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Spatial Variations in Vent Chemistry at the Lucky Strike Hydrothermal Field, Mid-Atlantic Ridge (37°N): Updates for Subseafloor Flow Geometry From the Newly Discovered Capelinhos Vent

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1 **Spatial Variations in Vent Chemistry at the Lucky Strike Hydrothermal Field, Mid**
2 **Atlantic Ridge (37°N): Updates for Subseafloor Flow Geometry from the Newly**
3 **Discovered Capelinhos Vent.**

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10 **Key points:**

- 11 • New hydrothermal vent named Capelinhos found at Lucky Strike hydrothermal vent
- 12 • New model of hydrothermal fluid circulation at Lucky Strike hydrothermal field
- 13 • Up to 65% of dissolved hydrothermal Fe stored along the hydrothermal up flow zone

14 **Abstract**

15 This study aims at characterizing the seafloor architecture of the Lucky Strike hydrothermal
16 field (LSHF) based on an extensive chemical database of the various vents. Our analysis is
17 motivated by the discovery in 2013 of a new active high-temperature site, named Capelinhos,
18 approximately 1.5 km east of the LSHF. Capelinhos fluids display particular chemical features
19 with chloride and metals (Fe, Mn) concentrations two times lower and four times higher,
20 respectively, compared to other vent sites. Trace element partitioning over the entire chlorinity
21 range indicates a single deep fluid source feeding all the venting sites. Applying the Si-Cl
22 geothermobarometer at Capelinhos, we find phase separation conditions at 435–440°C, and
23 370–390 bars (2500–2800 m below seafloor (mbsf)) consistent with former estimates for the
24 LSHF, while temperatures of fluid-rock last equilibrium are estimated at ~400°C for Capelinhos
25 and 350–375°C for the other sites based on the Fe-Mn geothermometer. We interpret these
26 discrepancies in thermodynamic conditions beneath the sites in terms of crustal residence time
27 which are likely related to permeability variations across the hydrothermal upflow zone. We
28 propose that conductive cooling of the up flowing fluids from the phase separation zone to the
29 seafloor, beneath the main field vent sites, lowers the T conditions of last fluid-rock equilibrium,
30 enabling ~65% of Fe mobilized in the reaction zone to be stored. In comparison, Capelinhos
31 fluids are transported more rapidly from the reaction zone to the seafloor along a high-angle
32 fracture system. The fluids venting at Capelinhos are more representative of the deeper part of
33 the hydrothermal reaction zone.

34

35 **1 Introduction**

36 Hydrothermal fluid circulation at mid-ocean ridges (MOR) is one of the major processes
37 controlling the cooling of the oceanic lithosphere (Chen & Morgan, 1990; Stein & Stein, 1994),
38 the geochemical composition of the crust (Kelley & Delaney, 1987; Kelley & Robinson, 1990;
39 Alt, 1995; Alt & Teagle, 2003; Barker et al., 2008; Brant et al., 2012), the chemical composition
40 of the ocean (Elderfield & Schultz, 1996; Resing et al., 2015) and the development of peculiar
41 chemo-synthetic ecosystems (Martin et al., 2008 and references therein). Increasing attention
42 has been drawn to the study of black smokers since their discovery along the East Pacific Rise
43 (EPR) off the Galapagos Islands in 1977. They have shown an unexpected diversity in
44 geological setting, hydrothermal ecosystems and fluid chemistry (Campbell et al., 1988; Von
45 Damm, 1988, 2000; Douville et al., 2002; Ludwig et al., 2006; Schmidt et al., 2007, 2011).
46 From the recharge zone to the discharge area, seawater is transformed into a high temperature
47 hydrothermal fluid, acquiring its chemical composition in a reaction zone, which is univocally
48 pictured on top of axial magmatic chambers (AMC) at least along magmatically robust sections
49 of fast (e.g. EPR, 9°50'N), intermediate (e.g. Endeavour segment, Juan de Fuca Ridge) and
50 slow spreading (e.g. Lucky Strike, 37°17'N) ridges. The chemical composition of high
51 temperature fluids can be further modified by either interaction with the rocks along the upflow
52 pathway to the seafloor or precipitation of secondary minerals, depending on the fluid velocity
53 (Saccocia & Seyfried, 1994; Lowell, 2003; Coogan, 2008; Steele-MacInnis et al., 2012). Thus
54 highlights that the P-T estimates for reaction zone conditions are sensitive to the geochemical
55 tracer used and clear depth boundaries of the reaction zone cannot be easily drawn. The water-
56 rock interactions which form hydrothermal fluid formation from seawater, occur over a wide
57 range of pressure and temperature conditions, i.e. from the deepest part of the hydrothermal cell
58 to the discharge at the seafloor, ΔP and ΔT can be a few hundred bars and °C, respectively.
59 Chemical variations can be identified depending on the nature of the substratum, i.e. extrusive

60 basaltic and/or mantellic substrate. The chemical compositions of focused hydrothermal fluids
61 are used to constrain the range of pressure and temperature conditions at which water-rock
62 interactions take place in the crust (Seyfried et al., 1998; Seyfried, 2003; Von Damm et al.,
63 2003; Mottl et al., 2011; Reeves et al., 2011). The purpose of the present study is to gain insight
64 on the seafloor flow geometry using chemical analysis on fluids from different sites at the
65 Lucky Strike hydrothermal field (LSHF).

66 LSHF was discovered at 37°N along the slow-spreading Mid-Atlantic Ridge (MAR) in 1992
67 during the FAZAR cruise (Langmuir et al., 1997). The discovery motivated several subsequent
68 research cruises with the objective of better characterizing the geological, geochemical,
69 geophysical and biological context of the vents (Fouquet et al., 1995; Langmuir et al., 1997;
70 Von Damm et al., 1998; Charlou et al., 2000; Ondréas et al., 2009; Barreyre et al., 2012; Pester
71 et al., 2012; Escartin et al., 2015). The first comprehensive study on the LSHF vent chemistry
72 (Von Damm et al., 1998) indicated that the discharge of focused fluids (sampled in 1993 and
73 1996) originated from deep rooted fluids that underwent near-surface processes, e.g. mixing
74 with seawater, in the upflow zone (Von Damm et al., 1998). Charlou et al. (2000) analyzed
75 fluids from ten active sites of the LSHF. All samples were depleted in chloride relative to
76 seawater (~420mM to ~520mM) with Cl increasing along a SE-NW transect across the LSHF.
77 These authors proposed that a relatively shallow reaction zone provided vapor-like fluids (i.e.,
78 Cl below 545 mM of seawater) to the upflow zone at subcritical conditions (temperature and
79 pressure lower than 407°C and 298bars respectively, equivalent to ~1300 mbsf; Bischoff,
80 1991). Chemical data presented by Charlou et al. (2000) also suggested that vapor-dominated
81 fluids underwent substantial subsurface mixing with altered seawater prior to venting as
82 indicated by variable Sr isotope signatures with Cl concentrations still below that of seawater.

83 Eleven years later, Pester et al. (2012) used trace element and chloride co-variation in fluids at
84 five active vents to infer P–T equilibrium conditions during upflow. The samples display both

85 chloride depletion and enrichment relative to seawater (from 414mM to 588mM). These authors
86 proposed that 1) a unique and deep source feeds the hydrothermal field and 2) fluids underwent
87 phase separation at 430–475°C and 410–480 bars (~2400–3100 mbsf), i.e. much higher P-T
88 conditions than those proposed by Charlou et al. (2000). They attributed the variability observed
89 in fluid chlorinity to subtle changes in pressure and/or temperature. These authors also argue
90 that after phase separation and prior to discharge, the hot fluids cooled conductively and
91 equilibrated with the greenschist facies of basaltic host rocks at lower temperatures (i.e. 350–
92 380 °C).

93 These different models of the plumbing circulation system below the LSHF highlight the
94 difficulties that arise when interpreting the chemical composition of vent fluids. This is
95 compounded by the lack of contemporaneous observations between the studied sites. The
96 EMSO-Azores program started in 2010 (FP6-ESONET; Person et al., 2009; Colaço et al., 2011)
97 and led to the set-up of a deep-sea observatory at LSHF. High temperature hydrothermal fluids
98 were collected yearly during maintenance cruises which permitted the discovery of a new, high-
99 temperature venting site, named Capelinhos (T=324°C, Table 1) in 2013 (Escartin et al., 2015;
100 Fig 1). Capelinhos lies approximately 1.5 km east of the LSHF and a few hundred meters from
101 the seafloor emergence one of the normal faults (referred as F2 in Combier et al., 2015) that
102 rifts the Lucky Strike axial volcano. All other active/inactive venting sites are located to the
103 west around a fossil lava lake on top of the volcano (Humphris et al., 2002; Ondréas et al., 2009;
104 Barreyre et al., 2012; Fig 1).

105 In this study, we report on the first chemical data acquired for the Capelinhos vent site and we
106 compare its chemical characteristics with those from 12 other active LSHF vents sampled
107 during the same cruise in 2013. We use this dataset to show that Capelinhos is a key site to
108 assess the role of subsurface mixing, phase separation, and conductive cooling processes on the

109 chemistry of the LSHF fluids and to propose an up-dated model of hydrothermal circulation
110 below the field.

111 **2 Geological setting**

112 The ~65 km long Lucky Strike segment is located south of the Azores islands along the MAR
113 between 37°03'N and 37°37'N (Detrick et al., 1995). The spreading rate is ~22 mm/yr (Cannat
114 et al., 1999; Miranda et al., 2005). The LSHF is located on top of a volcano at the center of the
115 segment. Seismic data have constrained the presence of an axial magma chamber (AMC) at a
116 depth of about 3500 m below the summit of the volcano (Singh et al., 2006). Microseismic
117 events recorded between 2007 to 2009 are located above the AMC at depths between 1800 and
118 2500 m below seafloor (mbsf), and interpreted as the product of active fracturing induced by
119 penetration of colder down-flowing fluids into the hot rocks (Crawford et al., 2013). These
120 microseismic events document the existence of two predominantly along-axis hydrothermal
121 cells, with a central upflow zone centered beneath the LSHF. Seismic data suggests that the
122 layer 2A depth is variable throughout the hydrothermal field (Arnulf et al., 2011).

123 The long known Lucky Strike hydrothermal field (Von Damm et al., 1998, Charlou et al., 2000,
124 Pester et al., 2012; Chavagnac et al., 2018 Fig.1) comprises 20-30 sites with high-temperature
125 black smokers (T= 200-340°C) and low-temperature diffuse venting (total area about 1800 m²,
126 Barreyre et al., 2012). These sites are all clustered around a fossil lava lake (300 m in diameter;
127 Fouquet et al., 1995) framed by two ancient volcanic cones, and one of which has been
128 truncated by N010°–N030° faults and fissures (Ondréas et al., 2009; Fig 1). The newly
129 discovered Capelinhos site is isolated and located 1.5 km to the east of the main field (Escartin
130 et al., 2015). There, chimneys form “candelabra-like structure” on top of 10 m high sulfide
131 mound and expel black smoker-type fluids at temperatures up to 324°C (Figure 2; Table 1).
132 Diffuse venting is limited to the close vicinity of the black smokers, at the base of the edifice.

133 **3 Sampling and Analytical Methods**

134 3.1 Fluid collection

135 The fluid samples studied in this paper were collected during the MoMARsat'13 EMSO-Azores
136 maintenance cruise on the French Research vessel *Pourquoi Pas?* in September 2013. The 13
137 sampling sites (12 on LSHF and Capelinhos) were chosen to document the fluid diversity
138 identified at LSHF by previous studies (Von Damm et al., 1998; Charlou et al., 2000; Pester et
139 al., 2012; Chavagnac et al., 2015). Each vent site was sampled four times in succession (total
140 duration of sampling <1 hour) to allow a better characterization of the end-member
141 hydrothermal fluid. In-situ temperatures were measured in each vent prior to fluid sampling
142 using the ROV Victor 6000 high temperature probe. High-temperature fluids were collected
143 with 200ml titanium gas-tight samplers handled and triggered by the hydraulic arm of the ROV.
144 The time between fluid samples at a given vent did not exceed 20 minutes. The samples were
145 processed on board immediately after the ROV recovery. First, gases were extracted from the
146 samplers and transferred into vacuumed stainless steel canisters or to ultra-clean water sealed
147 glass bottles, depending on the expected gas volume, for further analysis. The fluid samples
148 were then extracted, filtered through 0.45 μm Millipore filters and split into different aliquots
149 for onshore analysis and stored at 4°C. pH, Eh, salinity and conductivity were measured on-
150 board immediately after gas/fluid processing.

151 3.2 Analytical methods

152 All the chemical analyses were conducted at the Geosciences Environment Toulouse (GET)
153 laboratory. Ca, Na, K, Mg, Si, Fe, Mn and Li concentrations were determined with an
154 inductively coupled plasma atomic emission spectrometer (ICP-AES) Horiba Ultima2
155 instrument. Errors on analyses are indicated in Table 2. The instrument is calibrated using mono
156 elemental solutions, multi elemental solutions, and IAPSO standard solution (Besson et al.,

157 2014). The latter is a seawater standard solution provided by OSIL and certified for its salinity.
158 Salinity controls major element concentrations in seawater (Millero et al., 2008). Li
159 concentration measurements of IAPSO give an average concentration of $24.6 \pm 0.3 \mu\text{M}$
160 ($\text{rsd}=1\%$; $n=4$; $26.5 \mu\text{M}$ for seawater, Von Damm et al., 1998). The analytical drift is quantified
161 by the standard bracketing after every 8 samples. Analytical precision is better than 2%.

162 Sr, Rb and Cs concentrations were measured using an inductively coupled plasma mass
163 spectrometer ICP-MS Agilent500. The standard used was a NASS 6 solution (international
164 seawater standard certified for some trace metals) and give values of $79.1 \mu\text{M}$, $1.17 \mu\text{M}$, 1.19
165 nM compared to $87 \mu\text{M}$, $1.4 \mu\text{M}$ and 2nM of seawater values for Sr, Rb and Cs respectively
166 ($n=3$). Drift corrections are made using an in-house multi-elementary standard doped with In
167 and Re before analysis. Anions (Cl , SO_4 , Br) were determined by anionic chromatography
168 calibrated with a IAPSO standard seawater solution.

169 Sr isotopic composition was measured at the GET laboratory using a MAT FINIGAN 261
170 thermal ionization mass spectrometer. Analyses were performed on the samples which contain
171 the lowest Mg concentrations. Sr was isolated from the matrix using Sr-Spec resin (Eichrom,
172 USA). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio was defined as the average of 100 measurements of ion intensities
173 following the static multi-collection mode. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were normalized to $^{86}\text{Sr}/^{88}\text{Sr} =$
174 0.1194 . Measured values for NBS 987 standard (recommended values of 0.710250) was
175 $^{87}\text{Sr}/^{86}\text{Sr} = 0.710250 \pm 0.000011$ ($2\sigma_{\text{D}}$, $n = 14$).

176 We consider end-member hydrothermal fluids calculated by linear extrapolation to zero-Mg of
177 the least-square regression method (Table 2; Von Damm, 1988). The compositions of all
178 analyzed samples for Sr isotope signature are given in the Supplementary Materials (Fig S1 and
179 Table S1). For isotopic composition, the same method of linear extrapolation to a Mg/Sr equal
180 to zero is used.

181

182 **4. Results**

183 *Chloride, Sulfate, Bromide*

184 Hydrothermal fluids of the LSHF exhibit chloride concentrations between 415 mM for the
185 South-East (SE) sites (Montsegur, Aisics, and Tour Eiffel; Figure 1) and 574 mM for the North-
186 East (NE) sites (Y3 and Sintra) and the South-West (SW) sites (South Crystal, Crystal and
187 Sapins). White Castle, Isabel and Cyprès sites have Cl concentration closer to seawater one at
188 values ranging between 470mM and 520mM, also in line with previous data. Interestingly,
189 Capelinhos has the lowest chloride concentration ever measured at LSHF at 262 mM (Table 2).

190 All fluids have SO₄ values close to zero. However, some of them (Crystal and Tour Eiffel) have
191 calculated end-member value slightly higher than 0 probably due to anhydrite entrainment and
192 dissolution (Von Damm et al., 1998; Table 2). Br concentrations have variations similar to Cl
193 concentrations, whereby maximum Br concentrations are found in Y3 fluids at 0.88 mM close
194 to the seawater value of 0.84 mM while minimum values are found for Capelinhos fluids at
195 0.38 mM.

196 *Iron and Manganese*

197 Capelinhos end-member fluid shows very high Fe and Mn concentrations of 2789 μM and 639
198 μM, respectively. In comparison, Fe and Mn concentrations of LSHF end-member fluids vary
199 between 155 μM (Sintra) and 593 μM (South Crystal) for Fe (Fig 3a) and between 164 (Sintra)
200 and 232 μM (South Crystal) for Mn (Fig 3b).

201 *Silica*

202 Silica concentrations of hydrothermal end-members vary from 12.8 mM at Sintra (NE group)
203 to 18.6 mM at Cyprès (SW group). The Si concentration at Capelinhos is 14.1 mM, a value
204 similar to the one for the SE sites, despite distinct chlorinities (Fig 3c).

205 *Sodium, Calcium and Potassium*

206 Na, Ca and K constitute the major cations in the end-member fluids and are closely linked to
207 Cl concentration due to charge balance. Maximum Na concentrations (445mM) are found in
208 Crystal and South Crystal fluids, while the minimum Na concentration (205 mM) is obtained
209 for Capelinhos.

210 Maximum Ca concentration are found in Sintra, Y3, Crystal and South Crystal fluids with
211 values around 53 mM while at Capelinhos the Ca concentration is the lowest one at 18mM.

212 Maximum K concentrations (28.3 mM) are found in South Crystal and Crystal and the
213 minimum concentration (12.1 mM) is measured at Capelinhos.

214 *Rubidium, Strontium, Cesium and Lithium*

215 Rb, Cs and Li are ten to 20 times more enriched in the LSHF fluids compared to seawater while
216 Sr concentrations fluctuate around the seawater value at $\pm 20\%$. Concentrations in Rb, Li, Cs
217 and Sr are correlated to the fluid chlorinity (Table 2). Minimum and maximum Rb
218 concentrations are 18.7 μM and 50 μM for Capelinhos and Crystal, respectively. Minimum and
219 maximum Li concentrations are 197.1 μM and 367.9 μM , also for Capelinhos and Crystal,
220 respectively. Sr follows the same pattern with a maximum concentration of 180 μM for Crystal
221 and a minimum concentration of 35.9 μM at Capelinhos.

222 *Sr isotopes*

223 The least radiogenic end-member fluid compositions ($^{87}\text{Sr}/^{86}\text{Sr}=0.70384$; Table 2) are obtained
224 for Capelinhos. Y3, Crystal and South Crystal on the Northern and Western side of the fossil

225 lava lake have unradiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ ratios ranging between 0.7039 and 0.7040. $^{87}\text{Sr}/^{86}\text{Sr}$
226 signatures of the Aisics, Tour Eiffel, and Montsegur sites are the most radiogenic ones at 0.7042
227 - 0.7043 (Table 2). The Central sites display variable Sr isotope signatures, i.e. Cyprès at
228 0.70403 and White Castle at 0.70502.

229 **5. Discussion**

230 5. 1 Source(s) of hydrothermal fluids

231 Cl is the major anion present in hydrothermal fluids, and controls the cation abundances due to
232 the charge balance of the solution. We propose to define groups of sites based on Cl
233 concentrations and spatial distribution on the seafloor. With the lowest salinity, Capelinhos (Cl
234 = 262mM) defines its own group; the SE group of sites is composed of Tour Eiffel, Montsegur,
235 Aisics with a $\text{Cl}_{\text{average}}$ of $\sim 420\text{mM}$; the Central group characterized by $\text{Cl}_{\text{average}} \sim 495\text{mM}$,
236 comprises Isabel, White Castle, Cyprès. We define SW and NE groups for which salinity values
237 are all close to or above seawater, i.e. SW group (Crystal, South Crystal, and Sapin sites;
238 $\text{Cl}_{\text{average}}$: $\sim 580\text{mM}$) and NE group (Y3 and Sintra sites; $\text{Cl}_{\text{average}}$: 574 and 537 mM). Due to the
239 conservative behavior of Cl in hydrothermal system, this chemical tracer has been widely used
240 to infer the P-T conditions of the phase separation zone. Charlou et al. (2000) proposed that Cl
241 variability observed at the LSHF could be due to subcritical vapors mixing with altered seawater
242 in subsurface conditions prior to fluid venting. To address this hypothesis, we use the Sr isotope
243 compositions of the lowest Mg-rich hydrothermal fluids as a tracer of sources and mixing.

244 A hydrothermal fluid acquires its Sr isotopic signature from the rocks with which it interacts
245 (Albarède et al., 1981), i.e basalt at LSHF. The contrasting Sr isotope compositions of basalt at
246 0.70298 and seawater at 0.70916 are favorable to the detection of any input of modified
247 seawater in hydrothermal fluids. Any Sr isotopic signature of hydrothermal fluid end-member
248 that departs from that of the basalt, will suggest the contribution from an additional source

249 (Berndt et al., 1988; Palmer, 1992; Ravizza et al., 2001). The most vapor-dominated fluid at
250 Capelinhos exhibits the least radiogenic Sr isotopic composition at 0.70384. In comparison, the
251 SE, NE and SW groups which exhibit Cl concentrations at 420mM, 574mM and 570mM,
252 respectively, display similar Sr isotopic signatures at 0.7042-0,7044 for SE sites and 0.7040
253 and 0.7042 for NE and SW sites (Table 2). In contrast, the Central group (~495mM of Cl)
254 displays a wide range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, ranging from 0.70403 to 0.70502. The observed range
255 of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of fluid end-members do not correspond to the geographical distribution of
256 sites around the fossil lava lake. Moreover, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and the Cl contents of
257 hydrothermal end-members do not correlate positively and linearly. In addition, it is unlikely
258 that the variability of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios is related to local differences in the substratum, as basalts
259 from a wide selection of samples (including T- and E-MORB) across the Lucky Strike segment
260 have a well-constrained Sr isotopic signature of 0.70298 ± 0.00007 (n=13; Hamelin et al., 2013).
261 The tight but significant differences in Sr isotopic signature between the highest and lowest
262 chlorinity fluids means that Capelinhos fluid, compared to the other sites, has less interaction
263 with radiogenic Sr-rich material such as extensively altered rocks. The fact that $^{87}\text{Sr}/^{86}\text{Sr}$ is not
264 correlated with Cl rules out a modified seawater component that would mix with hydrothermal
265 fluid in subsurface. However, this does not rule out the contribution of a second deep-rooted
266 hydrothermal cell to produce the Capelinhos fluid.

267 We investigate the distribution of trace element concentrations (Br, Li, Sr, Rb and Cs) over the
268 LSHF chlorinity range, following the methodology of Pester et al. (2012) to detect the potential
269 occurrence of a second hydrothermal source. The elements considered are controlled by
270 vapor/brine partition coefficients during phase separation (Pester et al., 2015; Foustoukos &
271 Seyfried, 2007a, b; Foustoukos et al., 2004; Berndt & Seyfried, 1990; among others). Li and Cs
272 distributions exhibit a negative linear trend while Rb and Sr show a positive trend, indicating a
273 preferential partitioning to the vapor and brine phases, respectively (Figure 4). This is in line

274 with previous studies (Berndt & Seyfried, 1990; Pokrovski et al., 2005; Foustoukos & Seyfried,
275 2007a; Pester et al., 2015). This is also coherent with the LSHF study of Pester et al. (2012),
276 apart for Rb which shows less brine affinity with the extended Cl range of the present dataset.
277 Note, however, that without Capelinhos fluid, Rb presents a steeper linear slope at +0.38,
278 identical to the one at +0.35 reported in Pester et al. (2012). Element vapor/brine partitioning
279 can be summarized as follow $Sr < Rb \ll Br \ll Cs < Li$ with increasing affinities for the vapor
280 phase. Therefore, the linear correlation of each of these elements over the Cl range (from
281 262mM to 574mM; Figure 4) supports a phase separation process controlling overall trace
282 element abundances, i.e. a unique deep-rooted fluid source as proposed by Pester et al. (2012).
283 This implies that being 1.5 km away from the other sites, Capelinhos is nevertheless fed by the
284 same upflow zone and is part of the same hydrothermal cell.

285 5. 2 Chlorinity variability and geographical repartition

286 While the distribution of trace element suggests a unique source feeding the hydrothermal
287 upflow at LSHF, the end-member fluids nevertheless display a wide range of chlorinity, from
288 the most vapor-like, low-Cl fluids at Capelinhos to the brine-like, high-Cl fluids at Y3. Venting
289 of low-Cl fluids can unambiguously be ascribed to phase separation processes at depth and
290 flushing of buoyant vapor phase on the seafloor. However, the physics controlling the venting
291 of brine-like fluids is more complex as their thermodynamical (viscosity and density) and
292 surface properties (surface tension) counteracts their buoyancy, impacting therefore their rise
293 towards seafloor. In the literature, numerical models of two-phase flow including transport of
294 salt (considered as NaCl) have proposed scenarios for brine venting (Fontaine et al., 2007;
295 Coumou et al., 2009).

296 Phase separation leads to vapor and brine formation but due to different physical behavior, i.e.
297 the wetting effect of the liquid/brine phase, brines will segregate and be stored in backwater

298 porosity or will coat the walls of larger channels (Fontaine & Wilcock, 2006). As long as brine
299 saturation is lower than a threshold value, the brines do not form a continuous medium and
300 remain immobile. However, when this threshold is reached, and provided that the vertical
301 pressure gradients in the upflow are high enough compared to brine density (if not, the brine is
302 too dense and will sink down; Fontaine & Wilcock, 2006; Fontaine et al., 2007), then the brine
303 phase is able to move upwards. Fontaine and Wilcock (2006) showed that brine (<20-25wt%
304 NaCl) produced under supercritical conditions and stored within the rock backwater porosity
305 in the reaction zone, could still flow towards the surface. Meanwhile, vapor phases flow
306 preferentially through wider cracks (Goldfarb & Delaney, 1988; Fontaine & Wilcock, 2006)
307 and are flushed to the seafloor. Fontaine et al. (2007) also developed the hypothesis that vertical
308 permeability gradients could act as a barrier for brine upflow and tend to concentrate brine at
309 the base of layer 2A. When the backwater porosity is saturated with brine, or during cooling of
310 the system, brine can be entrained, and mixed with vapor or seawater salinity hydrothermal
311 fluid, which leads to fluid discharge with higher salinity than seawater.

312 Alternatively, Coumou et al. (2009) conducted numerical modelling without a permeability
313 gradient to investigate salinity variations at discharge zones for different bottom heat fluxes and
314 system pressures. For a pressure of 150 bars at the seafloor and ~250 bars at the bottom of the
315 system, i.e. phase separation zone ~1000mbsf, the simulations show temporal variations in vent
316 fluid salinity with several spikes at salinity higher than seawater over periods of several years.
317 These salinity spikes are also associated with temperature spikes of a few degrees (<10°C).
318 Coumou et al. (2009) explained these salinity pulses as due to brine mobilization occurring
319 when the porosity of the upflow zone becomes saturated. Further mixing of this brine with
320 vapor-dominated fluids during the upflow produces a transient spike in salinity. Salinity then
321 returns to the initial vapor-like one, while the brine layer builds up again and reduce saturation
322 of the backwater porosity. In summary, modeling studies infer that sustained phase separation

323 processes can drive brines to “over-saturate” the porous network causing their remobilization
324 and flushing to the seafloor without major changes in the thermodynamic state of the
325 hydrothermal cell.

326 Brine remobilization and flushing have been already observed and invoked in the natural
327 hydrothermal environment, to explain Cl variability related to periodic (tides) brine flushing at
328 the Main Endeavour Field (Larson et al., 2009). At LSHF, Y3 site exhibits Cl concentrations
329 which increase from vapor-like values at 436 mM in 1993 (Von Damm et al. 1998) to brine-
330 like ones at 574 mM in 2013 (this study, Table 2). We infer that this shift in fluid chlorinity of
331 Y3 site could be ascribed to brine over-saturation and mobilization in the porous/fractured crust
332 along the upflow zone, as venting temperature and reaction zone P-T conditions according to
333 the geothermobarometers did not fluctuate significantly over that time period (Pester et al.
334 2011). Based on a study of tidal forcing on LSHF vent fluid temperatures, Barreyre and Sohn
335 (2016) proposed that the permeability structure beneath the LSHF vents is variable, possibly
336 reflecting variations of layer 2A thickness from 300 to 600 mbsf between the west and east of
337 the fossil lava lake area. Such local variations would also be expected to impact brine storage
338 and remobilization beneath the field. Finally, the yearly monitoring of the Y3 vent in
339 comparison to other LSHF sites should provide key information of the temporal evolution of
340 brine venting. Such information is particularly important to better design numerical models of
341 hydrothermal flow dynamics and to gain more insight into the interrelationships between phase
342 separation, brine storage, remobilization and fate beneath the field.

343 5.3 Pressure and Temperature Conditions in the LSHF hydrothermal system

344 5.3.1 Conditions at the base of up-flow zone

345 Si and Cl end-member fluid concentrations have been used as a geothermobarometer to
346 determine the depth of the top of the two-phase zone in several hydrothermal systems

347 worldwide (Fontaine et al., 2009). This depth is considered a good estimate of the base of the
348 upflow zone as phase separation induces large variations in the fluid fluxibility (a measure of
349 the ability of buoyancy-driven water to transport energy), providing the necessary buoyancy
350 for the hydrothermal vapors to flow upwards (Jupp & Schultz, 2000; Coumou et al., 2009). Cl
351 concentration only changes in the two-phase zone, as soon as the vapor-dominated fluid starts
352 to rise it returns to a single-phase, and its Cl content is fixed for the remainder of the ascent to
353 the seafloor, provided no mixing with seawater occurs. The Cl content of flushed vapors are
354 thus representative of the conditions at the top of the two-phase zone. Using Cl-P-T solubility
355 relationships (e.g., Driesner, 2007), one can estimate a range of possible conditions (a iso-Cl
356 line) in P-T space for a given Cl. Now, considering that Si is in equilibrium with quartz during
357 upflow/cooling and using Si-Cl-T-P solubility relationships (e.g., Foustoukos & Seyfried,
358 2007b) one can use the Cl and Si contents of the vented vapors to estimate another range (an
359 iso-Si line) of possible T-P for a given Si. Intersection between these iso-Cl and iso-Si lines
360 gives a unique P-T condition for the roof of the two-phase area. When applying the Si-Cl
361 geothermobarometer to the Capelinhos fluids, we obtain a temperature of 438°C and a pressure
362 of 375 bars for the phase separation zone/base of the reaction zone, that corresponds to a depth
363 of 2600 mbsf (using a cold hydrostatic pressure gradient of $\rho=800\text{kg/m}^3$, Fontaine et al., 2009).
364 Using the Tour Eiffel chemical data for fluids collected in 1993 (Charlou et al., 2000), Fontaine
365 et al. (2009) determined similar P and T conditions at 390 bars and 440°C for the LSHF phase
366 separation zone. Thus, the deepest part of the upflow zone is quite well constrained with similar
367 estimates for fluids collected 20 years apart and at different site within the LSHF.

368 One should, however, be careful using this geothermobarometer as quartz may precipitate
369 during upflow. Quartz veins in the ocean crust are often associated to greenschist facies
370 minerals such as pyrite, chlorite, amphibole and epidosite at different depth levels (Delaney et
371 al., 1987; Honnorez, 2003; Heft et al., 2008; Alt et al., 2010). The vented Si concentration

372 should, therefore, be taken as a minimum value (the actual Si content of the rising vapor being
373 higher and not constrained) giving a minimum depth to the base of the upflow/roof of the two-
374 phase area. The Si-Cl geothermobarometer developed by Fontaine et al. (2009) predicts that the
375 estimated depth of the top of the two-phase zone would deepen by 70-100m per mmol of Si at
376 constant Cl. We note that the equilibrium pressure ranges of 375-390 bars derived with Si-Cl,
377 i.e. 2600-2800 mbsf is in good agreement with the 2300-2900 mbsf maximum depth range for
378 micro-seismic activity beneath the hydrothermal field (Crawford et al., 2013).

379 5.3.2 Conditions of last equilibrium in the reaction zone

380 Vent fluid Fe and Mn contents can be used to estimate the temperature of the fluid last
381 equilibrium with greenschist facies rocks (Pester et al., 2011). For Capelinhos end-member
382 fluids, which exhibit the highest Fe and Mn concentrations of all the LSHF fluids - equivalent
383 to Fe concentration found at the Broken Spur (29°N) and Snake Pit (23°N) fields on the MAR
384 (Campbell et al., 1988; James et al., 1995; Figure 5) - we find a minimum temperature of
385 greenschist facies equilibrium at 403°C. The Fe-Mn geothermometer does not provide a means
386 to calculate a corresponding equilibrium pressure. If we assume that both quartz and fluid-
387 greenschist (Fe and Mn bearing) minerals facies reached equilibrium concomitantly, i.e. that
388 the Capelinhos fluids interacted minimally with the surrounding rocks during their ascent from
389 the phase separation zone, then the equilibrium temperature is obtained from the Fe-Mn
390 geothermometer and the vent fluid Si concentration permits to calculate a corresponding
391 equilibrium pressure using Cl-T-P-dependent solubility relationships (Von Damm et al., 1991;
392 Foustoukos & Seyfried, 2007b). At Capelinhos (Si=14.1 mM, Cl=260 mM), the estimated
393 equilibrium pressure is 350 bars, i.e. a depth of ~2300 mbsf (Fig 5a). Applying the same
394 approach to the most brine-dominated hydrothermal fluids from the other LSHF sites, i.e. the
395 SW group and Y3 of the NE group, indicates a lower pressure (300 bars, i.e. ~1600 mbsf) and
396 cooler temperatures 350-375°C for the last fluid-rock equilibrium. These differences in P-T

397 conditions of last fluid-rock equilibrium between the Capelinhos and the other LSHF sites
398 probably indicates that the fluids venting at Capelinhos ascend more rapidly from the phase
399 separation zone than the fluids venting at the other sites so that they react less with the host
400 rocks upon ascent. This interpretation is also consistent with the least radiogenic Sr isotope
401 signature of Capelinhos end-member fluids which we interpret as the most representative of the
402 basaltic substratum (section 5.1). We thus propose that the observed variability in fluid
403 composition between the LSHF vent sites primarily results from variable fluid residence times
404 within the different areas that compose the upflow zone. We discuss a possible geological factor
405 controlling these processes at Lucky Strike in the next section.

406 5.4 Fluid circulation pathway at LSHF

407 Based on the Si-Cl geothermobarometer which is in agreement with the depth of micro-seismic
408 activity (Crawford et al. 2013), we propose that fluids at Lucky Strike start to upflow from a
409 depth/pressure of 2600-2800 mbsf/370-390 bars at 430-440 °C below the lava lake. Capelinhos
410 is located 300 m to the east of a west-dipping fault that has been imaged in seismic reflection
411 data and shown to extend at least down to a few hundred mbsf under the volcano (F2 in Combier
412 et al., 2015; Figure 6). We infer that small offset faults and fractures parallel to this fault and
413 belonging to the same network may intersect the seafloor near Capelinhos (Escartin et al.,
414 2015). We propose that this fault and fracture network intercepts the hydrothermal upflow zone
415 at depth, in the intrusive section (layer 2B), creating a fast-extraction pathway for hydrothermal
416 fluids venting at Capelinhos because of enhanced hydraulic properties (e.g., permeability,
417 porosity) due to tectonically produced faults and fissures (Caine et al., 2010) (Figure 6). This
418 “transverse” and longer (compared to a vertical flow) flow path leads to significant cooling
419 from 403°C in the phase separation zone to 324°C at the vent field but the fast fluid migration
420 and short residence time prevents extensive fluid-rock reactions, and the exiting fluids preserve
421 their metal content. On the other hand, the fluids exiting at the main LSHF sites distributed

422 around the lava lake would rise almost vertically, but more slowly due to the lower overall
423 permeability of younger intrusives beneath the fossil lava lake (Arnulf et al., 2011). This longer
424 residence time allows fluids to interact more extensively with the crust to temperature down to
425 350°C, and to precipitate a large part of their metal content, as secondary greenschist minerals
426 including Fe-bearing phases like chlorite.

427 The low Fe-Mn ratios of the main LSHF sites (~0.8 to 2.5), compared to Capelinhos fluids at
428 ~4.4 (Fig 4), reflects a significant Fe loss within the upflow zone. By using Capelinhos fluids
429 as a reference, we calculate that ~65% ($\pm 14\%$) of dissolved hydrothermal Fe from sites located
430 around the fossil lava lake is stored most likely as secondary minerals between the phase
431 separation zone and the seafloor. In other words, only ~35% of Fe mobilized in the reaction
432 zone is discharged as dissolved phase into the deep seawater mass at the LSHF.

433 **6 Conclusion**

434 In 2013, a new active site, named Capelinhos, was discovered at LSHF approximately 1.5 km
435 east of the main Lucky Strike axial graben and fossil lava lake, where the well-known LSHF
436 vents are located. Capelinhos vents high temperature black smoker fluids at 324°C. Its fluid
437 chemistry has two distinct characteristics compared to the other LSHF vent sites: 1) the lowest
438 Cl concentration at ~260 mM ever measured at LSHF and 2) the highest Fe and Mn
439 concentration (2800 μM and 640 μM respectively). The discovery of this site provides the
440 opportunity to reassess the thermodynamic condition within the reaction zone. Applying the Si-
441 Cl geothermobarometer of Fontaine et al. (2009), we show that Capelinhos fluids were formed
442 at supercritical conditions, as well as the other vapor-dominated fluids from LSHF. The
443 temperature of the phase separation zone is 438°C at 2600mbsf. These results are in agreement
444 with the maximum depth of microseismic events reported in Crawford et al. (2013) which are

445 interpreted as due to rapid heat exchange between hot rocks and down-going hydrothermal
446 fluids.

447 Applying other geochemical indicator, such as Fe–Mn geothermometer of Pester et al (2011),
448 a range between ~370°C for LSHF fluids and ~400°C for Capelinhos is obtained. By combining
449 quartz solubility with these calculated temperatures, the minimum equilibration temperature
450 and pressure in the reaction zone appears to be deeper for Capelinhos fluids (350 bars) than for
451 LSHF (300 bars). Because the fluids vent at temperatures that are still well within the
452 greenschist facies realm, these conditions are not considered as completely representative of
453 the top of the reaction zone. More accurately, they are indicative of significant differences in
454 the residence time of the fluid in the upflow zone between Capelinhos and the other LSHF
455 vents.

456 We propose a revised model of the hydrothermal path based on the discovery of Capelinhos.
457 When crossing the 2-phase boundary, at ~2600 mbsf, the fluid starts flowing upwards, with
458 brine entrainment to different extent from site to site. At the fossil lava lake, fluids (vapor and
459 brines) undergo more conductive cooling than Capelinhos vent along their upflow to the surface
460 due to longer residence time in the substratum. Brine mixing and cooling processes likely result
461 in the formation of secondary Fe-bearing minerals like chlorite beneath the fossil lava lake,
462 resulting in the storing of up to 65% of Fe mobilized in the reaction zone. Fluids discharging at
463 Capelinhos (the most vapor dominated fluid of the LSHF), reach the sea-surface by a high angle
464 normal fault zone, and undergo significant cooling but still preserve high dissolved Fe and Mn
465 concentrations.

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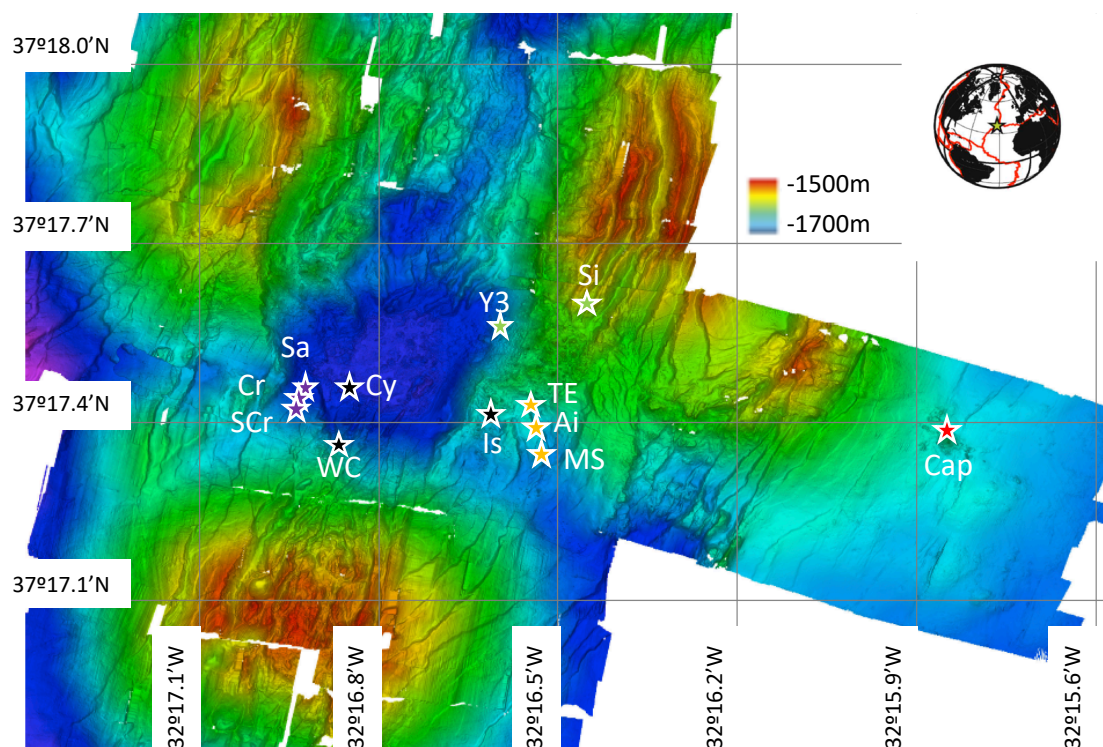
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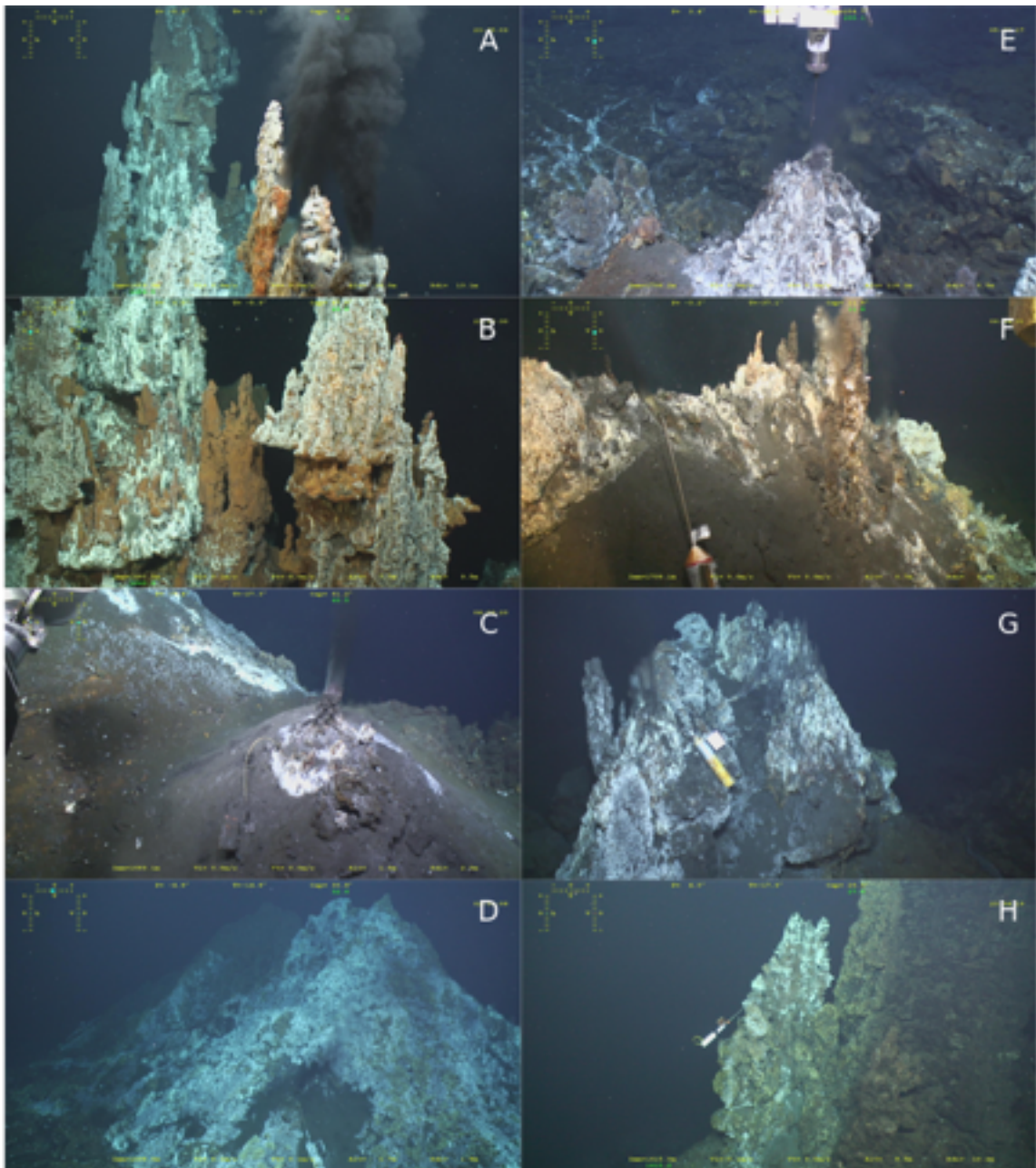
732 **Figure 1.** Bathymetric map of the Lucky Strike central volcano. Active hydrothermal vents are
 733 reported. Violet star: SW group composed of Sapins (Sa), Crystal (Cr) and South Crystal (SCr);
 734 Yellow star: SE group composed of Tour Eiffel (TE), Aisics (Ai) and Montsegur (MS); Green
 735 star: NE group composed of Sintra (Si) and Y3; Black star: Central group composed of Cyprés
 736 (Cy), White Castle (WC) and Isabel (Is); The red star locates Capelinhos (Cap). See text for
 737 details. Microbathymetry from Ondréas et al. (2009).



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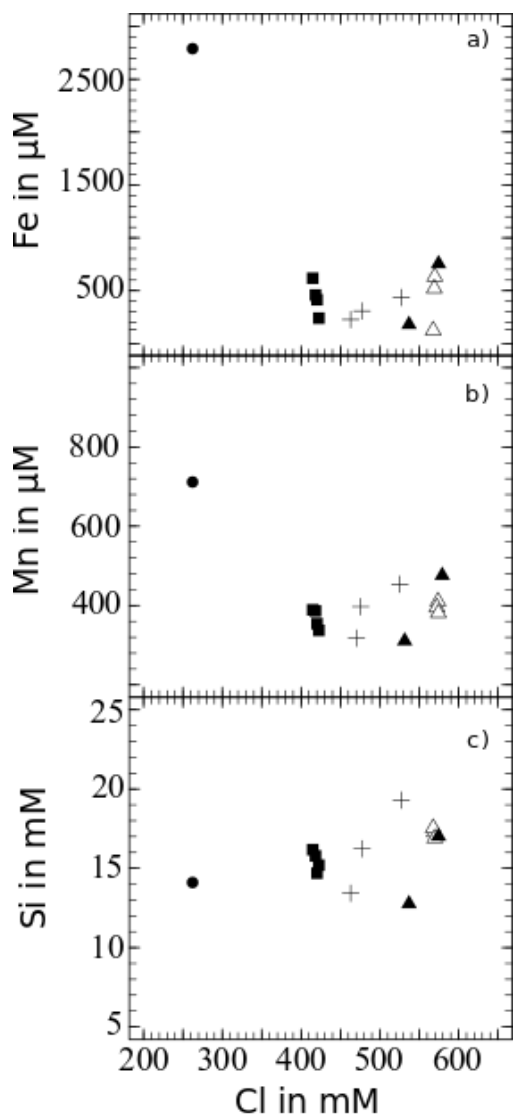
739 **Figure 2.** Overview of vent morphology. Snapshots of ROV videos (Ifremer-CNRS,
 740 MoMARSat 13), a), b) Capelinhos; c) Aisics; d) Montsegur; e) Cyprés; f) White Castle; g)
 741 Crystal; h) Sintra. a) and b) show candelabra-like structures that discharge focused high T fluid
 742 at Capelinhos vent site. c) and d) show hydrothermal mounds in the South Eastern hydrothermal
 743 area. The Aisics mound (c) is not as developed as the Montsegur mound (d) but both lack of
 744 tall indurated chimneys. e), f) and g) are situated in the South Western hydrothermal area.
 745 There, high-T vents are commonly set on elongated wall-like structures related to underlying

746 fissures. h) is situated in the waning North Eastern hydrothermal area and shows tall indurated
747 chimneys which are probably inherited from past intense activity.



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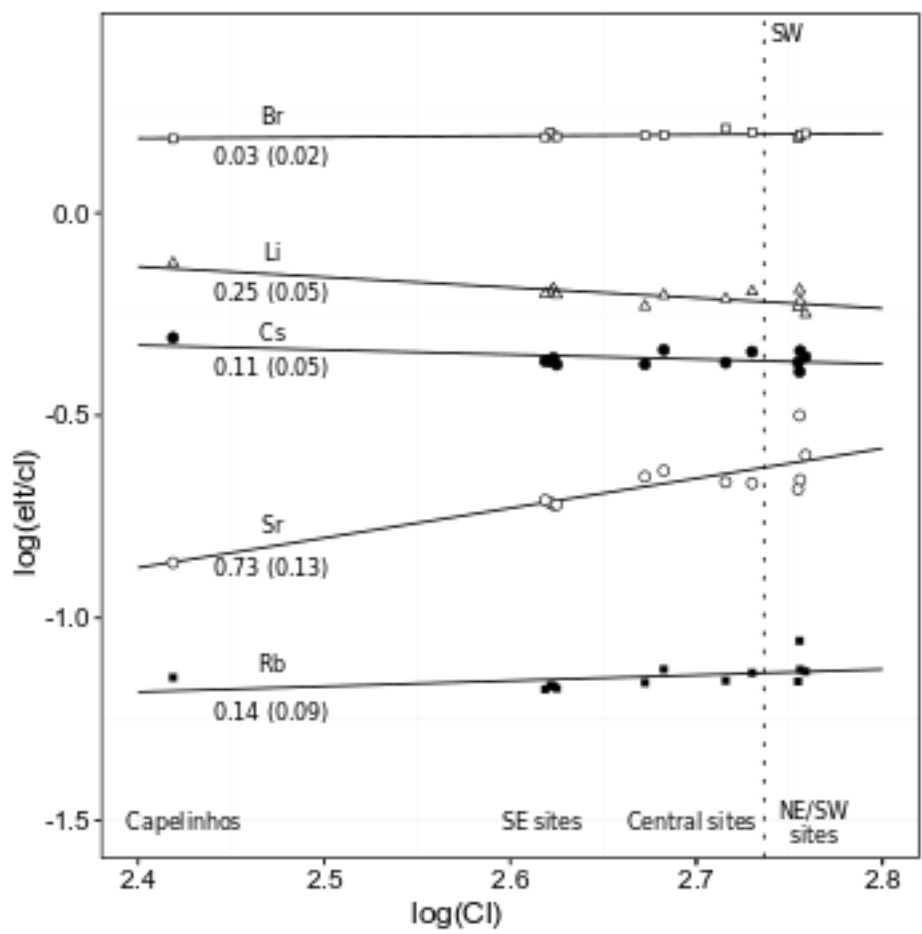
749 **Figure 3.** Fe, Mn and Si vs. Cl. in end-member fluids a) Fe versus Cl; b) Mn and versus Cl and
750 c) Si versus Cl. These diagrams show the lack of correlation of these elements relative to Cl.



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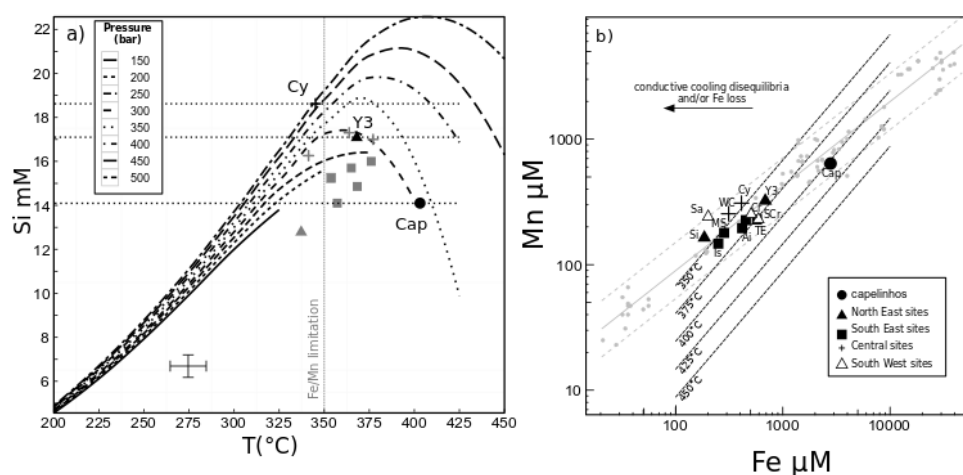
753 **Figure 4.** $\log(\text{Elt}/\text{Cl})$ vs $\log(\text{Cl})$ diagram. Diagram of elt/Cl versus Cl showing the linear
754 relationship between trace element and chlorinity, a proxy for the effect of phase separation.



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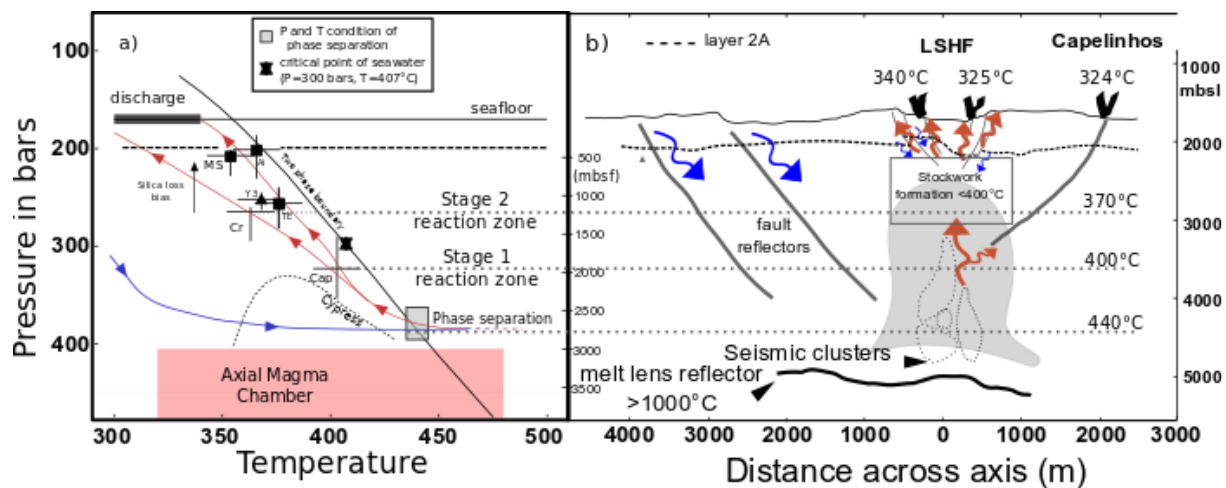
757 **Figure 5.** Geochemical information for P and T. a) Quartz geothermobarometer based on
 758 Foustoukos and Seyfried (2007b). Points represent Si measured in fluids with calculated
 759 temperature based on the Fe-Mn geothermometer. Grey dashed lines represent the “classical”
 760 approach to evaluate P and T (see text for details). b) Fe and Mn concentrations on a logarithm
 761 scale (Modified from Pester et al., 2011). Grey line represent basalt alteration line and black
 762 dashed lines are isotherms calculated from the Fe/Mn geothermometer.



763

764 **Figure 6.** Fluid circulation. a) Calculated P and T of some vents at equilibria with quartz are
 765 presented. P and T were calculated using the Fe/Mn geothermometer of Pester et al. (2011),
 766 quartz geothermobarometer (Foustoukos & Seyfried, 2007b) and NaCl-H₂O solution properties
 767 (Driesner et al., 2007) (See details in text). Dotted lines represent the depth of the base of layer
 768 2A for western and eastern sites (Arnulf et al., 2011). The red shaded domain represents the
 769 range of depth to the top of the axial magma chamber as identified by Singh et al. (2006) and
 770 Combiér et al. (2015). Grey lines represent fault labeled F1, F1b in Combiér et al. (2015), dotted
 771 grey line correspond to F2. The blue line represents the inferred P–T path in the recharge zone
 772 where cold seawater percolate through oceanic crusts and gradually interacts with rocks. Red
 773 lines represent the inferred P–T path in the upflow zone, based on the conditions estimated from
 774 fluid chemistry. The grey rectangle represents the phase separation zone estimated from the Si
 775 and Cl concentrations in the low salinity fluids (Capelinhos, TE) using the geothermobarometer

776 proposed by Fontaine et al. (2009). b) Modified from Escartin et al. (2015). Cartoon
 777 representing the hydrothermal cell under the LSHF. Blue arrows represent downward flow of
 778 seawater; red arrows are focused hot fluid moving upward through cracks and faults. Black star
 779 represents the hypothetic position of Capelinhos fluid extraction through a high angle fault.
 780 Seismic clusters are from Crawford et al. (2013). F1, F1b and F2 from Combier et al. (2015).



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782

787 **Table 2.** End-member composition of fluids from the LSHF. The concentration of end-member
788 fluids for each site is calculated based on linear least-square regression of element vs. Mg, at
789 Mg=0 (Albarède et al., 1981; Ravizza et al., 2001; Fig S1).

site	group	Ca mM	K mM	Na mM	Fe μM	Mn μM	Si mM	Cl mM	SO4 mM	Br mM	Rb μM	Sr μM	Li μM	Cs μM	87Sr/86Sr	T**	P**
Capelinhos	Cap	17.97±0.16	12.1±0.3	205.0±1.7	2789.4±84.8	639.5±27.6	14.1±1.1	262.3±0.1	-0.4±0.04	0.4±0.02	18.7±0.3	35.89±0.65	197.3±0.3	0.129±0.003	0.70384	403	350
Alarcs	SE	35.46±1.44	19.6±0.6	305.7±19.7	417.6±8.0	195.3±4.0	14.86±0.12	419.9±1.8	0.11±0.23	0.65±0.01	28.5±0.1	80.06±0.38	272.9±0.3	0.184±0.007	0.70422	368	225
Alarcs	SE	37.25±0.12	19.0±0.1	316.8±1.1	452.03±10.24	226.3±4.86	15.7±0.12	418.4±1.1	0.02±0.07	0.66±0.04	28.5±0.2	80.5±0.7	266.3±0.4	0.18±0.001	0.70428	365	n.d.
Montségur	SE	36.13±.46	19.6±0.1	325.3±0.8	283.7±20.7	179.2±8.4	15.25±0.04	421.7±1.3	-0.06±0.03	0.65±0.02	28.3±0.03	80.3±1.1	265±0.4	0.179±0.002	0.70423	354	225
Tour Eiffel	SE	39±0.64	19.7±0.4	334.4±6.5	574±68.19	229±15.51	15.1±0.82	415.7±4.7	0.9±1.29	0.64±0.02	27.7±1.3	81.16±0.53	262.6±0.4	0.179±0.008	0.70448	376	300
Cyprés	Central	43.24±0.03	23.4±0.1	395.34±2.2	411.33±16.64	309.01±11.58	18.62±0.27	519.7±2.7	-0.48±0.08	0.84±0.01	36.37±0.47	112.47±0.77	319±0.3	0.222±0.0074	0.70403	345	n.d.
Isabel	Central	39.6±0.37	22.3±0.2	373.8±5.7	250.42±21.29	147.05±1.92	14.1±0.3	470.3±11.2	-0.2±0.31	0.73±0.01	32.56±1.39	105.0±4.3	276.8±0.4	0.199±0.004	0.7043	357	n.d.
White Castle	Central	38.35±0.11	23.2±0.1	381.8±1.4	312.7±11.37	254.44±16.34	16.26±0.11	481.4±2.5	-0.2±0.01	0.75±0.01	35.95±0.1	111.19±0.28	301.6±0.1	0.221±0.003	0.70502	341	n.d.
Crystal	SW	53.55±3.9	28.4±0.1	445.9±0.4	507±68.55	260±19.4	17.3±0.77	569.5±0.3	2.56±2.69	0.88±0.01	50±5.65	180±45.85	367.9±0.3	0.231±0.004	0.70414	364	300
South Crystal	SW	49.3±0.8	28.3±0.1	445±1.5	593.4±31.7	231.8±10.04	17±0.12	569.9±1.3	0.1±0.02	0.89±0.05	42.5±0.11	125±0.42	346.1±0.4	0.260±0.004	0.704	377	300
Sarpins	SW	48.3±0.72	26.6±0.3	441.8±4.7	201.7±21.6	245.4±14.45	17.5±0.45	568.4±8.8	-0.28±0.97	0.87±0.01	39.6±0.87	118.1±2.36	332.9±.5	0.243±0.005	0.70429	322	n.d.
Sirtra	NE	53.7	27.1	433.2	185.5	164.1	12.8	537	0.35	0.85	39.3	115.5	343.5	0.244	0.70423	337	n.d.
Y3	NE	52.9±0.5	25.5±0.4	432.0±4.5	686.7±11.3	323.0±18.0	17.1±0.2	573.7±8.5	0.03±0.12	0.90±0.01	42.3±0.05	144.7±0.6	323.3±0.3	0.253±0.001	0.70401	368	300
Seawater		10.31	9.8	464	0	0	0.17	545	28.3	0.84	2.0	87.0	25.6		0.70916		

** : Temperature calculated by the Fe/Mn geothermometer and pressure estimated from Fe/Mn and Si concentrations (see text for details)

n.d. : not determined

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Supporting information for

Spatial Variations in Vent Chemistry at the Lucky Strike Hydrothermal Field, Mid Atlantic Ridge (37°N): Updates for Subseafloor Flow Geometry from the Newly Discovered Capelinhos Vent.

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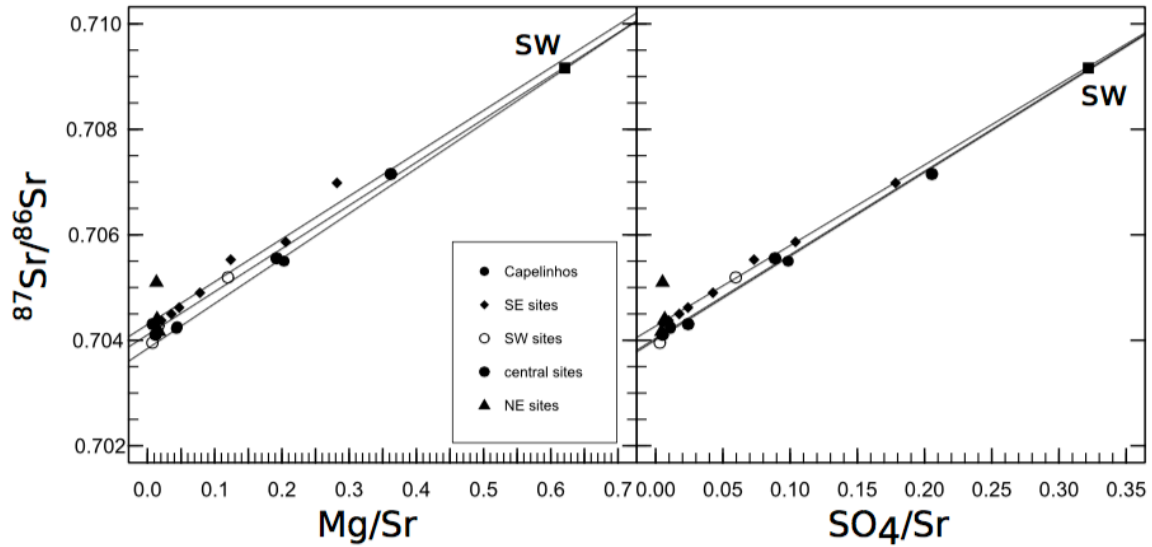
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- Figure S1
- Table S1

Introduction

This supporting information provide geochemical and Sr isotopic composition of hydrothermal fluids collected at LSHF.



811

812 **Figure S1.** Diagram of $^{87}\text{Sr}/^{86}\text{Sr}$ as a function of Mg/Sr in (a) and SO_4/Sr in (b). SW stands for
 813 seawater. a) shows the usual way of determining the Sr isotope signature of end-member fluids
 814 based on the Mg/Sr ratio. b) is an alternative method based on sulfate concentration. Sulfate
 815 contents of fluids will be dependent on seawater entrainment during sampling, as SO_4 is
 816 enriched in seawater compared to pure hydrothermal fluid. Sulfate will also be dependent on
 817 anhydrite. Dissolution of anhydrite may contribute to Ca, SO_4 and Sr concentrations measured
 818 in the fluid and may modify its Sr isotope signature. Assuming Sr isotope signatures of seawater
 819 and hydrothermal fluid, dissolution of anhydrite dissolution bias can be corrected as shown in
 820 Fig S1b. Consequently, endmember determined for South-West sites are better determined by
 821 SO_4/Sr least square regression. When no suspicion of anhydrite dissolution, Mg/Sr should be
 822 preferred instead of SO_4/Sr , i.e. for slightly negative end-member SO_4 value (e.g. Capelinhos).
 823 All the data are reported in Table S1.

824

825 **Table S1.** Geochemical compositions and Sr isotope signatures of hydrothermal fluids
826 collected during the MoMARSAT'13 cruise. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of hydrothermal end-members are
827 calculated by linear extrapolation to zero Mg/Sr or SO_4/Sr of the least-square regression method
828 (r^2 are provided). All geochemical measurements made aboard the cruise are available in the
829 cruise report at the following doi: <http://dx.doi.org/10.17600/13030040>.

site	groupe	sample	Cl mM	Mg mM	SO4 mM	Sr μM	Mg/Sr	SO4/Sr	$^{87}\text{Sr}/^{86}\text{Sr}$	r2
Capelinhos	cap	M13FLU028	322.7	8.69	4.22	42.78	0.203	0.099	0.705501	
Capelinhos	cap	M13FLU029	275	1.68	0.42	38.98	0.043	0.011	0.704226	
Capelinhos	cap	M13FLU031	265.4	1.63	0.40	36.86	0.044	0.011	0.704256	
		end-member					0		0.70384	0.999
		end-member						0	0.704034	0.999
Aisics	SE	M13FLU002	466.1	16.77	8.48	81.55	0.206	0.104	0.705865	
Aisics	SE	M13FLU003	435	2.86	1.41	80.42	0.036	0.018	0.704499	
Aisics	SE	M13FLU042	429.2	1.14	0.55	82.34	0.014	0.007	0.704386	
Aisics	SE	M13FLU043	442.3	6.28	3.43	80.44	0.078	0.043	0.7049	
Montségur	SE	M13FLU010	434.1	3.86	1.97	81.92	0.047	0.024	0.70462	
Montségur	SE	M13FLU011	431.7	1.63	0.76	79.51	0.02	0.01	0.704376	
Tour Eiffel	SE	M13FLU044	424.5	0.89	0.42	80.61	0.011	0.005	0.704324	
Tour Eiffel	SE	M13FLU045	487.1	23.74	15.03	84.21	0.282	0.178	0.706983	
Tour Eiffel	SE	M13FLU047	451.2	10.27	6.07	82.92	0.124	0.073	0.705528	
		end-member					0		0.70430	0.987
		end-member						0	0.704267	0.998
Cyprès	Central	M13FLU039	530.7	1.93	0.48	111.73	0.017	0.004	0.704174	
Isabel	Central	M13FLU008	497.6	1.55	0.73	108.61	0.014	0.007	0.704413	
White Castle	Central	M13FLU015	483.4	1.47	0.57	110.25	0.013	0.005	0.705108	
		end-member					0		0.704453	0,969
		end-member						0	0.704487	0.971
Crystal	SW	M13FLU022	563.0	19.16	8.85	99.66	0.192	0.089	0.705554	
Crystal	SW	M13FLU023	572.6	1.61	4.93	203.94	0.008	0.024	0.704307	
Sapins	SW	M13FLU049	576.6	33.87	19.22	93.54	0.362	0.205	0.707153	
South Crystal	SW	M13FLU017	573.9	1.49	0.64	123.70	0.012	0.005	0.704104	
		end-member					0		0.704102	0,997
		end-member						0	0.704004	0,998
Sintra	NE	M13FLU027	550.1	12.99	6.46	108.5	0.124	0.06	0.705191	
Y3	NE	M13FLU032	601.2	1.06	0.45	144.61	0.007	0.003	0.703953	
Y3	NE	M13FLU035	578.4	2.29	1.15	141.84	0.016	0.008	0.704275	
		end-member					0		0.704071	0,997
		end-member						0	0.70909	0,996
		seawater (SW)		54	28	87	0.621	0.322	0.709162	

830