47 Lesser Antilles arc), is, still challenging due to the high economic cost of exploratory drillings  
48 and poor knowledge of the mechanical properties of hydrothermalized materials at high P-T  
49 conditions (i.e.  $T > 300^{\circ}\text{C}$  and  $P > 2 \text{ Kbar}$ ). As frequently implemented in the study of oil  
50 resources, an effective alternative to address this type of issue is the study of exhumed  
51 analogues (e.g., Bouma, 1962; Middleton and Bouma, 1973; Beck and Lehner, 1974; Joseph  
52 and Lomas, 2004). This approach was successfully applied to epithermal systems coeval  
53 with magmatic activities, which are regarded as fossil geothermal systems (Bouchot and  
54 Genter, 2009; Liotta et al., 2010; Cooke D. R. and Simmons S. F. 2000; Garden et al., 2020).

55 On the west coast of Guadeloupe (Lesser Antilles magmatic arc), a high-energy geothermal  
56 system favourable for electricity production was identified in the 1970s in Bouillante Bay  
57 (Cormy et al. 1970; Demians d'Archimbaud and Munier-Jolain 1976; Demians d'Archimbaud  
58 and Surcin 1976). This geothermal field has been well characterized through geological,  
59 geochemical and geophysical investigations and several exploration and production wells  
60 (Iundt and Ouzounian, 1984; Abou Akar et al., 1992; Sanjuan and Brach, 1997; Guillou-  
61 Frottier, 2003; Lachassagne et al., 2009; Bouchot et al., 2010; Thinon et al., 2010; Calcagno  
62 et al., 2012; Gailler et al., 2013). However, despite these efforts, the pathways for fluid and  
63 heat transfer in the deepest part of this reservoir are still under debate (Navelot et al., 2018).  
64 A hydrothermal system considered a possible exhumed analogue of such a geothermal

65 reservoir has been identified in the Guadeloupe archipelago, precisely in Les Saintes Islands  
66 (Jacques and Maury, 1988; Verati et al., 2016). According to these authors, superposed fault  
67 networks structurally control the development of hydrothermal alteration zones, and in a  
68 previous paper, we investigated the role of faults, fractures and lithological discontinuities on  
69 fluid transfers, hydrothermal alterations and petrophysical properties of volcanic rocks in this  
70 area (Navelot et al., 2018). However, in the western arc of the Guadeloupe volcanic  
71 archipelago, ductile tectonic structures were recently discovered (Verati et al., 2018; Favier  
72 et al., 2019). As demonstrated by numerous authors, the tectonic and kinematic framework,  
73 and therefore the regional stress field, has remained unchanged since 5 Ma in this volcanic  
74 archipelago (Bouysse and Garrabé, 1984; Julien and Bonneton, 1989; Feuillet et al., 2002,  
75 2004; Mathieu et al., 2011; Corsini et al., 2011; Lardeaux et al., 2013; Münch et al., 2013;  
76 Leclerc et al., 2016). Consequently, the goal of this paper is to investigate the relationships  
77 between the mineralogical evolution of hydrothermalized rocks and ductile tectonic  
78 structures, in Les Saintes Islands, in order to document the roots of active geothermal  
79 systems developed in magmatic arcs and to develop new perspectives for deep geothermal  
80 exploration.

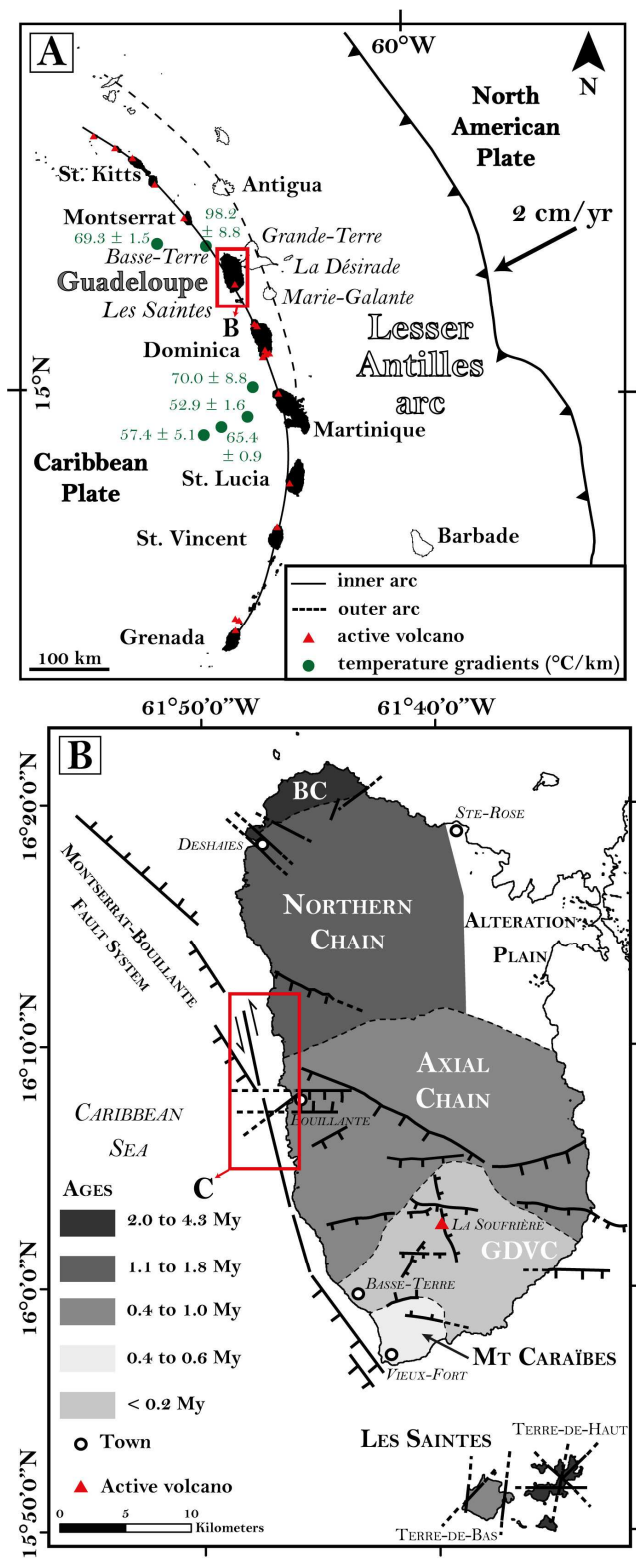
81

## 82 **2- Regional setting**

### 83 **2-1- The Guadeloupe archipelago in the Lesser Antilles arc**

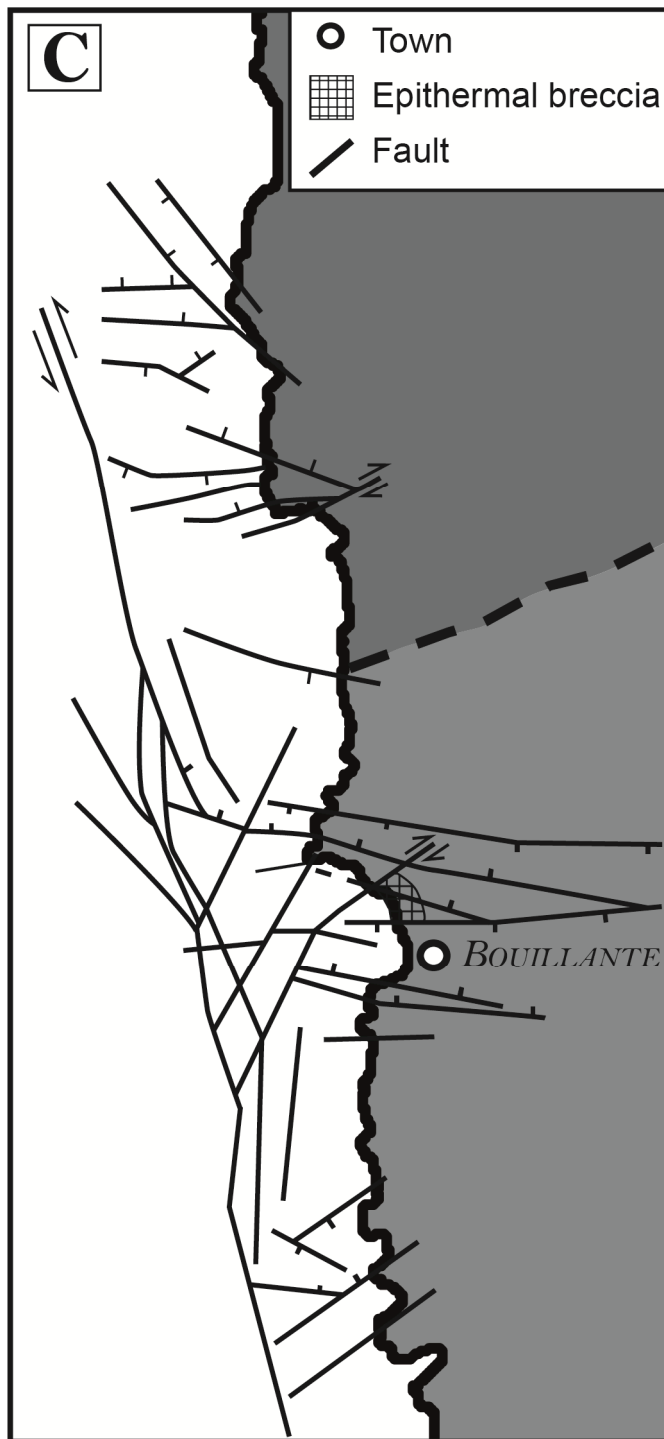
84 The 850 km-long curved Lesser Antilles arc is located at the convergent boundary between  
85 the Caribbean and American plates (Figure 1-A; Hawkesworth and Powell, 1980; Bouysse  
86 and Westercamp, 1988; Demets et al., 2000). This is the consequence of southwest-directed  
87 subduction of the American plates beneath the Caribbean plate with a velocity of  
88 approximately 2 cm/yr (Lopez et al., 2006). The crustal thickness of the upper plate  
89 supporting the volcanic arc was defined by geophysical investigations at approximately 28  
90 km (Kopp et al., 2011; Gailler et al., 2013). The conductive geothermal gradient of the Lesser

91 Antilles arc is now well established (Manga et al., 2012) and was measured around the  
92 Guadeloupe archipelago in a range between  $69.3 \pm 1.5$  and  $98.2 \pm 8.8$  °C/km (Figure 1-A).  
93 The northern part of the magmatic arc, including the Guadeloupe archipelago (Fig. 1-A), is  
94 divided into two subparallel ridges (Westercamp, 1979). The eastern arc is an ancient arc  
95 formed by late Oligocene to Pleistocene carbonate platforms (Grande-Terre), while the  
96 western arc is composed of a recent chain of volcanic islands (Basse-Terre and Les Saintes)  
97 active since 5 Ma (Bouysse and Westercamp, 1990). Basse-Terre Island consists of a cluster  
98 of volcanic complexes, frequently called “volcanic chains” (Figure 1-B). From north to south,  
99 Samper et al. (2007) recognized the following: the oldest, the Basal Complex, characterized  
100 by 4.28 to 2.67 Ma aged volcanism (Favier et al., 2019); the Septentrional Chain, formed  
101 between 1.81 and 1.16 Ma; the Axial Chain, active from 1.02 to 0.41 Ma; the Grande  
102 Découverte – Trois Rivières Complex (0.20 Ma-recent), including the present-day active La  
103 Soufrière volcano (Boudon et al., 1987, 2008); and the Monts Caraïbes Chain built at  
104 approximately 0.55-0.47 Ma (Blanc, 1983; Ricci et al., 2017). Southeast of Basse-Terre  
105 Island, the Les Saintes archipelago is composed of two islands: Terre-de-Haut Island with  
106 3.63 to 2.00 Ma aged volcanism and Terre-de-Bas Island younger than 1.00 Ma (Jacques et  
107 al., 1984; Jacques and Maury, 1988; Zami et al., 2014).  
108



109

110 Figure 1: A) Geodynamic setting of the Lesser Antilles arc (after Feuillet et al., 2001; heat  
 111 flow measurements from Manga et al., 2012), B) Map of the active volcanic arc in  
 112 Guadeloupe archipelago (i.e. Basse-Terre and Les Saintes) displaying the different volcanic  
 113 complexes and the main tectonic structures (modified after Boudon et al., 1987; Calcagno et  
 114 al., 2015; Ricci et al., 2017; Verati et al., 2018 and Favier et al., 2019), BC = Basal Complex;  
 115 GDVC = Grande Decouverte Volcanic Complex. (COLOR FOR ONLINE VERSION ONLY /  
 116 IMAGE SIZE: COLUMN WIDTH)



117

118 Figure 1: C) Tectonic sketch map of Bouillante area in Basse-Terre Island (modified after  
 119 Calcagno et al., 2012; Calcagno et al., 2015; Navelot et al., 2018). (COLOR FOR ONLINE  
 120 VERSION ONLY / IMAGE SIZE: COLUMN WIDTH)  
 121

122 **2-2- Brittle tectonic framework in the Guadeloupe archipelago:**

123 Regionally, four major brittle fault directions have been identified: N50-N70, N90-N110,  
 124 N120-N140 and N160-N10. Onshore and offshore investigations (Feuillet et al., 2002, 2004,

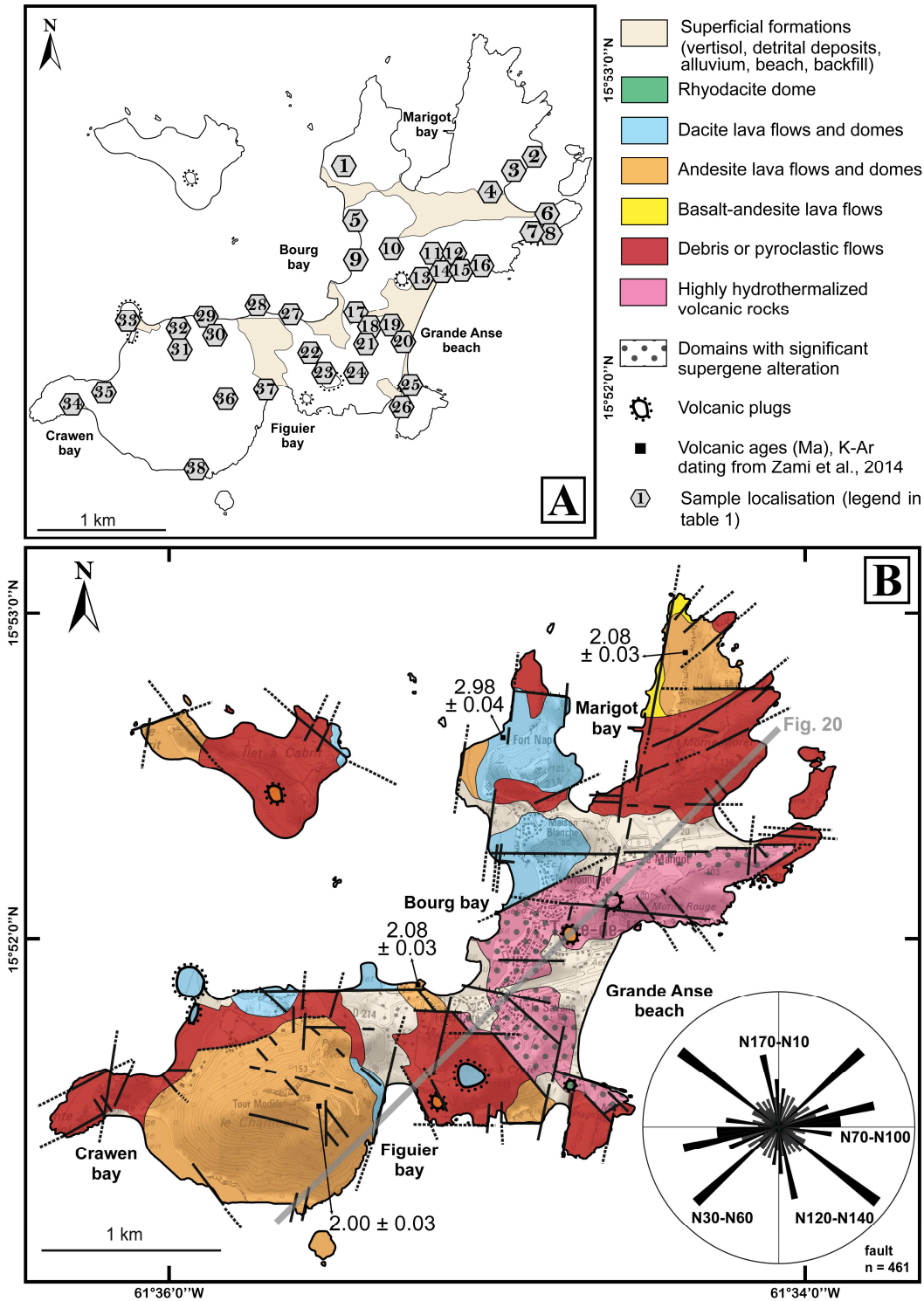
125 2010, 2011; Thinon et al., 2010; Mathieu et al., 2011; Corsini et al., 2011; Laigle et al., 2013;  
126 Lardeaux et al., 2013; Münch et al., 2013; De Min et al., 2015; Leclerc et al., 2016; Legendre,  
127 2018; Navelot et al., 2018; Leclerc and Feuillet, 2019) allow the reconstruction of the  
128 deformation history on the basis of relative chronology criteria (i.e., superposed geometries).  
129 Statistically, the N160-N10 structures are regarded as the youngest fault system, while the  
130 N40-N50 structures are the oldest. According to many authors, the N120-N140 and N90-  
131 N110 structures are synchronous, have been generated under a transtensive regime, and  
132 are presently active. Many studies (Bouysse, 1979; Bouysse and Westercamp, 1990; Feuillet  
133 et al., 2002, 2004; Corsini et al., 2011; Lardeaux et al., 2013; De Min, 2014; Legendre et al.,  
134 2018; Legendre, 2018; Verati et al., 2018; Navelot et al., 2018; Favier et al., 2019) have  
135 emphasized the importance of structural heritage, and thus tectonic reactivation, on the  
136 present-day active deformation pattern. Interestingly, the active Bouillante geothermal field  
137 (Figure 1-B) is located at an intersection between three of the main regional tectonic  
138 structures (Calcagno et al., 2012, 2015, Figure 1-C).

139

### 140 **2-3- Terre-de-Haut in Les Saintes Islands**

141 Terre-de-Haut, the oldest volcanic complex in the Les Saintes Islands, predominantly  
142 consists of calc-alkaline lavas, debris and pyroclastic flows, with abundant lava domes and  
143 volcanic plugs (Jacques and Maury, 1988; Verati et al., 2016). All volcanic rocks are  
144 frequently fractured and deformed by superposed fault systems. We have recently clearly  
145 established the main characteristics of the finite brittle field on Terre-de-Haut Island (Verati et  
146 al., 2016; Navelot et al., 2018; Favier, 2019). In summary, our structural analyses revealed  
147 the existence of four main directions for the orientation of fault networks characterized by the  
148 following statistical structural directions: N030-N060-, N070-N100-, N120-N140- and N170-  
149 N010-trending fault systems (Figure 2-B). All these faults are generally strongly dipping, and  
150 when kinematic criteria are visible, they are compatible with normal faulting with a strike-slip  
151 component. This brittle finite strain pattern is therefore consistent with the regional

152 transtensive tectonic framework currently recognized in the Guadeloupe archipelago (Feuillet  
 153 et al., 2002, 2004, 2010; Calcagno et al., 2012; Baird et al., 2015; De Min et al., 2015;  
 154 Leclerc et al., 2016).



155

156 Figure 2: A) Sample localisations map, legend in table 1. B) Revised geological map of Terre  
 157 de Haut Island showing the main geological formations and the volcanic lithologies defined  
 158 by their geochemistry and their mineralogy (modified after Jacques and Maury, 1988 and

159 Verati et al., 2016). The heavy dark lines represent the main brittle faults identified in the field  
 160 (modified after Verati et al., 2016 and Navelot, 2018). **(COLOR / IMAGE SIZE: FULL PAGE)**  
 161

162 Table 1: Samples location from Terre-de-Haut Island. **(BLACK AND WHITE / IMAGE SIZE:**  
 163 **COLUMN WIDTH)**  
 164

Localisation (fig. 2-A)	Samples
1	17TH26
2	17TH28
3	TH-23a,b,c,d,e
4	17TH29
5	TH-30a1,a2,b ; 16STH33
6	TH-32a,b, TH-33
7	GD-16-10
8	17TH30
9	17TH32a,b
10	17TH31
11	SA-14
12	17TH33
13	16STH01a,b; 16STH02; 16STH03; 16STH04a,b; 16STH05; 16STH06; GD16-06
14	16STH07; 16STH08; 16STH09a,b; 16STH10; GD-16-03; GD16-05
15	16STH11a,b; 16STH19; 16STH20; TH26a,b
16	16STH12; 16STH13; 16STH14; 16STH15a,b; 16STH16; 16STH17; 16STH18; 17TH16; 17TH17a,b; TH-27
17	16STH34; GD15-142
18	16STH35a,b; TH-24a,b,c; TH-25
19	17TH01; 17TH02
20	17TH03; 17TH04; 17TH15; 16STH21a,b
21	17TH14; 16STH22
22	17TH12
23	17TH11
24	17TH09; 17TH10
25	17TH05; 17TH06; 17TH07; TH-25'; SA-12-08; GD-17-08
26	17TH08; GD17-09
27	16STH23a,b
28	16STH24; 16STH25; 16STH26; 16STH27; 16STH28
29	16STH30; 16STH31; 16STH32
30	16STH29; 17TH24; TH-28
31	17TH23
32	17TH22
33	17TH21
34	17TH18; 17TH19
35	17TH20

<b>36</b>	17TH25a,b; DC-10-09a,b; GD17-11
<b>37</b>	17TH13; TH-29; GD-15-135
<b>38</b>	GD-15-139; GD15-140; GD15-141

165  
166 The central part of Terre-de-Haut Island is dominated by a large area (2 km<sup>2</sup>) of intensely  
167 hydrothermally altered rocks (Figure 3), within which the initial magmatic structures and  
168 mineralogy have been completely erased, making it impossible to distinguish between lava,  
169 debris or pyroclastic flows (Jacques and Maury, 1988; Verati et al., 2016). The recognition of  
170 high-temperature hydrothermal mineral associations as well as the importance of fault  
171 networks led Verati et al. (2016) to interpret this hydrothermal zone as an exhumed  
172 paleogeothermal reservoir. Such hypothesis is consistent with the age of volcanic activity (3-  
173 2 Ma) combined with the fact that the Guadeloupe archipelago is subjected to intense  
174 weathering and erosion processes (Dessert et al., 2015). Indeed, because effective  
175 mechanical and chemical erosion rates are approximately 1-2 mm/yr (Sak et al., 2010; Lloret  
176 et al., 2011; Rad et al., 2013; Ricci et al., 2015 a, b), the central part of this island was  
177 potentially buried by a minimum of 2 km of volcanic materials. Low-temperature mineral  
178 associations typical for subsurface zones of geothermal fields have been recently described  
179 in this area (Beauchamps et al., 2019). Altogether, these data suggest that the Terre-de-Haut  
180 hydrothermal system is the key target to study an exhumed analogue of a geothermal  
181 reservoir in the Guadeloupe archipelago.

182

183

184



185

186 Figure 3: Aerial view of a central portion of the Terre-de-Haut Island showing the extent of  
187 the highly hydrothermalized area. View from Pré Cassin. **(COLOR / IMAGE SIZE: FULL**  
188 **PAGE WIDTH)**  
189

### 190 **3- Material and methods**

#### 191 **3-1- Terminology**

192 The term “hydrothermal processes” strictly refers to the changes that occur when fluids  
193 circulate into and react with surrounding pre-existing rocks, producing physicochemical  
194 changes in these “country rocks” (see reviews in Meyer and Hemley, 1967; Guilbert and  
195 Park, 1986; Pirajno, 2009). Because a hydrothermal system requires a heat source, a fluid  
196 phase (supergene and/or hypogene fluids) and a transfer structure, there are different  
197 geological settings favourable for hydrothermalism. Consequently, the terms relevant to  
198 describing the chemical and mineralogical changes in rocks affected by hydrothermalism are  
199 not homogeneous. In particular, these chemical/mineralogical transformations are called  
200 “hydrothermal alteration” in the geothermal community but are described as “hydrothermal  
201 metamorphism” by endogenous petrologists, even if the pioneering and seminal works of  
202 Ramberg (1952) and Korzhinskii (1965) established that the physicochemical processes  
203 driving the fluid–rock interactions obey similar thermodynamic and kinetic laws. Since the  
204 late 2000s (Smulikowski et al., 2007), the International Union of Geological Sciences (IUGS)  
205 recommends using the term “hydrothermal metamorphism” for mineralogical and textural  
206 transformations of a local extent caused by the circulation of hot H<sub>2</sub>O-rich fluids developed in  
207 specific settings such as ocean-floor spreading centres, contact aureoles around igneous  
208 intrusions, or volcanic/magmatic arcs (see also Fyfe et al., 1978; Mason, 1978; Schiffman et  
209 al., 1986, 1991; Liou et al, 1987; Yardley, 1989; Frey and Robinson, 1999). During  
210 hydrothermal metamorphism, as is the case in geothermal systems or ore deposit formation,  
211 the percolation of volatile and sometimes overpressurized fluids may dissolve and remove  
212 chemical elements from the country rocks, significantly altering the country rocks’ chemical  
213 composition (Schiffman et al., 1984; Yardley and Cleverley, 2015, with references therein).

214 As underlined by Schiffman et al. (1984) and Pirajno (2009), high-temperature hydrothermal  
215 alteration and hydrothermal metamorphism are terms describing similar processes.  
216 Consequently, the term “hydrothermal alteration”, hereafter used, must be considered in its  
217 broadest acceptance.

### 218 **3-2- Mapping lithology, structural analysis and sampling strategy**

219 We mapped the various volcanic units distinguishing the following formations: well-developed  
220 soils, superficial quaternary sedimentary formations, debris flows, lava flows, lava domes and  
221 volcanic plugs.

222 More than a hundred measurements across the island of tectonic structures were acquired  
223 for this study during seven fieldwork campaigns between 2010 and 2017 to characterize the  
224 superposed finite ductile strain fields, the chronology of deformations and, when possible, the  
225 kinematics of deformation.

226 To assess the intensity and degree of hydrothermal alteration of the volcanic rocks, we  
227 sampled fresh, slightly altered, and highly altered volcanic rocks. The sampling was executed  
228 while taking into account the rock deformation in the regional finite deformation gradient. As  
229 a result, the undeformed fresh volcanic rocks are considered a standard reference in order to  
230 quantify the effects of hydrothermalism and deformation.

### 231 **3-3- Whole rock geochemistry**

232 Whole rock major and trace element analyses were performed on 108 samples (localized in  
233 figure 2-A and table 1; analytical results provided in the supplementary data in table A-1),  
234 first, to improve the definition of the lithotypes defined on the geological map and, second, to  
235 specify the magmatic characteristics of the sampled lava flows. We carefully selected the  
236 rock exposures to ensure that the main types of lava flows were sampled. Analyses were  
237 obtained by using inductively coupled plasma atomic emission spectrometry (ICP-AES) and  
238 inductively coupled mass spectrometry (ICP-MS) for major and trace elements, respectively,

239 on rock powders at the Geochemical and Petrographic Research Center in Nancy (SARM  
240 laboratory, CNRS-CRPG, with the procedure proposed by Carignan et al., 2001).

### 241 **3-4- Mineral chemistry**

242 In order to precise and quantify the mineralogical evolution of Terre-de-Haut, we investigated  
243 126 thin sections. Mineral chemistry data were obtained by electron microprobe analyses, X-  
244 ray diffractometry and Raman spectrometry.

245 Non-oriented powders of whole rocks (<70  $\mu\text{m}$ ) were analysed by an X-ray diffractor with an  
246 XPert-Pro MPD system (Bragg Brentano geometry, Cu anticathode, KAlpha1 and KAlpha2  
247 rays, PIXcel detector) at the CEMEF in Sophia-Antipolis Scientific Center.

248 Mineral major compositions were analysed using the "Service Commun de Microsonde" of  
249 the University of Montpellier using a Cameca-SX100 electron microprobe. The operating  
250 conditions were a 20 kV accelerating voltage and 10 nA beam current. The chemical  
251 analyses of hydrothermal phases are presented in the supplementary data in tables A-2, A-3,  
252 and A-4.

253 We also used Raman spectrometry to complete the mineral determination. The spectra were  
254 recorded with a LabRam HR800UV Jobin-Yvon™ microspectrometer equipped with 1800  
255 g/mm gratings and using 532 nm green laser excitation; the acquisition time span varied from  
256 20 to 70 s during three accumulating cycles.

### 257 **3-5- P-T-XH<sub>2</sub>O estimates for hydrothermal mineral associations**

258 The pressure and temperature conditions of hydrothermalism were quantified by the use and  
259 the comparison of:

260 - Stability fields of typical geothermal minerals, using the available detailed descriptions of  
261 phase distribution with respect to measured temperatures and depths in active geothermal  
262 fields (see, for example, Reyes, 1990 and White and Hedenquist, 1995).

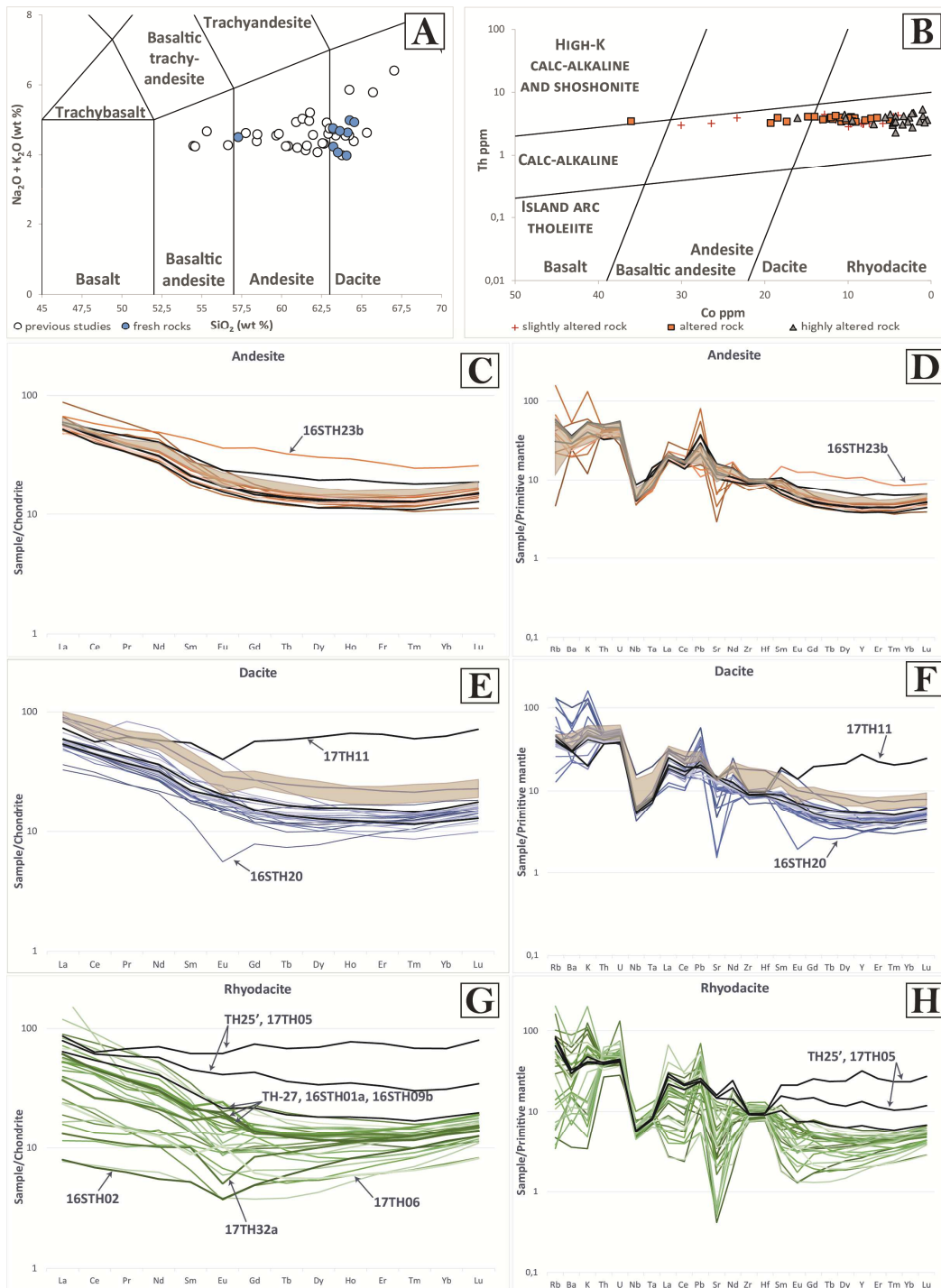
263 - Classical chlorite solid-solution thermometry, which has been successfully used (Lanari et  
264 al., 2014) since the pioneering works of Cathelineau and Nieva (1985) and Cathelineau  
265 (1988) in both geothermal and low-grade metamorphic systems.

266 - Thermodynamic modelling of representative samples using the THERIAK-DOMINO method  
267 (De Capitani and Petrakakis, 2010) with the Holland and Powell database (1996-2007).  
268 These calculations were carried out only on samples not affected or poorly affected by  
269 supergene alteration. THERIAK-DOMINO, similar to other mineral equilibrium modelling  
270 methods (for example THERMOCALC, Powell et al., 1998), allows us to approach, as closely  
271 as possible, the effects of all major chemical elements measured (i.e., real and effective  
272 whole rock chemistry) in a given bulk rock composition on the stability of the observed  
273 mineral assemblage in the thin section of interest. THERIAK-DOMINO is a free energy  
274 minimization program, taking into account available mixing models for mineral solid solutions  
275 and allowing calculation of pseudo-sections (i.e., isochemical phase diagrams) for a selected  
276 natural sample. Pseudo-section calculations allow exploration of not only the P-T stability  
277 fields of complex mineral assemblages (including the relative abundances of the different  
278 phases in a given mineral assemblage) but also the effects of the Fe-oxidation state on  
279 mineral stabilities as well as the influence of the amount and nature of fluids (particularly  
280 H<sub>2</sub>O) on the stability field of a given mineral association.

#### 281 **4- Diversity of lithologies**

282 The first geological map of Terre-de-Haut was produced in the 1980s (Jacques and Maury,  
283 1988) and was later improved by geochronological and structural investigations (Zami et al.,  
284 2014; Verati et al., 2016). To continue the refinement of Terre-de-Haut geology we produced  
285 a revised geological map (Figure 2-B). We refined the definition of volcanic rocks, sampled in  
286 domes or lava flows, by systematic geochemical and petrographic investigations taking into  
287 account their degree of hydrothermal alteration overprint.

288 SiO<sub>2</sub> content ranges from 54 to 67 wt.% (100% anhydrous) for lava flows, with a  
 289 compositional range from basalt-andesite to dacite (Figure 4-A). The studied samples have a  
 290 calc-alkaline affinity with an evolution from andesites to dacites and medium to low K series  
 291 (SiO<sub>2</sub> versus Nb/Y diagram, Winchester and Floyd 1977, and K<sub>2</sub>O versus SiO<sub>2</sub> diagram,  
 292 Peccerillo and Taylor 1976, in supplementary data in figure A-1).



293

294 Figure 4: Geochemical characterization of lava flows: A) Total alkali vs. silica diagram after  
295 Bas et al. (1986), white circles represent previous data from: Jacques et al., 1984; Jacques  
296 and Maury, 1988; Zami et al., 2014 and Navelot et al., 2018. Blue circles correspond to our  
297 new analyses. B) Co vs. Th plot after Hastie et al., 2007 showing the primary volcanic  
298 chemical characters of the studied rocks whatever their degree of alteration. B, E and G)  
299 Rare Earth Element (REE) patterns normalized to chondrite values (after Sun and  
300 McDonough, 1989) of the andesite, dacite and rhyodacite lava flows, respectively, used in  
301 this study. D, F and H) Multi-element patterns normalized to primitive mantle (after Sun and  
302 McDonough, 1989). For C to H : The shaded bands correspond to the range determined for  
303 Terre-de-Haut volcanic rocks (after Zami et al., 2014) and black bold lines for fresh samples.  
304 **(COLOR / IMAGE SIZE: FULL PAGE WIDTH)**

305 The Rare-Earth-Element (REE) diagram normalized to chondrite values (after Sun and  
306 McDonough, 1989; Figures 4-C, E and G) displays relatively constant slopes with light REE  
307 (LREE) enrichment for most samples, which is typical of insular arc-related calc-alkaline  
308 lavas (Hawkesworth and Powell, 1980; Labanieh, 2009). Moreover, the multi-element  
309 diagram normalized to primitive mantle (Sun and McDonough, 1989; Figure 4-D, F and H)  
310 also displays typical patterns of calc-alkaline arc lavas, i.e., with Th, Pb, U and large ion  
311 lithophile element (LILE) enrichments and Nb and Ta negative anomalies (Labanieh, 2009).

312 The rhyodacitic samples (Figures 4-G and H) show the most diversified geochemistry with  
313 especially strong Sr depletion, large K, Th, U enrichments, or K depletion. This geochemical  
314 variability can be explained either by different magmatic differentiation stages or by element  
315 mobility during alteration processes, especially for Sr and K.

316 However, the positive Y and heavy-REE (HREE) anomalies of samples from “Morne à Craie”  
317 (17TH11) (Figure 4-F) and the intrusion in the southern part of Grande Anse beach (TH-25’  
318 and 17TH05, Figure 4-H) reflect supergene processes (Cotton et al., 1995), as also identified  
319 in REE spectra (Figures 4-E and G).

320 Major and some trace elements (mainly LILEs and LREEs) are severely mobile in altered  
321 lavas. Therefore, to improve the geochemical characterization of our samples, particularly  
322 those affected by hydrothermal transformations, we selected diagrams based only on minor  
323 and trace elements considered the most immobile. Samples where the LOI (loss on ignition)  
324 is below 1-2% and in which the primary volcanic mineralogy is well preserved (see section  
325 6), demonstrating that hydrothermalism and/or weathering are insignificant, are named

326 slightly altered rocks, while when the LOI is above 3% (as suggested by Samper et al., 2007,  
327 to identify significantly altered volcanic rocks), and when secondary minerals are abundant,  
328 they are named altered or highly altered rocks. Hypogene and supergene hydrothermal  
329 alteration produce new mineral assemblages often associated with significant redistribution  
330 of chemical elements (Sturchio et al., 1986; Fulignati et al., 1999; Pandarinact et al., 2008;  
331 Salaün et al., 2011). For these reasons, we used the Th-Co diagram proposed by Hastie et  
332 al. (2007), which is an efficient tool for the characterization of magmatic protolith signatures  
333 in altered lava flows. In this diagram, the studied rocks define a typical medium-K calc-  
334 alkaline volcanic series (Figures 4-A and A-1) characterized by significant magmatic  
335 differentiation from basaltic andesites to rhyodacites. Many lava domes and volcanic plugs  
336 correspond to andesites or rhyodacites. Lava flows are more diversified and thus we  
337 distinguished, on the revised geological map, basalt-andesites, andesites, dacites and  
338 rhyodacites (Figure 2-B).

339 In common trace element diagrams (Figures 4 C to H), the lava flow samples are compared  
340 to fresh volcanic rocks from Terre-de-Haut Island (Zami et al., 2014)

341 Hydrothermally altered andesite samples (Figures 4-C and D) are similar to fresh volcanic  
342 andesites. A specific sample (16STH23b) displays enrichment in MREEs (middle-REEs) and  
343 HREEs, typical for weathering and leaching processes (Zhou et al., 2013).

344 Dacite samples (Figures 4-E and F) display a consistent trend for most samples with  
345 enrichment in LREEs. However, two samples (17TH11 and 16STH20) show chemical  
346 anomalies indicating supergene alteration processes (Cotten et al., 1995; Zhou et al., 2013;  
347 Ricci et al., 2017) and plagioclase fractionation or interaction with mildly acidic low  
348 temperature fluids (Bau, 1991; Taylor and McLennan, 1995) respectively.

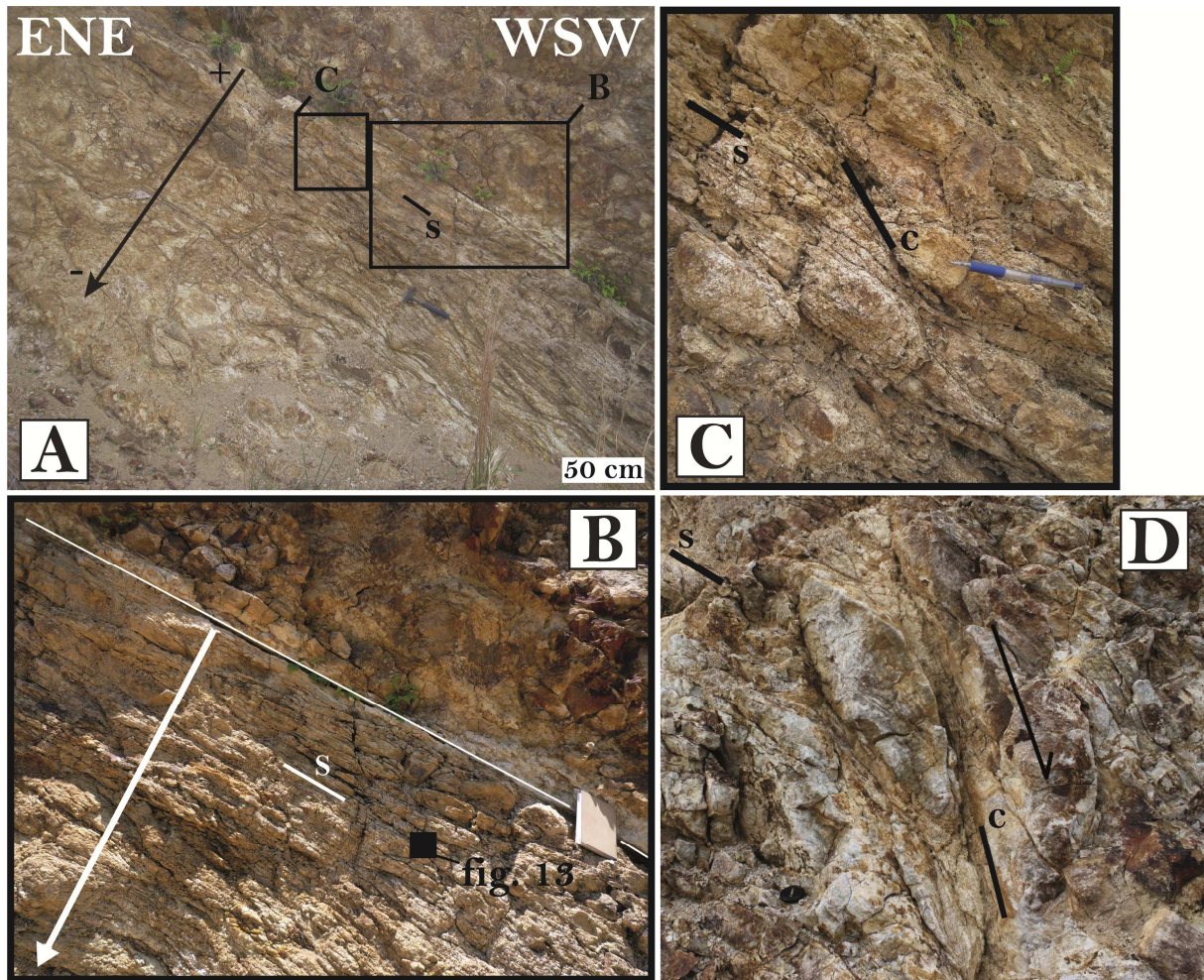
349 Rhyodacite samples (Figures 4-G and H) display the most varied spectra with different  
350 degrees of LREE enrichment. This feature can be either related to different magmatic  
351 differentiation stages or to secondary alteration events (hydrothermal and/or supergene

352 alteration). Some samples show especially negative or positive Eu anomalies, suggesting  
353 either plagioclase fractionation or interaction with secondary fluids (Bau, 1991). For example,  
354 two severely hydrothermalized samples from the southern part of the Bourg Bay area  
355 (17TH32a) and the northern part of Grande Anse beach (16STH02) reveal both LREE  
356 depletion and slightly negative Eu anomalies related to typical high temperature alteration  
357 under mildly acidic conditions (Bau, 1991). Other hydrothermalized samples from the  
358 northern part (TH-27, 16STH01a, 16STH09b) and southern part (17TH06) of Grande Anse  
359 beach present a positive Eu anomaly due to acidic high-temperature hydrothermal fluids  
360 (Michard et al. 1983; Cotten et al., 1995). Samples from the southern part of Grande Anse  
361 beach (TH-25' and 17TH05) show high HREE enrichment and slight negative Ce anomalies,  
362 reflecting weathering processes under tropical conditions (Cotten et al., 1995; Zhou et al.,  
363 2013; Ricci et al., 2017), as already observed for the dacitic sample (Figure 4-E).

#### 364 **5- Structural and microstructural analyses**

365 Despite previous structural analyses at Terre de Haut (Jacques and Maury, 1988; Verati et  
366 al., 2016; Navelot et al., 2018), we identify for the first time in Terre-de-Haut Island zones of  
367 ductile deformation (Figures 5-A and B), marked by the development of spaced cleavages  
368 produced by pressure-solution processes.

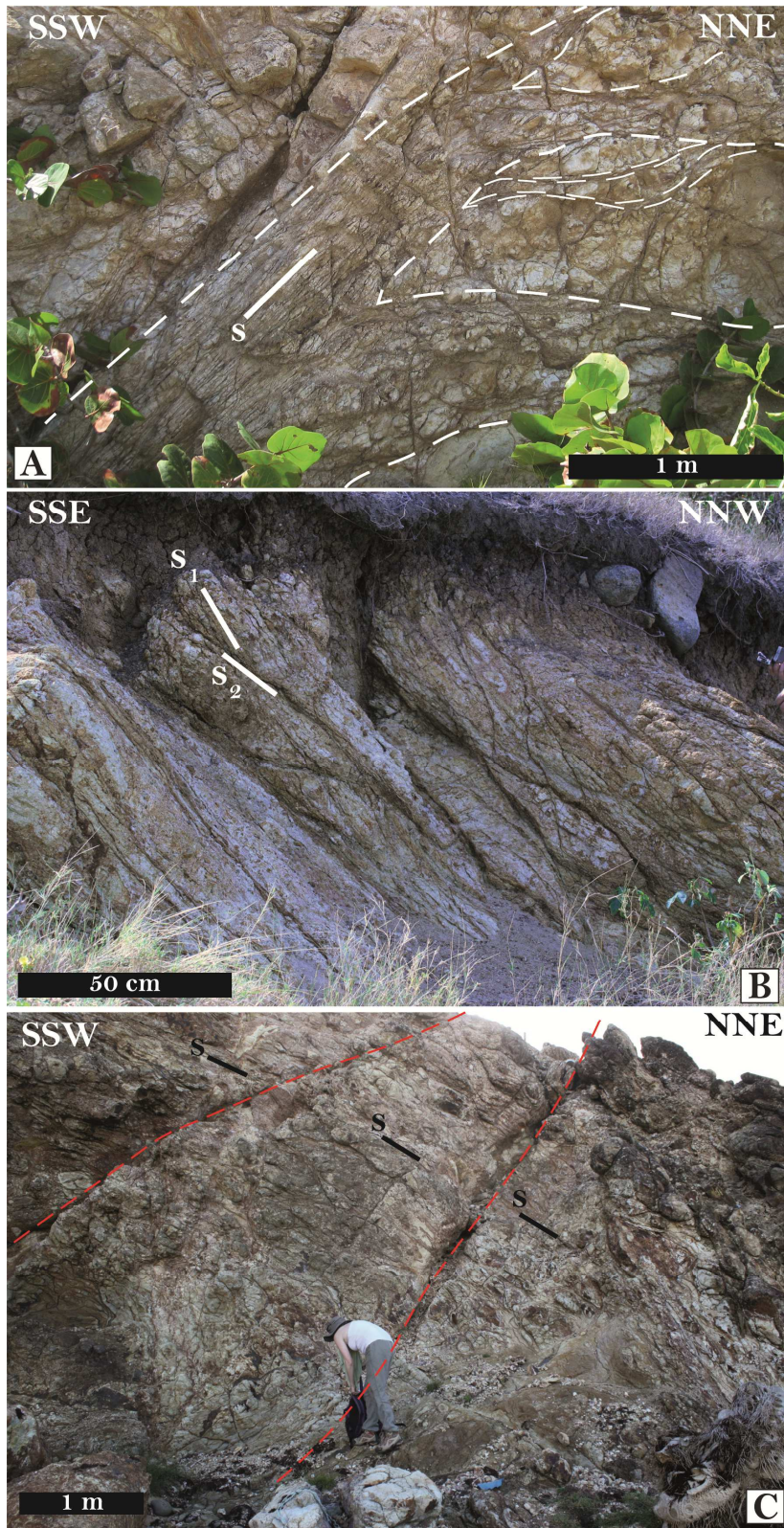
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370

371 Figure 5: A) Schistose corridors showing a finite deformation gradient (from the aerodrome  
 372 quarry). B) Zoom on well-developed schistosity planes. C) Local ductile strain pattern with S  
 373 (schistosity) and C (shear planes) structure compatible with normal sense of shear  
 374 (schistosity plane N160, 40W, stretching lineation N270, 35W, C plane N0, 75W). D) Typical  
 375 S (schistosity) and C (shear planes) structure compatible with a normal sense of shear in the  
 376 area of interest. (COLOR / IMAGE SIZE: FULL PAGE WIDTH)  
 377

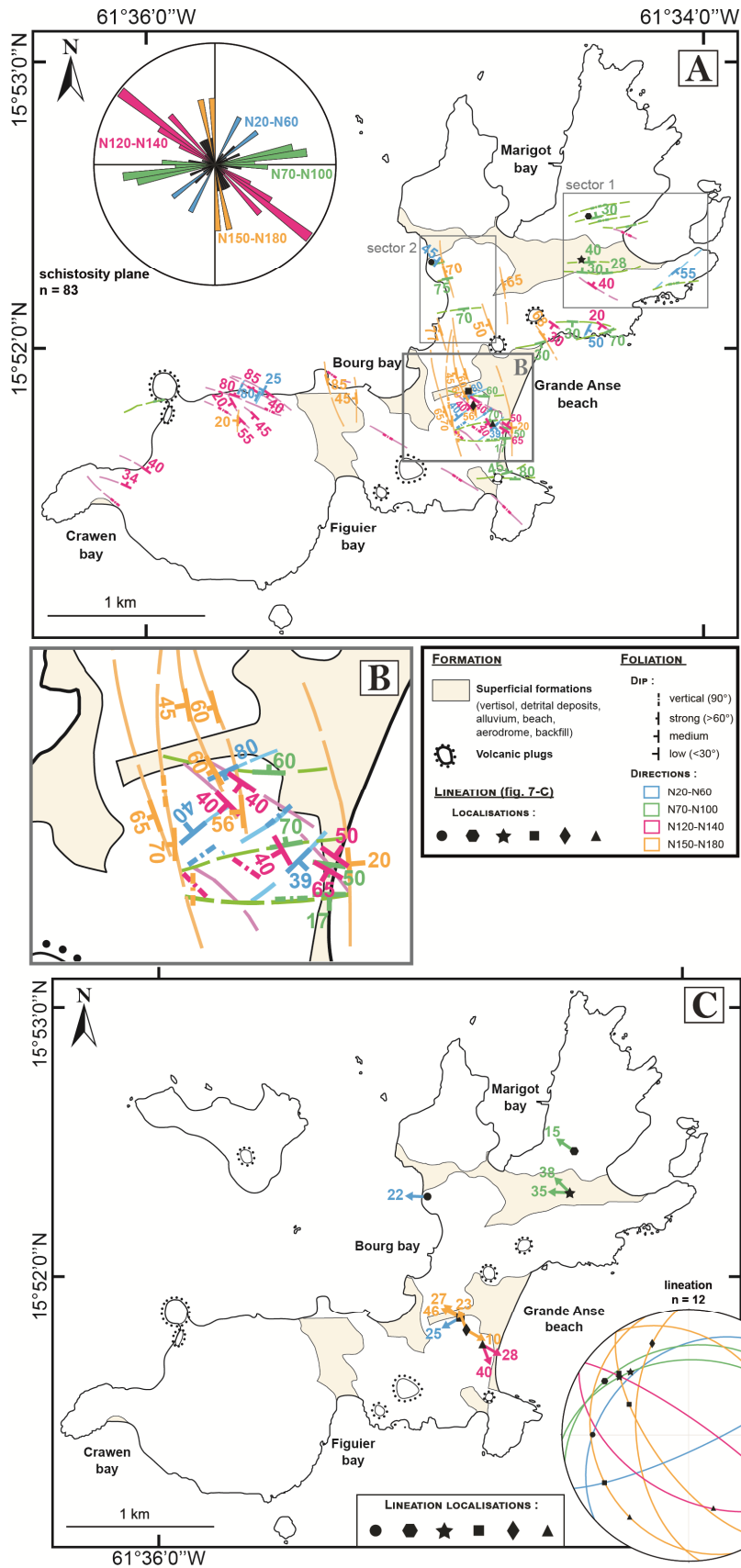
378 These planar rock structures are concentrated into decametre to hectometre corridors and  
 379 thus define “ductile shear zones” within the finite ductile strain pattern (Figures 5-A and B  
 380 and Figures 6-A and B). Stretching lineations are difficult to decipher, but in highly strained  
 381 domains, strongly dipping lineations and local C/S or C' structures indicate a normal sense of  
 382 shear (Figures 5-C and D). At both the outcrop and map scales (Figures 7-A and B), the  
 383 shear zones are heterogeneously distributed and tend to anastomose, forming lenticular  
 384 pods (Figure 6-A and sector 1 in Figure 7-A) or producing high-angle intersections (Figures  
 385 6-B and sector 2 in Figure 7-B).



386

387 Figure 6: A) Anastomosing schistosity planes forming lenticular pods. Outcrop from La  
 388 Savane. B) Intersection between two schistosity planes. Outcrop from north of Chameau  
 389 dome. C) Schistosity plane (S) intersected by fault planes (red dotted lines) and fractures.  
 390 Top of La Savane outcrop. (COLOR / IMAGE SIZE: FULL PAGE WIDTH)  
 391

392 Four directions of ductile shear zones (Figure 7-A) have been observed and measured in  
393 the field: (1) N020-N060, (2) N120-N140, (3) N070-N100, and (4) N150-N180. Both the  
394 abundance of anastomosed structures and the absence of coherent and repetitive criteria of  
395 relative chronology within the superimposed geometries (i.e., intersections) lead us to  
396 propose that the observed finite geometries are synchronously developed during progressive  
397 deformation.



398

399 Figure 7: A) Tectonic sketch map of schistosity trajectories on Terre-de-Haut Island. B) Zoom  
 400 on central part of Terre-de-Haut Island. C) Map of measured lineations on the Terre-de-Haut  
 401 Island. (COLOR / IMAGE SIZE: FULL PAGE WIDTH)

402 The planar fabrics display variable dips from 90 to 10° (Figures 6 and 8), and when  
 403 observable, the stretching lineations have dips ranging between 10 and 45° (Figure 7-C and  
 404 Table 2). The lineations' dip is compatible with the occurrence of a strike slip component  
 405 associated with ductile extensional shearing.

406 Table 2: Schistosity planes (S) and associated lineations (L) presented in Fig.7. **(BLACK**  
 407 **AND WHITE / IMAGE SIZE: FULL PAGE WIDTH)**  
 408

Site	Type	Strike	Dip	Quadrant	Trend	Plunge
●	S	N228	35	N		
	L				272	22° W
⬡	S	N258	30	N		
	L				305	15° NW
★	S	N268	40	N		
	L				315	38°NW
	L				272	23° W
■	S	N145	66	W		
	L				310	27° NW
	L				297	46° NW
■	S	N60	80	S		
	L				240	25° SW
◆	S	N175	56	W		
	L				340	23° NW
◆	S	N325	30	E		
	L				120	10° E
▲	S	N120	50	SW		
	L				160	40° S

409  
 410

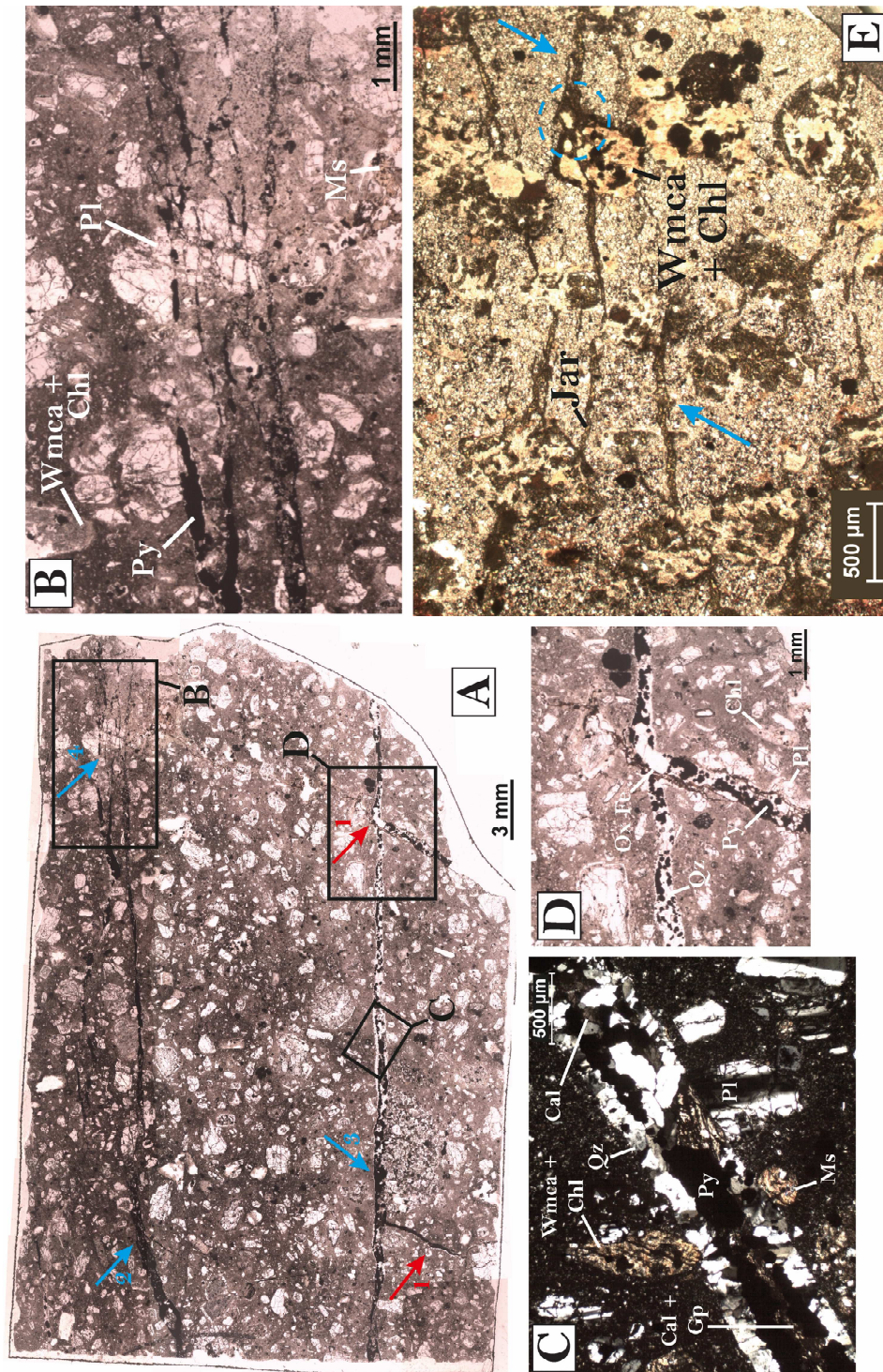


411  
 412 Figure 8: A) Highly hydrothermalized volcanic rock from North of Grande Anse beach with  
 413 strong dip schistosity plane (S) expressed by spaced disjunctive cleavages. B) Penetrative  
 414 schistosity plan (S) with low dip, planar fabric underlined by insoluble dark minerals (iron-  
 415 oxides). C) Example of a penetrative low dip schistosity plan (s) with concentration of  
 416 insoluble minerals in dark band. (COLOR / IMAGE SIZE: FULL PAGE WIDTH)  
 417

418 At the outcrop scale, the observed planar fabric is underlined by dark seams of insoluble  
 419 dark minerals (iron oxides) and corresponds to well-developed spaced disjunctive cleavages

420 (Powell, 1979; Borradaile et al., 1982; Rutter, 1983; Neuendorf et al., 2005; Passchier and  
421 Trouw, 2005; Brodie et al., 2007; Figure 8). In polymineral rocks, the passive concentration  
422 of insoluble minerals attests to stress-driven pressure solution processes (Durney, 1972;  
423 Green, 1984), a major mechanism of ductile deformation of the upper crust, which is  
424 frequently observed at the brittle/ductile transition zone (see Gratier et al., 2013, with  
425 references therein, for an extensive review).

426 In Terre-de-Haut Island, the spacing of the observed solution cleavage is variable (Figures 5,  
427 6-A-B and 8) and is locally associated with veins or fractures (Figures 6-C and 9-A). Most of  
428 the time, the veins or micro-cracks are perpendicular, or at high angles, to the spaced  
429 disjunctive cleavages (Figure 9-D).



430

431 Figure 9: A) Schistosity planes (thin section scale) underlined by insoluble minerals (blue  
 432 arrows). Schistosity plane reworked by opening of late veins (red arrows). B) Zoom on  
 433 schistosity planes, underlined by concentration of oxides and pyrite. Pressure-solution  
 434 cleavage is marked by partial dissolution of volcanic plagioclase. C) Zoom on a vein  
 435 reworking the schistosity plane and filled with quartz, pyrite, calcite and gypsum. D) Zoom on  
 436 an intersection of two veins. E) Spaced disjunctive cleavages (blue arrow) underlined by  
 437 jarosite (Jar). Sample from the aerodrome quarry. Blue arrow indicates schistosity planes  
 438 with jarosite. Blue dotted circle indicates the intersection between schistosity plane and  
 439 volcanic mineral replaced by hydrothermal phases (Wmca = white mica; Chl = chlorite).  
 440 (COLOR / IMAGE SIZE: FULL PAGE WIDTH)

441 In thin-section, the spaced cleavages are also underlined by concentrations of insoluble  
442 minerals, such as ilmenite and magnetite and also, in a restricted number of cases, by  
443 crystallization of pyrites or sulphates (gypsum, jarosite, barite; Figures 9-C and E). The  
444 spaced disjunctive cleavages, leading to the concentration of insoluble mineral species, are  
445 mostly developed in the volcanic groundmass of lava flows. When volcanic phenocrysts are  
446 intersected by spaced disjunctive cleavages, we frequently observe local dissolution of these  
447 primary volcanic minerals, mainly at their periphery (Figures 9-B, C and E). Moreover, in  
448 such a case, the volcanic phenocrysts, particularly the pyroxenes, are severely  
449 pseudomorphosed, with a gradient of alteration from rims to cores, and replaced by a  
450 secondary assemblage rich in white micas, chlorites, albite and quartz. Altogether, these  
451 microstructural observations confirm first that the observed spaced cleavages are the result  
452 of intergranular pressure-solution mechanisms and second that solution cleavage formation  
453 is contemporaneous with fluid and mass transfers responsible for hydrothermal alterations of  
454 pyroxenes. Moreover, in the studied lavas, as is the case in the whole volcanic arc of the  
455 Guadeloupe archipelago, the volcanic groundmass is rich in infra-millimetre microlites  
456 (particularly Fe or Fe-Ti), oxides and anorthoclases.

## 457 **6- Petrography and mineralogy**

458 Because of the heterogeneity of the hydrothermal transformations, the finite deformation  
459 state, and finally, the degree of supergene alteration depicted in Terre-de-Haut Island, we  
460 need to establish clear petrographic distinctions between fresh, slightly altered, altered, and  
461 highly altered volcanic rocks. Mineral chemistry was performed on all rock types, and  
462 selected microprobe analyses are presented in table 3 and in supplementary data tables A-2,  
463 A-3 and A-4. Except for fresh volcanic rocks, two stages of hydrothermal alterations, and a  
464 late stage of supergene alteration, were recognized on the basis of classical microstructural  
465 overprinting criteria.

### 466 **6-1-Fresh rocks**

467 The well-preserved lava flows and volcanic plugs are basaltic andesite, andesite, dacite and  
468 rhyodacite. Numerous infra-millimetre microlites of Fe-Ti oxides are present in all lavas, while  
469 anorthoclase microlites are also observed in dacites and rhyodacites.

470 All fresh rocks show porphyric textures with various amounts and natures of phenocrysts  
471 depending on the lithology.

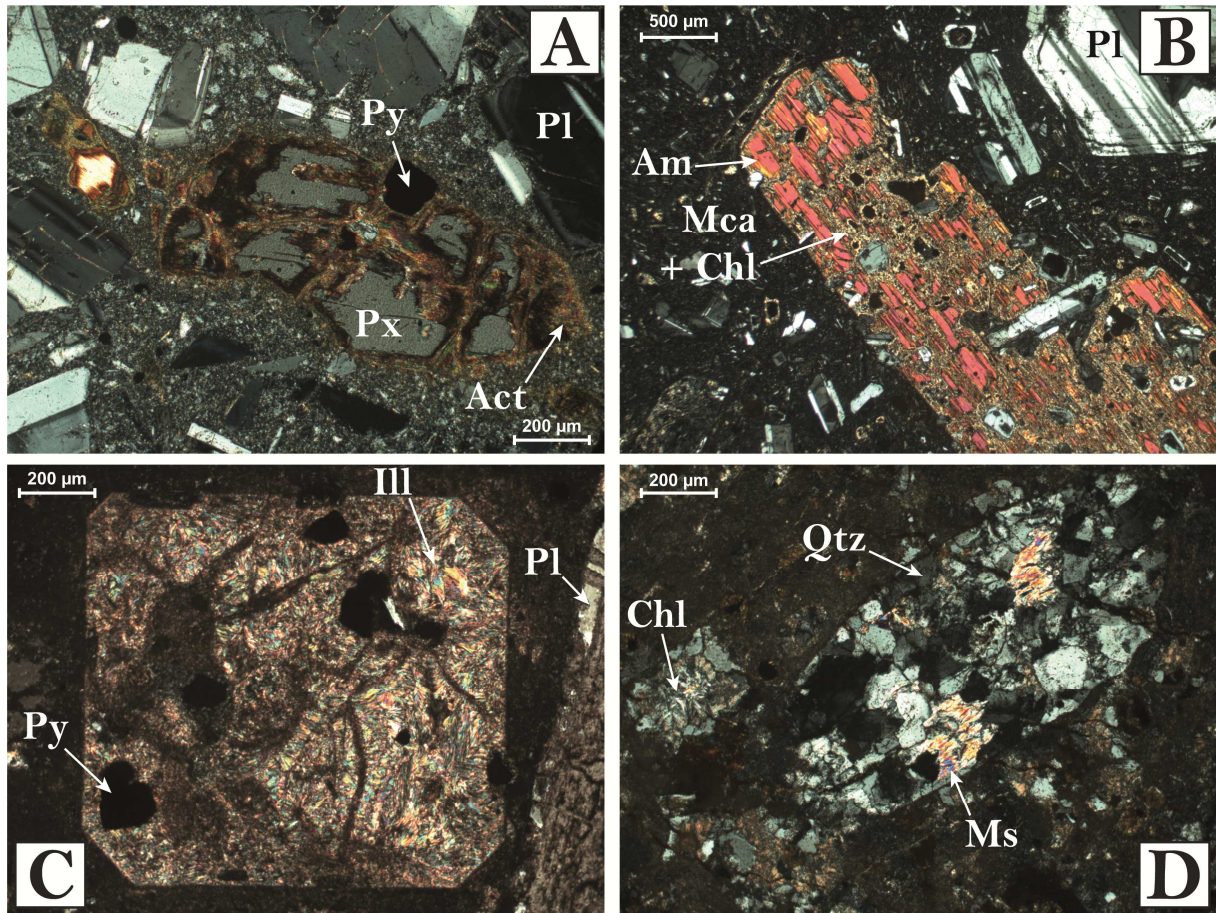
472 - Plagioclases (70-75% of the observed phenocrysts): The phenocrysts are present in all  
473 lithologies and are mostly labradorite and less frequently bytownite. As previously underlined  
474 by Jacques and Maury (1988), in basaltic andesites and andesites, some plagioclases are  
475 zoned, with bytownite in the core and labradorite towards the rim.

476 - Ferromagnesian phases (20-25% of the observed phenocrysts): Brown amphiboles are  
477 observed in basaltic andesites, andesites and dacites, and their compositions vary from  
478 hornblende to edenite or pargasite depending on the considered bulk-rock chemistry (cf.  
479 Jacques and Maury, 1988). Pyroxenes are presenting all rock types. The orthopyroxenes are  
480 hypersthene, while the clinopyroxenes are augite.

481 - The oxides (5% of the observed phenocrysts), are ilmenite and/or titanomagnetite.

## 482 **6-2-Slightly hydrothermally altered rocks**

483 In these rocks, the primary volcanic textures are preserved, but we observed the limited  
484 development of coronas around ferromagnesian volcanic phases (Figures 10-A and B),  
485 mainly around orthopyroxenes and clinopyroxenes. In the coronas, the secondary mineral  
486 phases are actinolite, chlorite, muscovite and calcite. The most affected lithologies are dacite  
487 and debris or pyroclastic flows, within which the secondary phases represent 5 to 10% of the  
488 observed mineral composition.



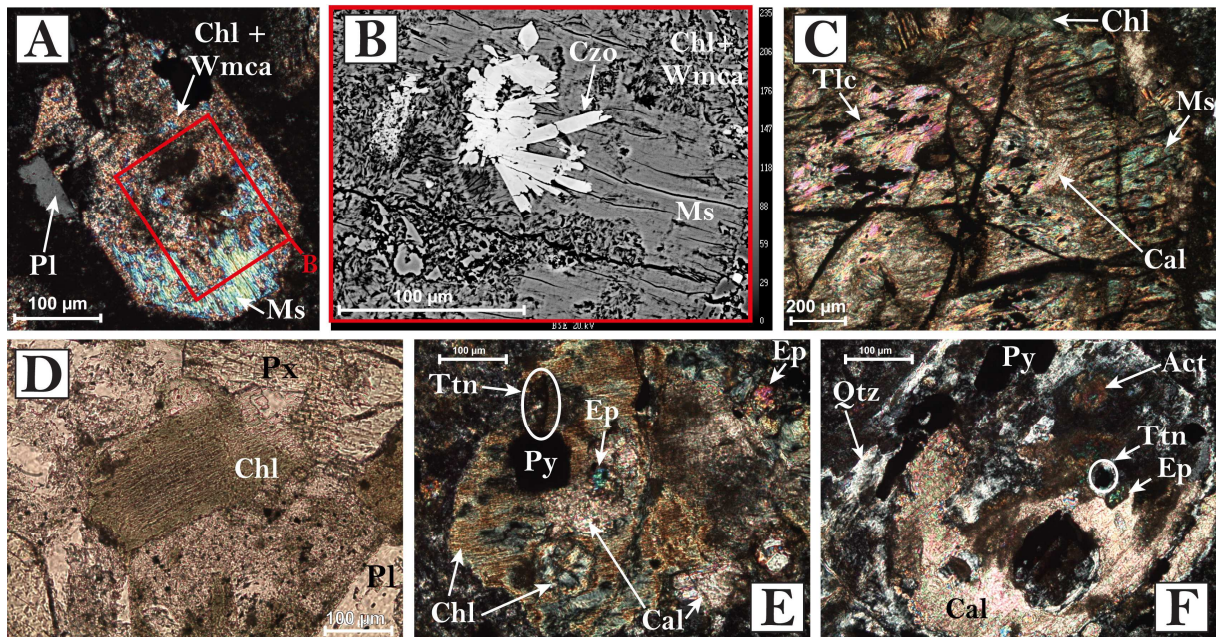
489

490 Figure 10: High-temperature (first hydrothermal stage, T above 300°C) pseudomorphic  
 491 transformations of volcanic minerals. A) Reaction coronas around pyroxene, highlighted by  
 492 actinolite, in an altered andesite. Sample from south of Bourg bay (16STH23a). B)  
 493 Replacement of volcanic amphibole by a mix of chlorite and mica, in altered rhyodacite from  
 494 a pyroclastic flow. Sample from north of Crawen bay (17TH20b). C) Volcanic pyroxene  
 495 replaced by white mica, in an altered rhyodacite. Sample from north of Grande Anse beach  
 496 (16STH19). D) Volcanic phase replaced by quartz, chlorite and/or muscovite, in an altered  
 497 rhyodacite. Sample from La Savane (16STH21a). Act = actinolite, Am = amphibole, Chl =  
 498 chlorite, Qtz = quartz, Ill = illite, Mca = mica, Ms = muscovite, Pl = plagioclase, Px =  
 499 pyroxene, Py = pyrite. (COLOR / IMAGE SIZE: COLUMN WIDTH)  
 500

### 501 6-3-Hydrothermally altered rocks

502 In this category of rocks, we observe relicts of volcanic textures: more than 70% of volcanic  
 503 pyroxenes or amphiboles and more than 50% of volcanic plagioclases are transformed. The  
 504 hydrothermal phases can completely replace the initial volcanic phenocrysts (pseudomorphic  
 505 transformations, Figures 10-C and D), and the most altered lithologies are dacite and  
 506 rhyodacite lava flows. The observed secondary minerals are quartz, albite, epidote  
 507 (clinozoisite in small sticks in Figure 11-B and pistacite in colourless grains in Figure 11-E),

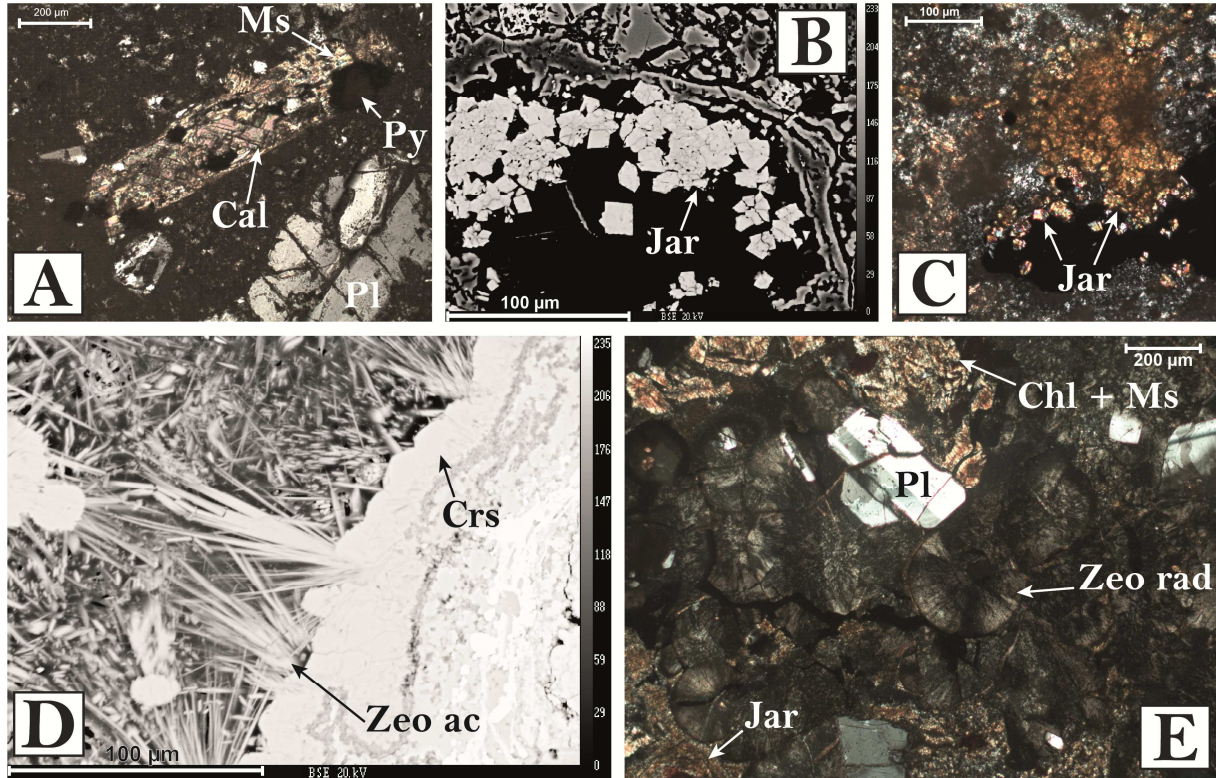
508 muscovite (Figure 11-A), biotite, chlorite (Figures 11-C and D), actinolite (Figure 11-F),  
 509 titanite (Figure 11-E), and, in a lesser amount, talc (Figure 11-C), and pyrite. In many cases,  
 510 grey automorphous chlorite grains are rimmed by green-grey to yellow fibrous grains.  
 511 Subautomorphous muscovites can be replaced by pyrophyllite or fibrous overgrowth acicular  
 512 crystals in sheaf-like aggregates of illite and green to yellow chlorites (Figure 10-C). In late  
 513 veins, small-sized cracks or vacuoles, carbonates (microcrystalline calcite grains with  
 514 polysynthetic twins in Figure 12-A and subautomorphous calcite grains in Figure 11-F),  
 515 sulphates (Figures 12-B and C), silica polymorphs (quartz, chalcedony, cristobalite, Figure  
 516 12-D), acicular or radial zeolites (Figure 12-D and E), and illite in sheaves or spherules can  
 517 also be observed. Finally, in a restricted number of samples, a strong supergene alteration is  
 518 marked by the complete transformation of volcanic phases by clay minerals, particularly  
 519 kaolinite.



520

521 Figure 11: Diversity of secondary new phases developed at the expense of primary volcanic  
 522 minerals. A) Pyroxene replaced by muscovite and a fine-grained mix of chlorite-white mica,  
 523 in an altered rhyodacite. Sample from north of Grande Anse beach (16STH16). B) BSE  
 524 Back-scattered electron image (EPMA acquisition). Zoom on clinozoisite and displaying  
 525 contrast between fibrous muscovite and the mix chlorite-white mica. C) Volcanic phenocryst  
 526 pseudomorphosed by multiple secondary new phases, in an altered andesite. Sample from  
 527 north of Grande Anse beach (SA-14). D) Fibrous green chlorite, in an altered andesite.  
 528 Sample from north of Grande Anse beach (SA-14). E) Pyroxene phenocryst replaced by  
 529 numerous secondary new phases, in an altered andesite. Sample from north of Grande Anse  
 530 beach (SA-14). F) Crystallization of various secondary new phases at the expense of a

531 volcanic phenocryst, in an altered dacite. Sample from north of Grande Anse beach  
 532 (17TH16). Act = actinolite, Cal = calcite, Chl = chlorite, Czo = clinozoisite, Ep = epidote, Ms =  
 533 muscovite, Qtz = quartz, Pl = plagioclase, Px = pyroxene, Py = pyrite, Tlc = talc, Ttn =  
 534 titanite, Wmca = white mica. (COLOR / IMAGE SIZE: FULL PAGE WIDTH)  
 535



536

537 Figure 12: Examples of low-temperature (second hydrothermal stage, 220<T<300°C) mineral  
 538 crystallizations. A) Ferromagnesian phenocryst replaced by calcite, in an altered dacite.  
 539 Sample from north of Grande Anse beach (16STH11b). B) BSE image. Jarosite, on the edge  
 540 of a cavity, in an altered dacite from a pyroclastic flow. Sample from north of le Chameau  
 541 volcanic dome (17TH24). C) Jarosite on the edge of a cavity, in an altered dacite. Sample  
 542 from north of Grande Anse beach (16STH20). D) BSE image. Cristobalite and acicular  
 543 zeolite on the edge of a cavity, in an altered dacite from a pyroclastic formation. Sample from  
 544 north of le Chameau volcanic dome (16STH29). E) Jarosite and radial zeolite in matrix of an  
 545 altered dacite from pyroclastic flow. Sample from north of le Chameau volcanic dome  
 546 (16STH29). Cal = calcite, Chl = chlorite, Crs = cristobalite, Jar = jarosite, Ms = muscovite, Pl  
 547 = plagioclase, Py = pyrite, Zeo ac = acicular zeolite, Zeo rad = radial zeolite. (COLOR /  
 548 IMAGE SIZE: FULL PAGE WIDTH)  
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550 The most hydrothermalized rocks are located in the central part of the island. Here, the  
 551 primary volcanic textures have been completely erased, and in some cases, phantoms of  
 552 phenocryst shapes can be recognized. In most cases, the whole rock is recrystallized  
 553 (phenocrysts and matrix) into a fine-grained association of new phases including quartz-  
 554 epidote-white micas-chlorite-actinolite-titanite +/- talc.

555 In the central part of the island, a significant amount of supergene clay minerals, particularly  
556 illite-smectite partly altered in smectite, kaolinite and, to a lesser extent, halloysite have been  
557 recently recognized and described in detail (Beauchamps et al., 2019). In this area, gypsum  
558 and calcite are also frequently observed.

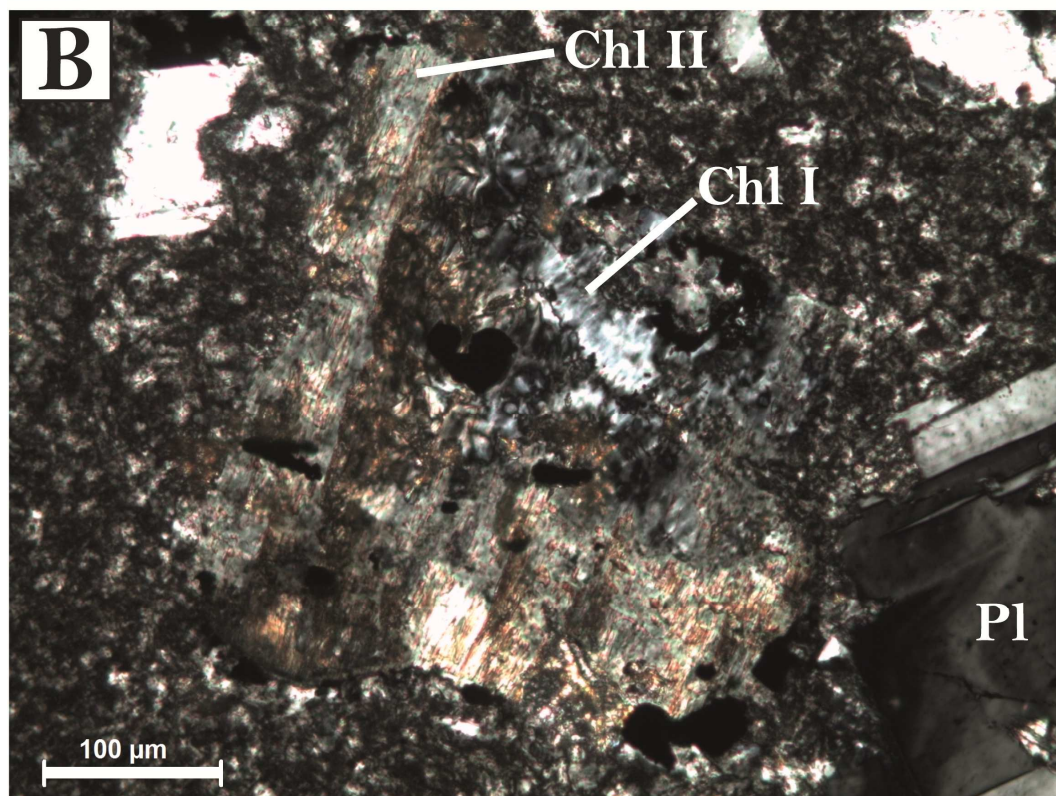
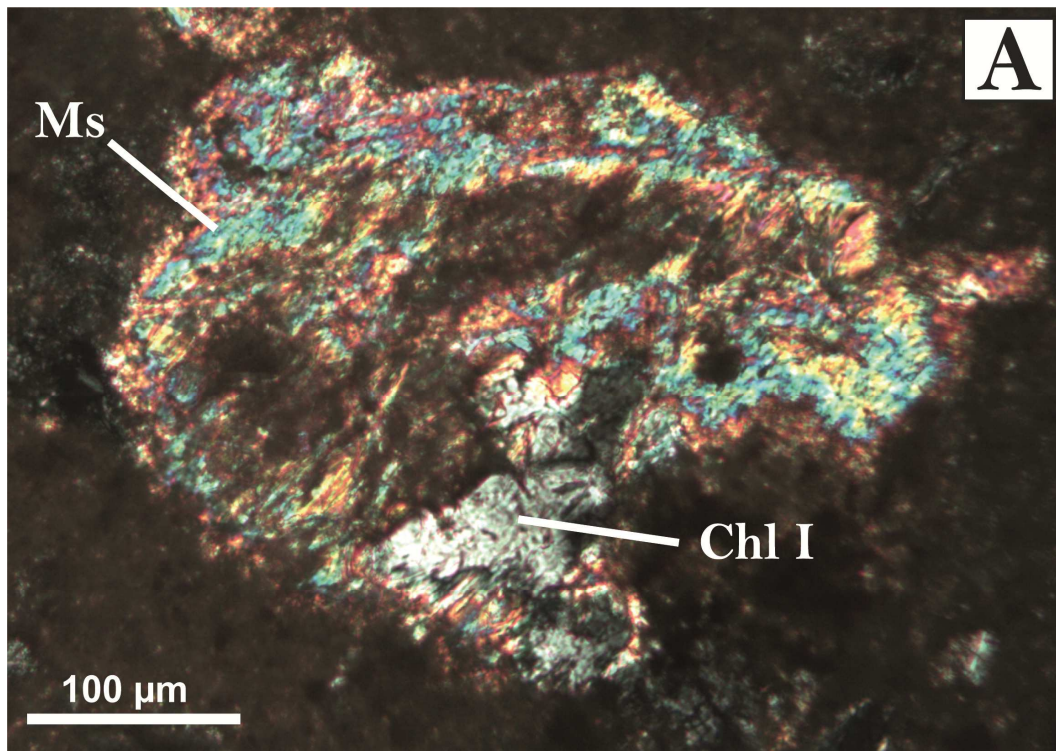
559 **6-4-The different stages of hydrothermalism - a synthesis**

560 Altogether, the microstructural overprinting criteria observed in the Terre-de-Haut  
561 hydrothermalized rocks attest to a polyphased hydrothermal alteration history with the  
562 following stages:

563 - A first stage with the mineral association of quartz + albite + epidote + muscovite + grey  
564 chlorite (Figures 13-A and B) + biotite + actinolite + titanite +/- talc and pyrite.

565 - A second stage with the crystallization of illite + green to yellow chlorite (Figure 13-B) +  
566 silica polymorphs + pyrophyllite + calcite + sulphates + zeolites.

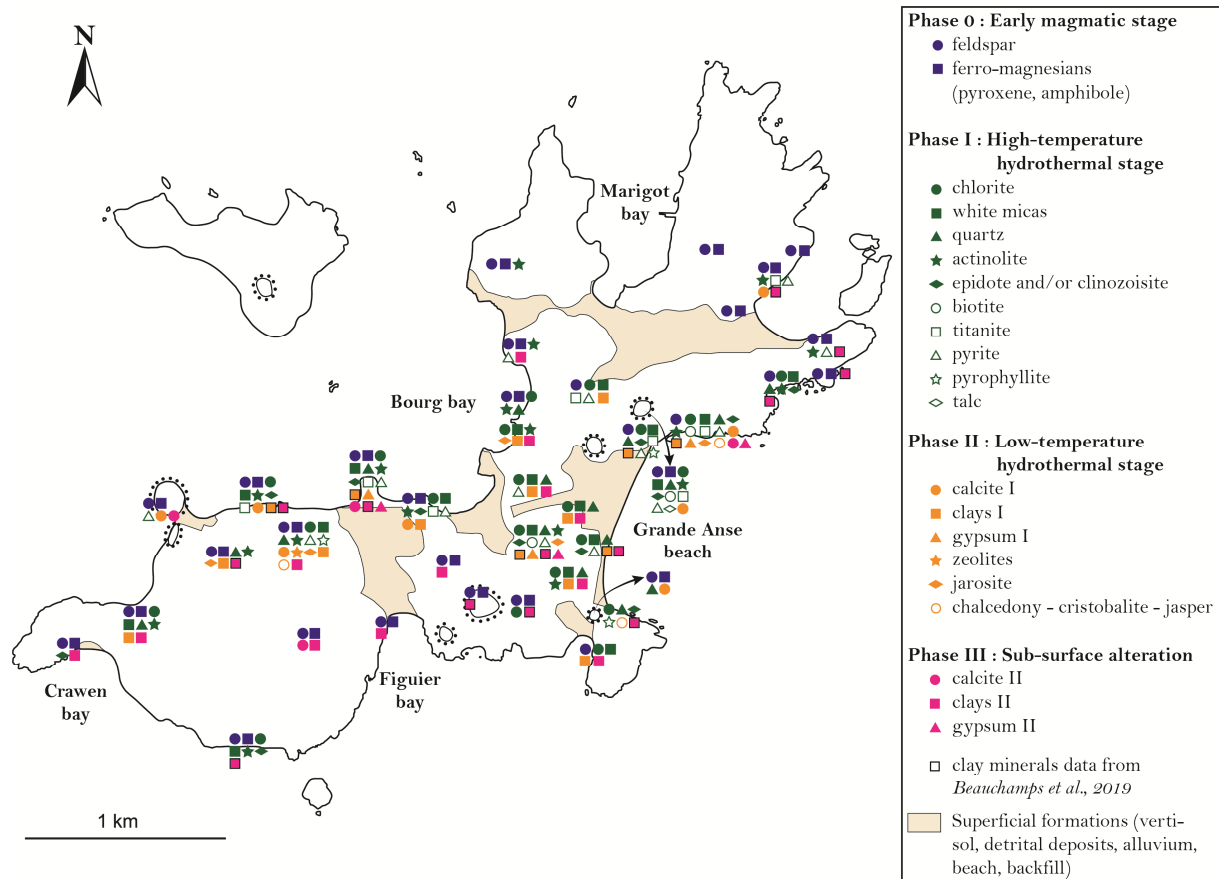
567 - A third stage with a supergene mineral association including illite-smectite or smectite +  
568 kaolinite + calcite + gypsum +/- halloysite.



569

570 Figure 13: Chlorite morphologies. A) Ferromagnesian phenocryst pseudomorphosed by  
 571 fibrous muscovite and grey automorphous crystals of chlorite (type I), in a transformed  
 572 rhyodacite. Sample from north of Grande Anse beach (16STH19). B) Ferromagnesian  
 573 phenocryst pseudomorphosed by grey automorphous crystals of chlorite (type I) and green-  
 574 grey to yellow fibrous grains (type II), in a transformed dacite. Sample from north of Grande  
 575 Anse beach (17TH16). (COLOR / IMAGE SIZE: COLUMN WIDTH)  
 576

577 To highlight the importance and spatial distribution of the previously described  
 578 transformations, we propose a map of mineral occurrences at the scale of Terre-de-Haut  
 579 Island (Figure 14), distinguishing the two hydrothermal stages and supergene alterations.



580

581 Figure 14: Distribution of different generations of mineral phases (volcanic, high and low  
 582 temperature hydrothermal and sub-surface alteration) within Terre-de-Haut Island. **(COLOR /**  
 583 **IMAGE SIZE: FULL PAGE WIDTH)**

584

585 **7- P-T-XH<sub>2</sub>O estimates for hydrothermalism**

586 **7-1- Geothermal thermometry**

587 The mineral association (Figure 14) representative of the first hydrothermal stage is  
 588 observed in multiple geothermal systems for temperatures above 300°C, while the  
 589 mineralogy developed during the second hydrothermal stage is typically described for  
 590 temperatures ranging between 300 and 220°C (Browne, 1978; Henley et al., 1986).

591 The late alteration stage reflects the progressive cooling history of the paleo-hydrothermal  
592 system with crystallization of a sequence of minerals commonly reported for temperatures  
593 below 200 °C (smectite and kaolinite, Ji and Brown, 2000; Lagat, 2014).

#### 594 **7-2- Chlorite geothermometry**

595 Cathelineau and Nieva (1985) and Cathelineau (1988) first proposed a geothermometer  
596 based on the chemical composition of chlorite. Later, more elaborated versions of this  
597 thermometer have been proposed that are particularly well-adapted for hydrothermalized  
598 rocks (Lanari et al., 2014; Bourdelle and Cathelineau, 2015). Following these recent  
599 investigations, for Si-poor and Al-rich chlorites, the more appropriate calibration is the one  
600 proposed by Lanari et al. (2014), while for Si-rich chlorites, the calibration proposed by  
601 Bourdelle and Cathelineau (2015) is better adapted. We thus selected a given calibration as  
602 a function of the measured chlorite chemistry.

603 Because chlorite is stable over a wide temperature range in geothermal systems (< 100°C to  
604 > 350°C, Henley and Ellis, 1983; Reyes, 1990; White and Hedenquist, 1995; Lagat, 2014), it  
605 is critical to distinguish the possible variations in chlorite compositions with respect to their  
606 various microstructural crystallization sites. Indeed, as presented in the previous section, we  
607 found the occurrence of chlorite in mineral associations representative of both the first and  
608 the second hydrothermal stages.

609 To quantify the past temperature range of the Terre-de-Haut hydrothermal system, we  
610 selected three samples from north of Grande Anse beach (altered rhyodacite 16STH19,  
611 altered dacite 17TH16, altered andesite SA-14) in a significantly hydrothermalized domain,  
612 with well-developed solution cleavage corridors. We also selected this area because it is a  
613 domain without evidence of supergene alteration (Figures 2-B and 14). Moreover, in these  
614 selected samples, chlorite developed in different microstructures as (1) grey automorphous  
615 crystals (Figure 13-A), associated with muscovite, quartz and epidote, developed at the  
616 expense of magmatic pyroxenes, and (2) green-grey to yellow fibrous grains (Figure 13-B),

617 associated with fibrous white micas, quartz, calcite, pyrite and/or talc, preferentially located at  
618 the rims of pseudomorphosed pyroxenes.

619 Interestingly, chlorite compositions evolve in relation to their microstructural position (see  
620 table 3), and even the type (I) grey automorphous grains display, in many cases, composition  
621 zoning from cores to rims. An increase in Si content and a decrease in Al content are  
622 depicted from type I automorphous to type II fibrous chlorites.

623

624 Table 3: Microprobe analyses and chlorite thermometry on three samples from north of Grande Anse beach: 16STH19 (altered rhyodacite),  
 625 17TH16 (altered dacite) and SA-14 (altered andesite). **(COLOR FOR ONLINE VERSION ONLY / IMAGE SIZE: FULL PAGE)**

Sample	16STH19							17TH16							SA-14					
Chlorite analysis	grain 1		grain 2		grain 3	grain 4	grain 5	grain 6			grain 7		grain 8	grain 9	grain 10	grain 11		grain 12	grain 13	
Chlorite type	I	I	I	I	I	I	I	I	II	II	I	I	II	II	II	II	II	II	II	
Structural position	core	rim	core	rim	core	core	core	rim	rim	rim	rim	rim	rim	rim	rim	core	rim	rim	rim	
Mineral %wt																				
Al <sub>2</sub> O <sub>3</sub>	20,57	20,30	20,08	20,22	20,18	20,11	20,27	17,15	17,12	16,27	16,98	16,77	16,05	15,40	17,04	16,95	15,83	15,01	14,99	
SiO <sub>2</sub>	27,32	27,01	25,66	25,51	25,45	25,69	25,53	31,02	30,94	31,60	29,71	29,69	31,37	32,50	31,94	30,16	32,15	31,33	32,98	
TiO <sub>2</sub>	0,01	0,06	0,00	0,00	0,01	0,01	0,02	0,01	0,04	0,04	0,16	0,02	0,01	0,02	0,02	0,06	0,05	0,01	0,03	
Na <sub>2</sub> O	0,00	0,00	0,19	0,00	0,05	0,04	0,02	0,03	0,02	0,04	0,02	0,00	0,02	0,05	0,05	0,11	0,11	0,08	0,14	
MgO	16,71	16,50	13,73	13,65	13,94	13,71	13,95	18,43	16,48	17,02	17,83	18,19	18,48	18,93	17,65	17,63	17,01	18,83	17,78	
MnO	0,29	0,33	0,37	0,23	0,30	0,34	0,26	1,82	1,57	1,44	1,75	2,15	1,37	1,56	1,83	0,69	0,75	0,73	0,66	
FeO	23,64	22,48	27,85	26,98	24,36	27,91	27,82	21,52	23,05	20,82	22,75	21,60	21,71	20,47	21,10	24,21	23,55	22,38	23,66	
K <sub>2</sub> O	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,01	0,02	0,08	0,02	0,01	0,02	0,02	0,02	0,04	0,04	0,01	0,05	
CaO	0,09	0,10	0,00	0,00	0,00	0,00	0,00	0,28	0,37	0,46	0,28	0,20	0,33	0,31	0,44	0,18	0,76	0,23	0,57	
<i>Total</i>	<i>88,63</i>	<i>86,78</i>	<i>87,88</i>	<i>86,59</i>	<i>84,29</i>	<i>87,81</i>	<i>87,87</i>	<i>90,28</i>	<i>89,60</i>	<i>87,77</i>	<i>89,50</i>	<i>88,64</i>	<i>89,36</i>	<i>89,26</i>	<i>90,09</i>	<i>90,03</i>	<i>90,26</i>	<i>88,62</i>	<i>90,87</i>	
Structural formulae																				
Al	2,50	2,50	2,53	2,57	2,60	2,54	2,55	2,02	2,05	1,97	2,04	2,03	1,91	1,82	2,01	2,03	1,88	1,81	1,76	
Al <sup>IV</sup>	1,19	1,17	1,26	1,25	1,22	1,26	1,28	0,76	0,68	0,54	0,88	0,89	0,70	0,55	0,61	0,87	0,57	0,69	0,55	
Al <sup>VI</sup>	1,31	1,33	1,27	1,32	1,38	1,28	1,27	1,26	1,37	1,43	1,16	1,14	1,21	1,27	1,40	1,16	1,31	1,12	1,21	
Si	2,81	2,83	2,74	2,75	2,78	2,74	2,72	3,11	3,14	3,24	3,03	3,05	3,17	3,27	3,19	3,06	3,24	3,20	3,29	
Ti	0,00	0,01	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,01	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	
Na	0,00	0,00	0,04	0,00	0,01	0,01	0,00	0,01	0,00	0,01	0,00	0,00	0,00	0,01	0,01	0,02	0,02	0,02	0,03	
Mg	2,56	2,57	2,19	2,19	2,27	2,18	2,22	2,75	2,50	2,60	2,71	2,79	2,79	2,84	2,63	2,67	2,55	2,87	2,65	
Mn	0,03	0,03	0,03	0,02	0,03	0,03	0,02	0,15	0,13	0,12	0,15	0,19	0,12	0,13	0,15	0,06	0,06	0,06	0,06	
Fe	2,04	1,97	2,49	2,43	2,23	2,49	2,48	1,80	1,96	1,78	1,94	1,86	1,84	1,72	1,76	2,06	1,98	1,91	1,98	
K	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,01	0,00	0,00	0,00	0,00	0,00	0,01	0,00	0,00	0,01	
Ca	0,01	0,01	0,00	0,00	0,00	0,00	0,00	0,03	0,04	0,05	0,03	0,02	0,04	0,03	0,05	0,02	0,08	0,02	0,06	
Total (cations)	9,94	9,92	10,02	9,97	9,92	9,99	10,00	9,88	9,83	9,79	9,93	9,93	9,87	9,82	9,81	9,93	9,83	9,91	9,83	
Ox. number	14	14	14	14	14	14	14	14	14	14	14	14	14	14	14	14	14	14	14	
Calculated temperature (°C)	328	289	375	360	318	376	379	168 +/- 18	149 +/- 14	123 +/- 23	215 +/- 35	210 +/- 30	173 +/- 28	133 +/- 18	138 +/- 23	230 +/- 20	135 +/- 15	180 +/- 20	139 +/- 16	
	After Lanari et al., 2014							After Bourdelle and Cathelineau, 2015												

626 The core compositions of type I chlorites are Si-poor; thus, we used the Lanari et al. (2014)  
627 calibration, which yields a temperature range of 320-380°C, with a calculated average  
628 temperature, taking into account the different samples, of 355°C (standard deviation=27). For  
629 the rims of type I chlorites a calculated temperature range is 180-350 °C, and the calculated  
630 average temperature, considering the different samples, is 252°C (standard deviation=57).

631 Type II fibrous chlorites also have higher Si contents; in this case, as previously underlined,  
632 the Bourdelle and Cathelineau (2015) calibration is the most adapted. With this calibration,  
633 type (II) chlorite formed between 125 and 230°C with a calculated average temperature,  
634 considering the different samples, of 140 °C (standard deviation=33).

635 Altogether, chlorite thermometers provide coherent estimations of the progressive cooling  
636 registered by the various generations of chlorites developed during the two hydrothermal  
637 stages. However, as frequently observed with the use of mineral chemistry-based  
638 geothermometry, the calculated temperature ranges are large, mainly because of possible  
639 continuous chemical exchanges between minerals during a given P-T path (Spear, 1993).  
640 We therefore used thermodynamic modelling to try to improve P-T estimates for Terre-de-  
641 Haut hydrothermalism.

### 642 **7-3- Thermodynamic modelling**

643 The intensive parameters (pressure and temperature) corresponding to the high-temperature  
644 hydrothermal stage were calculated with the free energy minimization program THERIAK-  
645 DOMINO (De Capitani and Petrakakis, 2010, updated software v. 4 February 2017) and the  
646 internally consistent thermodynamic “tcd55c2d” database (Holland and Powell, 2004).  
647 Mixing models used for solid solutions are presented in the appendix (table A-5). This  
648 approach is a powerful tool to quantify the P-T-XH<sub>2</sub>O conditions for hydrothermalism.  
649 Considering the goal of this paper, we used thermodynamic modelling instead of reactive  
650 transport modelling, the latter being more oriented on quantification of fluid-rock reaction for  
651 a specific surface area and thus calculation of reaction rates.

652 To be as close as possible to the equilibrium conditions at the scale of the whole rock, which  
 653 is essential for the application of this type of method, we selected a highly transformed  
 654 rhyodacite (16STH19) with a maximum of secondary phases expressed.

655 P-T pseudosections were calculated in the  $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-TiO}_2\text{-FeO-MnO-MgO-CaO-Na}_2\text{O-K}_2\text{O-}$   
 656  $\text{H}_2\text{O}$  system using the whole rock composition of the selected sample (table 4). The observed  
 657 mineral assemblage involves albite + chlorite + muscovite + biotite + clinozoisite + epidote  
 658 s.s. + quartz. Under tropical conditions, the iron oxidation state is always difficult to estimate.  
 659 To test the effect of  $\text{Fe}_2\text{O}_3$  in our models, we calculated pseudosections with various  
 660 amounts of  $\text{Fe}^{3+}$ . When the latter reaches 5%, we never find the mineral assemblage  
 661 observed in the natural sample. Consequently, we believe a model with total iron considered  
 662 FeO is the best approximation for pseudosection calculations.

663 Table 4: Whole-rock composition of 16STH19 sample and corresponding input in Theriak-  
 664 Domino program. (COLOR FOR ONLINE VERSION ONLY / IMAGE SIZE: COLUMN  
 665 WIDTH)  
 666

Sample 16STH19			
Composition (%)		Input Theriak-Domino (mol)	
<b>SiO<sub>2</sub></b>	59.555	<b>Si</b>	50.489
<b>Al<sub>2</sub>O<sub>3</sub></b>	16,070	<b>Al</b>	16.058
<b>Fe<sub>2</sub>O<sub>3</sub></b>	1.772	<b>Fe</b>	1.129
<b>MnO</b>	0.103	<b>Mn</b>	0.074
<b>MgO</b>	3.055	<b>Mg</b>	3.862
<b>CaO</b>	5.531	<b>Ca</b>	5.024
<b>Na<sub>2</sub>O</b>	1.691	<b>Na</b>	2.780
<b>K<sub>2</sub>O</b>	1.299	<b>K</b>	1.405
<b>TiO<sub>2</sub></b>	0.558	<b>Ti</b>	0.365
<b>P<sub>2</sub>O<sub>5</sub></b>	0.151	<b>P</b>	0.109
<b>LOI</b>	10.214	<b>H</b>	57.761
26.46 % <i>CO<sub>2</sub>total</i>	2.703	<b>O</b>	?
58.97 % <i>H<sub>2</sub>Ototal</i>	6.023		
7.04 % <i>FeO</i>	0.719		
7.53 % <i>S<sub>total</sub></i>	0.769		

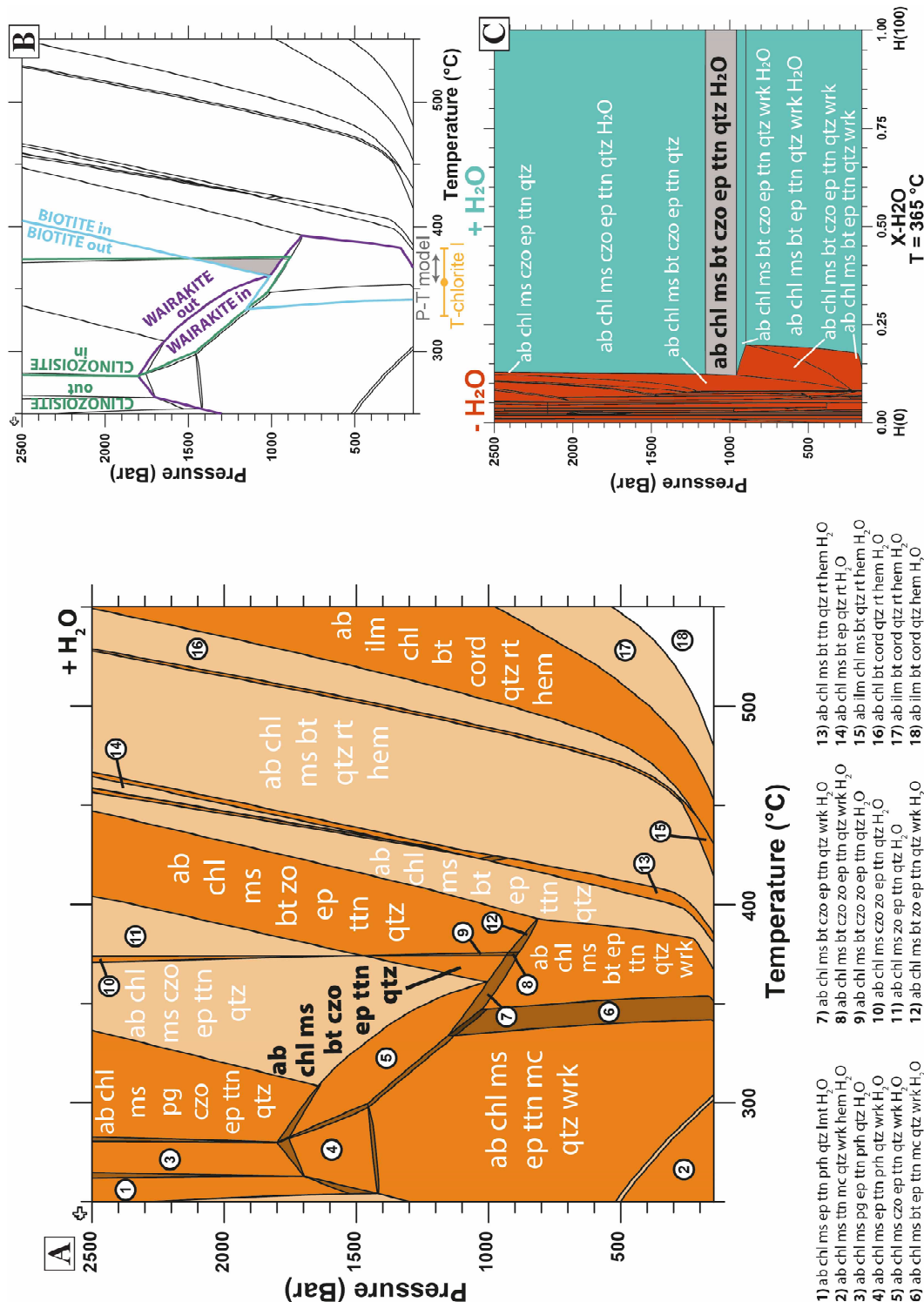
667

668 We considered the fluid phase in excess to be consistent with the conditions of a fluid-  
669 saturated hydrothermal system. However, in high-temperature geothermal systems, fluids  
670 have different origins (meteoric waters with or without sea-water components, volcanic fluids  
671 and gases with variable amounts of CO<sub>2</sub>, H<sub>2</sub>S or SO<sub>2</sub>); thus, it seems critical to test the effect  
672 of fluid composition on the modelled system. In the first approach, a pseudosection was  
673 calculated with 100% H<sub>2</sub>O for the fluid composition. In this case, the observed mineral  
674 assemblage corresponds to the calculated field (360-375°C and 0.90-1.40 kbar) identified  
675 with bold-black characters in figure 15-A and is in agreement with the highest temperature  
676 values obtained by chlorite geothermometry on type-1 chlorite cores from this sample  
677 (16STH19, see figure 15-B and table 4). This stability field is indeed limited by the  
678 appearance of wairakite and the disappearance of clinozoisite towards low temperatures and  
679 pressures. A temperature of approximately 365°C is an estimation consistent with both  
680 methods. In a second test, a specific P-XH<sub>2</sub>O pseudosection (with XH<sub>2</sub>O representing the  
681 variability of amount of water, in moles, in the considered system) was calculated to decipher  
682 the effect of the amount of H<sub>2</sub>O in the fluid present in excess on the pressure estimates  
683 (Figure 15-C). The observed mineral assemblage is stable for a large range of XH<sub>2</sub>O values,  
684 i.e., between 12% and 100%. The chemical components of the LOI (see table 3) allows an  
685 estimation of the components in the fluid phase with respect to the bulk-rock chemistry of the  
686 analysed sample. In the 16STH19 sample, the amount of H<sub>2</sub>O in the fluid phase is estimated  
687 at approximately 59%, with a CO<sub>2</sub> content of approximately 27%. The result of our  
688 thermodynamic modelling is completely consistent with this estimation and indicates that  
689 regardless of the XH<sub>2</sub>O value, as long as is greater than 12%, the observed mineral  
690 assemblage remains stable. Consequently, in the studied sample, for the XH<sub>2</sub>O values of  
691 interest and for a temperature of approximately 365°C, the modelled pressure range is  
692 restricted to between 0.95 and 1.15 kbar.

693 The results obtained from various hydrothermal and/or geothermal fields demonstrate that an  
694 increase in the CO<sub>2</sub> concentration in the hydrothermal fluid would have displaced the

695 equilibrium into the calcite-stable field at the expense of epidote and/or wairakite stability  
696 (Browne and Ellis, 1970; Hedenquist, 1986; Hedenquist and Browne, 1989; Browne, 2003).  
697 In our model, the addition of CO<sub>2</sub>, which is present in our system as previously underlined,  
698 shifts the wairakite stability towards lower temperatures, on the order of approximately fifteen  
699 degrees, and pressures, on the order of approximately 0.05 kbar. Therefore, the best P-T  
700 estimate for the highest temperature stage of hydrothermalism corresponds to a temperature  
701 of approximately 350°C for a pressure between 0.9 and 1.1 kbar.

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Figure 15: A) Calculated P-T pseudosection for a rhyodacite sample (16STH19) with H<sub>2</sub>O saturation and total iron as Fe<sup>2+</sup> with Theriak-Domino program (De Capitani and Petrakakis, 2010, updated software v. 4 February 2017) and "tcd55c2d" database (Holland and Powell, 2004). Rock composition is in mol.% oxide. Fields are coloured with according to their variances, darker colours indicating higher-variance assemblages. The stability field written in bold correspond to the observed assemblage. B) Simplified representation of the P-T pseudosection with the stability field of the observed mineral association in grey. C) P-X(H<sub>2</sub>O) equilibrium phase diagram calculated at 365°C, with similar composition in Figure 15-A, and modelled between 0 (H = 0) and 100 moles (H = 100). The field coloured in grey corresponds to the observed assemblage. Mineral abbreviations are from Kretz, 1983. (COLOR / IMAGE SIZE: FULL PAGE WIDTH)

## 715 **8- Discussion**

### 716 **8-1- Relationship between deformation and hydrothermalism: implications for fluid** 717 **transfer pathways**

718 In a general cross-section of Terre-de-Haut Island, the main altered zone is localized within a  
719 WNW-ESE graben (Figure 16-A). However, we discovered the occurrence of ductile tectonic  
720 structures, precisely pressure solution cleavages concentrated in specific shear zones,  
721 clearly intersected, and thus overprinted, by numerous brittle faults (Figure 16-A). Moreover,  
722 we established that the crystallization of the high-temperature hydrothermal phases (i.e., the  
723 first stage of hydrothermal alteration) is contemporaneous with the development of ductile  
724 tectonic structures. At the regional scale, this is highlighted by the comparison between the  
725 structural and mineralogical maps (Figures 7 and 14). Our microstructural observations  
726 indicate that a stress-induced pressure solution process is the mechanism leading the  
727 development of ductile tectonic structures. Numerous studies have shown that pressure  
728 solution creep involves grain dissolution in the zones of highest stress, mass transfer of the  
729 most soluble chemical elements away from dissolution sites in a free-fluid phase, and  
730 deposition of these elements in the zones of smallest stress (veins, micro-cracks, pores, etc.;  
731 Sorby, 1863; Ramsay, 1967; Rutter, 1972, 1976; Siddans, 1972; Elliot, 1973; Kerrich, 1977;  
732 Beach, 1979). This implies fluid circulation in the open spaces (pores and cleavages) of the  
733 rocks located in the ductile corridors. Similar structural pathways for fluid transfers are well  
734 documented in various cases of actively deforming ductile crust (Mc Caig, 1988; Oliver,  
735 1996; Famin et al., 2004; Mulch et al., 2006).

736 In the study area, fluids necessarily react with volcanic minerals to produce the observed  
737 high-temperature secondary phases. Indeed, to be produced, the observed microstructure  
738 requires:

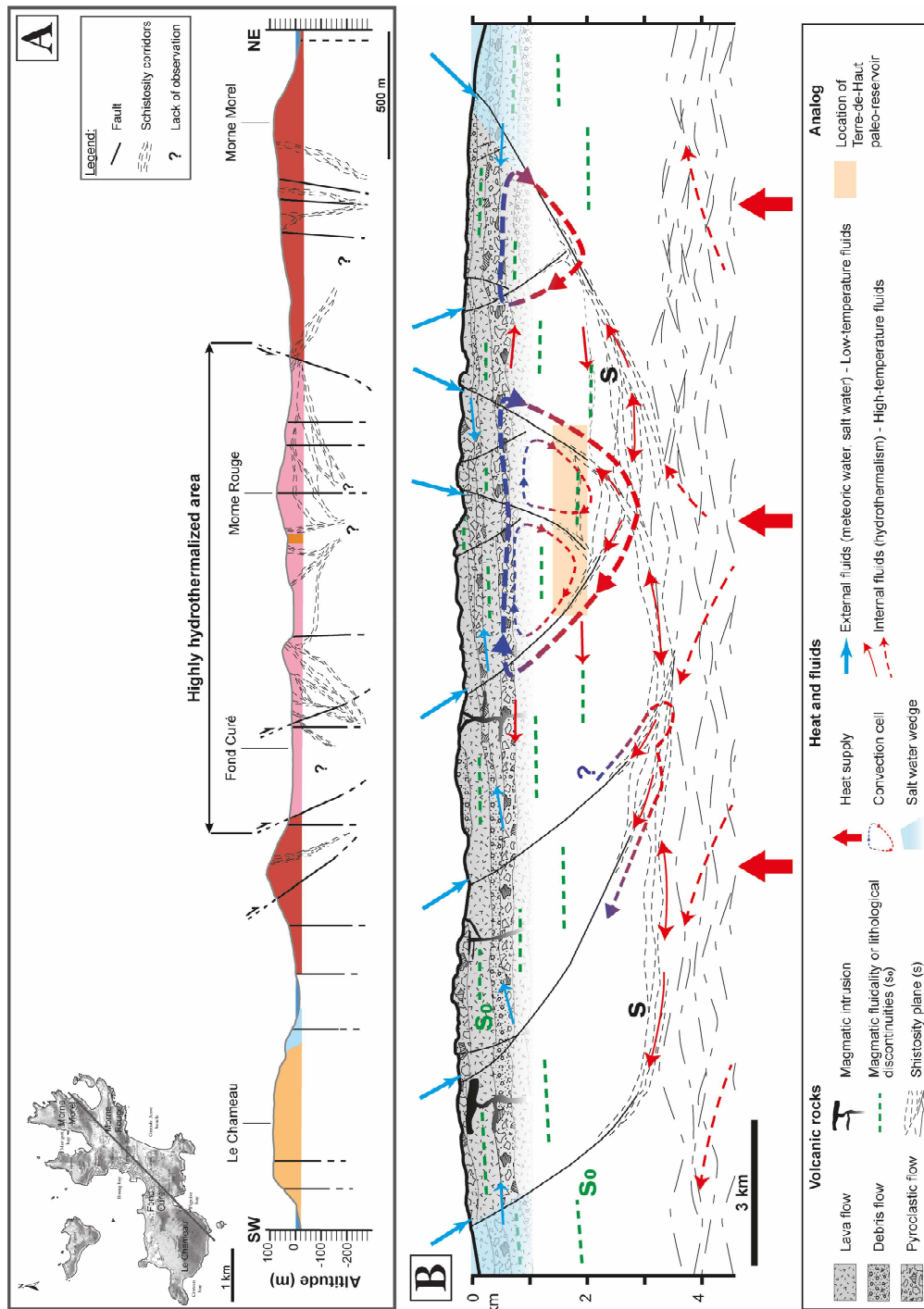
739 (1) H<sub>2</sub>O-rich fluid infiltration in the volcanic rock, stress-induced dissolution of anorthoclase  
740 microlites and, to a lesser extent, some phenocryst rims, leaving space for concentration of  
741 insoluble minerals;

742 (2) transfer of at least K, Si, Al and Na in the free fluid film in the pore space of the rock;  
743 (3) reaction of pyroxenes with a part of this fluid phase trapped at phenocryst contacts and  
744 crystallization of micas, chlorite, albite and quartz at the expense of pyroxenes;  
745 (4) expulsion of the fluid phase in excess through pores and/or micro-cracks and veins.

746 In such a case, the ductile corridors are not sites of fluid production but only pathways for  
747 fluid transfer. However, progressive mineral crystallization tends to close spaces, and a given  
748 shear zone can be sealed by secondary phases (Navelot et al., 2018). Because the whole  
749 process implies different steps, i.e., dissolution, transfer and crystallization, if the rate of a  
750 given step is slower than the others, it will control the kinetics of the whole process and thus  
751 control the fluid circulation. Therefore, a structure being a fluid pathway at a given increment  
752 of hydrothermal system development can act as a barrier to fluid transfer later. In this case,  
753 there must be relays between the transfer zones to maintain the activity of the hydrothermal  
754 system. In dealing with this issue, the geometry of the finite strain pattern is a crucial  
755 element. In the area of interest, ductile shear zones with different dips tend to anastomose  
756 (sector 1 in Figure 7-A, east of Fond Curé in Figure 16-A), and in other cases (sector 2 in  
757 Figure 7-A, west of Morne Rouge in Figure 16-A), subvertical ductile corridors clearly  
758 intersect other moderately dipping zones. If connected, the geometric relays between  
759 anastomosed shear zones with contrasted dips are particularly favourable to both vertical  
760 and lateral fluid transfers, while the intersections between superposed ductile corridors allow  
761 the persistence of fluid transfer through time to be envisaged.

762

763



764 Figure 16: A) NE-SW cross-section from Terre-de-Haut Island displaying the superposition of  
 765 brittle faults upon the network of schistosity planes. The highly hydrothermalized area is  
 766 located in the central part of the cross-section within a WNW-ESE graben. The colours are  
 767 similar to the ones of the geological map in figure 2-B (localisation of the cross-section). B)  
 768 Conceptual model of the geothermal reservoir in the Guadeloupe magmatic island arc. This  
 769 north-south cross section displays the possible plumbing system of the geothermal reservoir,  
 770 from the surface to the deeper parts, with the different fluid flow contributions. The  
 771 brittle/ductile transition is represented as a sub-horizontal horizon acting as a crustal  
 772 scale shear zone. The geometry of faults and schistose zones pattern is purely indicative. The  
 773 geological formations are only illustrated schematically over the first kilometre of the  
 774 cross-section. (COLOR / IMAGE SIZE: FULL PAGE WIDTH)

775 During the exhumation and thus the thermal evolution (i.e., cooling) of the paleo-  
776 hydrothermal system, low-temperature hydrothermal phases continue to crystallize in  
777 fractures, veins and brittle faults, as well as in some shear zones. Indeed, if some veins are  
778 contemporaneous with the solution cleavage formation and indicate incremental extension  
779 parallel to the planar structures, late veins and fractures are related to a second and  
780 independent deformation episode under brittle conditions. This recent to active brittle  
781 extension is well documented in the area of interest (Feuillet et al., 2001, 2002, 2004; Bazin  
782 et al., 2010; Leclerc et al., 2016; Verati et al., 2016; Navelot et al., 2018). These late tectonic  
783 structures contain, or are underlined by, supergene clay minerals (illite, smectite) or zeolites.  
784 This means that during the cooling history of the hydrothermal system, tectonic structures  
785 continue to be preferential drains, allowing the circulation of fluids. These observations are  
786 consistent with different works indicating that in tectonically active regions, such as the  
787 Guadeloupe archipelago, earthquake faulting is possibly associated with seismic pumping, a  
788 process that can enhance channelized fluid flow and thus fluid migration from production  
789 zones towards open spaces, i.e., dilatation zones (Sibson et al., 1975; Garven and  
790 Ruffensperger, 1997; Famin et al., 2005).

## 791 **8-2- The paleo-depth of the Terre-de-Haut exhumed hydrothermal system**

792 Pressure is a crucial parameter that must be quantified to constrain the dynamics of  
793 geological systems. Thermodynamic laws applied to mineralogy are powerful tools to  
794 address this question, and pressure quantification is constrained by the stability fields of  
795 specific mineral assemblages (Spear, 1993; Bucher and Frey, 2002, with references therein).  
796 In such a case, and by definition, the calculated pressure, often called “thermodynamic  
797 pressure” (Kondepudi and Pigogine, 1998), is an intensive parameter independent of the  
798 physical parameters that control the “effective pressure” applied to a given rock (Fermi, 1936;  
799 Korzhinskii, 1965). In crustal rocks, within which both solid and fluid phases are present, both  
800 hydrostatic and lithostatic pressure must be considered physical components of the effective  
801 pressure (Jaeger, 1969; Fyfe et al., 1978; Jaeger and Cook, 1979; Yardley, 2009; Moulas et

802 al., 2019). In the uppermost part of the crust (first few kilometres), as observed in boreholes  
803 located worldwide, there are enough fractures, veins and open connected pores that the free  
804 fluid phase within these voids is under hydrostatic pressure. With increasing depth (between  
805 2 and 3 km), the interconnected networks of pores and fractures tend to close, and the pore  
806 pressure increases and tends to approach lithostatic pressure (Coyle and Zoback, 1988;  
807 Ehrenberg, 1993; Buller et al., 2005; Nadeau and Ehrenberg, 2006; Taylor et al., 2010;  
808 Nadeau, 2011; Sachau et al., 2015).

809 In shallow crustal hydrothermal systems, as in geothermal systems, both hydrostatic and  
810 lithostatic pressures are physical parameters that control the effective pressure (Nor and  
811 Walder, 1992; van Ruth and Hillis, 2000; Rowland and Simmons, 2012; Sachau et al., 2015;  
812 Chambefort et al., 2017). Pore pressure acts against minerals in the opposite sense of  
813 lithostatic pressure, and consequently, the effective pressure is lithostatic pressure reduced  
814 by hydrostatic pressure (Batchelor, 1967),

815 To restore the paleodepth of the Terre-de-Haut paleohydrothermal system, we have to  
816 consider a lithostatic gradient calculated with a mean dry bulk density adapted to the  
817 lithology of a volcanic arc (basaltic andesites, andesites, dacites and rhyodacites) that is  
818 approximately  $2.6 \text{ kg/m}^3 \cdot 10^{-3}$  (Klein and Johnson, 1983). Thus, the lithostatic gradient  
819 increases by 26 MPa/km. Because, in volcanic arcs, and particularly in the case of the  
820 Guadeloupe arc, fluids in hydrothermal and geothermal systems are dominated by H<sub>2</sub>O, the  
821 hydrostatic gradient must be approximately 10 MPa/km. Consequently, in the uppermost part  
822 of the arc crust,  $P_{\text{fluid}} \approx 1/2.6 P_{\text{lithostatic}}$  and effective pressures in the case of interest range  
823 between 0.55 and 0.68 kbar. This was the case for the Terre-de Haut paleo-reservoir 3 or 2  
824 Ma ago (i.e., during island volcanic activity, Zami et al., 2014), at depth between 1.4 and 1.8  
825 km. The timescale for exhumation of this paleo-reservoir is therefore consistent with erosion  
826 rates proposed for the Guadeloupe archipelago (Sak et al., 2010; Lloret et al., 2011; Rad et  
827 al., 2013; Ricci et al., 2015a, b).

828 **8-3- A conceptual scheme for high-energy geothermal systems in the Guadeloupe**  
829 **archipelago**

830 **8-3-1- The uppermost boundary of the conceptual scheme**

831 To constrain the uppermost part of our scheme (Figure 16-B), we have a large number of  
832 published surface data, some of which was obtained in the active geothermal system of  
833 Bouillante and some was obtained in the last five years on Terre-de-Haut Island.

834 We want to stress that an important result of the last two decades of tectonic and  
835 geophysical research is the fact that the tectonic pattern of the whole Guadeloupe  
836 archipelago results, since the last 5 Ma, from arc-parallel extension combined with  
837 transcurrent faults (i.e., transtensional regime, Julien and Bonneton, 1989; Feuillet et al.,  
838 2002, 2004, 2010; Mathieu et al., 2011; Laigle et al., 2013; Münch et al., 2013; De Min et al.,  
839 2015; Leclerc et al., 2016). In this tectonic framework, the Bouillante active geothermal  
840 system is located in a “horsts and grabens” system bounded by E-W oriented normal faults  
841 associated with a N160 oriented transcurrent fault (Bouchot et al., 2010, 2014; Thinon et al.,  
842 2010; Calcagno et al., 2012, and Figure 1-C). Numerous investigations have established that  
843 (1) brittle faults contribute to fluid flow through the recharge of the system in supergene fluids  
844 (dominant meteoric water and minor seawater, Bouchot et al., 2010; Millot et al., 2010;  
845 Sanjuan et al., 2010), (2) vertical clay mineral zonation, typical of hydrothermally altered  
846 systems and utilized in geothermal fields worldwide, is well developed along the available  
847 boreholes (Mas et al., 2006) and (3) temperatures in the range of 230-255°C are measured  
848 at depths between 475 m and 1.2 km (Guillou-Frottier, 2003; Bouchot et al., 2010; Mas et al.,  
849 2006; Sanjuan et al., 2001, 2004).

850 In our previous works on Terre-de-Haut Island, we provide datasets on brittle fault geometry  
851 and kinematics as well as on thermo-physical properties of hydrothermalized rocks (Verati et  
852 al., 2016; Navelot, 2018; Navelot et al., 2018; Favier, 2019). We observed, here also, that  
853 fault intersections are suitable for fluid transfers, particularly when the faults are connected to

854 fractured lavas or cooling joints in lavas. In this case, the intersections between highly  
855 dipping faults or fractures and sub-horizontal volcanic structures are favourable geometries  
856 for both vertical and lateral fluid transfers.

### 857 **8-3-2- The lowermost boundary of the conceptual scheme**

858 In the Guadeloupe archipelago, a recent study (Manga et al., 2012) revealed a steady-state  
859 conductive gradient between  $69.3 \pm 1.5$  and  $98.2 \pm 8.8$  °C/km. As demonstrated by  
860 mineralogical and petrological investigations on the oldest, and thus most eroded, volcanic  
861 units of the considered archipelago, this conductive gradient did not change during the last 5  
862 Ma (Verati et al., 2018; Favier, 2019; Favier et al., 2019). Similar thermal conditions are well  
863 known and generalized in numerous examples of recent and active as well as old and  
864 exhumed magmatic arcs located on the upper plates of subduction zones (Miyashiro, 1961;  
865 McKenzie and Sclater, 1968; Hasebe et al., 1970; Oxburgh and Turcotte, 1970; Watanabe et  
866 al., 1970; Ernst, 1971; Sclater, 1972; Furukawa and Uyeda, 1989; Ribe, 1989; Cloos, 1993;  
867 Furukawa et al., 1998; Fukahata and Matsu'ura, 2000; England and Wilkins, 2004; Ranalli  
868 and Rybach, 2005). In the area of interest, temperatures close to or higher than 300°C are  
869 thus reached at depths between 4 and 5 km, and consequently, since 5 Ma, the brittle-ductile  
870 transition is necessarily localized at that depth in the arc crust. This shallow depth rheological  
871 horizon constitutes the lowermost boundary of our conceptual scheme (Figure 16-B), and its  
872 position is consistent with the thermomechanical conditions recognized in subduction zones'  
873 upper plates, for example, in the Mediterranean region, in the Tuscany magmatic province in  
874 Italy, where the Larderello geothermal field is located (Batini et al., 1983; Cameli et al., 1998;  
875 Liotta and Ranalli, 1999; Bellani et al., 2004; Bertini et al., 2006; Liotta and Brogi, 2020), or in  
876 Anatolia (Menderes Massif), which is also recognized as an active geothermal domain even  
877 in a different arc setting (Aydin et al., 2005 ; Erkan, 2015; Roche et al., 2018). In the  
878 thermomechanical framework of a magmatic arc, it is thus necessary to consider that the  
879 steeply dipping brittle faults, well developed in the uppermost part of the arc crust, will

880 become more horizontal and will progressively root on the frictional-viscous boundary (Hobbs  
881 et al., 1976; Jaeger et al., 2009; Nicolas and Poirier, 1976; Kirby, 1983).

882 **8-3-3- Terre-de-Haut hydrothermal system in the conceptual scheme: an analogue of**  
883 **geothermal systems' roots in magmatic arcs**

884 The paleo-depth of the Terre-de-Haut exhumed hydrothermal system was estimated to be  
885 between 1.4 and 1.8 km. At these depths, the occurrence of hydrothermal mineral  
886 assemblages that are stable under a temperature of approximately 350 °C suggests a  
887 significant thermal anomaly with respect to the regional standard geothermal gradient. This  
888 paleotemperature is even higher than the temperatures proposed for the active Bouillante  
889 field (Mas et al., 2006) but is compatible with downhole temperature ranges in different  
890 geothermal fields around the world (Browne, 1978; Moro et al., 1982; Bertini et al., 1991;  
891 Battaglia et al., 1991; White and Hedenquist, 1995; Bogie et al., 2008; Bouchot and Genter,  
892 2009; Bertani et al., 2018; Cherkose and Mizunaga, 2018). Furthermore, in the area of  
893 intensely altered rocks located in the WNW-ESE graben, the primary volcanic mineral  
894 assemblages are frequently replaced beyond recognition. Indeed, the anhydrous minerals of  
895 the volcanic rocks (mainly pyroxenes, plagioclases and anorthoclases) are replaced by  
896 hydrated phases (white micas, chlorites, biotites, pyrophyllites, etc.) until the formation of  
897 clay minerals at the end of the alteration process. Such a mineralogical evolution requires  
898 significant H<sub>2</sub>O-rich fluid infiltrations in the volcanic rocks during the progressive exhumation,  
899 and thus cooling, of the paleo-hydrothermal system. Additionally, high-temperature  
900 hydrothermal fluid circulation was effective enough during the first stages of the alteration  
901 history to frequently erase the primary volcanic structures and to allow tectonic structures  
902 (i.e., ductile shear zones), driven by stress-induced pressure solution processes, to be  
903 developed.

904 When taken together, these observations lead us to consider that the Terre-de-Haut  
905 hydrothermal system represents a piece of arc crust that was located, 2 or 3 Ma ago, at a  
906 depth of 1.5-2 km, and submitted to a significant thermal anomaly and to important fluid

907 percolations. This system offers the opportunity to characterize the transfer structure of  
908 hydrothermal fluids in a position equivalent to the root of the geothermal systems in the  
909 Lesser Antilles magmatic arc. In this area of interest, we discovered the occurrence of ductile  
910 shear zones generated by pressure solution processes coeval with the development of high-  
911 temperature hydrothermal mineral associations at the expense of primary volcanic phases.  
912 This demonstrates that, at shallow crustal depths under fluids' present conditions, pressure  
913 solution creep interacts and is in competition with brittle deformation, as established in the  
914 last forty years by natural observations and experimental or theoretical modelling (Rutter,  
915 1983; Renard et al., 2000; Dysthe et al., 2002, 2003; Sibson and Rowland, 2003; Gratier et  
916 al., 2009, 2013). Therefore, stress-driven fluid and mass transfer creep laws accommodate  
917 ductile shear zones development in the upper arc crust. In Terre de Haut Island, the  
918 observed shear zones must be considered pathways for fluid infiltration, and because they  
919 have variable dips, they constitute a structure favourable for both vertical and lateral fluid  
920 transfers. They also represent the geometric connection between the near-surface brittle  
921 faults and the brittle-ductile, sub-horizontal, transition horizon.

922 In the proposed conceptual scheme of a magmatic arc evolving under an extensional  
923 tectonic regime, the abnormal heat flow is the result of arc-related magmatism, which  
924 transfers differentiated magmatic melts, heat and high temperature fluids towards the upper  
925 crustal levels. In this context of the supra-subduction zone, magmatic sources can be  
926 envisaged for volatile components. These deep, hot fluids can be channelized and/or trapped  
927 in the brittle-ductile transition horizon and expelled in the more permeable sectors of this  
928 horizon.

929 In the uppermost part of our scheme (Figure 16-B), as demonstrated by numerous  
930 investigations in the active Bouillante geothermal system, the downward flow of supergene  
931 cold fluids is efficient along conjugated normal faults (Figure 1-C), which act as preferential  
932 strongly dipping pathways (Sanjuan and Brach, 1997; Guillou-Frottier, 2003; Bouchot and  
933 Genter, 2009; Lachassagne et al., 2009; Bouchot et al., 2010; Thinon et al., 2010; Calcagno

934 et al., 2012). Lithological contacts and volcanic structures can also account for lateral  
935 movements of fluids (Navelot et al., 2018). Between the first kilometre of arc crust and the  
936 brittle-ductile transition horizon, the extensional shear zones observed in the Terre-de-Haut  
937 analogue act as transfer zones where circulation of geothermal fluids is possible. The  
938 geometry of the finite shear zone pattern suggests that geothermal fluids can be vertically or  
939 laterally channelled and/or stored in tectonic structures.

940 Finally, in the proposed conceptual scheme (Figure 16-B), the shear zones, driven by stress-  
941 induced pressure solution processes, may have an important enhancing role in establishing  
942 an advective regime in the whole geothermal system.

943

944 Addressing deep geothermal exploration, our investigations suggest that in magmatic arcs:

945 - Ductile tectonic structures are efficient pathways for geothermal fluid flows, as already  
946 proposed by other authors (Baldi et al., 1995; Liotta and Ranalli, 1999; Brogi et al., 2003,  
947 2005; Bellani et al., 2004; Kaya, 2015; Roche et al., 2018; Liotta and Brogi, 2020),

948 - Tectonic and volcanic structures contribute to fluid flows and could be efficient drains for  
949 both vertical and lateral fluid circulations and/or storage,

950 - The brittle-ductile transition horizon, located at shallow depths, could act as a permeable  
951 zone where the transfer of deep geothermal fluids is facilitated. It may potentially be  
952 considered a deep-seated reservoir hosting an important geothermal resource.

953

## 954 **9- Conclusions**

955 We document and describe, for the first time in Terre-de-Haut Island (Les Saintes volcanic  
956 archipelago), the occurrence of ductile deformation, i.e., shear zones generated by pressure  
957 solution processes coeval with the development of high-temperature hydrothermal mineral  
958 associations.

959 (1) Using conventional thermometry and thermodynamic modelling, we demonstrate that  
960 the hydrothermal system of Terre-de-Haut Island developed at a temperature of  
961 approximately 350°C and at depths in the range of 1.4 – 1.8 kilometres.

962 (2) The link between mineral transformations and ductile deformation suggests first that  
963 shear zones are efficient pathways for geothermal fluid flows and second that both  
964 vertical and lateral fluid transfers are possible in the crust of the Lesser Antilles  
965 volcanic arc.

966 (3) Together with preceding available structural, mineralogical and petrophysical data,  
967 this study clearly establishes that the highly hydrothermalized area of Terre-de-Haut  
968 Island corresponds to an eroded and exhumed geothermal paleo-reservoir. It is thus  
969 a key analogue for understanding the roots of current active geothermal systems in  
970 supra-subduction volcanic arcs.

971

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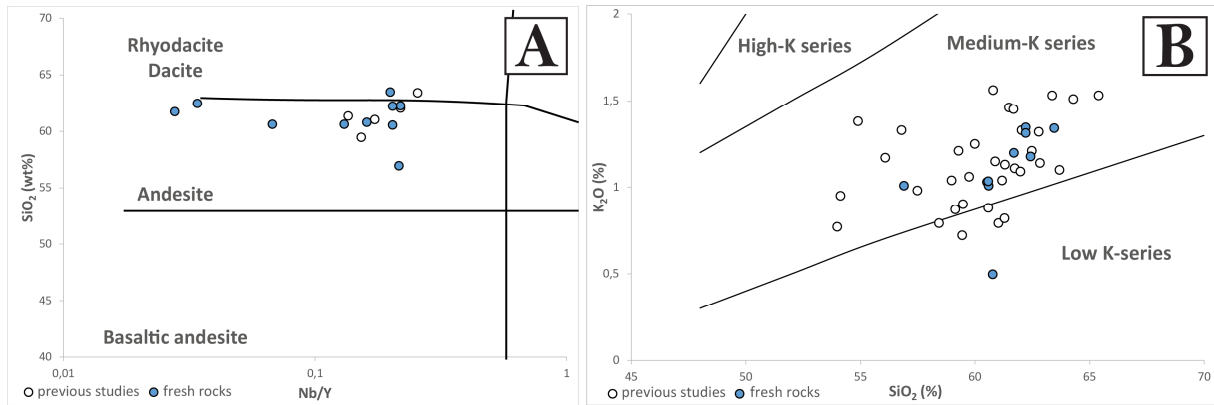
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981

982

983 **11- Appendices**



984

985 Figure A-1: A) SiO<sub>2</sub> vs. Nb/Y diagram after Winchester and Floyd 1977, white circles  
986 represent previous data from Jacques and Maury, 1988; Zami et al., 2014 and Navelot et al.,  
987 2018. Blue circles correspond to our new analyses. B) SiO<sub>2</sub> vs. K<sub>2</sub>O plot after Peccerillo and  
988 Taylor (1976), white circles represent previous data from Jacques et al., 1984; Jacques and  
989 Maury, 1988; Zami et al., 2014 and Navelot et al., 2018. Blue circles correspond to our new  
990 analyses.  
991

992 Table A-1: Table of whole rock major and trace element analyses. Total major element  
993 analyses not recalculated to 100%.  
994  
995 *Excel file*

996 Table A-2: Electron microprobe analyses and structural formula of volcanic phases  
997  
998 *Excel file*

999 Table A-3: Electron microprobe analyses of low-temperature hydrothermal phases  
1000  
1001 *Excel file*

1002 Table A-4: Electron microprobe analyses and structural formula of high-temperature  
1003 hydrothermal phases  
1004  
1005 *Excel file*

1006 Table A-5: Table of mixing models solid solution used in tcd55c2d database of Theriak-  
1007 Domino software  
1008  
1009 *Excel file*

1010

1011

1012 **12- References**

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