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Gaëtan Launay, Stanislas Sizaret, Laurent Guillou-Frottier, Eric Gloaguen, Filipe Pinto. Deciphering fluid flow at the magmatic-hydrothermal transition: A case study from the world-class Panasqueira W–Sn–(Cu) ore deposit (Portugal). *Earth and Planetary Science Letters*, 2018, 499, pp.1-12. 10.1016/j.epsl.2018.07.012 . hal-01955965

HAL Id: hal-01955965

<https://brgm.hal.science/hal-01955965v1>

Submitted on 14 Dec 2018

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1 **Deciphering fluid flow at the magmatic-hydrothermal**
2 **transition: a case study from the world-class W-Sn-(Cu) ore**
3 **deposit of Panasqueira (Portugal)**

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18 Manuscript submitted to Earth & Planetary Science Letters, the 26th of January 2018
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38 **Abstract:** Deciphering the behavior of fluid flow at the magmatic-hydrothermal transition is
39 crucial to understand physical processes leading to the formation of intrusion-related ore deposits
40 and hence to predict the localization of mineralized bodies. However, the hydrodynamics
41 (direction and velocity) of the hydrothermal fluid flow related to this transitional stage remains
42 poorly constrained. Here we present a coupled textural and chemical study performed on
43 tourmaline growth bands to constrain fluid flow during the initiation of the hydrothermal system
44 of the W-Sn-(Cu) Panasqueira deposit. This exceptional deposit consists of a dense network of flat
45 wolframite and cassiterite-bearing quartz veins intensely developed above a well-known hidden
46 greisen cupola. The W-Sn mineralization is preceded by a tourmalinization stage occurring as
47 metasomatic halos around veins and as selvages developed along the vein-wall-rocks contacts.
48 Results emphasize the key role of the greisen cupola on fluid focusing and the role of fluid
49 overpressure during the vein opening. Velocity values highlight an efficient transport of metals in
50 veins largely dominated by advective process (10^{-4} to 10^{-3} m.s⁻¹), whereas fluid flow in the altered
51 wall-rocks is slow and pervasive (10^{-6} to 10^{-5} m.s⁻¹), suggesting that the element transport through
52 the metasedimentary host rock was low and limited to the alteration haloes. LA-ICP-MS analyses
53 of tourmaline growth bands reveal that fluids coming from the cupola are enriched in Na, K, Li, Sr
54 and Sn, thus emphasizing the contribution of magmatic fluids during the vein formation and the
55 metasomatic alteration of the wall-rocks. More generally, this study demonstrates that the apical
56 portions of granite bodies play as emanative centers of mineralized fluids, and highlights the
57 usefulness of mineral growth band analysis in the search for intrusion-related mineralization.

58
59 **Keywords:** magmatic-hydrothermal activity, Panasqueira, tourmaline, growth bands, flow
60 direction, flow velocity

66 **1. Introduction**

67 The formation of hydrothermal ore deposits and the development of alteration halos in rocks
68 involve the transfer of a large amount of fluids through permeable rocks during several episodic
69 pulses integrated over a long period of time (Barnes, 1997; Robb, 2005). To form a high-grade
70 deposit, the flow of metal bearing fluids must be focused in a restricted space and coupled with an
71 efficient precipitation and trapping mechanism. Hence, understanding hydrothermal fluxes
72 constitutes a major issue of metallogeny and requires resolving some fundamental questions
73 related to: (i) the driving force of fluid flow and (ii) the direction and the velocity of fluid flow.
74 The search for answers to these issues involves a combination of approaches including recognition
75 of permeable pathways (Sibson, 1992; Cox 2005), mineralogical and geochemical analysis
76 (Cartwright and Buick, 1996; Ferry et al., 2002) and fluid hydrodynamics (Ingebristen and
77 Appold, 2012).

78 Among hydrothermal ore deposits, those triggered and driven by magmatic felsic intrusions
79 represent a main primary source of economically important metals, including copper (Cu),
80 tungsten (W), tin (Sn), molybdenum (Mo) and gold (Au) (Hedenquist and Lowenstern, 1994;
81 Černý et al., 2005). There is general agreement that these magmatic-related deposits are partly
82 formed by releasing and expulsion of metal-bearing fluids during the emplacement and the cooling
83 of magmatic intrusions (Hedenquist and Lowenstern, 1994; Černý et al., 2005). Another important
84 process for metal transport in magmatic-hydrothermal systems is the development of large
85 convective cells around plutons driven by the thermal contrast between intrusive bodies and the
86 host rocks. This convective fluid flow is able to mobilize a large amount of fluids from the
87 surrounding host rock and has a marked effect on the fluid flow patterns (Norton and Cathles,
88 1979; Eldursi et al., 2009). More specifically, it has been commonly observed that mineralized
89 systems are usually centred on small plutons, apices or feeder zones (Dines, 1956; Dilles and
90 Proffett, 1995; Gloaguen et al., 2014) suggesting a strong control of the pluton roof geometry on
91 mineralizing fluid flow. Indeed, some numerical models have demonstrated that fluid flow is
92 optimal above the apical part of intrusions and predict the formation of ore deposits in these areas

93 (Sams and Thomas-Betts, 1988; Eldursi et al. 2009; Weis et al. 2015). Despite these elegant
94 numerical results, this expected fluid flow pattern (flow direction and velocity) needs to be
95 verified by field investigations.

96 Moreover, it is widely recognized that magmatic-hydrothermal systems integrate a complex
97 history encompassing episodic stages of fluid flow related to polyphase intrusion emplacement,
98 which have important effects on the geometry of hydrothermal systems. In this scheme, the
99 magmatic-hydrothermal transition is a key event that marks the initiation of the hydrothermal
100 system and defines the future behavior of fluid flow. Indeed, it is usually during this transitional
101 stage that permeable structures open and drain mineralized fluids. However, fluid flow related to
102 this specific hydrothermal stage is difficult to access and thus remains poorly constrained due to
103 the lack of recognized methods to implement in the field.

104 The textural analysis of mineral growth bands has provided significant information about the
105 direction and velocity of fluid flow involved in the formation of hydrothermal ore deposits
106 (Kessler et al. 1972) and during metasomatic processes (Sizaret et al., 2009; Majhoubi et al.,
107 2016). This direct approach constitutes a powerful tool to constrain fluid flow related to a specific
108 hydrothermal event such as the magmatic-hydrothermal transition. In the present study, we
109 perform a textural analysis of tourmaline growth bands to decipher fluid flow at the initiation of
110 the magmatic-hydrothermal stage of Panasqueira (Portugal). This textural analysis is combined
111 with trace elements analysis of tourmaline growth bands to constrain the chemical characteristics
112 of fluids from which the tourmalines have crystallized. Panasqueira is an historic world-class W
113 vein deposit characterized by a well-preserved magmatic-hydrothermal plumbing system. This
114 deposit is one of the best-documented and characterized in the world and represents a reference
115 site to study magmatic-hydrothermal processes leading to the formation of a Sn-W ore deposit
116 (Thadeu, 1951; Kelly and Rye 1979; Poyla, 1989; Poyla et al., 2000; Foxford et al., 2000;
117 Lecumberri-Sanchez et al., 2017; Codeço et al., 2017). From the results of estimated fluid
118 directions and velocities, we discuss the key parameters controlling the behavior of fluid flow at
119 different scales, such as: (i) the role of granitic cupolas on the localization of mineralized areas by
120 fluid flow focusing, (ii) the role of fluid overpressure conditions on vein opening and propagation

121 of alteration halos in wall-rocks, and (iii) the required duration of fluid flow during the W-Sn
122 mineralization stage.

123 **2. Geological and metallogenic framework**

124 *2.1 Regional geology*

125 The W-Sn-(Cu) Panasqueira ore deposit is located in the Central Iberian Zone (CIZ), which
126 constitutes the axial zone of the Iberian Variscan belt (Fig. 1a) (Julivert et al., 1972). The CIZ is
127 composed of the Schist-Greywacke-Complex (SGC) intruded by a large volume of granitoid
128 intrusions resulting from an extensive magmatic event during the latest stages of the Variscan
129 orogeny (Dias et al., 1998). The SGC consists of a thick (8-11 km) sequence of late
130 Neoproterozoic schists and greywackes affected by a regional greenschist metamorphism
131 (Martínez-Catalán et al., 2007). Deformation of the SGC consists of tight and upright folds with
132 NW-SE sub-vertical axial planes produced by a NE-SW shortening related to the earliest
133 compressive stage of the Variscan orogeny (Dias and Ribeiro, 1995 and Martínez-Catalán et al.,
134 2007). This folding is associated with a regional sub-vertical penetrative axial planar cleavage (S1)
135 (Fig. 1a and 1b). The CIZ host numerous Sn-W ore deposits and occurrences related to granitic
136 intrusions and formed by magmatic-hydrothermal processes (Derré, 1982).

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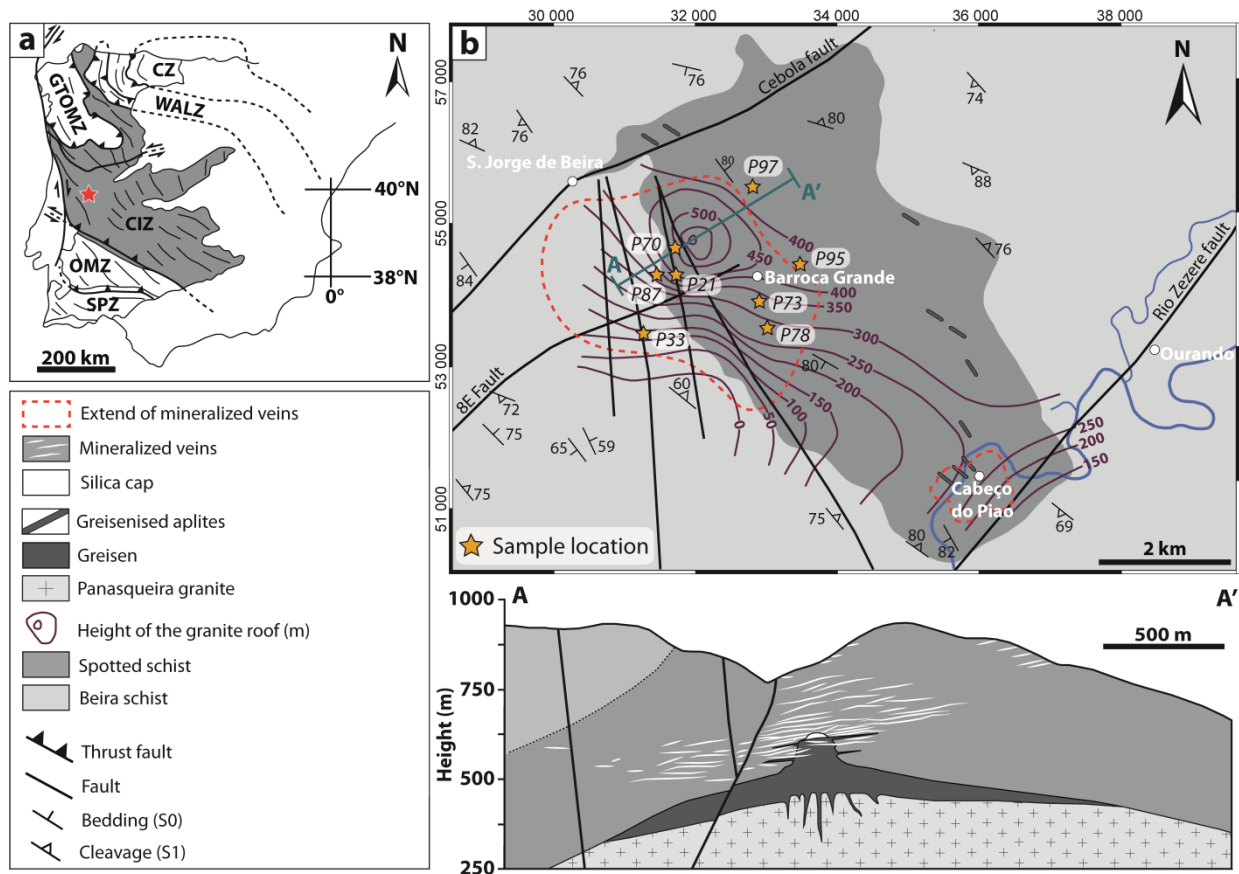


Figure 1 (a) Tectonic map of the Variscan Iberian massif showing the main tectonostratigraphic domains defined by Julivert *et al.*, (1972). The location of the Panasqueira deposit is marked by a red star. CZ: Cantabrian Zone, WALZ: West Asturian-Leonese Zone, GTOMZ: Galicia Tràs-os-Montes Zone, CIZ: Central Iberian Zone, OMZ: Ossa-Morena Zone and SPZ: South Portuguese Zone (b) Geological map of the Panasqueira ore deposit including the height of the granite roof. The (A-A') geological cross section displaying the spatial relationship between mineralized veins and the greisen cupola (data source Beralt Tin & Wolfram S.A.).

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139 2.2 The W-Sn-(Cu) Panasqueira ore deposit

140 The Panasqueira ore deposit is located in the Beira Baixa province in the southern flank of the
 141 Serra da Estrela massif constituted by late to post tectonic granitic intrusions. In this region the
 142 late Neoproterozoic folded and metamorphosed SGC is locally named Beira Schist. At
 143 Panasqueira, spotted schists mark the presence of a thermal metamorphic aureole related to an
 144 underlying S-type granite (Thadeu, 1951; Clark, 1964). The intrusive body is limited by the
 145 Cebola fault to the north and the Rio Zêzere fault to the south (Fig. 1b) (Clark, 1964). As exposed
 146 by underground mining works and drill cores, the upper part of this granite includes a quartz-

147 muscovite greisen cupola formed by interaction between granitic rocks and acidic F-B and Li-rich
148 fluids (Shcherba, 1970; Bishop, 1989) (Fig. 1b and 2a).

149 The W-Sn mineralization is hosted by a dense network of low dipping veins crosscutting the
150 vertical foliation of the Beira Schist and the greisen cupola (Fig. 2a). The veins system is centered
151 above the greisen cupola and extends over an area of 6 km² for a depth extension of about 200-300
152 meters (Fig. 1b) (Kelly and Rye 1979; Poyla et al., 2000). This unusual shallow dipping
153 orientation of mineralized veins is compatible with the regional stress field that involved NE-SW
154 shortening with a vertical σ_3 leading to the formation of tight and upright folds accompanied by
155 the development of the regional NW-SE sub-vertical foliation (Dias and Ribeiro, 1995; Martínez-
156 Catalán et al., 2007). Based on textural and structural analysis of the vein swarms, Foxford et al.,
157 (2000) proposed a crack-and-seal model involving episodic vein dilation and filling induced by
158 injections of fluids at supralithostatic pressure conditions in a compressive crustal regime.
159 Although this mechanism is quite appealing, these conditions of fluid overpressure in veins remain
160 unproven. The structural control of the vein formation has probably induced permeability
161 anisotropy with a strong horizontal component which could affect the pattern of fluid flow at the
162 mineralization time. A late NNW-SSE and ENE-WSW sub-vertical fault system crosscuts the
163 veins and the granite intrusion (Fig. 1b) (Thadeu 1951, Kelly and Rye 1979, Foxford et al., 2000).
164 As evidence in the field, these faults postdate mineralization event and have not controlled fluid
165 flow during the W-(Sn) mineralization (Thadeu, 1951).

166 The complex paragenetic sequence of veins was described in detail by Kelly and Rye (1979)
167 and improved by Poyla et al., (2000). At least five mineralization stages occurred in the veins
168 from oldest to youngest: the Quartz-Tourmaline (QTS) stage, the Main Oxide Silicate Stage
169 (MOSS), the Main Sulfide Stage (MSS), the Pyrrhotite Alteration Stage (PAS) and the Late
170 Carbonate Stage (LCS). The MOSS (Fig. 2b) and the MSS (Fig. 2c) are the most economically
171 relevant ore stages. The MOSS carries the W-Sn mineralization and consists in muscovite and
172 quartz accompanied by wolframite and cassiterite. The two earliest mineralization stages (QTS
173 and the MOSS) constitute a continuous sequence which is then superimposed and/or cross-cut by
174 the late sulfide stage (MSS). The MSS, which carries the Cu mineralization, is mainly composed

175 of arsenopyrite, chalcopyrite, sphalerite pyrite and pyrrhotite (Thadeu 1951, Kelly and Rye 1979,
 176 Poyla *et al.* 2000). Based on fluid inclusions analyses, Kelly and Rye (1979) and Bussink (1984)
 177 have reported that fluids related to the MOSS and the QTS have been trapped at temperatures
 178 between 300 and 350°C. Although the tourmaline stage (QTS) is volumetrically of minor
 179 importance (mm to cm of thickness), it marks the initiation of the Panasqueira hydrothermal
 180 system (Poyla *et al.*, 2000).

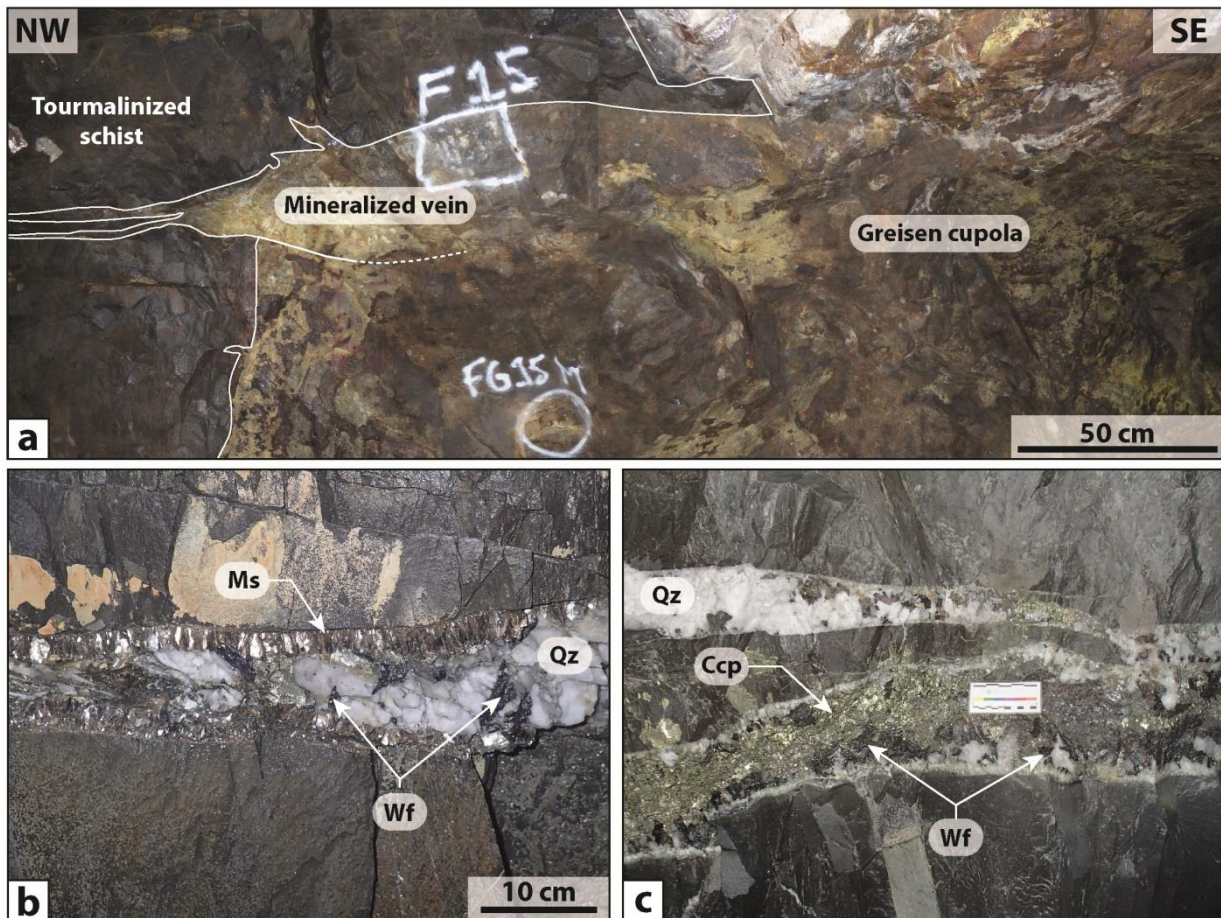


Figure 2 The mineralized system of the W-Sn-(Cu) Panasqueira deposit. (a) Contact between the greisen cupola and the tourmalinized metasedimentary host rock at the level 1 of the mine. Note the presence of mineralized veins which crosscut this contact. (b) Typical Quartz-Wolframite veins corresponding to the main oxide stage. (c) Quartz-Wolframite veins superimposed by the late sulfide stage carrying the Cu mineralization. Abbreviation from Whitney and Evans (2010) Ccp: Chalcopyrite, Ms: Muscovite, Qz: Quartz and Wf: Wolframite.

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185 **3. Tourmaline occurrence**

186 At Panasqueira, tourmaline is common, especially in metasedimentary host rock surrounding
187 mineralized veins (Fig. 3a). Tourmaline is present within alteration halos, which are developed
188 along schist-vein contacts within the lower and the upper wall-rocks. These halos result from
189 metasomatic reactions induced by fluid–rock interactions during hydrothermal fluid flow in the
190 veins. Locally, it is possible to observe alternation of quartz-rich and tourmaline-muscovite rich
191 layers inherited from lithological heterogeneities of the Beira Schist. These tourmalines are fine
192 and their trigonal axes are randomly oriented in the vertical cleavage planes of the schist.

193 Tourmaline is also present in veins along schist-vein contacts
194 (Fig. 3b and 3c). They occur as fine acicular crystals (1-3 mm) with $\langle c \rangle$ axes vertical and
195 perpendicular to the vein edge and consequently to the inferred plane of fluid flow (Fig. 3c). These
196 tourmalines grains are commonly associated with apatite, rutile and muscovite (Fig. 3d), while
197 their relation with the ore minerals (wolframite and cassiterite) is difficult to observe. However, in
198 some veins tourmaline appears to shortly predate wolframite and can be texturally associated with
199 cassiterite (Fig. 3e). Consequently, tourmaline represents a good candidate to track W-Sn
200 mineralized fluid flow related to the QTS and the MOSS in the Panasqueira veins system.

201 For both types of tourmaline, the sections normal to the $\langle c \rangle$ axis are characterized by growth
202 band zonation with a core and well-defined rims (Fig. 3d and Fig. 3f). These bands emphasize an
203 anisotropic growth and highlight a symmetry breakdown due to a directional fluid flow (Fig. 4a
204 and 4b).

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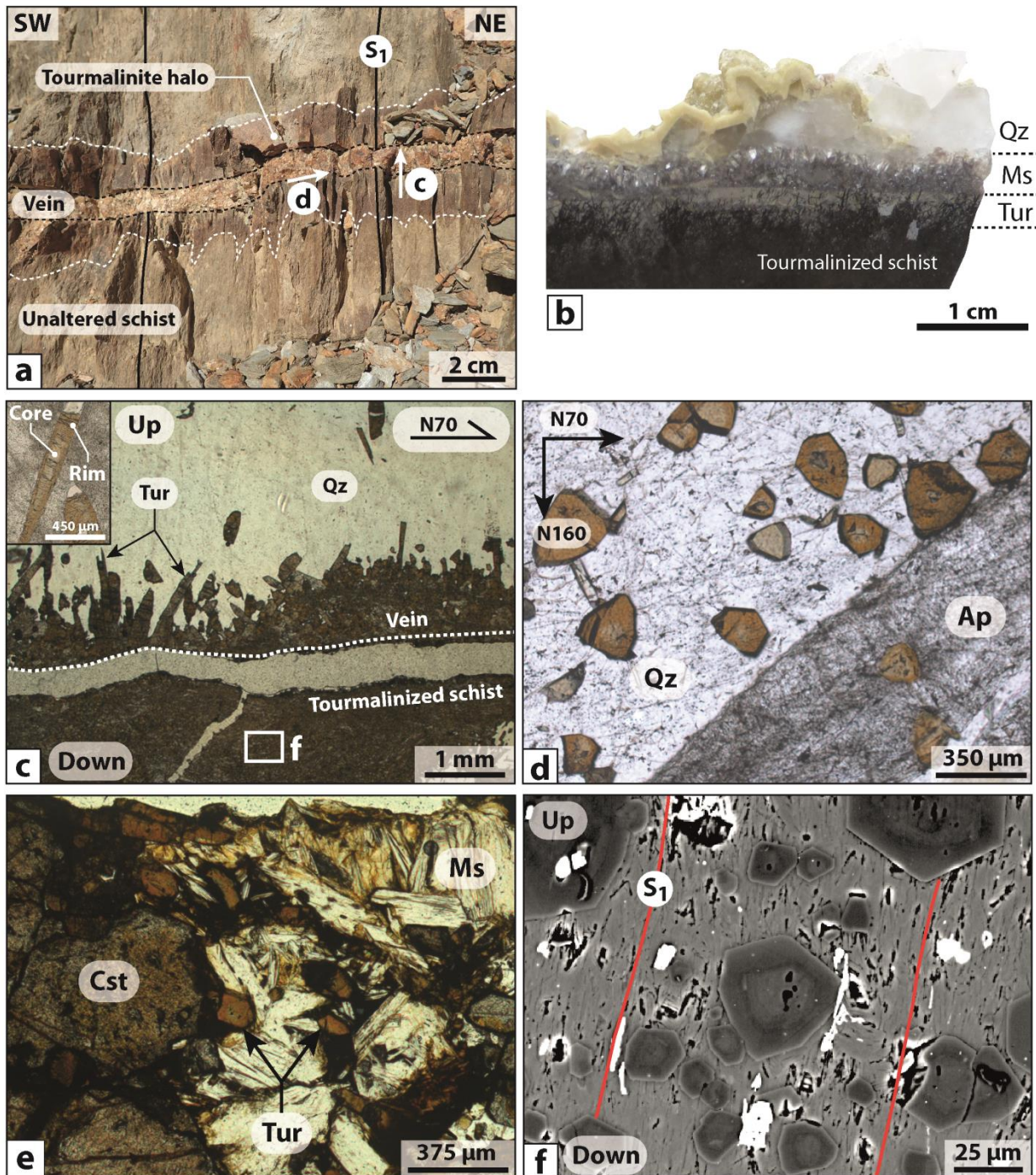


Figure 3 Textural and petrographic characteristics of tourmalines present in the mineralized system of Panasqueira. (a) Quartz-tourmaline vein with tourmalinization of the metasedimentary wall-rocks. The white dashed line indicates the tourmalinization front in the metasedimentary host rock. c and d arrows give orientation of the thin sections c and d. Note the preferential migration of tourmalinization front in the vertical foliation planes S_1 (b) Sample of mineralized veins showing the vertical growth of acicular tourmalines along the schist-vein contact. (c) and (d) photomicrographs of vein tourmaline showing sections parallel (c) and normal (d) to the trigonal axis of tourmaline. (e) Photomicrograph of mineralized vein showing tourmaline grains associated with cassiterite. (f) SEM photomicrograph of altered schist perpendicular to the foliations planes showing growth band zoning in tourmaline basal sections. Abbreviation from Whitney and Evans (2010) Ap: Apatite, Cst: Cassiterite, Ms: Muscovite and Qz: Quartz.

4. Sampling and methodology

Owing to its remarkable mechanical and chemical stability, tourmaline is able to record significant information about its host environment (Henry and Dutrow, 1996; Van Hinsberg et al. 2011a). In particular, tourmaline provides a powerful tool to constrain hydrodynamics (direction and velocity of fluid flow) of geological processes involving fluid flow (Sizaret et al., 2009; Mahjoubi et al., 2015). Moreover, tourmaline incorporates a diversity of trace elements and constitutes a geochemical monitor of fluid compositions (Marks et al., 2013; Van Hinsberg et al., 2017). In this study, tourmaline growth bands were used to track both flow and chemical composition of hydrothermal fluids involved during the earliest stages of the vein formation at Panasqueira.

Oriented vein samples with tourmaline were collected in the mine and at the surface, at different places around the cupola, both horizontally and vertically to have a full spatial distribution, and to establish a paleo-fluid flow map around the granitic intrusion (Fig. 1b). Unfortunately, no samples were available in the northwestern part of the studied area.

The vertical migration of the alteration front (tourmalinization) within the metasedimentary host rocks appears to be driven by the sub-vertical foliation planes (S1). To study the vertical fluid flow related to the alteration of the wall-rocks, samples of alteration halos have been collected above and below veins perpendicularly to the vertical foliation planes (NE-SW section). The orthogonal NW-SE sections are parallel to the foliation planes and hence do not permit to perform fluid flow reconstruction in the wall rock due to the preferential crystallization of tourmaline in parallel within the foliation planes.

4.1 Fluid flow reconstruction by growth band measurements

In hydrothermal systems, the crystal shape is controlled by its intrinsic symmetry and the effects of external forces, such as fluid flow, that induce anisotropic growth (Curie 1908). The method used for reconstruction of local fluxes is based on the notion that crystal faces exposed to the chemical flux (i.e. the upstream faces) grow faster than downstream faces (Kessler et al. 1972 and Sizaret et al., 2006 and 2009). The relationship between flow velocity and the relative growth

234 rate of upstream and downstream faces can be modelled and quantified (Sizaret et al. 2006 and
235 2009). From this postulate, it is possible to deduce local fluid flow directions and velocities in the
236 hydrothermal system. Oriented thin sections were cut parallel to the expected stream-line, i.e.
237 parallel to veins, within the vein, and vertically and perpendicular to foliation planes in altered
238 host rocks. Tourmaline crystals were examined on polished thin sections using optical transmitted
239 light and scanning electron microscope. Then, on tourmaline basal section (i.e., perpendicular to
240 the trigonal axis) the growth band thickness of equivalent crystal faces (d_1 , d_2 and d_3) and their
241 respective directions (α_1 , α_2 and α_3) have been measured (Fig. 4c). The three normalized
242 thicknesses d_{\max} , d_{int} (intermediate) and d_{\min} were determined. Assuming that the thickest band
243 (d_{\max}) received the largest chemical flux, the orientation of d_{\max} gives the direction of the paleo-
244 fluid flow recorded by the crystal (Fig. 4c).

245 The relationship between fluid velocity and growth ratios (d_{\max}/d_{\min}) was established by
246 solving the coupled Navier-Stokes and chemical transport equations with the Comsol Multiphysics
247 finite element code (Sizaret et al., 2009). In this modeling the edge size of tourmaline is 50 μm
248 and the fluid (water) viscosity is chosen at realistic mineralizing conditions, taken here at 350°C
249 ($7.3 \cdot 10^{-5}$ Pa.s source NIST WebBook, 2017). Varying flow velocity provides a relationship
250 between chemical flux ratio (upstream/downstream) and fluid velocity (Fig. 4d). The growth band
251 thicknesses measured on minerals can be considered as chemical fluxes integrated over a given
252 period of time. Consequently, the fluid velocity can be determined by inverse method by reporting
253 the measured d_{\max}/d_{\min} ratio on the flux ratio (Upstream/downstream) vs velocity curve (Fig. 4d)
254 (Sizaret et al. 2006 and 2009).

255 To obtain representative measurements, basal sections of tourmaline were chosen from the
256 following criteria: absence of contact with another crystal of tourmaline, absence of
257 recrystallization and corrosion and easily identifiable equivalent faces.
258 For each site, flow directions and velocities deduced from each tourmaline section were
259 implemented and treated with the free meteorological software WRPLOT View™ (Lake
260 Environmental, 2016) in order to establish “wind roses” of local paleo-fluid flow. These rose
261 diagrams are then reported on the map.

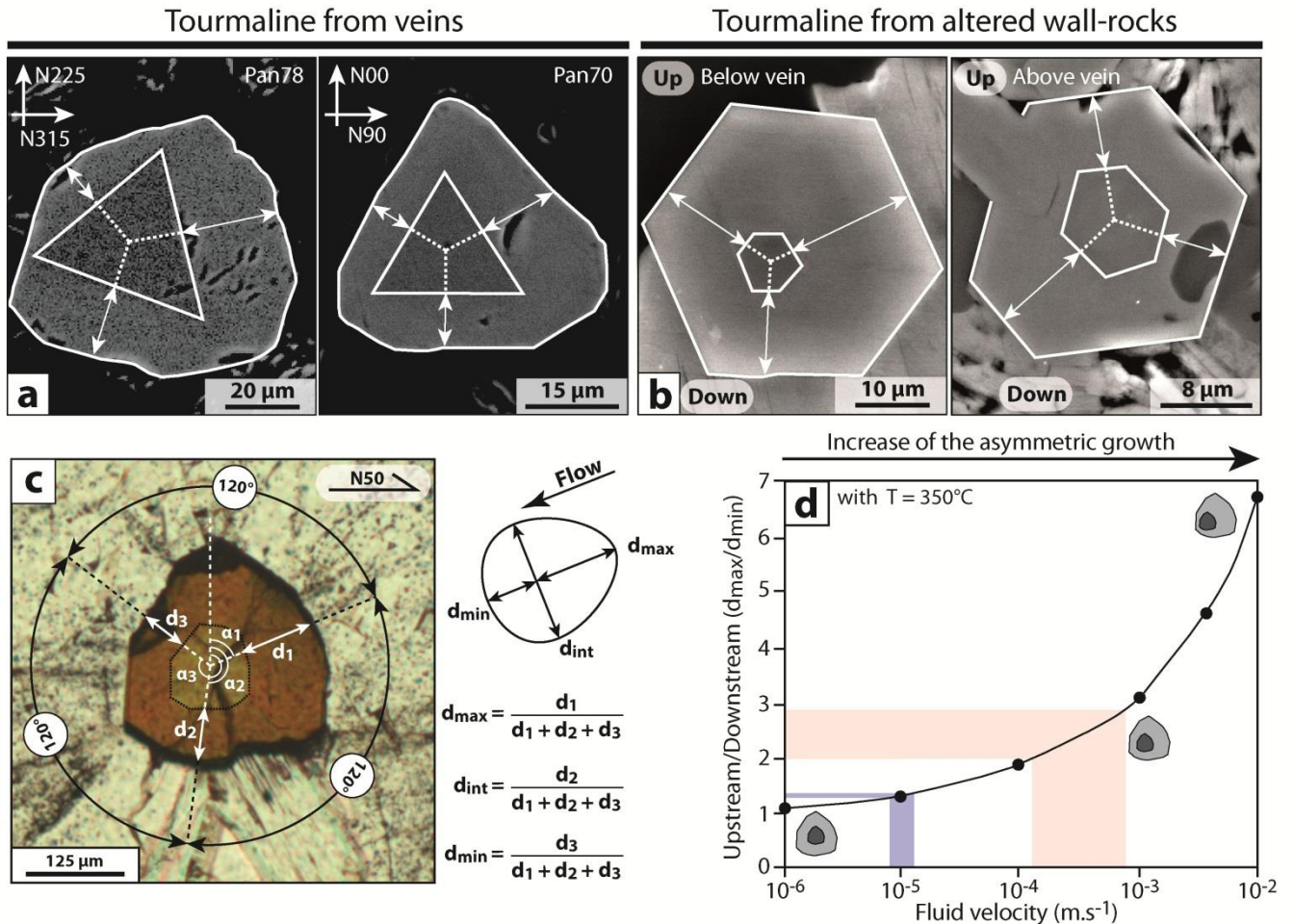


Figure 4 Examples of basal sections of tourmaline from veins (a) and altered wall-rocks (b) displaying an asymmetric shape due to an anisotropic growth (c) Measurements of growth bands thickness and orientation on a basal section of tourmaline. (d) Curve deduced from numerical modeling showing relationship between fluid velocity and flux ratio (see text for details). The filled boxes indicate range of averages of fluid velocities derived from d_{\max}/d_{\min} ratio measured on tourmalines from altered wall-rocks (blue) and veins (red).

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263 4.2 Tourmaline growth band chemistry

264 4.2.1 Major and minor element analysis

265 The major and minor element compositions of tourmaline were determined on polished thin
 266 sections using a Cameca SX-Five electron probe micro-analyzers (EPMA) at the Institut des
 267 Sciences de la Terre d'Orléans (ISTO). Analyses were performed using an accelerating voltage of
 268 15 kV, a beam current of 6 nA with a diameter of 2 μm. The elements analyzed were Si, Ti, Al,
 269 Fe, Mn, Mg, Ca, Na, K, F and Cl. For calculation of the structural formulae and site allocation in
 270 tourmaline, analyses were normalized to 15 cations (T + Z + Y-sites) and B₂O₃ and H₂O contents

271 were calculated by stoichiometry, considering B = 3 apfu and OH + F + Cl = 4 apfu. Analytical
272 conditions and calibration are detailed in electronic supplementary materials (ESM1).

273 4.2.2 Trace element analysis

274 Trace element concentrations in tourmaline were determined by laser ablation inductively
275 coupled plasma-mass-spectrometry (LA-ICP-MS). These analyses were performed at BRGM
276 (French Geological Survey) using a ThermoScientific X series II quadrupole ICP-MS coupled
277 with a Cetac Excite 193 nm laser ablation system. Ablations were performed with a repetition rate
278 of 8Hz and laser energy of 3.5 J/cm² with a beam size of 40 μm. Analyses comprise 20 seconds of
279 gas blank measurement following by 60 seconds of ablation. Elements analyzed were ⁷Li, ²³Na,
280 ²⁴Mg, ²⁷Al, ²⁹Si, ⁴⁷Ti, ⁵¹V, ⁵⁵Mn, ⁶⁶Zn, ⁸⁸Sr and ¹¹⁸Sn. Data reduction was performed with the
281 software Glitter 4.0 using the NIST SRM-612 glass standard for external standardization and ²⁹Si
282 as the internal standard. Concentrations in Mg and Al obtained by LA-ICP-MS data reduction
283 were compared with EPMA analyses to check the accuracy of the results. Analyses of certified
284 standard NIST-612 SRM give a good accuracy with a relative standard deviation (RSD) below
285 10%. Detection limits for most elements range between 0.2 and 5 ppm. Higher detection limits
286 were achieved for Zn, Ti and Na (respectively 5.5 ppm, 15.4 ppm and 37.4 ppm).

287 5. Results

288 5.1 Large scale fluid flow

289 Fluid flow directions and velocities are summarized in Table 1 (complete measurements in
290 electronic supplementary materials ESM2) and illustrated in Figure 5. For veins-hosted
291 tourmaline, an average ratio of d_{\max}/d_{\min} was determined for each of the 8 sample sites. These
292 average ratios range from 2.0 to 2.9 implying a fluid velocity varying from 10⁻⁴ to 10⁻³ m.s⁻¹ in
293 veins (Table 1 and Fig. 4d). Figure 5c shows the histogram representation of all velocity values,
294 which are characterized by an asymmetric distribution centered on 10⁻⁴ - 5.10⁻⁴ m.s⁻¹. This wide
295 variable distribution is probably due to the sinuous nature of the fluid flow path. Indeed, a
296 narrowing of the pathway can be responsible of the local high velocity value recorded by some
297 tourmalines. For each sample of veins, the flow directions are dispersed with a significant mean

298 direction (Fig. 5a). In the “rose” diagrams, these directions generally host the highest velocity
299 values (10^{-4} to 10^{-3} m.s⁻¹). Conversely, lowest fluid velocity values are not aligned with these mean
300 directions of fluid flow (Fig. 5a). Considering all the sites, the mean orientations of flow depict a
301 large-scale streamline structure with radial directions coming from a center located on the greisen
302 cupola (Fig. 5a and 5b). The vertical fault system crosscutting the veins does not influence flow
303 direction in the veins (Fig. 5b). The pitch of flow direction vectors on the vein plane is not aligned
304 to the dip direction of veins (Table 1). Consequently, flow directions are not influenced by veins
305 orientation.

306 *5.2 Fluid flow in host rock*

307 In altered schist, tourmaline growth bands show a fluid flow escaping sub-vertically from
308 veins toward the host rock, with an averaged angle of 55° above and 60° below the vein (Fig. 6).
309 This symmetrical behavior may be related to the horizontal component of fluid flow within the
310 vein, as suggested by measurements on the P21 sample. Mean velocities obtained in altered wall-
311 rocks are 1 to 2 orders of magnitude lower than fluid velocity obtained in the veins (Fig. 4d and
312 Fig. 6). In addition, it would appear that velocities are higher above (10^{-5} - 5.10^{-5} m.s⁻¹) than below
313 veins (5.10^{-6} to 10^{-5} m.s⁻¹) (Fig. 6).

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| | Coordinates (ETR S89/Portugal TM06) | | | Veins orientation | | d_{\max}/d_{\min} | | | Fluid flow | | |
|-----------------------------|--|---------|-------|-------------------|-----|---------------------|------|------|----------------|------------------------------------|-------|
| | X (m) | Y (m) | Z (m) | Strike | Dip | N | Mean | SD | Mean direction | Mean velocity (m.s ⁻¹) | Pitch |
| Veins: | | | | | | | | | | | |
| Pan70 | 31675.9 | 54649.3 | 620 | N136 | 7° | 30 | 2.06 | 0.66 | N28 | 2.76E-04 | 77° |
| Pan87 | 31410.9 | 54261.6 | 620 | N300 | 5° | 41 | 2.50 | 0.95 | N64 | 6.65E-04 | 60° |
| Pan21 | 31675.1 | 54253 | 560 | N200 | 5° | 31 | 2.06 | 1.06 | N27 | 5.53E-04 | 10° |
| Pan33 | 31217.9 | 53385.2 | 470 | N130 | 15° | 24 | 2.24 | 0.39 | N40 | 3.33E-04 | 90° |
| Pan78 | 33043.4 | 53490.3 | 618 | N138 | 7° | 31 | 2.29 | 1.13 | N331 | 6.66E-04 | 26° |
| Pan73 | 32933.2 | 53875.3 | 662 | N127 | 10° | 25 | 2.75 | 1.10 | N321 | 9.46E-04 | 24° |
| Pan95 | 33058.4 | 54287 | 741 | N127 | 5° | 18 | 2.92 | 1.27 | N300 | 1.38E-03 | 8° |
| Pan97 | 32833.3 | 55559.5 | 732 | N160 | 10° | 25 | 2.74 | 0.96 | N201 | 9.47E-04 | 42° |
| Altered wall-rocks : | | | | | | | | | | | |
| P20 Up | 31675.1 | 54253 | 560 | - | - | 28 | 1.36 | 0.18 | 55 | 1.44E-05 | - |
| P20 Down | 31675.1 | 54253 | 560 | - | - | 28 | 1.29 | 0.11 | 60 | 6.95E-06 | - |

327 **Table 1 Results of directions and velocities of fluid flow deduced from tourmalines growth bands in veins and in altered wall-rocks (SD: Standard Deviation and N:**
328 **number of measurement).**

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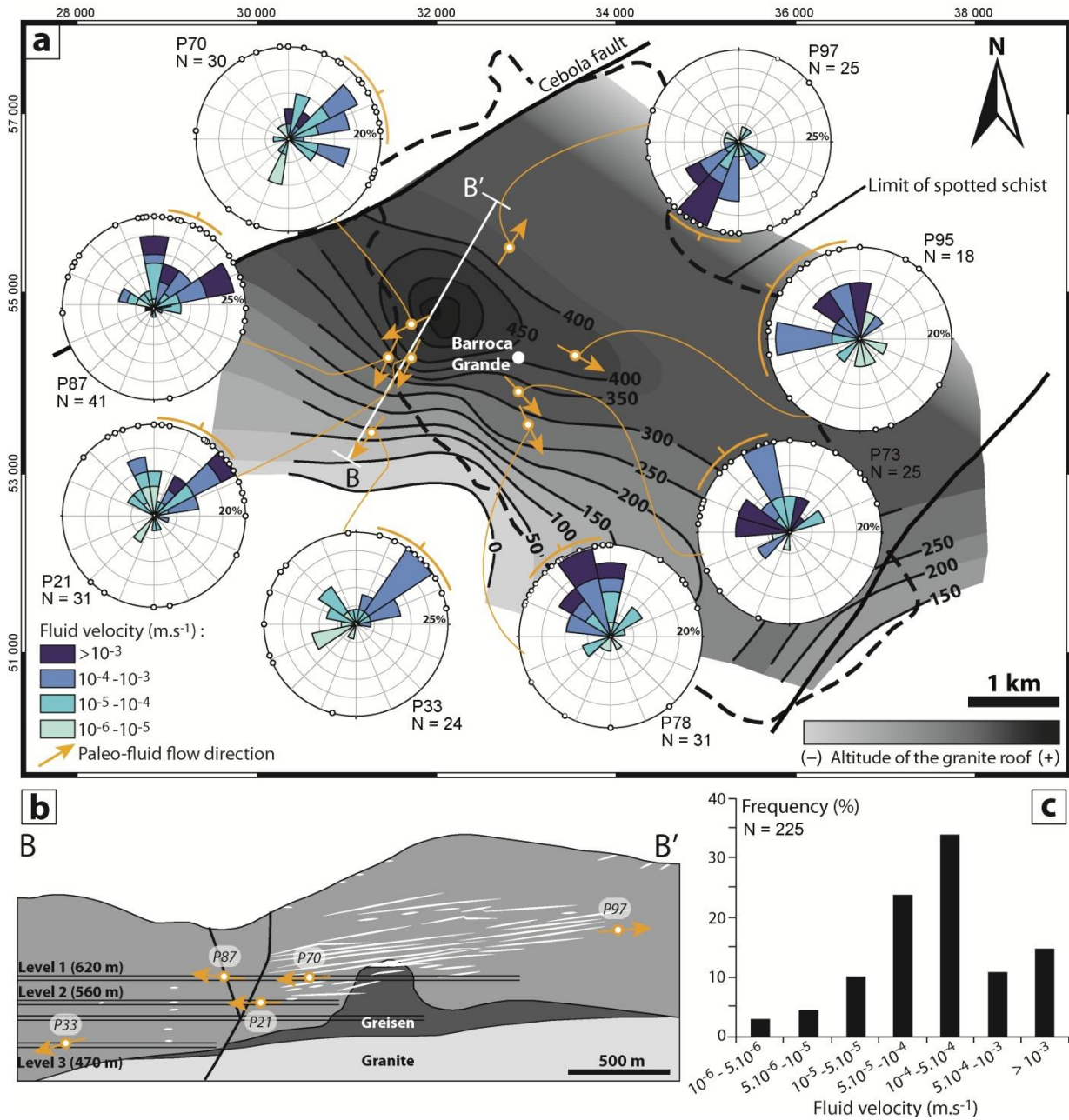


Figure 5 Results of fluid flow at the ore deposit scale deduced from tourmaline growth bands in mineralized veins. (a) Large scale map of paleo-fluid flow around the hidden granite of Panasqueira. Local fluid flow direction and velocities are represented with “rose” diagrams, which give direction of tourmaline faces exposed to the fluid flow (d_{max}) and directional distribution of fluid velocities. The orange arcs give the 95% interval of confidence, from where the fluid comes. The orange arrows on the map correspond to the mean directions of flow determined for each sample site. N corresponds to the number of measurements performed for each sample sites. (b) Results of fluid flow projected on the geological cross section (B-B') showing flow moving away from the cupola. (c) Histogram showing distribution of all fluid velocity values.

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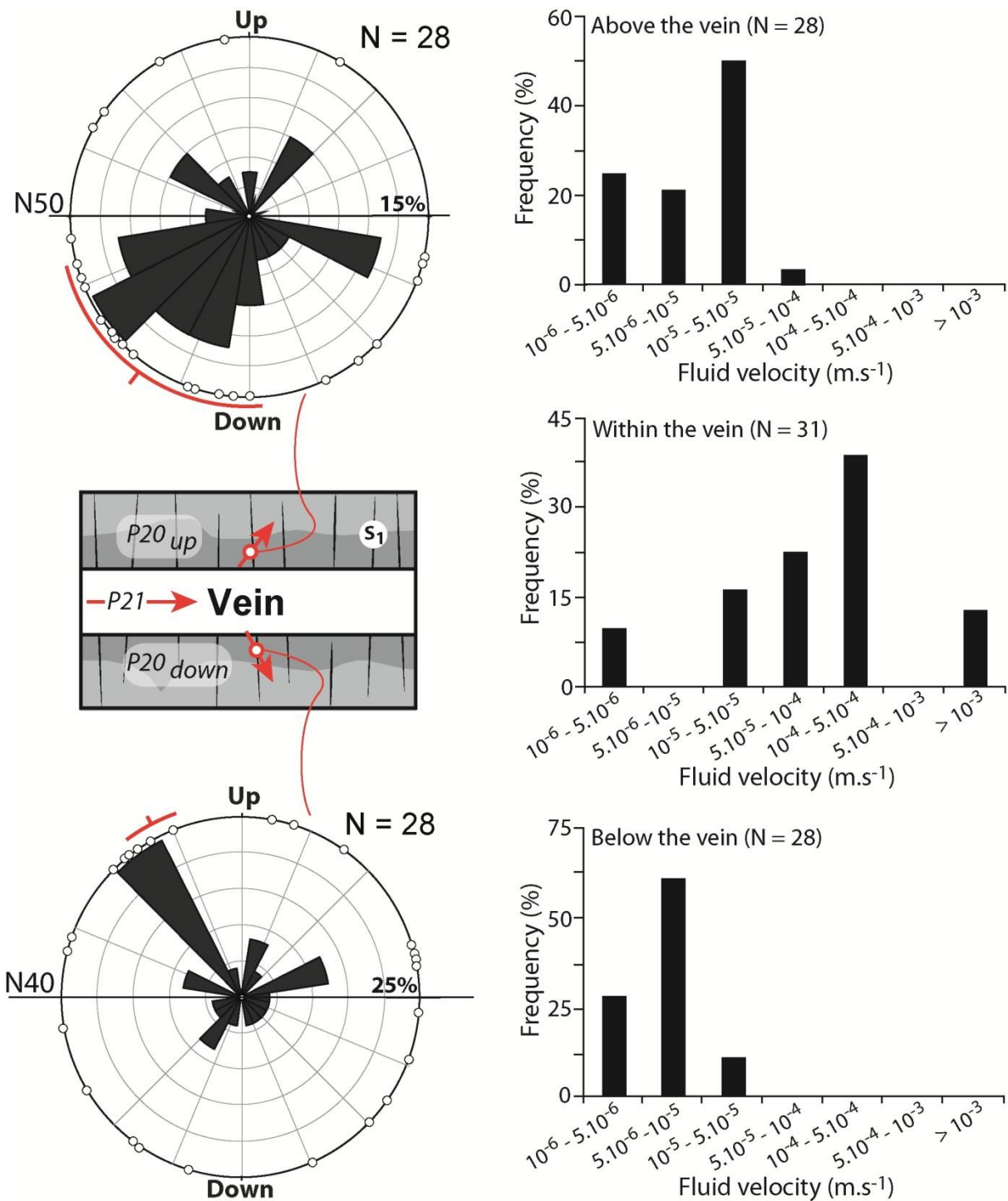


Figure 6 Sketch showing vertical paleo-fluid flow deduced from tourmaline in altered wall-rocks. Fluid flow directions are represented with rose diagrams giving direction of faces exposed to fluid flow. The red arcs indicate the 95% interval of confidence, from where the fluid comes. Velocity values obtained above, below and within the vein are presented in histogram diagrams. N corresponds to the number of measurement performed for each sample.

334

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337 *5.3 Tourmaline growth band compositions*

338 Chemical compositions of tourmalines are summarized in Table 2 (complete analyses in
339 electronic supplementary materials ESM3 and ESM4). General features and classification of
340 Panasqueira tourmalines have already been described by Codeço *et al.*, (2017). Here, we focus on
341 compositional differences in minor and trace elements between core and growth band used for
342 fluid flow reconstruction. It appears that this growth band zoning is systematically associated with
343 chemical variations in major, minor and trace elements (Table 2). In the veins, cores and rims of
344 tourmaline exhibit compositions that suggest equilibration with Li-depleted granitic rocks (Fig.
345 7a). However, rims are richer in Fe and poorer in Mg and Al than cores. In altered wall-rocks,
346 cores are characterized by a wide range of compositions suggesting equilibration with
347 metasedimentary host rocks and Li-depleted granitic rocks; whereas rims exhibit a more restricted
348 composition identical to rims of veins-tourmalines and that suggest equilibration with Li-depleted
349 granitic rocks (Fig. 7a). This range of core compositions (in altered wall-rocks) reflects the
350 contribution of hydrothermal fluids and the metasedimentary host rocks during the core
351 crystallization of tourmaline.

352 It is generally accepted that magmatic fluids contain significant amounts of Na, K and Cl. As a
353 result, the trace element concentrations in cores and rims of tourmaline contents of core and rims
354 of tourmaline are plotted as a function of the Na concentration to show the possible evolution of
355 the magmatic contribution during tourmaline crystallization. Cores of tourmalines from veins and
356 from altered wall-rocks are characterized by the same compositional ranges. This observation is
357 also valid for rims (Table 2 and Fig. 7b). Consequently, crystallization of tourmalines in the veins
358 is probably contemporaneous with the development of tourmalines in the altered wall-rocks. In
359 addition, results show strong compositional differences between core and rims of tourmalines (Fig.
360 7b). Indeed, the core to rim enrichment in Na is positively correlated with core to rim enrichments
361 in K, Ti, Li, Mn, Sn, and Sr (Fig. 7b). Average concentrations in rims display high contents of Li,
362 K, Mn, Ti and Zn (several hundreds to thousands of ppm) and moderate concentrations of Sn and
363 Sr (several tens of ppm). Conversely, cores are characterized by lower trace element abundances
364 (< 100 ppm for most elements).

365 These compositional differences between cores and rims, which are similar in the veins and in
366 the wall-rocks, suggest that tourmalines were formed during two distinct stages. The first stage is
367 related to core crystallization (both in veins and in wall-rocks) whereas the second stage is related
368 to the rim overgrowths.

369 The absence of experimental partitioning coefficients between tourmaline and hydrothermal
370 fluids do not permit to quantify the chemical signature of the hydrothermal fluid from which
371 tourmaline has crystallized. Nevertheless, according to Van Hinsberg *et al.*, (2011a; 2017), the
372 compositional changes between cores and growth bands provide a robust tool to track external
373 changes in the trace-element budget of fluids from which tourmalines have crystallized. The
374 present study suggests that rims have crystallized from a fluid richer in Na-K-Sn-Li-F-Zn than the
375 fluid from which cores crystallized.

376

| | Tourmaline from veins | | | | Tourmaline from altered wall-rocks | | | |
|--------------------------------|-----------------------|-------|-------------|-------|------------------------------------|-------|--------------|-------|
| | Core | | Rim | | Core | | Rim | |
| | Ave (n= 20) | Stdev | Ave (n= 20) | Stdev | Ave (n = 20) | Stdev | Ave (n = 20) | Stdev |
| wt% | | | | | | | | |
| SiO ₂ | 35.74 | 0.54 | 35.14 | 0.53 | 34.94 | 1.43 | 35.34 | 0.46 |
| TiO ₂ | 0.18 | 0.04 | 0.48 | 0.08 | 0.19 | 0.07 | 0.46 | 0.08 |
| Al ₂ O ₃ | 33.79 | 0.56 | 31.10 | 0.82 | 34.40 | 1.07 | 30.18 | 0.47 |
| FeO | 10.86 | 0.70 | 14.23 | 0.59 | 8.89 | 0.83 | 13.29 | 0.95 |
| MnO | 0.05 | 0.05 | 0.11 | 0.08 | 0.04 | 0.06 | 0.08 | 0.06 |
| MgO | 2.60 | 0.62 | 1.31 | 0.36 | 3.82 | 0.54 | 2.62 | 0.63 |
| CaO | 0.04 | 0.05 | 0.08 | 0.04 | 0.09 | 0.05 | 0.03 | 0.05 |
| Na ₂ O | 1.39 | 0.10 | 2.07 | 0.20 | 1.69 | 0.14 | 2.26 | 0.13 |
| K ₂ O | 0.02 | 0.00 | 0.05 | 0.01 | 0.02 | 0.01 | 0.06 | 0.01 |
| F | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| Cl | 0.00 | 0.00 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 |
| Total | 84.68 | | 84.57 | | 84.09 | | 84.34 | |
| ppm | Ave (n= 16) | Stdev | Ave (n= 17) | Stdev | Ave (n= 11) | Stdev | Ave (n= 14) | Stdev |
| Li | 28 | 9.6 | 175 | 82 | 43 | 24 | 211 | 33 |
| Na | 8399 | 1049 | 17190 | 1739 | 6843 | 1887 | 14514 | 1463 |
| Ti | 1146 | 211 | 2465 | 390 | 2096 | 660 | 3318 | 269 |
| V | 84 | 29 | 98 | 27 | 61 | 63 | 55 | 16 |
| Mn | 413 | 42 | 698 | 125 | 171 | 63 | 542 | 43 |
| Zn | 545 | 80 | 908 | 181 | 302 | 105 | 853 | 78 |
| Sr | 14 | 6.3 | 40 | 9.3 | 29 | 18 | 41 | 5.5 |
| Sn | 9.6 | 2.9 | 18 | 6.1 | 9.1 | 6.0 | 14 | 3.8 |

Table 2 Summary of EPMA and LA-ICP-MS results for core and rim of tourmaline from veins and altered wall-rocks

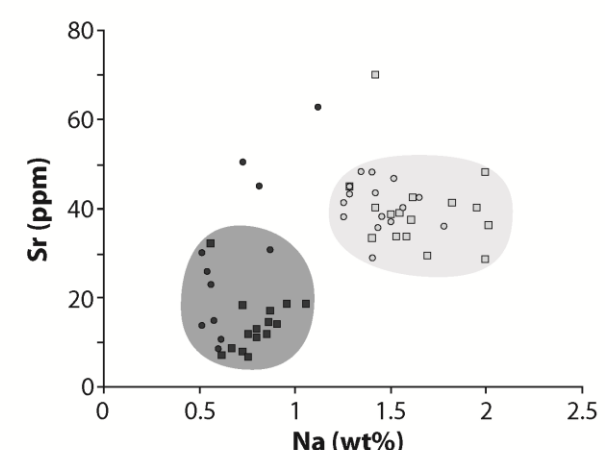
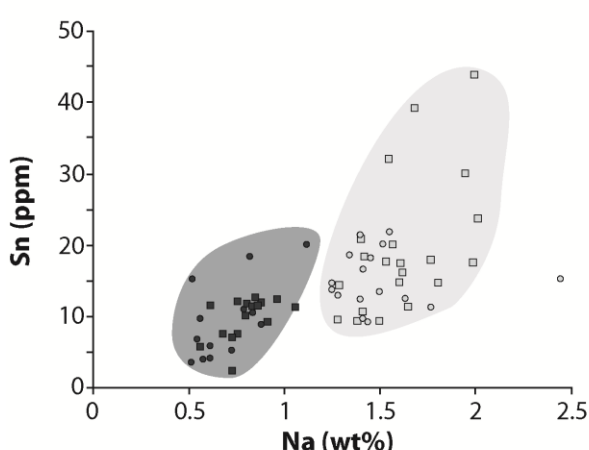
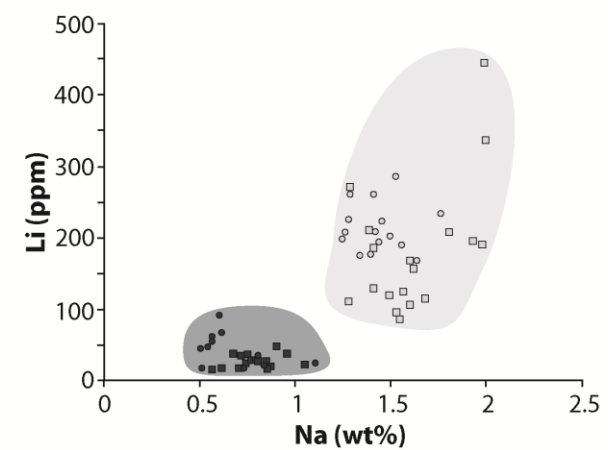
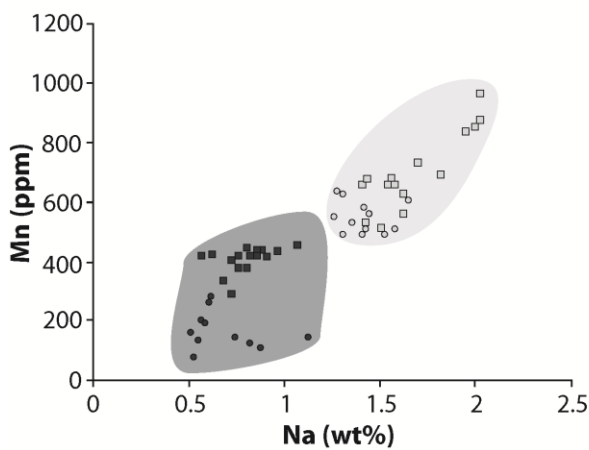
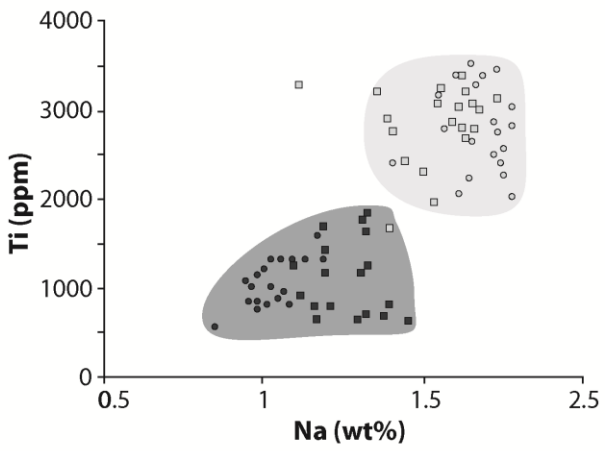
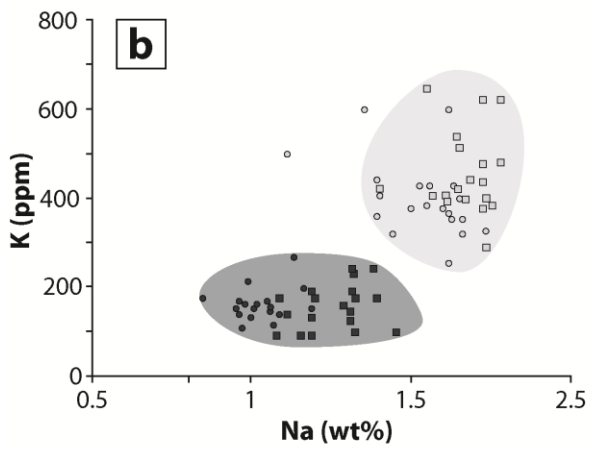
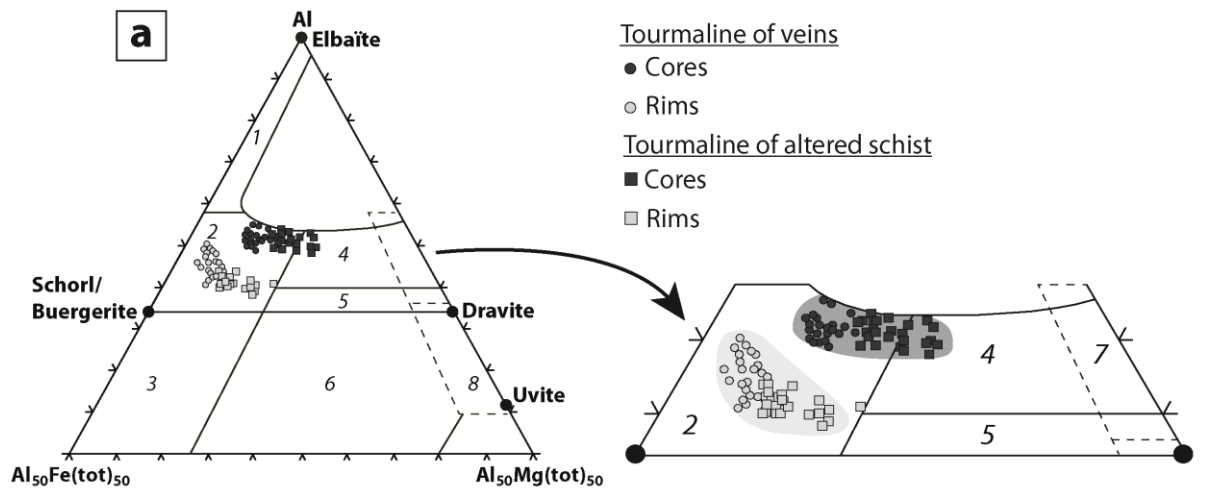


Figure 7 Geochemical characteristics of tourmalines used for fluid flow reconstruction in veins and in altered wall-rocks. (a) $Al_{50}Fe_{50}-Al_{50}Mg_{50}$ ternary diagram showing relation between tourmaline compositions and crystallization environment (from Henry and Guidotti, 1985). (1) Li-rich granitoids, pegmatites and aplites; (2) Li-depleted granitoids, pegmatites and aplites; (3) Fe^{3+} -rich hydrothermally altered granitoids; (4) Metapelites and metapsammites with Al-saturating phase; (5) Metapelites and metapsammites without Al-saturating phase; (6) Fe^{3+} -rich metapelites and calc-silicate rocks with quartz and tourmaline; (7) Cr-V-rich metapelites and Ca-depleted meta-ultramafics rock; (8) Meta-carbonates and meta-pyroxenites. (b) Elements variation diagrams showing chemical core-rim evolution of tourmalines in minor (Na, K and Ti) and trace (Li, Mn, Sn and Sr) elements.

394

395 6. Discussion

396 6.1 Role of the granitic cupola on focusing fluid flow

397 In hydrothermal systems, fluid velocity is proportional to pressure gradient and its average
398 direction is parallel to this gradient. Here, our spatial distribution and orientations of flow
399 velocities suggest a high-pressure zone centered on the greisen cupola with a lateral pressure
400 gradient triggering fluid flow in veins. Hence, hydrothermal fluids have probably been injected
401 into the vein system from the cupola, which, consequently, represents the emanative center from
402 where mineralizing fluids rose and spread laterally in veins. Moreover, the $\Sigma Na/\Sigma K$ ratio
403 determined by Poyla et al., (1989) for the ore-forming fluids (W-Sn stage) suggests an extensive
404 interaction of these fluids with the underlying granite at 350-400 °C and 1 kbar. The rim
405 compositions of tourmaline confirm an equilibration of fluids with granitic rocks. Furthermore, the
406 extensive greisenisation in the apical part of the granite provides evidence of this equilibration and
407 corroborates our interpretation of a fluid flow focused by the cupola.

408 This fluid flow pattern can explain the spatial correlation commonly observed between
409 localization of mineralized systems and emplacement of granitic cupolas (Dines, 1956; Dilles and
410 Proffett, 1995). Indeed, it has been proposed that magmatic fluids are preferentially expelled from
411 cupolas, and that can disturb the pattern of convective fluid flow around intrusions by relocating
412 convective cells and discharge zones (Norton and Cathles, 1979; Eldursi et al., 2009).
413 Hydrothermal fluxes determined in this study provide direct evidence of the control of apices on
414 focusing of mineralizing fluids. This behavior of fluid flow can be explained by a preferential
415 localization of thermo-mechanical instabilities around apices (Guillou-Frottier and Burov, 2003),

416 which, when coupled with a preferential accumulation of fluids, leads to opening of permeable
417 structures in which mineralizing fluids are then discharged.

418 Hence, the apical parts of plutonic intrusions represent convergence spots for hydrothermal
419 fluids and thus are favorable zones for deposition of high grade mineralization. As a result, the
420 approach applied in this study can be useful for mining exploration as pathfinder to discover
421 hidden granitic cupolas and thus locate the emanative centers of mineralizing fluids.

422 *6.2 Fluid escape in wall-rocks and role of fluid on vein opening*

423 The vertical escape of fluids from veins toward metasedimentary host rock involves a vertical
424 pressure gradient with overpressured fluid conditions in veins, causing pervasive fluid flow in the
425 wall-rocks. This interpretation is consistent with the presence of alteration fronts observed around
426 veins (Fig. 3a). Besides, Lecumberri-Sanchez et al., (2017) have demonstrated that schists
427 proximal to the veins are systematically enriched in K, B, Sn and W. The rims of tourmalines
428 present in altered wall-rocks are also enriched in certain of these elements (K and Sn). This is
429 consistent with extensive fluid–rock interaction triggered by fluids coming from veins.

430 According to the rim compositions of vein-and schist-tourmalines, fluid flow in veins can be
431 considered contemporaneous with pervasive fluid flow in altered wall-rocks. As shown above,
432 fluid flow in wall-rocks is related to overpressured conditions in veins. Thus, our results suggest
433 that veins opening and dilation have been induced by injection of fluids at supralithostatic
434 conditions such as proposed by Foxford et al., (2000). The low velocity values (not exceeding
435 5.10^{-5} m.s⁻¹) obtained in altered wall-rocks imply a low permeability in the host rocks. As a result,
436 the element transport through the host rock was probably low and limited to the alteration haloes.

437 Based on the removal of iron (Fe) from the wall-rocks during fluid-rock interactions,
438 Lecumberri-Sanchez et al., (2017) suggest that some elements such as Fe have migrated from the
439 host rocks toward veins. At first sight, this chemical flux seems to be incompatible with the
440 direction of fluid flow discussed above. However, iron migration through the host rocks was
441 probably driven by diffusion following a strong chemical gradient between fluid in veins and
442 metasedimentary host rocks, whereas fluid flow was driven by pressure gradient. These two

443 processes have not involved the same driving force, and it is, therefore, possible to observe a
444 chemical flux that is characterized by a vector that is opposed to the fluid flow direction.

445 *6.3 Velocity analysis and estimation of fluid flow duration during the QTS-MOSS*

446 In this study, the mean fluid flow velocity deduced from veins tourmalines (10^{-4} m.s⁻¹) is high
447 and close to values obtained with the same method in the same type of deposit by Sizaret *et al.*,
448 (2009) and Mahjoubi *et al.*, (2016), and suggests an efficient metal transport in veins. Considering
449 a porous media with an averaged porosity of 1%, the conversion of this mean velocity to a Darcy
450 velocity ($V_{\text{Darcy}} = V \cdot \text{porosity}$) gives a value of 10^{-6} m.s⁻¹. Darcy velocities obtained in contact
451 metamorphic aureoles from mineral reactions and by numerical modeling range respectively from
452 10^{-9} to 10^{-11} m.s⁻¹ (Ferry *et al.*, 2002), and from 10^{-8} to 10^{-11} m.s⁻¹ (Eldursi *et al.*, 2009). It appears
453 that velocities obtained in this study are 2 to 5 order of magnitude higher than those deduced from
454 mineral reactions and numerical modeling. This difference may be due to the fact that velocities
455 obtained from mineral growth bands are valid for a specific hydrothermal stage occurring over a
456 short period of time, while those deduced from other methods are integrated over a larger scale
457 and for the total duration of the hydrothermal system (Ingebritsen and Manning, 1999).

458 It is widely agreed that the overall time span of hydrothermal activity encompasses multiple
459 fluid pulses, each of them occurring over only a short period of time. If geochronological methods
460 allow dating hydrothermal activity, the uncertainties of these analytical methods do not always
461 permit the estimation of the fluid flow duration for a specific stage. As in most cases, the
462 Panasqueira deposit was formed during a long period of time lasting about 4.2 Myr (Snee *et al.*,
463 1988) but subdivided into several pulses. Here we propose the use of the mean fluid velocity value
464 obtained from veins-tourmaline to estimate the fluid flow duration required for the vein formation
465 during the QTS and MOSS. To do this, we assume that the QTS and the MOSS were formed
466 during the same continuous hydrothermal event with a constant fluid flow velocity. According to
467 Poyla *et al.*, (1989) the total volume of fluid that flowed through the vein system during the MOSS
468 is about 1000 km³ with 0.2 ppm of dissolved WO₃. Considering a total volume of vein about 0.003
469 to 0.006 km³ extending over an area of 6 km² (Hebblethwaite and Antao, 1982) characterized by
470 an elliptical shape, we can calculate the surface of the vertical section of fluid flow. From this

471 surface and the mean fluid flow velocity (10^{-4} m.s^{-1}) we obtain a fluid flow duration ranging from
472 35 000 to 70 000 years. This is consistent with the short time duration of mineralization
473 emplacement estimated for a single magmatic-hydrothermal event (spanning only about 10 000 to
474 200 000 years) from numerical modeling (Cathles *et al.*, 1997; Chelle-Michou *et al.*, 2017) and
475 radiometric dating (Parnell, 1998).

476 *6.4 Hydrodynamic model of the Panasqueira deposit*

477 Our results must be considered as ‘snapshot’ of the fluid flow at the incipient phase of the
478 hydrothermal system of Panasqueira, corresponding to the magmatic-hydrothermal transition.
479 Considering chemical characteristics of tourmalines and our fluid flow reconstruction, a
480 hydrodynamic model subdivided into two stages is proposed for the early hydrothermal event
481 preceding the main stage of W-Sn mineralization (Fig. 8). The first stage is related to the core
482 crystallization of tourmalines and involves an initial flow of boron-rich fluids. These fluids are
483 partly buffered by the metasedimentary host rocks for Al, Mg and Fe during the crystallization of
484 tourmaline in the wall-rocks (Fig. 7a). This first pulse does not seem to be associated with any
485 W-Sn mineralization. This is consistent with the low contents in trace elements in cores of
486 tourmaline. We have no information about directions of fluid flow related to this first pulse.
487 However, the chemical equilibration of cores of veins-tourmalines with Li-depleted granitic rocks
488 suggests that the first stage can be related to a first expulsion of magmatic fluid during granite
489 crystallization.

490 The second stage is recorded by tourmaline growth bands and is related to the flow of the
491 mineralized (Li-K-Na-Sn)-rich fluid in the veins system. During this second stage, the external
492 convecting fluids and magmatic fluids released by granite crystallization are both focused by the
493 cupola, which causes the greisenization in the apical part of the granite (Fig. 8). This focused fluid
494 flow is injected into the vein system, whose the significant horizontal permeability anisotropy
495 induces a horizontal fluid flow along the vein planes. The low permeability of the
496 metasedimentary host rock promotes fluid pressure increase leading to the vein opening when
497 fluid pressure reaches supralithostatic conditions (Fig. 8). The velocity contrast between veins (10^{-4}
498 to 10^{-3} m.s^{-1}) and altered wall-rocks (10^{-6} to 10^{-5} m.s^{-1}) emphasize this strong contrast of

499 permeability and suggests that fluid flow was mainly channeled in the vein system. This
 500 channelization of mineralizing fluids in veins could contribute to form a high grade deposit by an
 501 efficient metal transport in structural trap sites. The overpressured condition in veins enhances
 502 fluid-rock interaction and triggers a pervasive fluid migration in the wall-rocks (Fig. 8). According
 503 to Lecumberri-Sanchez *et al.*, (2017), the metasedimentary host rock is mainly affected by
 504 tourmalinization and muscovitization reactions, which are crucial for the wolframite deposition by
 505 iron releasing into the fluid. Finally, the escape of fluids toward the wall-rocks observed in this
 506 study can constitute a crucial mechanism for the wolframite deposition by promoting fluid-rock
 507 interactions.

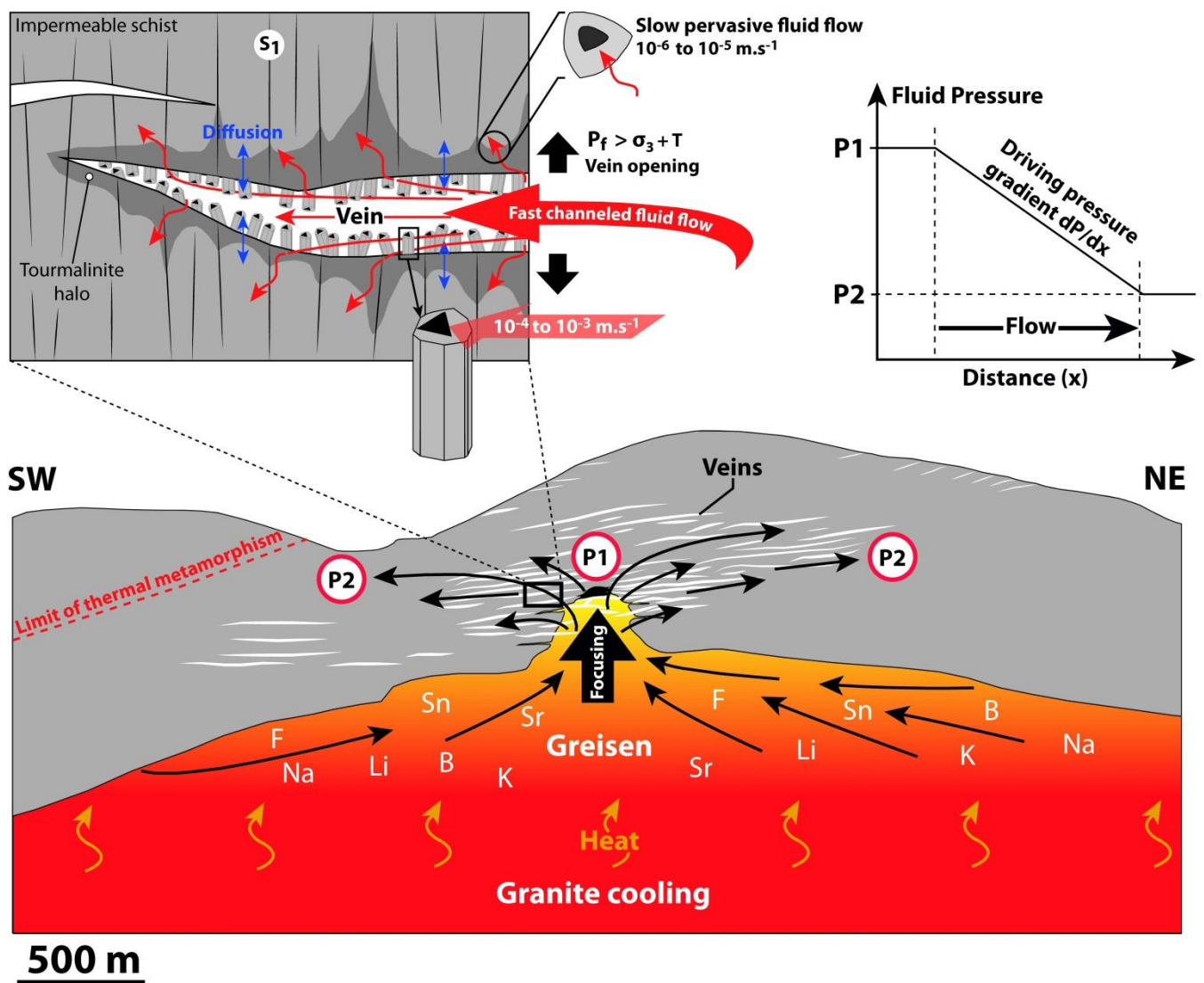


Figure 8 Hydrodynamic model of fluid flow at the magmatic-hydrothermal transition of the W-Sn-(Cu) Panasqueira deposit (drafted on simplified cross section from Thadeu, 1951). With P_f : the fluid pressure condition, σ_3 : the vertical stress and T: the tensile strength.

508 **Acknowledgments**

509 We are greatly thankful to Beralt Tin & Wolfram S.A. for the possibility to access to the
510 Panasqueira underground mine. We thank Paolo Ferraz for help during the collection of samples
511 in mine and for discussions about our results. We further would like to thank P. Lach for
512 assistance in LA-ICP-MS analyses and proceeding of data. We are grateful to anonymous
513 reviewers for their constructive remarks, which have greatly improved this manuscript. This
514 research was financially supported by the French Geological Survey (BRGM), Region Centre, the
515 Labex Voltaire (ANR-10-LABX-100-01) and has also taken part of the ERAMIN project “New
516 Ores”.

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