Cover Letter

2 Bologna, May 5th 2019

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5 Dear editor,

- 7 we herewith submit to your attention the revised version of our manuscrip "Fluid-mediated, brittle-ductile deformation
- 8 at seismogenic depth: Part I Fluid record and deformation history of fault-veins in a nuclear waste repository
- 9 (Olkiluoto Island, Finland)", hoping that you will find it improved toward its publication in Solid Earth. As you know,
- 10 the open discussion has been a very fruitful and constructive process, which has greatly helped us to sharpen our work and
- 11 improve it in light of some constructive criticisms that were made by the three reviewers.
- 12 Our detail rebuttals to each of them were already prepared and a point-by-point reply was prepared to the purpose of the
- 13 open discussion. In that occasion we discussed openly each point made and explained how we would implement the
- 14 required changes in the final, amended version.
- 15 We have now done it and there is no point for us to repeat all the content of the initial rebuttals. We confirm here that we
- 16 changed the manuscript and the figures as per discussion, doing exactly what we said we would do.
- 17 To help you appreciate the revision work, we submit a version of the file in review mode, with all changes highlighted.
- 18 Please note that all minor comments and requests of changes were basically taken care of in the final version.
- 19 To sum up our revision work, we can repeat here the main changes:
- 20 -We have tried to shorten and streamline the manuscript by polishing the language and removing unnecessary text sections
- 21 and even some repetitions.
- 22 -We have modified some of the figures to comply with the request of one reviewer. We have prepared a Supplementary
- 23 Material section with some extra data, figures and information.
- 24 -We have acquired extra EBSD data to better support our interpretation of some unclear microstructures of Qtz I.
- 25 -We have acquired extra FI data to strengthen the analysis of the fluids involved during deformation. This has brought us
- 26 to a more open discussion as to the number and chemical composition of the fluid batches that ingressed the fault during
- 27 its evolution.
- 28 -After the second reviewer comments, we added to the newversion of Figure 12 hydrostatic and lithostatic pressures,
- 29 reconstructed in accordance with regional gradients at the time of vein emplacement. These gradients are used to constrain the
- 30 upper and lower bounds to physically possible fluid pressure values.
- 31 -We added the chlorite compositional diagram on Figure 11 with the aim to argue about the composition of the fluid
- 32 batches.

-In accordance with the first reviewer, we have shorten the Abstract. -We also added to the main text most of the bibliography suggested by the reviewers. Please address all the correspondence to: Barbara Marchesini PhD Student Department of Biological, Geological, and Environmental Sciences (Bi.Ge.A.) University of Bologna Via Zamboni, 67, I-40126 Bologna. E-mail: barbara.marchesini2@unibo.it. We look forward to hearing from you at your earliest convenience. Best regards, Barbara Marchesini, corresponding author.

3 Fluid-mediated, brittle-ductile deformation at seismogenic depth:

Part I- Fluid record and deformation history of fault-veins in a

nuclear waste repository (Olkiluoto Island, Finland)

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Abstract. The dynamic evolution of fault zones at the seismogenic brittle-ductile transition zone (BDTZ) expresses the delicate interplay of between numerous physical and chemical processes that occur at the time of strain localization. Deformation and fluid flow of aqueous fluids within these zones, in particular, at the BDTZ are closely related and mutually dependent during repeating and transient cycles of repeating, transient frictional and viscous deformation. Despite numerous studies documenting in detail seismogenic faults exhumed from the BDTZ, uncertainties remain as to the exact role of fluids in facilitating broadly coeval brittle and ductile deformation at that structural level in this zone, particularly with regard to the mechanics of broadly coeval brittle and ductile deformation. We combine here structural analysis, fluid inclusion data and mineral chemistry data from synkinematic and authigenic minerals to reconstruct the temporal variations in fluid pressure (P_f), temperature (T), and bulk composition (X) of the fluids that mediated deformation and steered strain localization in along BFZ300, a strike-slip fault from originally active at the BDTZ. This is a fault formed within BFZ300 deforms the Paleoproterozoic migmatitic basement of southwestern Finland and, hosting-hosts in its core two laterally continuous quartz veins formed by two texturally distinct types of quartz-types - Qtz I and Qtz II, where with Qtz I is demonstrably older than Qtz II. Veins within the diffuse damage zone of the fault are formed infilled exclusively by Otz I. Meso- and microstructural Multi-scalar structural analysis combined with fluid geochemistry compositional data indicate s-recurrent cycles of mutually overprinting brittle and ductile deformation triggered by fluid pressure oscillations of fluid pressure, with documented peaking at pressure of 210 MPa. Fluid inclusion microthermometry and mineral pair geothermometry indicate that both the two documented quartz types precipitated from a distinct different fluid batches phases, that was in a homogeneous state during the recurrent cycles of faulting, and whose with bulk salinities werey was at first in the 1-50-5 wt% NaCleg range for Qtz I and then evolved in in the 6-11 wt% NaCleg range for Qtz II, for Qtz I and Qtz H respectively. The temperature of the fluids phases involved with the various episodes of initial strain localization and later fault reactivation changed with time evolved through time, from >e. 350 °C or even higher temperature 240 °C in the damage zone to e. 350 °C in the core during Qtz I precipitation to < 3200 °C at the time of Qtz II crystallization. PThe peak values of fFluid pressure estimates show-constrain an oscillation pore pressure oscillations in pore pressure comprised between 80 and -210160

96 and 10 MPa during the several recorded documented fault activitying stages episodes. Our results suggest significant variability in 97 of the overall-physicoal-chemical coorditions of the fluids steering deformation -phase (Pf, Tf, Pf-X)- reflecting during the fault 98 deformation history, reflecting the ingress and interaction effects of several multiple batches of different fluid compositions fluid 99 possibly reflecting the interaction of several batches of compositionally similar fluids ingressing in the dilatant fault zone at 100 different stages of its evolution, each with specific T and P_t conditions. Initial, fluid-mediated embrittlement of the faulted rock 101 volume generated a diffuse network of joints and/or hybrid/shear fractures in the damage zone, whereas; progressive subsequent 102 strain localization led to more localized deformation within the fault core. Localization was guided by cyclically increasing fluid 103 pressure and transient embrittlement of a system that was otherwise at-under overall ductile conditions.

Our analysis <u>implies</u> suggests that fluid overpressure at the <u>brittle duetile transitionBDTZ</u> can play a key role in the initial embrittelment of the <u>metamorphic basementdeforming rock</u> and <u>steer subsequent strain localization mechanisms</u>.

1 Introduction

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The pPhysical and chemical properties of fault systems play a fundamental role in controlling the rheological behaviour of the Earth's crust and in steering channelled fluid flow (e.g. Caine et al., 1996). Deformation and fluid flow are closely related and mutually dependent via a number of feedbacks, such as the control that fluids exert upon the effectiveness of deformation processes and the development of fault systems at all scales, and the control by rock heterogeneities and/or fracture system topology on the net fault transmissivity (e.g. Crider and Peacock, 2004). The nucleation and development of permeable fault systems and the mechanisms whereby individual faults may weaken and eventually fail are, therefore, complex functions of a number of processes. In this perspective, the multiscalar interaction between fluid and mineral phases within fault rocks needs to be studied with a system approach in order to single-out the roles and importance of all processes involved (Kaduri et al., 2017). The most evident An obvious effect of fluid involvement, particularly in crustal volumes that have experienced large deformationcontrolled fluid fluxes, is the precipitation of authigenic and hydrothermal minerals within faults (Oliver and Bons, 2001; Viola et al., 2016) and their immediately adjacent host rock (Mancktelow and Pennacchioni, 2005; Garofalo, 2004). In the seismogenic region of the crust, where fluids may even be the primary driver of the seismic cycle (e.g. Miller, 2013), faults have been shown to have the potential to function like a "fluid-activated valve", whereby they experience transient and cyclic fluid pressure buildup before sudden fluid venting, pore pressure- and mechanical strength drop concomitant with seismic failure (e.g. Sibson, 1989, 1992b, 1993; Cox, 1995; Viola et al., 2006; De Paola et al., 2007; Wehrens et al., 2016). Hydrothermal ore deposits, where fault networks focus relatively large volumes of ore fluids and precipitate economic minerals (Cox et al. 2001; Boiron et al., 2003; Moritz et al., 2006; Scheffer et al., 2017a) are also pertinent examples of significant deformation-controlled fluid ingress. The seismogenic depth down to 10-15 km (e.g. Kohlstedt et al., 1995) is thus a key region of the crust where to study the whole range of fluid-rock interaction processes occurring within fault zones. Deformation at that depth might be accommodated under overall brittle-ductile conditions along fault systems crossing or rooting into the brittle ductile transition zone (BDTZ). In detail, the deformation style in the BDTZ is generally characterized by the cyclicity, also at the short time scale, between brittle and

129 ductile behaviour (Famin et al., 2004; Famin et al., 2005; Siebenaller et al., 2013). This is induced and regulated by the complex 130 and transient interplay of numerous parameters, among which the lithological composition and transient variation of temperature, 131 pore pressure and strain rate within the deforming system. Field studies have documented unequivocally that ductile and brittle 132 deformation may even be simultaneously active during deformation as a function of the transient and spatially heterogeneous 133 evolution of the chemical and physical parameters steering deformation, leading to the broad coexistence of geological features 134 expressing frictional deformation and viscous creep, mechanisms and to mutual crosscutting relationships thereof (e.g., Guermani 135 and Pennacchioni, 1998; Kjøll et al., 2015; Pennacchioni et al., 2006; Wehrens et al., 2016; Scheffer et al., 2017b). 136 Veins are particularly important in this context because they attest to the relative abundance of aqueous fluids in the deformation 137 history (e.g. Cox et al., 2001). Portions of the seismogenic crust that experience large fluid fluxes host pervasivelarge and 138 vertically extensive vein networks (Sibson et al., 1988), within which up to several millions of m³ of hydrothermal minerals may 139 deposit from the flowing fluid (e.g. Heinrich et al., 2000; Cox, 2005; Bons, 2001; Garofalo et al., 2002). In contrast, portions of 140 the crust deforming in the absence of significant fluid flow would show scarce evidence of- or no veining, with only synkinematic 141 H₂O-rich minerals within the fault rock attesting to hydrous conditions (cf. Mancktelow and Pennacchioni, 2004; Menegon et al., 142 2017). 143 The physical-chemical conditions of fluid-rock interaction in the BDTZ have been-extensively studied within exhumed faults by 144 applying a set of geochemical tools that include fluid inclusion data-analysis (e.g. Morrison, 1994; Morrison and Anderson, 1998; 145 Mulch et al., 2004; Ault and Selverstone, 2008; Garofalo et al., 2014; Siebenaller et al., 2016; Compton et al., 2017), determination 146 of the isotopic compositions of fault fluids, and mass transfer calculations between host rock and fault rocks (e.g. Goddard and 147 Evans, 1995; Garofalo, 2004; Mittempergher et al., 2014; Spruzeniece and Piazolo, 2015). These data is approach yields important 148 constraints on the PT conditions of fluid-rock interaction within the studied faults BDTZ, on the source region of the fluids 149 reaching and flowing within the BDTZ deformation zones, and on element mobility during syn-tectonic fluid flow. These studies, 150 however, provide only limited information ondo not specifically address the role of fluids on the potentially complex mechanisms 151 that trigger and permit the aforementioned cycles of brittle-ductile deformation. Open questions thus remain, such as, for example, 152 which pPressure, -tTemperature, -cComposition (P, T, X) conditions are best for a fluid to trigger brittle-ductile deformation 153 cycles in a fault system within the BDTZ $_{7}$ and which fluid property is specifically most effective in controlling the cycles. 154 In this work, we follow a multidisciplinary approach by combininge the meso- and microstructural observations with the 155 geochemical analysis of fluids, petrographic documentation of fault rocks and veins, microthermometric properties of fluid 156 inclusion assemblages, electron probe microanalyses (EPMA) of fault minerals, Raman spectrometry of fluid inclusions, and 157 electron probe cathodoluminescence imaging to study the effects of numerous cycles of fluid-rock interaction that have occurred 158 in a vein-rich deformation zone from within at the seismogenic BDTZ of the seismogenic region of and now exhumed as part of 159 the Paleoproterozoic continental crust of southwestern Finland. The studied deformation zone belongs to an exhumed conjugate 160 fault system that experienced a complex history of structural reactivation and fluid flow. Deformation zone BFZ300, the target of our study, crops out at c. 426 m below sea level within the deep <u>Onkalo</u> nuclear waste repository that is presently being built in the island of Olkiluoto (Fig. 1a).

163 We Our results allow us to constrain and describe the progressive evolution of the deformation processes and the role of fluids fluids 164 activity during involved both at fault initiation and during the subsequent reactivation phases. We propose that fluid pressure 165 activity-fluctuation cycles combined with a general within an overall ductile environment at the BDTZ where deformation 166 occurred by crystal plastic processes triggered the here proposed brittle-ductile cyclicity encompassing fracturing, vein 167 precipitation and crystal-plastic deformation before renewed and fluid-induced embrittlement. Our multitechnique approach made 168 it possible to determine many of the actual chemical and physical properties of the fluids involved in the deformation process, 169 leading to a well-constrained conceptual mechanical model for the fault nucleation and subsequent development. Quartz 170 precipitation in opened fractures plus crystal plastic processes and viscous recovery helped the system to regain strength and 171 pressurize, triggering a new hydrofracturing event. The adopted integrated approach provides detailed and new insights into the 172 mechanisms steering deformation within the BDTZ. We propose a mechanical conceptual model that accounts for the constraints 173 derived from our multidisciplinary approach.

The study area is located in southwestern Finland, on the island of Olkiluoto (Fig. 1a) within the Paleoproterozoic Svecofennian

2 Geological setting

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176 orogenic province, which is characterized-formed by supracrustal high-grade metamorphic sequences and plutonic rocks. The 177 most abundant lithologies in the study area are variably migmatitic metasedimentary rocks interleaved with up to several meter 178 thick levels -of metavolcanic rocks, in addition to calc-alkaline synorogenic TTG-type granitoids, and as well as late orogenic 179 leucogranites (Figs. 1a, 1b). For a detailed lithological characterization of the area, we refer the reader to Hudson and Cosgrove 180 (2006) and Aaltonen et al. (2016). 181 Numerous studies carried out on Olkiluoto have highlighted the long geological evolution of the region, which is commonly 182 summarised by tectonic models for the Paleoproterozoic evolution of southern Finland proposing either an evolution during a 183 single and semi-continuous Svecofennian orogenic event (Gorbatschev and Bogdanova, 1993) or, insteadalternatively, a sequence 184 of up to five distinct accretion events leading to the amalgamation of several microcontinents and island arcs at the margin of the 185 Archean craton between 1.92 and 1.79 Ga (e.g. Lahtinen et al., 2005). In this scenario, several subduction systems developed, 186 and the collision of the involved microcontinents and island arc complexes resulted in conspicuous continental growth, forming 187 the major part of the Paleoproterozoic domain of the Fennoscandian Shield (1.89-1.87 Ga). According to Lahtinen et al. (2005), 188 this "Fennian accretionary event" ended with a phase of orogenic collapse associated with regional extension and remarkable 189 crustal thinning between c. 1.86 and 1.84 Ga. Renewed compression ensued during collision of the "Sarmatian Plate" with the 190 previously consolidated Svecofennian Shield, causing major crustal shortening, high temperature regional metamorphism 191 (Kukkonen and Lauri, 2009) and the emplacement of S-type granites (e.g. Ehlers et al., 1993). Tectonic activity ascribable to this 192 orogenic phase ceased with a new-distinct orogenic collapse phase at 1.79-1.77 Ga (Lahtinen et al., 2005).

193 Pervasive reworking of the Svecofennian domain took place in the Mesoproterozoic when the crust underwent significant 194 stretching and was intruded by voluminous Rapakivi granites and diabase dykes resulting from the widespread melting of the 195 lower crust at c. 1.65-1.50 Ga. This tectonic phase was probably due to the development of a rift along the present Baltic Sea 196 (Korja et al., 2001). Crustal thinning caused also the formation of the "Satakunta Graben", a NW-SE trending graben located c. 197 50 km to the north of Olkiluoto, which was later filled by Mesoproterozoic sandstone (Jotnian sandstones, Fig. 1a). The latest 198 stage of crustal evolution in southern Finland is expressed by the intrusion of 1.27-1.25 Ga, N-S striking -olivine diabase dikes 199 (Fig. 1a; e.g., Suominen, 1991). 200 As to the structural evolution of the study area, the bedrock was affected by complex, polyphase ductile deformation between 201 1.86 and 1.81 Ga. According to the evolutionary deformation scheme proposed by Aaltonen et al. (2010) the results of up to five 202 different phases, referred to as D₁-D₅, are preserved in the local structural record, each characterised by structures with distinctive 203 mineral composition, metamorphic grade, geometry and kinematics. The most relevant phases to our study are D₂ to D₄. During 204 these ductile episodes, a regional and pervasive NE-SW striking and moderately SE-dipping foliation developed, strain localized 205 along mesoscopic shear zones parallel to subparallel to the foliation and extensive migmatization occurred under amphibolite-206 facies metamorphic conditions. NNE-SSW and N-S striking mylonitic shear zones also formed under those conditions, whereas 207 later ductile events developed under progressively lower-grade metamorphism until c. 1.7 Ga ago, when brittle deformation 208 became the dominant deformation style in response to progressive regional exhumation and cooling (Mattila and Viola, 2014; 209 Aaltonen et al., 2016). The penetrative, inherited ductile grain that by then characterised the crystalline basement and that was 210 suitably oriented with regard to the prevailing stress field was invariably reactivated. This is the case for several NNE-SSW 211 striking faults mapped underground in the Onkalo repository, which clearly overprint earlier D4 shear zones and fully exploit the 212 pre-existing ductile precursors. Other faults, such as BFZ300, do not show any clear genetic relation to the older ductile 213 fabric and cut it discordantly. 214 As will be shown in the following section, BFZ300 belongs to a set of subvertical, conjugate brittle-ductile to fully brittle strike-215 slip faults characterized by N-S-trending sinistral and NW-SE dextral faults. Both sets show document a complex history of 216 reactivation and contain evidence for cyclic and transient switches between brittle and ductile deformation at all scales. Meso-217 and microstructural studies show that the sinistral faults overprint and probably reactivate a dextral viscous-mylonitic precursor 218 related to earlier, localized ductile deformation (Prando et al., in prep.). These faults locally contain pseudotachylyte injections, 219 which-potentially suggests seismic behaviour during deformation (Menegon et al., 2018). In contrast, dextral faults cut across the 220 foliation, do not exploit any ductile precursors and do not host pseudotachylytes. The fault zone studied here BFZ300 belongs to 221 this second group of faults. In the following, we describe its architecture, reconstruct its deformation history and constrain the 222 deformation mechanisms and faulting conditions that acted prevailed during its nucleation and subsequent development. The 223 architecture and deformation history of the remarkably different conjugate structure of to BFZ300, which is a sinistral 224 brittle-ductile deformation zone, whose seismic brittle failure was steered by the presence of a penetrative ductile 225 precursor, will be is described in a the separate Part II companion paper (Prando et al., in prep.).

226 3 MApplied methods: Fluid inclusion, mineral chemistry and EBSD analyses

227 Field documentation and sampling were carried out at the underground BFZ300 exposures of Onkalo, which are necessarily

228 limited in extent but that, together with the logged diamond drill holes from the underground exploration, allow a well-constrained

3D reconstruction of the local geology. The studied fault section is located at a depth of 426 m b.s.l. (Fig. 1b) and is about 8 m

long. To characterize the fault architecture and constrain the spatial and temporal association of fault rocks and the type of fluid

involved in the deformation, several outcrop samples, each representative of a distinct structural domain, were collected at the

232 outcrop (TPH 2, TPH 3, TPH 4, TPH 5and TPH 6), in addition to samples PH 21 and PH22 from a diamond

233 154 drill core that intersects BFZ300 at the same depth in an area that is currently not excavated (Fig. 3). From these samples we

234 prepared 10 petrographic thin sections (samples: TPH120 2, TPH 120 4, TPH 120 6, PH 21 and PH 22) and 9 doubly polished

sections for fluid inclusion analysis (thickness: ~150 μm, samples: TPH120-2, TPH 120-4, TPH 120-6, PH-21 and PH-22). Due

to the extensive reactivation of the fault zone and the consequent obliteration of the FI record, FI study was carried out only in

237 samples TPH 120-4, TPH 120-6, PH-21. Hand samples and drill cores localities are specified in Fig. 2.

238 Microstructural work was carried out on oriented petrographic thin sections cut orthogonally to the foliation and parallel to the

239 striae that track the overall strike slip kinematics of the deformation zone. Strie are defined by elongated trails of chlorite grains,

240 at the vein host rock boundary.

241 Field documentation and sampling were carried out at the underground Onkalo BFZ300 exposures of BFZ300 (Fig. 1b) Onkalo

which are-were necessarily limited in extent to the actual excavated volume of rocks at the time of our study but that, together

with the logged diamond drill holes-cores from the underground exploration, allow a well-constrained 3D reconstruction of the

244 local geology.

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246 3.1 Fluid inclusions and mineral chemistry

Fluid inclusion measurements were conducted on "fluid inclusion assemblages" – FIAs, i.e. on petrographically discriminated, cogenetic groups of fluid inclusions located along trails or (less commonly) within clusters (Bodnar, 2003a; Goldstein, 2003). By definition, FIAs are groups of inclusions that have been trapped together (i.e., they are cogenetic) at a specific stage of mineral formation (i.e. co-genetic), and, as such, give the highest level of confidence when characterizing the properties of trapped fluids and discriminating possible stages of post-entrapment re-equilibration (Bodnar, 2003b, and references, therein). We identified appropriate FIAs that constrain the deformation history of BFZ300, but also applied the Roedder's identification criteria of FIAs according to the timing of entrapment (i.e., primary, secondary, pseudosecondary) in order to link stages of fluid entrapment with stages of brittle and ductile deformation of quartz. In this regard, we considered as co-genetic, and therefore representative of aone specific stage of brittle deformation and fluid circulation, only those FIAs that exhibited both similar orientation and petrographic characteristics at the scale of the thin section, can consider FI trails as synkinematic features, where FIAs are entrapped during stages of brittle deformation and fluid circulation, such that FIAs arranged along trails of the same orientation and with similar petrographic features might be representative of the same deformational event.

259 In the selected samples, we studied 2812 FIAs entrapped within two distinct generations of quartz (named Qtz I and Qtz II) 260 infilling forming two different generations of veins (named Otz I and Otz II) and exhibiting the least petrographic evidence of 261 post-entrapment overprinting by later ductile and/or brittle deformation, which provided c. 800400 microthermometric properties. 262 Due to the well-documented tendency of fluid inclusions to modify their shape, volume, and composition after their initial 263 entrapment even at low deviatoric stress conditions (e.g. Diamond et al., 2010; Kerrich, 1976; Tarantola et al., 2010; Wilkins and 264 Barkas, 1978), working on FIAs that show the are similar as possible to those preserving the pristine fluid conditions is essential 265 when aiming at the study of the original physical and chemical properties of the fluid involved in the fault activity, lowest-least 266 possible degree of textural re-equilibration is essential when aiming at the study constraining of the original physical and chemical 267 properties of the fluid(s) involved in the fault activity. 268 Microthermometric properties of fluid inclusions were determined at the Department of Biological, Geological and 269 Environmental Sciences of the University of Bologna using a Linkam THMSG 600 heating/freezing stage coupled with an 270 Olympus BX51 polarizing microscope. The microthermometry stage was calibrated by using synthetic fluid inclusion samples at 271 -56.6, 0.0, and 374 °C, which correspond to the melting of CO₂, ice melting, and final homogenization of H₂O inclusions, 272 respectively. Obtained accuracies were ±0.3 °C for final ice melting temperature (Tm_{ice}) and ±3 °C for final homogenization 273 temperature (Th_{tot}). In order to produce an internally consistent dataset, all phase transitions were exclusively collected for 274 individual FIAs and measured by with the same standard procedure. Samples were first rapidly cooled to c. -180 °C and then 275 slowly heated to detect the potential formation of a solid carbonic phase, eutectic phases, salt hydrates, ice, and clathrates. The 276 Th_{tot} were later determined in the FIAs by heating the samples from room temperature and recording the mode of homogenization 277 (i.e., by bubble or liquid disappearance). All phase transitions were measured by using the cycling method described by Goldstein 278 and Reynolds, (1994), and care was taken also into-recording the minimum and maximum values for each assemblage. Volume 279 fractions of individual fluid inclusions determined as % of the ratio φ=V_v/V_{tot} (cf. Diamond, 2003), were estimated optically at 280 room temperature using calibrated charts. Salinity, bulk densities and isochores were computed from the measured Tm_{ice} values 281 using the HokieFlincs Excel spreadsheet (Steele-MacInnis et al., 2012 and reference therein). 282 Fluid inclusions were also analysed using mMicro-Raman spectrometric analysis of fluid inclusion was spectrometry. Analyse is 283 were carried out at the Department of Mathematical, Physical and Computer Sciences of the University of Parma (Italy) using a 284 Jobin-Yvon Horiba LabRam spectrometer equipped with He-Ne laser (emission line 632.8 nm) and motorized XY stage. The 285 spectral resolution of the measurements was determined as nearly 2 cm⁻¹. The confocal hole was adjusted to obtain a spatial 286 (lateral and depth) resolution of 1–2 μm. Most spectra were obtained with a 50× objective (N.A. 0.75), although for shallow inclusions also a 100× objective (N.A. 0.90) was used. The calibration was made using the 520.7 cm⁻¹ Raman line of silicon. A 287 wide spectral range (100-3600 cm⁻¹) was scanned for each inclusion for the presence of CO₂, N₂, CH₄, and H₂S, but the final 288 289 acquisitions were made mainly between 1100 and 1800 cm⁻¹ for the study of CO₂ spectra, and between 2500 and 3300 cm⁻¹ for 290 CH₄ and H₂S. The acquisition time for each spectral window was 120-240 s, with two accumulations. The power on the sample

- surface is nearly 1 mW but the power on the analysed inclusions has to be considered lower due to reflections and scattering.
- Analyses were carried out on the vapour bubbles of the fluid inclusions.
- 293 After the calculation of representative fluid inclusion isochores for each FIA, the pressure corrections were assessed by using the
- 294 crystallization temperatures of two mineral pairs namely chlorite-quartz and stannite-sphalerite as independent input
- 295 parameters for Qtz I and Qtz II veins, respectively. Chlorite-quartz temperatures were calculated by using the method of Bourdelle
- and Cathelineau (2015), which assumes quartz-chlorite equilibrium and uses ratios of chlorite end-member activities to link the
- 297 chlorite compositions with the corresponding formation temperatures through the quartz-chlorite equilibrium constants. This
- 298 method is based on the measurements of the concentrations of the major chlorite components (Si, Fe, Mg) and can only be applied
- 299 to chlorites with (K₂O + Na₂O + CaO) < 1wt%, indeed the case of our chlorites. To estimate the formation temperature of
- 300 cogenetic sulphides associated with Qtz II we used the stannite-sphalerite formation temperature following the method proposed
- 301 by Shimizu, and Shikazono (1985). This geothermometer uses the temperature dependency of iron and zinc partitioning between
- 302 stannite and sphalerite (Nekrasov et al., 1979) as a useful temperature indicator of the association Qtz II-stannite and sphalerite.

3.2 Electron Probe Microanalysis Microanalysis

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- 304 Electron Probe Microanalysis (EPMA) of fault minerals were-was carried out by using with a JEOL-8200 wavelength-dispersive
- 305 electron microprobe housed at the Department of Earth Sciences of the University of Milan, Italy. The instrument fits 5 WDS
- 306 spectrometers utilizing lithium fluoride (LiFH), pentaerythritol (PETJ and PETH), and thallium acid pthalate (TAP) analysing
- 307 crystals and an optical microscope. Samples were probed with a beam size of ~1 μm at 15 keV and 5 nA beam current. Synthetic
- 308 and natural materials were used as calibration standards at the beginning of each session. Analytical 1- σ errors are typically < 4%
- 309 for major elements and for the minor elements.
- 310 Panchromatic cathodoluminescence (CL) imaging was also performed by using the CL CCD detector adjacent to the optical
- 311 microscope of the JEOL-8200 on the sections used for microstructural work. The electron beam was focused on the sections with
- 312 an accelerating voltage of 15 kV and 30 nA beam current. Black/white digital images were collected with a 40x magnification by
- beam mapping with the CCD detector at a spatial resolution of 1 µm (beam resolution), which resulted in imaged areas of 27.8 x
- 314 22.2 mm. The exposure time for image acquisition was 120 s.
- 315 Petrographic thin sections were lateralso analysedused at the Scanning Electron Microprobe (SEM) to investigate the
- 316 crystallographic preferred orientation (CPO) of the selected sites of the quartz veins from the fault core (sample name TPH-
- 317 120-4, see Figure 2 for sample location). Sample-Samples wereas analysed using-with a JEOL 6610 SEM equipped with a
- 318 Nordlysif UF 1000 Nano EBSD detector, hosted at the Electron Microscopy Centre School of Geography, Earth and
- 319 Environmental Sciences of the University of Plymouth, UK, EBSD analysis details and the acquired images detailed results
- 320 are reported in the Supplementary Mmaterials.

322 4 Results

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4.1 BFZ300 fault architecture

324 The studied fault-BFZ300 section is located at a depth of 426 m b.s.l. and is about 8 m long (Fig. 2a). ItBFZ300 strikes NNW-325 SSE and dips very steeply to subvertically to the southwest (Fig. 2b). It cuts through high-grade veined migmatite, interlayered 326 with gneiss and pegmatitic granite. At the studied underground outcrop with a length of 8 m, tThe fault is a strike-slip fault system 327 formed by two main subparallel fault segments connected by a mesoscopic sinistral step-over zone. Subhorizontal striae defined 328 by elongated trails of chlorite grains and kinematic indicators such as chlorite slickensides (Fig. 2c) and R and R' planes invariably 329 indicate invariably dextral strike-slip kinematics. The most striking mesoscopic characteristic of BFZ300 is the presence in the 330 fault core of a composite set of almost continuous quartz veins (between 1 and 20 cm in thickness) along the entire exposed strike 331 length. A schematic representation of the fault zone is shown in Figure 32. 332 The fault contains a 0.5-2 m thick damage zone separated by from the undeformed host rock by two discrete bounding surfaces 333 (Y_I planes according to Tchalenko, 1970 Fig. 2a). The damage zone can be defined in the field on the basis of the presence of a 334 fractured volume containing sets of conjugate dextral and sinistral hybrid fractures (Fig. 3a) intersecting to form a tight acute 335 angle of c. 38° (Figs. 2b., 3a). Laterally continuous, NNW-SSE striking quartz filled-Mode I fractures (joints) -invariably bisect 336 this angle (Figs. 2b,3a), helping to constrain the stress field orientation at the time of fracture formation, with the greatest 337 compressive stress axis σ_1 parallel to the Mode I fracture strike and oriented c. NNW-SSE. Joints are sharp and have a regular 338 spacing of c. 10 cm. Quartz-filled The joints and the hybrid fractures of the damage zone forms contain quartz, referred to as Otz 339 I hereinafter, forming veins up to 1-1.5 cm in thickness thick and is referred to as Otz I hereinafter (Fig. 3a). Fractures and faults 340 decorated by Qtz I have a translucid look that reflect the generally fine grain size of Qtz I (< 1 cm, Fig. 3b₇). Locally they are 341 formed by en-echelon tensional segments connected by shear planes not decorated by any quartz infill (Fig. 3b). Joints occur also 342 as barren fractures defining a penetrative sympathetic fracture cleavage (sensu Basson and Viola, 2004; green lines in Fig. 2b). 343 Field evidence also suggests that fracture density within the damage zone tends to increase towards the fault core. 344 The fault core is bounded by two main discrete slip surfaces (Y_{II}, Figs. 2a, and 3d, f, h). It contains, and is defined by, two distinct 345 generations of quartz veins (Fig. 3c) that are interrupted and offset laterally by a metric sinistral step-over zone (Fig. 3d-f). The 346 main quartz vein of the core is infilled by quartz exhibiting the same mesoscopic appearance of Qtz I in the damage zone; we 347 therefore refer to it as a Qtz I vein. It is accompanied by a younger, subparallel vein formed by a milky-white type of quartz with 348 a significantly larger quartz grain size than Qtz I (>1 cm) that we refer to as Qtz II (Fig. 3c). Locally, pockets of cataclasite and 349 breccia formed at the expense of the host migmatitic gneiss are also observed along and in between the two veins (Figs. 3g. -i). 350 These shears are formed by cataclastic bands formed at the expense of the host migmatitic gneiss. The Qtz II vein shows exhibits 351 a quite irregular, curved geometry (Figs. 3c₂₋₇h) and a variable thickness up to a maximum of c. 20 cm. The minimum Qtz II vein 352 thickness coincides spatially with an lateral apparent lateral displacement of the vein. The BFZ300 core varies in thickness 353 between 20 and 30 cm along most of the exposed fault length, but becomes thicker (up to 50 cm) in the compressional step-over

- 354 zone that connects the two fault segments that are offset laterally by c. 1 m. The sinistral step-over zone is defined by synthetic T
- 355 fractures (Figs. 3d, e) and contains a decimetric brecciated lens (Fig. 3d). T fractures are generally filled by Qtz I veins (Fig. 3e).
- 356 Chlorite is present as a secondary phase, with a modal abundance between 5 and 10 vol% in both Qtz I and Qtz II veins. In Qtz I
- yeins it occurs as euhedral/subhedral crystals that are up to 1-2 mm in size (Fig. 3g). Chlorite is present mostly as a disseminated.
- interstitial phase, concentrated mainly in the internal part of the Qtz I veins (Fig. 3g). In the Qtz II vein, however, it occurs as
- elongated crystals (5-8 mm in length) arranged perpendicularly to the walls of the vein, which suggests orthogonal dilation at the
- configured crystals (5.6 min in longar) arranged perpendicularly to the wans of the vent, which suggests of mogoniar distribution at the
- time of opening (Fig. 3h). The Qtz II vein contains also small (1-2 cm) aggregates of sulphides (sphalerite, pyrite, galena, and
- 361 chalcopyrite) mainly concentrated in the central part of the vein (Fig. 3g).
- 362 As observed in the field, the presence of Qtz I veins along the joints in the damage zone and the continuity of the fault core Qtz I
- 363 vein suggest Mode I fracturing during Qtz I emplacement (Figs. 2a, 3a, -c, and 2a). The semi-continuous parallelism of Qtz I and
- 364 Qtz II veins in the fault core, combined with the location of the Qtz II vein along the walls of the Qtz I vein, suggest the partial
- 365 reactivation of the Qtz I vein during Qtz II emplacement. Dilation leading to Qtz II emplacement exploited and further reworked
- 366 the Qtz I-host rock contact, that seemingly had a lower tensile strength than the pristine migmatite. The reconstructed time
- 367 relationship between the two vein generations is also consistent with local evidence of the Qtz II vein partially partly crosscutting
- 368 parts of the Otz I vein (Fig. 3f).

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4.2 BFZ300 microstructural analysis

- 370 To constrain the spatial and temporal association of fault rocks and the type of fluid involved in the deformation, several outcrop
- 371 samples, each representative of a distinct specific structural domain, were collected at the studied underground outcrop (TPH-
- 372 120-2, TPH-120-3, TPH-120-4, TPH-120-5 and TPH-120-6), in addition to samples PH-21 and PH-22 from diamond drill cores
- that intersect BFZ300 at the same depth in an area that is currently not excavated. From these samples we prepared 10 petrographic
- thin sections (samples: TPH-120-2, TPH-120-4, TPH-120-6, PH-21 and PH-22) and 9 doubly-polished sections for fluid
- inclusion analysis (thickness: ~150 µm, samples: TPH-120-2, TPH-120-4, TPH-120-6, PH-21 and PH-22). Due to the extensive
- 376 reactivation of the fault zone and the consequent obliteration of the FI record, the FI study was carried out only in samples TPH-
- 377 120-4, TPH--120-6 and PH-21. Hand samples and drill cores localities are specified shown in Figure 3.
- 378 The mMicrostructural work was carried out on oriented petrographic thin sections cut orthogonally to the migmatitic foliation
- and parallel to the striaeslickenlines.
- 380 In the following we provide a description of the microstructural characteristics of BFZ300 by detailing our findings and
- 381 observations separately for the main structural domains of the fault zone.
- 383 *4.2.1 Damage zone*
- 384 Qtz I veins within the damage zone cut across the migmatitic host rock and form the infill of conjugate sets of hybrid fractures,
- 385 which, when studied at the microscale, appear as formed by dilatant segments joined by cataclastic shear fractures (Fig. 4a).

386 Shearing on the latter is well documented by the asymptotic bending into the shear surfaces of foliation planes formed by the 387 alignment of chlorite and muscovite, both partly altered to sericite and chlorite, respectively (Fig. 4a). Qtz I infilling the tensional 388 segments has an average grain size between 200 µm and 3 mm and exhibits a rather heterogeneous texture, from purely blocky 389 to mixed elongated-blocky (Figs. 4b, c). The largest crystals (800 µm to 1 mm) are elongated and stretched from the vein walls 390 towards the inner part of the vein (Figs. 4c, and 5a), which is consistent with a syntaxial growth mechanism (Bons et al., 2012). 391 At least two episodes of vein growth/renewed dilation, as indicated by the presence of median lines (ML), are elearly visible 392 within one of the studied veins and confirm a syntaxial growth mechanism for the vein (Fig. 5; e.g. Bons et al., 2012). Medial 393 lines are defined by the alignment of chlorite, sericite, and carbonate aggregates (Figs. 5a, b, d). Blocky euhedral quartz crystals 394 are also found varying in , with a grain size between 300 andto 600 µm. These crystals are juxtaposed against to-very fine grained 395 quartz (<200 µm) within sericite-rich cataclastic bands (Fig. 4b). These cataclasites contain also hydrothermally alterated host-396 rock fragments including pervasively altered K-feldspar-bearing lithic fragments and phyllosilicates.

397 With the exception of the blocky variety, Otz I crystals exhibit various degrees of crystal-plastic deformation and recovery. They 398 contain widespread evidence of undulose extinction and extinction bands (Fig. 5b), and incipient bulging along grain boundaries 399 is also evident (Fig. 5c) indicating distributed internal plastic deformation. Millimetric intracrystalline barren fractures are also 400 recognized in the samples (e.g. Fig. 5c). Cathodoluminescence imaging of Otz I from the damage zone also confirm shows the 401 presence of a diffuse dense network of healed quartz microfractures (Fig. 4d), which demonstrates healing subsequent to brittle 402 deformation and fracturing.

403 Chlorite occurs as a disseminated phase occurs along the median lines ML of the veins, secondary cracks, at along grain boundaries and as as inclusions within quartz crystals within the Otz I veins of the damage zone and in textural equilibrium with quartz It 405 has a peculiar vermicular texture (Fig. 5db) and crystal dimensions of aboutup to 50100 µm., and displays interference colours 406 ranging from violet to Berlin blue. Vermicular chlorite forms small pockets mainly located in the central part of the veins and at the triple junctions of blocky quartz crystals.

408 4.2.2 Fault core

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409 In the BFZ300 fault core, the Qtz I grain size reaches the smallest observed value (range: 30-800 µm, Fig. 6a) grain size, although 410 it of Qtz I is strongly variable within the vein, suggesting the presence of heterogeneous and complex structural sub-domain, s. 7 411 of deformation. Qtz I has the smallest observed grain size (range: 30-800 µm, Fig. 6a) and documents multiple and cyclic episodes 412 of mutually overprinting brittle and ductile deformation leading to a complex microstructural record. The earliest post-vein 413 emplacement recognised deformation stage is reflected by the low-temperature, intracrystalline deformation of the largest crystals 414 (400-800 µm in size). Typical microstructures, such as Uundulose extinction, wide extinction bands (WEBs, Derez et al., 2015), 415 and bulging along grain boundaries are the most common microstructures ascribable to this deformation stage (Figss. 6a, b, b). 416 A distinct, first -brittle deformation event is documented by narrow, intracrystalline fractures that crosscut the largest quartz 417 crystals (Figs. 6b, and c)), and which locally contain new grains of quartz ranging in size between 20-100 μm (Fig. 6d). More in

418 detail, these new grains form parallel bands that are oriented at low angle (<30°) to the vein walls and that can be up to 2 mm in 419 length and 200 µm in thickness. Plastically deformed Qtz I crystals hosting these intracrystalline bands of new grains are cut 420 across by another distinct-later set of subparallel intercrystalline fractures, which are interpreted as the expression of yet another 421 deformation event that occurred under overall brittle conditions. These fractures are parallel to the strike of BFZ300 and are in 422 turn sealed by partly recrystallized new quartz grains (grain size: 50-150 µm; Fig. 6e). Petrographic analysis on intercrystalline 423 fractures also show that they are also locally decorated by trails of fluid inclusions (Fig. S2 Supplementary Materials) and that 424 they can be up to 2.5 cm in length and up to 500 µm in width (Fig. 6a). Their cathodoluminescence imaging of these fractures 425 shows that they are sealed and healed, yielding an homogeneous dark eathodoluminescence signal (Figs. Sla, b) of in the 426 Supplementary Materials). They are locally decorated by trails of fluid inclusions (Figs. S2a, d of the Supplementary Material) 427 and can be up to 2.5 cm in length and up to 500 µm in width (Fig. 6a). EBSD maps were performed acquired along some of these 428 intercrystalline bands, and results suggest that the new grains sealing the fractures reflect the combined effect of initial cracking, 429 grain nucleation and subsequent partial dynamic recrystallization filled by quartz new grains (EBSD maps and their location 430 across the thin section are reported in Figs.ure S2b, c of the Supplementary Materials). The EBSD results highlithed indicate that 431 recrystallization of in Otz I occurred prevalently by bulging and subgrain rotation recrystallization but also suggest that along the 432 sealed intercrystalline fractures express the delicate interplay of both brittle and crystal plastic recrystallization processes are 433 competing deformation. This is also in accordance with the petrographically contiguity of brittle fractures marked by fluid 434 inclusions and recrystallized intercrystalline fractures.

Qtz II within the fault core is typically coarse grained (individual crystals: 300 µm-1 cm in size) and exhibits a regular blocky texture devoid of any shape or crystal preferred orientation (Fig. 7a). Locally, these large crystals display primary growth textures, such as primary FIAs oriented parallel to specific crystallographic planes. With the exception of undulose extinction, Qtz II does not show clear evidence of plastic deformation, although cathodoluminescence imaging of optically continuous Qtz II has also shown that a dense network of healed quartz microfractures locally crosscuts Qtz II crystals (Fig. 7c). These are relatively thin (hundreds of µm thick) networks that are poorly visible to invisible by standard petrographic analysis. The only petrographic evidence for these healed microfractures within quartz are well defined trails of fluid inclusions crosscutting primary growth bands (Fig. 7d).

443 Chlorite is the second most abundant phase within the fault core Qtz I and Qtz II veins and occurs with a variety of textures.

444 Aggregates of vermicular chlorite similar to that occurring in the damage zone (Fig. 5d) are also present in the Qtz I from the

445 core (Fig. 8e), although chlorite with flaky and radiate textures (Fig. 8f) is also present. The latter $\frac{100-300}{100}$ $\frac{100-300}{100}$ $\frac{100-300}{100}$

in size and is in textural equilibrium with quartz and rare calcite. Radiate chlorite overgrowing fractured Qtz II (Figs. 7b_e)

447 suggests late Qtz II precipitation.

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Associated with Qtz II, a sulphide assemblage made of pyrite, sphalerite, galena, and chalcopyrite (Figs. 7d, and e, see also Fig.

449 , see also Fig. 3gb) forms aggregates that are commonly located along quartz grain boundaries. The studied sections, these

- 450 aggregates have dimensions between 10 and 600 μm. Chalcopyrite occurs as μm_ sized, irregular inclusions within sphalerite
- 451 forming the typical "chalcopyrite disease" texture (e.g., Barton and Bethke, 1987; Fig. 7e).
- 452 Multiply reworked breccias and cataclasites occur within and crosscut BFZ300. In the studied sections, a cataclastic band between
- 453 5 and 8 mm thick crosscuts both Qtz I and Qtz II veins (Fig. 8a), but is in turn crosscut by a different quartz-radiate chlorite vein
- 454 displaying evidence of syntaxial growth. This cataclasite contains poorly sorted and angular quartz clasts varying in size between
- 455 8 and 12 mm in size set in a finer (20-200 μm in size) white mica-quartz matrix. The largest quartz fragments show irregular,
- 456 lobate grain boundaries and are affected by undulose extinction. We interpret these textures as the product of dissolution and
- 457 cataclastic reworking of Otz I. vein.a previous generation, plastically deformed quartz.
- 458 Parallel sets of stylolitic seams strike-trend c. N-S, parallel to the strike of BFZ300, and mark the two sides of the cataclastic band
- 459 (Figs. 8a, c). They host anhedral sphalerite, stannite, galena, pyrite, and chalcopyrite (Fig. 8d), which are coeval with the formation
- 460 of the Qtz II vein. We interpret the presence of these anhedral sulphide minerals along the stylolite as the product of passive
- 461 concentration by pression-solutionn processes. We use the stannite sphalerite mineral pair as a geothermometer for the Qtz II
- 462 emplacement (see below).

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4.3. Fluid inclusion data

465 <u>4.3.1 Fluid inclusion petrography</u>

- 466 The studied FIAs contain invariably a two-phase fluid (liquid-vapour) and are mainly arranged in secondary trails within Qtz I
- 467 crystals in the damage zone (Type S1) and also within Qtz I fault core, where they form dismembered (Type S2) trails and also
- 468 appear as individual clusters inside the crystals affected by intracrystalline viscous-crystal-plastic deformation (Type S3). Within
- 469 the Otz II fault core, FIAs are arranged as pseudosecondary (Type PS) and secondary (Type S4) trails. Representative examples
- 470 of FI petrographic features are shown for each BFZ300-structural domain in Fig. 9. Table 1 gives-provides a schematic
- 471 representation of the location of the FI types-presented above, in addition to their location within the fault architecture and their
- 472 fluid properties.
- 473 Damage Zone: Within Otz I grains (Figs. 9a, and b), secondary FIAs are found as trails (Fig. 9a) that parallel what we interpret
- 474 as healed, old intracrystalline microfractures. These microfractures are likely to be old joints and hybrid fractures whose
- 475 orientation mimics that of the mesosopic BFZ300 structural features. In these assemblages, FIs have a maximum size ranging
- 476 between 2 and 20 μm, a regular equidimensional shape (i.e. negative crystal morphology), and arelatively uniform volume
- 477 fraction, φ ($\varphi = V_v/V_{tot}$, see section 3) ranging between (volume fractrions) \exists of 5 and -15% (Fig. 9b).
- 478 Fault Core: Otz I grains host secondary FIAs (Type S2), which are transgranular trails (Fig. 9c) representing along healed joints
- 479 and hybrid fractures. These trails are locally interrupted and dismembered by aggregates of new, fine-grained quartz grains (Fig.
- 480 9c), and generate a texture that is indeed typical of Qtz I from the fault core (ef. Fig. 6a). Fluid inclusions entrapped along these
- 481 trails (Type S2) vary in size between 1 and 10 μm, have a φ of 10-20%, and show a negative crystal morphology (Fig. 9d). Fluid
- 482 inclusions are also found as isolated clusters inside intensely recrystallised quartz domains (Fig. 9c). FIAs inside these

483 recrystallized quartz domains were pervasively obliterated during later episodes of ductile deformation. The development of 484 WEBs, intercrystalline bands and bulging (ef. Fig. 6) resulted in the transposition remobilization (i.e., "transposition" sensu 485 Anderson et al., 1990) of these assemblages. This is invariably regularly observed and is documented, for instance, by the presence 486 of short, secondary trails of regularly shaped inclusion oriented at a high angle with respect to a longer, parent trail (Fig. 9c). 487 Morphologically, these trails resemble the transposed trails documented in high-grade metamorphic rocks (Andersen et al., 1990; 488 Van den Kerkhof et al., 2014). Different types of fluid inclusion morphologies are found within the intensely recrystallized quartz 489 domains (Fig. 9f). Negative crystal morphology is observed in some areas of the selected samples, but it is uncommon. More 490 typical is instead the "dismembered" morphology (cf: Vityk and Bodnar, 1995; Tarantola et al., 2010), which is observed in the 491 relatively large inclusions (> 20 µm). This morphology is made of a central (often empty) inclusion, showing several tails and re-492 entrants, surrounded by a three-dimensional clusters of small "satellite" inclusions. These clusters might be arranged with a quasi-493 planar geometry inside the host (i.e. in a trail-like fashion). Another typical texture found in most assemblages is the "scalloped" 494 morphology of small- to medium-sized inclusions (<10-15 µm), which is defined by the presence of indentations, embayments, 495 irregularities, and sharp tips of the inclusion walls (Fig. 9f). Small inclusions (<1 \mum) are also found at the edge of the straight, 496 regular boundaries of new quartz grains; they are mostly dark, i.e. they are vapour-rich or empty, and are equant in shape (Fig. 497 9e). Although small inclusions do not allow a microtermometric study of the fluid-phase behaviour in this structural domain, they 498 confirm the complex reactivation history of BFZ300. 499 Qtz II contains both pseudosecondary (Type PS) and secondary (Type S4) assemblages (Figs. 9g, -ii, hi). The first type is arranged 500 in trails, that cut at low angle the hosting quartz but not the neighbouring phases (e.g., chlorite,). In these assemblages, FIs are 501 relatively large (2-45 µm). They and exhibits how elongated shape and their of varies values between 15 and 30 % (enlargement 502 in-Fig. 9 gh). Type -S4 FIAs (Fig. 9hi) have host two-phase inclusions whose size (5-35 μm) is similar to that of PS trails, but 503 show a φ between 30 and 40 % (Fig. 9 ii). 504 PRare primary FIAs are also present along growth planes of Otz II and are best observed predominantly in the least deformed 505 Otz II crystals, where they have a relatively large size (20-50 μm; Figs. S3a, b, c of the Supplementary Material). Telsewhere 506 they present irregular and "dismembered" textures, which suggest intense post-entrapment re-equilibration. Primari FI textures 507 are shown in Figure S3 of the Supplementary Materials. 508 In summary, our microtextural study shows that the FIAs to be selected for the microthermometric study are only those hosted 509 within Qtz I and Qtz II crystals with the minimum degree of little to no recrystallization and whose inclusions have textures

513514 4.3.2 Microthermometry

the host quartz exhibits only undulose extinction (S1, S2, S4 and PS).

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corresponding to the least intense post-entrapment re-equilibration (Bodnar, 2003b, and references, therein; Tarantola et al.,

2010). These are the pseudosecondary and secondary FIAs in which dendritic or transposed inclusions are absent, and in which

515 Damage Zone: The majority of s Secondary FIAs hosted within Qtz I from the damage zone (Type S1) show a range of T_{mice} 516 between -5.9 and -0.1 °C, which corresponds to a salinity of 0-9 wt% NaCleq (Fig. 10a). In these FIAs, final homogenization 517 (Thto) occurs into the liquid phase (i.e. by disappearance of the vapour bubble) and mainly between 150-400 330 °C (Fig. 10e). 518 Fault Core: The secondary FIAs hosted within Qtz I of in the fault core (Type S2) show a range of T_{mice} between 3.9 and 0 -8.2 519 and -0.4 °C, which corresponds to salinities between 0 and 614 wt% NaCleq (Fig. 10b), and final homogenization occurs into the 520 liquid phase by bubble disappearance is between 150130 and 420410 °C (Fig. 10f). 521 Pseudosecondary FIAs entrapped within Otz II The (Type PS) show a range of T_{mice} between -4113.6 and 0-0.1 °C, which 522 corresponds to a salinity range betweenof 0 and -18 15.2 wt% NaCleq (Fig. 10c); , and final homogenization occurs into the liquid 523 phase and is comprised between 150 and 440 °C (Fig. 10g). Secondary FIAs in Qtz IIThe (Type S4) show a range of T_{mice} between 524 -7.3-11 and 0 °C, which corresponds to a 0-15-10.9 wt% NaCleq range of salinity (Fig. 10d), while final homogenization into the 525 liquid phase is comprised between 150–130 and 430 °C (Fig. 10h). 526 As no gases were detected determined during microthermometric analysis (i.e. melting of carbonic phase or clathrate hydrates 527 were not detected during the freezing experiments), additional micro Raman analysis has been was performed on a set of 528 representative FIAs (samples: TPH-120-4; TPH-120-6; PH21; PH22).on several FIAs for the detection of gases into the studied 529 agueous inclusions. Aqueous fluid inclusions hosted both by the Otz I and Otz II show peaks at the characteristic wavenumbers 530 of CH₄ (2917 cm-1), and CO₂ (1388 cm-1). These peaks were determined as weak in all spectra, and CO₂ detection was only 531 sporadic in a few inclusions in of only one one of the sample of the fault core (TPH-120-4A). Such These spectroscopic 532 determinations are consistent with the lack of microthermometric evidence for CO2 or CH4 occurrence in the FIAs, i.e., with the 533 failure to detect melting of a carbonic phase or clathrate hydrates during the freezing experiments (cf. Rosso and Bodnar, 1995; 534 Dubessy et al., 2001). Although spectroscopic detections, the CO₂- and CH₄--bearing inclusions are not systematically associated 535 with distinct specific quartz vein generations or specific microstructures (i.e. intracrystalline healed cracks, WEB's planes, 536 intercrystalline fractures). WTherefore, we can not therefore associate the presence of CO₂ and/or CH₄ to any specific deformation 537 stages of the fault. 538 Such spectroscopic determinations are consistent with the lack of microthermometric evidence of carbonic phase or clathrate 539 hydrates during the freezing experiments (cf. Rosso and Bodnar, 1995; Dubessy et al., 2001). The impossibility to detect CO₂-540 and CH₄-bearing fluids during the freezing experiments indicate a gas pressure that is systematically lower than that required to 541 observe clathrate dissociation (e.g., 1.4 MPa in CO₂-H₂O fluids, Rosso and Bodnar, 1995), i.e. it shows low gas concentrations. 542 As a consequence, we have modelled the fluid phases as simple H₂O-NaCl systems... 543 The impossibility to detect CO₂ and CH₄ bearing fluids via microthermometric determinations indicates a gas pressure in the 544

The impossibility to detect CO₂—and CH₄ bearing fluids via microthermometric determinations indicates a gas pressure in the analysed inclusions that is systematically lower than that required to observe clathrate dissociation (e.g., 1.4 MPa in CO₂ H₂O fluids, Rosso and Bodnar, 1995), i.e. it shows low gas concentrations, systemsOlkiluoto fluid. Considering the broad salinity range of 0.1-14 wt% NaCleq for the BFZ300 fluids (which corresponds to NaCl concentrations of 1.7-10-3—2.4 M), we cannot estimate a maximum CH4 concentration.

4.4 Chlorite and sulphide geothermometry

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549 Chlorite composition has been determined for several generations of chlorites in association with Otz I and II, namely vermicular 550 chlorite associated with Otz I from the damage zone, vermicular and radiate chlorite associated with Otz I from the fault core, 551 and radiate chlorite associated with Otz II from the fault core (Table 2). Chlorite compositions are plotted in the classification 552 diagram of Hey (1954) are shown in Figure 11a and they are expressed as function of the Fe/(Fe+Mg) -ratio. (i.e. XFe). Chlorite 553 compositional data are presented for according toeach the structural domain of the fault they are associated with and for the 554 distinct chlorite to the corresponding textures. Vermicular chlorite associated with Qtz I in the damage zone has a XFe range 555 between 0.50 and 0.55, while vermicular chlorite associated with Qtz I from the fault core has a XFe of 0.53. Radiate chlorite 556 associated with Qtz I from the fault core has a XFe range between 0.71 and 0.81 while radiate chlorite associated with Qtz II from 557 the fault core is between 0.65 and 0.80, constraining. Then, dominant compositions are within the ripidolite and aphrusiderite 558 end-members. This plot highliths distinct clusters of chlorite compositions across the fault architecture as possible due to the 559 distinct fault reactivation stages. The EPMA data show that, in general, the BFZ300 chlorites are Fe-rich (XFe = Fe/(Fe+Mg) between c. 0.65 and 0.90), have concentrations of (Na2O+K2O+CaO) <1 wt%, and result mainly from a solid solution of the 560 561 sudoite and daphnite end members, i.e., of Mg2Al3(Si3Al)O10)(OH)8 (Fe,Mg)5Al(Si,Al)4O10(OH)8. The dataset shows in 562 particular that the compositions of the distinct chlorite generations vary systematically with vein generation, as shown by the 563 vermicular chlorite associated with Qtz 417 I veins (Fig. 8e) with a XFe between 0.65 and 0.67 and the radiate chlorite associated 564 with Otz I from the fault core and Otz II (Figs. 8f and 7b) with a XFe between 0.76 and 0.86.

565 Temperature-composition relationships for the quartz-chlorite pair portraved in the T-R2+Si diagram of Bourdelle and 566 Cathelineau (2015) show that, in the hypothesis of quartz-chlorite equilibrium, the precipitation of vermicular chlorite within the 567 Otz I of the damage zone took place in the 175-2450 °C range (green diamonds of Fig. 11a). This range is distinct from that of 568 the vermicular and radiate chlorite from Otz I of the fault core, which is probably c.-350 °C because the measured R²⁺-Si compositional parameters ($R^{2+} = Mg + Fe$) plot at the edge of, or slightly outside, the calibrated region of the Bourdelle and 569 570 Cathelineau plot (red diamonds of Fig. 11a). We stress that the high-T chlorite plots parallel to the 350 °C isotherm, suggesting 571 that it precipitated most probably at the same, or at a similar, temperature. Radiate chlorite associated with Otz II from the fault core is instead compatible with an equilibrium precipitation at 160-220 °C (light-blue diamonds in Fig. 11a).

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573 The collected EPMA data show that the sulphides associated with Qtz II have compositions that approach those of pure phases 574 (Table 3). Pyrite has trace element concentrations (Cu, As, Pb, Ni, Zn) that are in general below the EPMA detection limit, while 575 galena, sphalerite, and chalcopyrite show only some significant trace contents of Fe and Zn (e.g., Fe: 0.22-1.00 wt% in galena; 576 Zn: 0.11-3.95 wt% in chalcopyrite). Pyrite and sphalerite from the Qtz II veins (Fig. 7e) have trace element concentrations that

577 are, again, mostly below detection limits.

578 The stylolites bordering the cataclasite bands described above and formed at the contact between the Otz I and Otz II vein contain 579 pyrite, galena, and the sphalerite-stannite pair (Figs. 8a, c, d), with the latter showing the largest compositional variation. This 580 pair represents a mineral geothermometer because the partitioning of Zn and Fe between sphalerite and stannite was demonstrated

- to be temperature dependent but pressure independent (Nekrasov et al., 1979; Shimizu, M.-and Shikazono, 1985). In the fourteen analysed pairs, stannite shows a range of Zn concentrations varying between 0.48 wt% and 3.25 wt%, while those of Fe, Cu and Sn vary within narrow ranges (Fe: 12.74±0.56 wt%; Cu: 28.30 ±0.33 wt%; Sn: 27.65 ±0.71 wt%). Sphalerite in the pair has concentrations of Fe and Zn of 7.63±0.87 wt% and 56.68 ±1.17 wt%, respectively. These ranges allow the calculation of the partition coefficient (K_D) of the reaction: Cu₂FeSnS₄ (in stannite) + ZnS (in sphalerite) = Cu₂ZnSnS₄ (in stannite) + FeS (in sphalerite). We have used the logK_D-T relationship of Shimizu and Shikazono (1985) to calculate the formation temperature of the pair, which is portrayed in the (X_{Cu2FeSnS4}/X_{Cu2ZnSnS4})-(X_{FeS}/X_{ZnS}) plot of Shimizu and Shikazono (Fig. 11b). The resulting 220-
- 588 305 °C interval lies at the low end of, or slightly outside, the 250-350 °C interval of the geothermometer.
- Therefore, we consider hile the 250-305 °C interval can be taken as an estimation of the formation T of sphalerite stannite in the stylolite, the 220-250 °C interval should be taken with caution. as anthe best estimation of the formation T of sphalerite-stannite in the stylolite.

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5 Discussion

Our work constrains the structural architecture and the environmental conditions at which BFZ300 deformation took place. Field and petrographic observations support the idea of transiently elevated fluid pressures, cyclic frictional-viscous deformation and progressive, yet discrete strain localization (Figs. 2, and 3). Analytical data suggest that these deformation cycles took place at the BDTZ. In the following, we discuss these constraints by systematically considering our different analytical results.

5.1. Fluid inclusion data and mineral-pair geothermometry

- 599 Field evidence combined with microstructural observations, fluid inclusion analyses and the documented distinct generations of
- 600 synkinematic chlorites confirm that Qtz I and Qtz II veins precipitated from distinct generations batches of aqueous fluid (i.e.
- 601 H₂O-NaCl)pulses, repeatedly and actively injected into that ingressed the BFZ300-fault zone during different stages of its
- 602 evolution. Microthermometric results suggest that these fluids were in a homogeneous liquid state at the time of entrapment, as
- 603 testified by the consistent final homogenization into the liquid phase (i.e. by bubble disappearance).
- Microthermometric and Raman spectrometry data show that the fluid entrapped within the studied FIAs at the time of formation
- of the damage zone and fault core during precipitation of Qtz I and Qtz II veins can be represented by a H2O-NaCl model fluid.
- The fluid was in a homogeneous state at the time of entrapment, as testified by the consistent final homogenization by bubble
- disappearance. It also had a low bulk salinity, as shown by the distribution of >80% of the ice melting (Tmice) measurements
- 608 skewed towards values of 3 °C or higher, which corresponds to bulk salinities of 5 wt% NaCleq or less (Fig. 10a d).
- 609 We documented a wide range of bulk salinity range in for each FIAs entrapped within the quartz veins in each structural domain
- 610 (Figs. 10-a-d-e-e-g). This suggests post-entrapment re-equilibration of fluid inclusions (cf. Bakker and Jansen, 1990; Diamond et
- al., 2010. The Th_{tot} varies between c. 130 and 440 °C without a clear mode or a skew (Figs. 10e-he-h) indicating and shows that

612 no common range of entrapment temperatures can be identified in the dataset. Therefore, we conclude that even the properties of 613 individual, petrographically intact FIAs do not correspond with to chemically well-preserved assemblages. Indeed, the ranges of 614 Thtot in individual FIAs are typically of the order of 150-200 °C (Figs. 10e-h $\frac{10e-h}{10e-h}$), i.e. a value that is much higher than the ~10 615 °C range expected for homogeneous FIAs entrapped isochorically and isoplethically (Fall et al., 2009; Vityk and Bodnar, 1998) 616 and that demonstrates post-entrapment re-equilibration (cf. Vitik and Bodnar, 1998; Bodnar, 2003b; Sterner and Bodnar, 1989; 617 Invernizzi et al., 1998). A major implication of fluid inclusion re-equilibration in our study is that the calculated fluid properties 618 do not rigorously reflect those of the pristine fluid originally entrapped within BFZ300, but rather that of a fluid that modified its 619 properties during the fault activity. This is comparable to the results of other fluid inclusions studies from faults (Boullier, 1999; 620 Garofalo et al., 2014; Roedder, 1984). 621 Then, a possible approach to interpret our FI dataset is the comparison with the experimental work on synthetic fluid inclusions 622 subjected to a range of post-entrapment re-equilibration conditions (Bakker, 2017; Bakker and Jansen, 1990, 1991, 1994; Vityk 623 and Bodnar, 1995, 1998; Vityk et al., 1994; Invernizzi et al., 1998). Such comparison A straight comparison to the experiments is 624 in our case difficult because most experimental work was carried out at high TP conditions (500-900 °C; 90-300 MPa) and also 625 only few experiments were carried out under deviatoric stress conditions that approach those of natural rocks (Diamond et al., 626 2010; Tarantola et al., 2010). Despite these limitations, however, some key experimental results provide fundamental constraints 627 on our dataset. First, both hydrostatic and uniaxial compression experiments showed that in each re-equilibrated FIA a number 628 of inclusions survive virtually intact the modified post-entrapment PT conditions, showing that only severe deformation brings 629 to total re-equilibration and complete obliteration of pristine inclusions (i.e., $\Delta \sigma > 100$ MPa in uniaxial compression experiments; 630 >400 MPa change of confining P in hydrostatic experiments). Second, under conditions leading to only low to moderate re-631 equilibration, the bulk chemical composition of the fluid inclusions does not change significantly from that of the pristine 632 inclusions. 633 All of this implies that natural quartz samples with microstructures typical of moderate T deformation, such as deformation 634 lamellae, deformation bands, undulose extinction and bulging, and hosting FIAs with moderately re-equilibrated textures, should 635 still contain a number of inclusions whose properties resemble those of the pristine fluid. In this scenario, our microthermometric 636 dataset can be used to constrain the more probable salinity ranges of the fluid batches which trigger BZ300 reactivation stages. 637 Two possible interpretations of the microthermometric dataset can be follow and we can give accordingly different salinity ranges 638 for the fluids. 639 One possibility is that the different quartz veins and the fluids trapped within the fluid inclusions originated from multiple pulses 640 of a single, low—to intermediate salinity fluid, with a salinity between 0 and 7 wt%NaCleq, as shown by the distribution of >70% 641 of the bulk salinities skewed towards values of 7 wt% NaCleg or less (Fig. 10a-d). Thus, it is possible that an aliquots of the 40-642 75 wt% NaCleq FIAs from Qtz I and II crystals from both the damage zone and fault core is still representative of the pristine 643 sampled fluid. These inclusions would be those that survived or were relatively less affected by deformation events postdating 644 their entrapment. Inclusions falling outside the most typical 40-5-7 wt% NaCleq salinity range would instead correspond to those which progressively modified their properties as a consequence of fluid-rock interaction during faulting and to those that 645

646 experienced significant H₂O loss and consequent salinity increase during the successive stages of fault deformation (cf. Bakker 647 and Jansen, 1990; Diamond et al., 2010). The large documented range of Thiot lacking a specific mode observed in individual 648 FIAs is the product of fluid density changes caused by fluid inclusion re-equilibration during post-entrapment deformation. This 649 would have happened repeatedly and cyclically within the host quartz during all ductile and brittle stages of deformation of the 650 multi-stage deformation history of BFZ300. 651 Alternatively, multiple batches of fluids with different salinities (from low to intermediate salinity) may have ingressed and 652 evolved within BFZ300 during its activity. In fact, considering the salinity dataset presented for each structural domain, fluid 653 salinity can be seen clustering in restricted ranges typical for each domain: 1) the salinity of 60% of secondary fluid inclusions in 654 Otz I from the damage zone is between 0 and 1 wt%NaCleq; 2) > 80% of the secondary inclusions in Qtz I from the fault core 655 preserve a salinity in the 1 to 5 range wt%NaCleq; 3) 75% of pseudosecondary inclusions in Qtz II show salinity values between 656 6 and 11 wt%NaCleg and 4) ~70% of the secondary inclusions trapped within Otz II show salinity values between 0 and 3 657 wt%NaCleq. These clusters may best represent the original compositional ranges of different batches of fluids, each involved 658 during a different faulting stage. Salinities outside these clusters may instead be explained again as resulting from the post-659 entrapment re-equlibration of those fluids with different salinities. This hypothetical scenario, in which chemically distinct fluids 660 ingressing the fault and interacting with the rock at different times (e.g. Selverstone et al., 1992; Boiron et al., 2003; Famin et al., 661 2005) is also reinforced by several lines of observation such as: the variation of chlorite composition, the slight change in 662 paragenesis/redox state with Quartz II and Quartz I (i.e. the absence of massive sulphides) and by the prolonged history of faulting 663 (see below). 664 Fully aware of these interpretative uncertainties of our dataset limitations, we have combined the microthermometric data of the 665 studied FIAs with the independent quartz-chlorite and sphalerite-stannite geothermometers to constrain the most probable fluid 666 pressure during the faulting events. With this approach, we use the formation temperatures of the mineral pairs as independent 667 geothermometers and consider the intersection between these values and the FIA isochores to derive the ranges of trapping 668 pressure (cf. Roedder and Bodnar, 1980). In Fig. 12, we present the Pf ranges that are calculated using the entire salinity range of 669 the studied FIAs (cf. Fig. 10); however, we highlight the most probable Pf ranges that are consistent with what we consider best 670 preserved salimity range (0.5 wt% NaCleq). Accordingly, for the damage zone we estimate a Pf interval of 20.90 MPa (Fig. 12a) 671 by intersecting the range of T obtained from the chlorite-quartz pair in the damage zone Qtz I (T c.170-240 °C, Fig. 11a) with the 672 range of isochores from the same quartz. As to fluid pressure estimations in the fault core, we combine the 350 °C obtained from 673 the chlorite quartz pair from the fault core Otz I (T>350 °C are outside the calibrated range of the geothermometer) with the 674 ranges of isochores from the same quartz, from which we obtain Pf ranging between c. 140 and 120 MPa (Fig. 12b). Similarly, 675 the intersection between the equilibrium T of the sphalerite stannite pair in the Qtz II fault core (250 305 °C) and the range of 676 isochores of the Type PS FIAs of Qtz II (Fig.9) defines Pf values ranging between 10 and 140 MPa (Fig.12e). Estimations from 677 Type S4 FIAs (Fig.9) constrain a range between 40 and 160MPa (Fig. 12d). We propose that these values are sufficiently accurate to constrain multiple stages of fault slip, each one triggered by a fluid pulse having a distinct pressure. Hence, fault activity started 678

679 at 200 °C and at Pf varying between 20 and 90 MPa and continued through higher temperatures (305-350 °C) and Pf (120-160 680 MPa). 681 In Figure 12, we present the ranges of the possible fluid-pressure (P_f)s of the fluids involved during faulting as $\frac{1}{2}$ -calculated by 682 combining from the fluid inclusion analyis data and with contrained the constraints by provided by the pair-mineral 683 geothermometry and the hydro- and lithostatic pressure gradients and a possible geothermal gradient reconstructed regional geothermal gradients(e.g. Van Noten et al., 2011; Selverstone et al., 1995; Jaques and Pascal, 2017). The reconstructed regional 684 685 gradients- present at the time of vein emplacement -are derived related from peak metamorphic conditions (4-5 kbar; 650-700 °C leading to c. 40 °C/km;, from Kärki and Paulamäki, 2006). We used the geothermal gradient-Hydrostatic and lithostatic pressures 686 687 are then calculated by using pure water density and assuming a rock density of 2700 kgm⁻³, respectively, to calculate the idrostatic 688 and lithostatic pressure assuming a rock density of 2700 kg m³. These gradients are used to give constrain the upper and lower 689 bounds to physically possible fluid pressures. We computed the maximum and minimum isochores ealeulated by using the entire 690 salinity and T_{htot} ranges obtained for from the FIAs in each structural domain (ef. Fig. 10). We also computed the isochores of the 691 inclusions with the most representative salinity estimatesies evaluate for each structural domain, that we considered as indicative 692 of the most probable active fluid phase obtained by. To estimate the most pProbable compositions of the distinct batches of fluids 693 wes were determined compared ving the ing frequency diagrams (Fig. 10) with the and Thiot vs. salinity plots (see Supplementary 694 Materials Fig. S4). Considering the peak temperature of each structural zone obtained from the geothermometric estimations 695 combined in combination with the computed isocores, the estimated peak conditions of the fluid pressure are: 1) 80 MPa for Otz 696 I from the damage zone, 2) 210 MPa for Otz I from the fault core—Otz I: 3) 140 MPa from pseudosecondary inclusions in Otz II 697 from the core and 4) 180 MPa from secondary inclusions in Otz II, still from the core (Fig. 12; Table 1). 698 In addition to the $P_{\rm f}$ peak conditions we can also constrain the physically possible Other possible fluid pressure ranges for each 699 stage of fluid ingress, which are given-derived by considering the temperature range estimated for each structural domain. These 700 pressure may be interpreted as the result of re-equilibration and progressive reactivation of the system recorded both by fluid 701 inclusions and geothermometric estimations on authigenic minerals. Accordingly, Thus, for the damage zone, we estimate a Pf 702 interval of 50-80 MPa (Fig. 12a) can be derived by intersecting the range of T obtained from the chlorite-quartz pair in the Otz I from the damage zone Qtz I (T e.175 240 °C, Fig. 11b) with the range of isochores from the same quartz. As to fluid pressure 703 704 estimations in the fault core, we combine the 350 °C constraint obtained from the chlorite-quartz pair from Qtz I in the fault core 705 Otz I (T>350 °C are outside the calibrated range of the geothermometer) with the ranges of isochores from the same quartz, from 706 which we obtain which yields P_f ranging between c. 30 and 210 MPa (Fig. 12b). Similarly, the intersection between the equilibrium 707 T of the sphalerite-stannite pair in the Qtz II from the fault core (250 305 °C) and the range of isochores of the pseudosecondary 708 FIAs of Otz II (Type PS, Fig. 9g) defines P_f values ranging between 50 and 140 MPa (Fig. 12c). Estimations from secondary 709 FIAs in Qtz II-(Type S4, Fig.9i) constrain a range between 40 and 180 MPa (Fig. 12d). 710 As also illustrated supported by the microstructures described above, we propose that these values are sufficiently accurate to 711 constrain at least four stages of fault reactivation, each one-triggered by a fluid pulse having awith distinct phisico chemical 712 conditionsphysical and compositional properties.

- 713 As suggested shown by the pressure T vs. P plots of Figure 12, the secondary FIAs entrapped in Qtz I from the damage zone 714 show constrain the lowest value of P_f (i.e. 50-80 MPa) in of the entire dataset. We interpreted this not as representative of the 715 early BFZ300 localisation, but rather as the possible result of possibly resulting from -fluid entrapement during the latesta later 716 stages of fault activity reactivation at T, occurred at lower temperature (~200 °C). This is also also consistent with the estimated 717 calculated temperature of crystallization of the formation range of vermicular chlorite associated with Otz I from the damage 718 zone (175-240 °C, Fig. 11b) and with the secondary nature of the entrapped FIAs. Also, the most abundant salinities observed in 719 the Otz I from the damage zone (0-1 wt%NaCleq) coincide with the lowest Thiot measured in the same structural domain. The 720 latest Later ffracturing of Qtz I in the damage zone were may thus have been coeval correlated with the formation of vermicular 721 chlorite preserved therein, which is generally arranged found along secondary cracks and median lines (Fig. 5d).
- In the light of all these considerations, we interpreted propose that initial BFZ300 localization occurred fault activity started in the presence of a fluid with T and P of at at least 350 °C or even higher temperature and at P_{ff probably} higher than 210 MPa, respectively. Later faulting and continued by cyclic brittle-ductile switches induced and assisted by fluid batchesthrough attemperatures and fluid pressure.

5.2. Structural evolution and fluid flow: a conceptual model

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727 BTherefore, bBased on the integration of our field, microstructural, thermometric and fluid inclusions constraints (summarized 728 in Table 1), we propose a conceptual model for the structural evolution of BFZ300 (Fig. 13). The fault's finite strain results from 729 several slip episodes mediated by multiple events of fluid ingress and fluid-rock interaction. A first constraint provided by our 730 study is that the analysis of the bulk chemical composition of the fluids that repeatedly circly cally flowed within ingressed the fault 731 are suggests characterized by specific values of T, Pr and salinity, did not change significantly during the documented fault activity, 732 as the best preserved 0.5 wt% NaCleg salinity range points to a compositionally homogeneous fluid. This suggestings the likely 733 presence of several a-batches of fluids of varying salinity and compositioneompositionally heterogenous homogeneous source 734 region of the fluids, which also their own evolve and modify their properties as the results of fluid rock or, alternatively, that the 735 studied section of the fault did not interactions, with fluids of substantially different composition. 736 The embrittlement of the Olkiluoto metamorphic basement (time t₁ of Figs.g. 13a, b) represents the initial stage of the 737 deformational history of BFZ300, when conditions for brittle dilation and fracturing of the Paleoproterozoic basement were first 738 met in a transient fashion. We propose that brittle failure under still ductile environmental conditions was caused by transiently 739 elevated P_f (> 210 MPa) (probably under peak pressure major than 210 MPa), as also demonstrated by field evidence of 740 hydrofracturing (pure tensional en enchelon veins at the BDTZ depth₂) Figss. 2 and 3), and high fluid temperature (~350 °C or 741 even higher), and the pore pressure estimations (Fig. 12 and Table 1). Hydrofracturing of the host basement is also expressed 742 indicated by the emplacement of Qtz I veins along within the diffuse network of joints and conjugate hybrid/shear fractures of 743 the damage zone (Figss. 13a, and 3a, b2g). These brittle features are quite evenly broadly distributed within the damage zone 744 suggesting an initial volumetrically diffuse strain distribution. Their formation caused the overall mechanical weakening of the

actively fracturing host rock volume, which in turn facilitated later strain localization. Brittle structures formed during this stage 746 are discordant to the ENE-WSW striking metamorphic foliation (Fig. 1b), which they cut at high angle (Fig. 13a). Conditions for tensional and hybrid failure require low differential stress, i.e. σ_1 - σ_3 ~ 4T, where T is the tensional strength of the rock. Opening 747 748 of fractures caused a stress drop, sudden increase of permeability, fluid venting and inhibited further build-up of Pf. Dilatant 749 fractures were partially infilled by Qtz I, which precipitated from a first pulse of the low salinity fluid, with inferred low salinity 750 (in the range between 1 and 5 wt%NaCleq fluid for comparison with the salinity estimated from Otz I fault core). 751 Precipitation Crystallization of Otz I and formation of veins within these fractures caused hardening of the system. The progressive 752 recovery of shear stresses concomitant with the progressive sealing of dilatant fractures altered the overall background stress 753 conditions such that failure, after causing initial pure dilation, was later accommodated by hybrid extensional failure—and, 754 eventually, by shear fracturing (Fig. 13b), thus forming laterally continuous and interconnected shear fractures associated to-with 755 breccia pockets and cataclasites (Figs. 3d, g, i)2e g i and 3bd). Conjugate shear fractures connected the previously formed 756 extensional fractures through a fracture coalescence mechanism (e.g. Griffith, 1921; Sibson, 1996); fracture coalescence 757 mechanism in Otz I is showed by a straight, red line in Fig. 13a). At the micro-scale this is demonstrated by the elongated blocky 758 texture of Otz I crystals from the damage zone (Figs. 4c and 5a44), where crystals grew at high angle to the vein boundaries (thus 759 suggesting initial near-orthogonal dilation) and are physically connected by cataclastic shear bands to form a fault-fracture mesh 760 (e.g. Sibson, 1996; Figures 4ab). Cataclastic bands formed at the expenses of the migmatitic host rock are enriched in authigenic, 761 synkinematic sericite, likely due to the interaction between K-feldspar and fluids circulating in the dilatant fault zone (Fig. 4b). 762 Shear fractures thus deformed the migmatitic host rock to connect dilatant and mostly Otz I-filled tension gashes during a 763 continuum of deformation. The conjugate shear fractures ascribable to this stage invariably define tight acute angles (Figs. 2b, 764 3a), which we take as further evidence of overall low differential stress conditions at the time of failure (Fig. 13b). 765 In synthesis, Otz I veins from the damage zone are interpreted as the expression of the earliest stage of fault nucleation, before 766 strain localization affected a progressively narrower rock volume to eventually form the main fault core. Indeed, the meso- and 767 microscale features observed in Qtz I-in the damage zone, lacking of pervasive crystal-plastic recrystallization deformation as 768 otherwise occurred in Qtz I-fault core, are used to document the initial stage of embrittlement. preserves mostly brittle 769 microstructur, twithbatcherebyes and lacks a pervasive ductile overprint, which is instead prevalent within the fault core. As a 770 consequence, we interpret the chemical properties of the fluid derived from these veins as the closest to the initial conditions of 771 the first fluid involved in BFZ300 nucleation. Fluid inclusion and geothermometric estimations from the sinkinematic chlorite 772 crystals associated with the damage zone Otz I (Figs. 5a and 11a), suggest chlorite precipitation at a T of c. 200° C and P_c between 773 e. 90 and 20 MPa at the time of fault nucleation. Based on geometric, kinematic and deformation style characteristics, we 774 tentatively assign this deformation episode to Stage 1 by Mattila and Viola (2014,)-(their Fig. 18), i.e. to a discrete brittle episode 775 that they considerconsidered the expression of the earliest onset of brittle conditions in southwestern Finland c. 1.75 Ga ago, 776 under overall NW-SE to NNW-SSE transpressive conditions. 777 Further deformation of the BFZ300 (time t₂ of Fig. 13c) occurred by progressive inward strain localization and narrowing of the 778 actively deforming volume of the deformation zone (from a wide damage zone to a narrow fault core). The early BFZ300 core,

779 consisting of the main Qtz I vein is interpreted as having formed at this stage, within an overall dextral strike-slip kinematic 780 framework. Emplacement of the Qtz I vein in the core represents the last pulse of this brittle deformational episode (Fig. 13b). 781 Major fluid venting was likely associated with it, such that the system, once brittle failure in the core had occurred by 782 hydrofracturing, moved back to a more diffuse deformation style typical of the still prevailing ductile conditions. Microscopic 783 evidence of ductile crystal-plastic deformation by and dynamic recrystallization (Figs. 6-a, -b₃. Table 1) overprinting the early 784 brittle structures of Otz I in the fault core supports slow strain rate conditions during deformation. However, this viscous ductile 785 background deformation was punctuated by renewed and cyclically transient embrittlement as documented by healed fractures 786 shown by trails of secondary fluid inclusions cutting across both the ductile fabrics and the earlier brittle deformational features 787 (Figs. 6c₅, -d, -and e). In accordance, EBSD results performed on analysis of the new grains documented along healed microcracks 788 also suggests that they likely nucleated from fluids circulating in the early fractures before being later deformed result from quartz 789 deformation in the low-temperature plasticity regime. In such regime. Thus, we show that at the BDTZ 'neocrystallisation' by 790 nucleation and growth in fractured fragments and dynamic recrystallisation (typically by bulging and subgrain rotation) and 791 'neocrystallisation' by nucleation and growth in fractured fragments coexist and compete in the overall microstructural evolution 792 of quartz (e.g. Kjøll et al., 2015). Accordingly, the microstructures showed in Otz I from the fault core show evidence for both 793 processes being active during deformation of Otz I grains. Initial nucleation from circulating fluids along now sealed cracks is 794 proposed to have caused fracture healing and sealing. At the same time, and in light of targeted EBSD analysis that we have 795 performed to better understand Qtz I crystallization in the fault core (see below), we can also document the local importance of 796 dynamic recrystallisation by bulging and subgrain rotation. The combination of both mechanisms recalls the results by Kiøll et 797 al. (2015), which proposed the combination of these mechanisms after a detailed microstructural analysis in quartz veins 798 associated with a thrust, formed at the brittle ductile transition. Repeated pulses of high P₁ (peak conditions: 210120-140 MPan) 799 likely triggered these brittle ductile oscillations. Repeated fluid ingresses and related deformation would, in addition, also have 800 caused some of the post-entrapment equilibration of the FI, as discussed above. 801 The cycles of brittle and viscous deformation may be explained as follows. Cyclic brittle failure would have repeatedly lowered 802 Ps. which lowered the background stress and strain rate and favoured ductile deformation by dynamic recrystallization at T > 300 803 °C between the slip events (e.g. Passchier and Trow, 2005). The fault regained cohesive strength after each brittle failure episode 804 through vein formation and sealing/healing of the fracture networks. Porosity destruction by mineral crystallization and fracture 805 sealing, as clearly shown by CL imaging (Fig 4d), induced a progressive reduction of permeability and mechanical healing of the 806 fault, which promoted an increase of P. and ultimately triggered a new brittle failure. Therefore, pore pressure build up promoted 807 episodic brittle fracturing followed by cementation and plastic deformation/recovery. The compelling evidence for this 808 deformation occurring at T

350° C indicate that the described processes identify the BDT of the quartz feldspathic crust 809 (Kohlstedt et al., 1995). 810 Mattila and Viola (2014) described a second brittle stage (referred to as Stage 2, their Fig. 18) during which a c. N-S to NNE-

SSW-oriented episode of transpressional deformation affected southwestern Finland. Geometric and temporal relationships

between structures of Stages 1 and 2 (see also Viola et al., 2009) were used to infer a clockwise rotation of the horizontal

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813 compression direction from NW-SE (Stage 1) to NNE-SSW (Stage 2). Consistent with the kinematic framework of Stage 2, we 814 propose here that during progressive regional exhumation and cooling to entirely brittle conditions, the BFZ300 deformation 815 continued through a further, distinct deformation phase (t₃ of Fig. 13e). This stage accommodated the selective reactivation of the 816 BFZ300 core, with renewed dilation due to the rotated σ_1 during Stage 2 acting subparallel to the strike of the Qtz I vein in the 817 BFZ300 core. Localised dilation in a still fluid-rich system allowed the emplacement of the Qtz II vein (Fig. 13e). Our estimations 818 indicate that peak conditions of P_r and T conditions at that time were 140 between 140 and 10 MPa and $T \approx 305^{\circ}$ C, respectively. 819 The BFZ300 core was reactivated by an intermediate salinity fluid (in the range between 6 and 11 wt%NaCleq) under overall 820 hybrid conditions (Fig. 13f), as suggested by the irregular thickess and curved geometry of the Qtz II vein therein, and by the 821 synkinematic chlorite crystals that are stretched orthogonally to the vein boundaries (Fig. 3he). The Qtz II vein invariably 822 localized along at the contact between Qtz I and the host rock (Figs. s.3f, 2, 3 and 13e) suggesting selective reactivation along 823 the pre-existing principal slip zones (Riedel shears and boundary shears, Tchalenko, 1970), which represented the weakest part 824 of the fault (strength profile Fig. 13h). Evidence for mesoscale hybrid fracturing and our P_f estimates (Fig. 4112) suggest that P_f 825 was lower than that of the earlier deformation stages during Otz I emplacement. 826

827 crystals (Fig. 7c). The Ffluid pressure peack value estimations for this structural stage is around. 180 phase are between 160 and 828 40-MPa. 829 A possible latest, very late BFZ300 reactivation stage (time, t_x), of unknown age is also documented by the secondary chlorite 830 associated with Otz I in the damage zone (Figs. 5a, d). The lowest temperature estimated from chlorite geothermometry (~ 200°C). 831 consistent with the lowest homogenitazion temperature of the greatest part of FIAs petrographically discriminated in this

BFZ300 underwent one or more events of brittle fracturing and induration (Fig. 13g), as suggested by the CL imaging of Otz II

832 structural domain, suggest that they probably represent a latest reactivation of the system, triggered by a batch of fluid with at 833 lower temperature (~ 200°C), lower pressure (peak condiiotns: 80 MPa) and lower salinity (0.1 wt%NaCleg). This deformation

834 stages may probably be not consistent with the deformation cycle here presented.

835 TAlso, the stylolitic seams having a strikestriking parallel to the BFZ300 fault zone suggest a direction of maximum compression 836 (6) oriented c. E-W, i.e. subparallel to the inferred Sveconorwegian main shortening direction (e.g., Viola et al., 2011). The 837 sphalerite-stannite mineral pairs arranged along these structures are supposed to bewere possibly concentrated through a pression-838 solution mechanism-during this deformational stage.

839 Skyttä and Torvela (2018) proposed that the BFZ300 is a brittle structure localized onto a zone of incomplete structural 840 transposition inherited from the earlier ductile history of the Olkiluoto basement. However, in our mesoscale and microstructural analysis we did not find evidence of any ductile precursor, and we note that BFZ300 cuts the ductile structural grain at high angle,

842 which excludes any reactivation of precursor ductile fabrics.

843 5.3. Implications for seismic deformation at the base of the BDTZ

- This study demonstrates the role of overpressured fluids on strain localisation during the incipient stages of fault nucleation and
- subsequent reactivation(s) at the BDTZ. The maximum estimated fluid pressure and fluid temeperature conditions derived in
- 846 this study (peak conditions of 210160 MPa and 350 °C) are indeed realistic for the base of the seismogenic zone in the continental
- 847 lithosphere (e.g., Scholz, 1990, and references therein) where the brittle-ductile transition for quartz occurs.
- 848 Mechanical models of long-term deformation (Rolandone and Jaupart, 2002) propose that deformation at the brittle-ductile
- 849 transition can be reasonably described as being mostly accommodated by intermittent and concomitant coseismic slip and ductile
- 850 flow. HMajor hydrofracturing, as that documented in this study by the Qtz I and II veins, is possibly related in that context to
- 851 seismic failure. Faults accommodating hydrofracturing are indeed commonly interpreted as seismogenic (e.g._Sibson, 1992a;
- 852 Cox, 1995) particularly at depth., where the reactivation of misoriented faults is only possible for fluid pressures exceeding σ_3
- 853 (e.g. Sibson, 1985).
- 854 Our study confirms this view because BFZ300 contains not only brittle fault rocks overprinting and overprinted by veins, but also
- 855 clearcut evidence of mutually overprinting brittle and ductile deformation (Fig. 6). In the light of the field observations discussed
- 856 and of the constraints derived, we suggest therefore that BFZ300 behaved in a seismic way at least during the emplacement of
- 857 the principal Qtz I and Qtz II veins. Hydrofracture veins are largely interpreted in the literature as the evidence of earthquake in
- 858 fluid-rich faults (Cox, 1995).
- 859 In this perspective, two possible scenarios can be considered to explain the genetic relationships between BFZ300 and a possible
- 860 seismic behaviour of the crust during the Svecofennian orogeny. In a first scenario, the quartz veins of the fault core would
- 861 represent the result of coseismic rupture during the mainshocks of a fully developed seismic cycle. Pore pressure fluctuations
- 862 caused the repeated transient embrittlement of the rock mass, which was otherwise under overall ductile conditions. The
- 863 documented brittle-ductile cycles are thus the expression of coseismic fracturing and aseismic creep between the individual
- shocks, as shown by viscous deformation overprinting the brittle features, guided by the residual differential stress.
- 865 A second possibility is that faulting occurred in the absence of a well-defined sequence of main- and aftershocks. As in the case
- 866 of man-induced earthquakes triggered by high-pressure fluids during injection of fluids (e.g. Healy et al., 1968), where
- 867 deformation is typically accommodated by diffuse swarms of low magnitude seismicity rather than well-defined mainshock-
- aftershock sequences (Cox, 2016), we propose that BFZ300 might have localised strain by diffuse veining with crack and seal
- 869 textures (Cox, 2016). Breccias and cataclasites (Fig.s.s 3, and 8) mutually overprinting with veins show that failure and veining
- 870 were indeed broadly coeval (e.g. Cox, 1995; Cox, 2016). Healing in fluid-rich environments can occur over short periods of time
- 871 (days-months) when compared with recurrence time of large earthquakes (10-100 years) (Olsen et al., 1998; Tenthorey and Cox,
- 872 2006). Therefore, the documented repeated switches between brittle and ductile deformations would then be steered again by
- 873 transient episodes of fluid overpressuring but in this case would express the accommodation of swarms of minor background
- 874 earthquakes within overall ductile conditions.

- Microstructures of fault-rocks exhumed from the brittle-ductile transition in other geological settings, are mostly in agreement with our hypotheses of seismic deformation. Transient and short term high-stress deformation followed by phases of stress
- 877 relaxation, which is prevalently characterized by recovery and recrystallization processes, has been documented by several
- 878 authors in deformed quartz (Trepmann and Stöckhert, 2003; Trepmann et al., 2007; Bestmann et al., 2012; Trepmann and
- 879 Stöckhert, 2013; Trepmann et al., 2017).
- 880 To conclude, BFZ300 represents an interesting case of likely seismic deformation within a fluid rich system at the base of the
- 881 seismogenic crust. The absence of later, thoroughgoing and high strain, potentially obliterating deformation episodes allows the
- 882 documentation of a complex structural evolution, from the earliest localisation to the mature structural stage.

6 Conclusions

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884 This workstudy Our analysis shows that a a multi-scale and disciplinary, multi-technique approach, based on the 885 combination leading to the generation of several independent constraints offers the potential to, gives an high degree of confidence 886 when used to r reconstruct in detail the evolutionary historyolution stages of also -a-fault zones that, which have experienced 887 multiple events of reactivation triggered by fluid overpressure and in which intense fluid-rock re-equilibration processes have 888 taken place. In accordance, Wwe documented the localised, initial embrittlement -stage-of the Olkiluoto-Paleoproterozoic 889 basement of southwestern Finland at the BDTZ, which occurred by brittle brittle failure under stilloverall ductile environmental 890 conditions eaused by in response to transiently elevated high fluid pressure and temperature (peak conditions: $P_f \ge 210 \text{ MPa}$; 891 T~350 °C). Latest events of reactivation and strain localization occurred by several brittle ductile deformation cycles, triggered 892 again by multiple pulses of high pressure fluids channelled into the system, study of faulting initiation and evolution has indeed 893 the potential to provide useful insights into the complex and cyclic processes of fluid fault interaction and effects thereof at the 894 base of the seismogenic crust. It Our results further constrains, moreover, the importance of cyclic seismicity and fluids in the 895 fragmentation of Precambrian cratons when deformed at the brittle ductile transition zone BDTZ, something that is not yet that 896 well understood for the Fennoscandian Shield. Our study, moreover, provides potentially important inputs to many modern 897 geological applications, including site characterization of deep geological disposal facilities for spent nuclear fuel. Results from 898 the detailed geological characterization of faults at the Olkiluoto site can thus be used toward the continuous updating of the 899 geological site description and yield further constraints on the mechanics of faulting at the BDTZthose conditions and at that 900 time.

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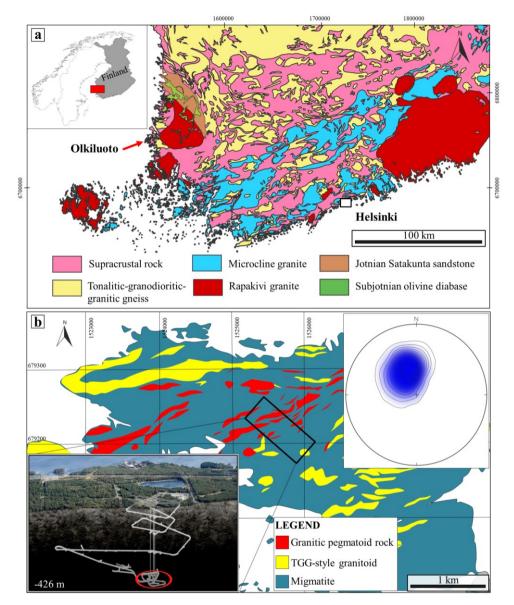


Figure 1. (a) Simplified geological map of southwestern Finland modified after Mattila and Viola (2014). (b) Geological sketch of the Olkiluoto Island. The upper right inset shows the poles to foliation planes measured from all available Olkiluoto drill cores (N = 4479, equal area, lower hemisphere projection; Mattila and Viola, 2014). The lower left inset is a panoramic photograph with an overlay drawing of the underground infrastructure (photo courtesy of Posiva Oy, Finland). The red circle shows the depth location of BFZ300. Coordinates are given in the local KKJ1 coordinate system.

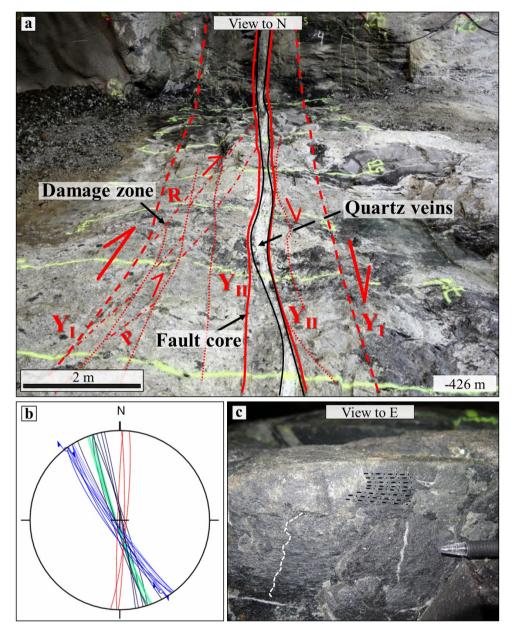


Figure 2. (a) View to the north and interpretation of the structural elements of the fault, whose core hosts BFZ300 two generations of quartz-ehlorite veins (thicker black lines). (b) Lower-hemisphere, equiangular projection of conjugate fault segments (blue great circles: lines are used for dextral faults; while blue onested great circles: indicate_sinistral faults), cleavage (green great circles) and quartzQtz I-chlorite veins infilling joints are presented with green and black coloured lines respectively(black great circles). (cb) SlickenfibersSlickensides (white dashed line) and slickenlines (black dashed lines) on a chlorite-decorated, -NW-SE striking fracture plane at the vein_host rock boundaryinterface_and the geometry of the R and P shears suggestindicating dextral strike-slip kinematics. Slickenfibers and slickenlines are observed at the vein host rock boundary. Sample location is reported in panel a with a white square.

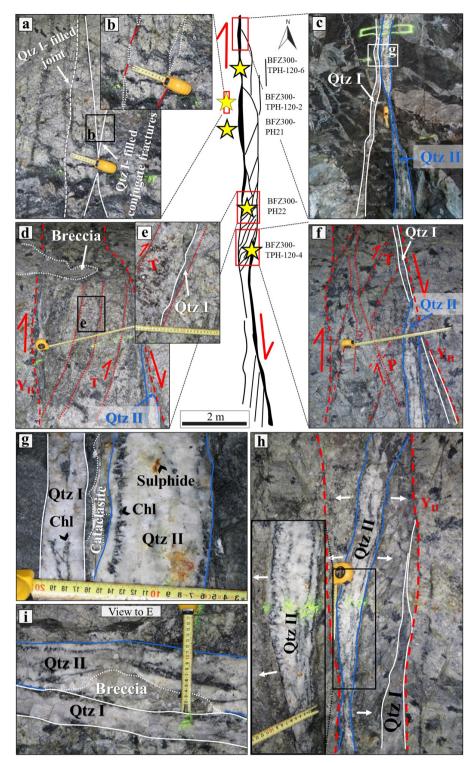


Figure 3. BFZ300-fault geometry and architecture (centre of figure) with examples of representative structural outcrop-features. The red rectangles locate the areas along the fault segments where detailed outcrop photos were taken. Stars locate hand and drill core samples. Stars with a black layout outline identify samples used for the microthermometric study. Note that the fault is made of two main segments offset laterally at a sinistral compressive step-over zone with overall dextral kinematics. Fault core quartz veins are shown by thicker black lines in the schematic model (centre of figure), while blue and white lines highlight the positions of the two types of quartz veins in the outcrop pictures. (a) Damage zone made of mm-thick, en-echelon veins connected by conjugate shear segments. (b) Detail of (a) showing fractures filled by the first quartz generation (Qtz I). (c) Two distinct generations of quartz-chlorite veins recognised in the fault core (Qtz I and Qtz II). (d) Detail of the sinistral compressional step-over zone characterized by multiple and parallel T fractures, filled by Qtz I. A brecciated body is crosscut by the Y planes. (e) TDetail of a tTensional fracture infilled by Qtz I. (f) Compressional structures (P shears) from the step-over zone and relationships between Qtz I and Qtz II-within the fault. The Riedel geometry suggests that the Qtz II vein formed due to the reactivation of the internal principal principal slip zones (Y_{II}). Note the Qtz II vein cutting the Qtz I vein. (g) Juxtaposed Qtz I and Qtz II veins. Qtz I veins are thinner and made of a translucid, small grained quartz. In contrast, Qtz II veins, which contain pockets of sulphide aggregates, are thicker and made of larger and euhedral quartz. Chlorite occurs as minor phase in both veinstypes of veins, but only in Qtz II veins it forms long and prismatic aggregates growing perpendicular to the fracture walls. In Qtz I veins, chlorite is small grained and forms thin levels within the quartz. Notice the presence of a cataclastic band between the two veins. (h) Spatial continuity of the chlorite aggregates within the Otz II veins, which grow always orthogonal to the vein boundaries. The ilnset shows the detail of the prismatic aggregates forming long and parallel ribbons. This open space filling texture suggests hybrid conditions of reactivation of the older Otz I veins. (i) Small quartz breccia formed between the two generations of quartz veins.

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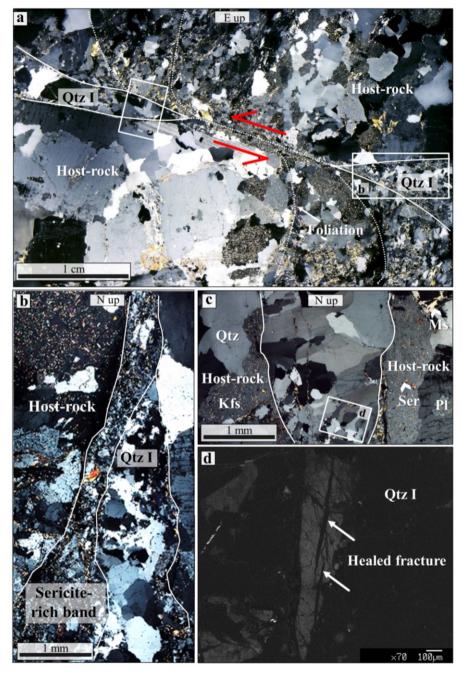


Figure 4. Microtextural characteristics of Qtz I from the damage zone of BFZ300 (sample: TPH-120-2). (a) Composition of Stitched photomicrographs of a Qtz I vein interconnecting with a sinistral shear band (crossed nicols). Faulting kinematics is suggested by drag folds in the host rock. (b) Tip of Qtz I vein hosted by a sericite-rich cataclastic band of the host rock. (c) Detail of panel a showing open-space filling texture in the Qtz I vein. Notice the sericite microfractures crosscutting Qtz I. (d) Panchromatic cathodoluminescence image of Qtz I showing healed microfractures crosscutting the crystal.

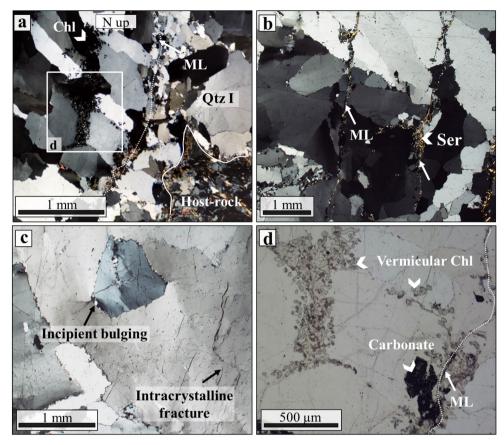


Figure 5. Microtextural characteristics of Qtz I from the damage zone of BFZ300 (samples PH21 and TPH-1202). (a) Stitched microphotographs of a Qtz I vein showing elongate-blocky texture with crystals growing obliquely with respect to the vein boundaries, which suggests growth under oblique dilatation. A series of median lines (ML) are marked by (b) sericite crystals suggesting repeated crack-and-seal. Quartz crystals show low temperature plasticity crystal-plastic deformation by undulose extinction and extinction bands. (c) Detail of plastic deformation in damage zone quartz veins: distorted crystals showing incipient bulging recrystallization and intracrystallinegranular fracturing. (d) Detail (plane polarized light) of the median linea ML and secondary fractures both decorated by vermicular chlorite and aggregates of REE-bearing carbonate.

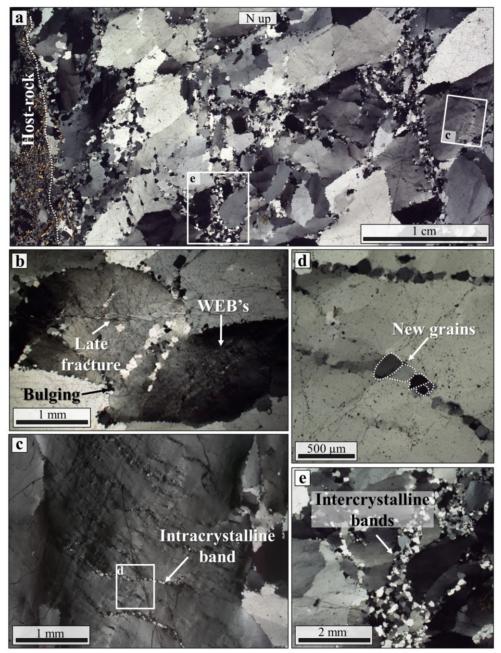


Figure 6. Microtextural characteristics of Qtz I from the BFZ300 core (sample TPH-120-4). (a) Stitched photomicrographs showing the typical heterogeneous grain size of Qtz I (30-800 μ m). (b) Evidence of plastic deformation of Qtz I from the fault core given by bulging of the largest crystals, wide extinction bands and undulose extinction. Note the late brittle fractures crosscutting all the previously formed plastic features. (c) Intracrystalline deformation bands within a large crystal. (d) Detail of (c) showing the typical grain size of the band (50-250 μ m). Intracrystalline deformation bands are oriented at <30° with respect to the BFZ300 vein walls and can be up to 2 mm in length. (e) Intercrystalline deformation band showing a thickening at the triple junction of larger grains. These intercrystalline bands are parallel to the strike of BFZ300.

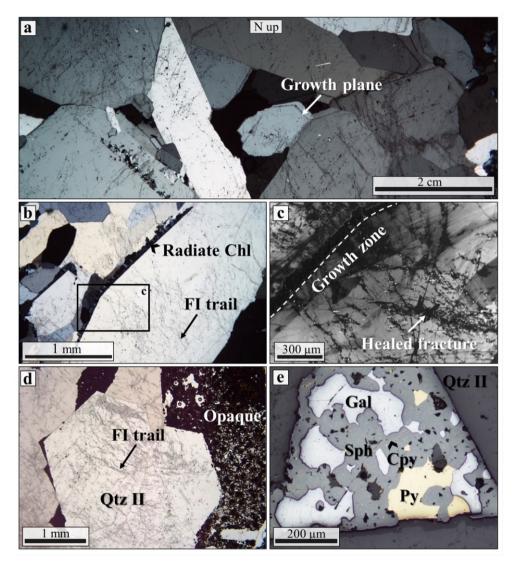


Figure 7. Microstructural characteristics of Qtz II from BFZ300 (samples TPH-120-6, PH22). (a) Stitched photomicrographs of Qtz II vein from the fault core. Notice the coarse quartz crystals and their elongated-blocky texture. Primary growth textures are sometimes visible and are marked by solid inclusions and decrepitated FIAs. (b) Radiate chlorite crystals along a prismatic Qtz II crystal boundary. Note that Qtz II is crosscut by numerous trails of FIs. (c) Panchromatic cathodoluminescence image of the same large Qtz II crystal from panel b, showing radiate chlorite along the crystal boundary and a primary growth zone cut by a set of healed fractures. (d) Euhedral quartz crystals set within opaque phases and crosscut by a network of thin microfractures. (e) Reflected light photomicrograph showing the opaque mineral assemblage typically associated with Qtz II, i.e. subhedral to anhedral sphalerite, pyrite, and galena. Chalcopyrite is a minor phase and occurs as small round inclusions within sphalerite (chalcopyrite "disease") or as large subhedral/anhedral masses together with galena.

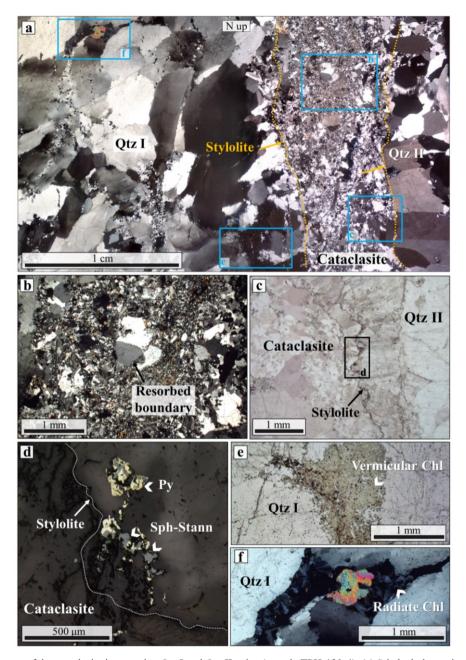


Figure 8. Microstructures of the cataclasite juxtaposing Qtz I and Qtz II veins (sample TPH-120-4). (a) Stitched photomicrographs covering the contact between the two quartz veins and the intervening 5 mm-thick cataclastic band. (b) Cataclastic band eonstituted bycontaining large Qtz I fragments (8-12 mm) embedded within a finer matrix (20-200 µm in size) of sericite and recrystallized quartz. The largest crystals show lobate boundaries, suggesting dissolution and local resorption along the clast-matrix interface. (c) Stylolite seams at the boundary of the cataclasite that strike parallel to the BFZ300. (d) Reflected-light photomicrograph showing anhedral to subhedral pyrite, chalcopyrite, stannite, and sphalerite arranged along the stylolite as residual products of pressure solution. (e) Vermicular and radiate (f) chlorite aggregates associated with Qtz I close to the cataclastic band.

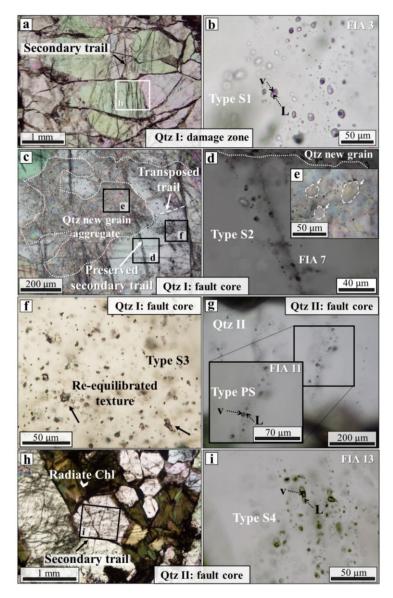


Figure 9. Characteristics textures of FIAs hosted within the BFZ300 quartz (samples PH21, TPH-120-4, TPH-120-6). (a) Secondary trails crosscutting large Qtz I crystals of the damage zone. (b) Detail of (a) showing the phase ratios of one of the studied secondary assemblages (FIA3), most representative of Type S1 FIA. (c) Long secondary transgranular trails crosscutting Qtz I of the fault core, dismembered by intercrystalline fractures, infilled by quartz new grains. Qtz I fault core also hosts set of short sub-trails developed at high angle with respect to the long trails. (d) Detail of Type S2 FIA entrapped along a preserved secondary fracture trail. (e) Small inclusions (<1µm) arranged along the boundaries of new polygonal quartz. (f) Example of Type S3 FIA arranged as isolated clusters inside ductile deformed fault core Qtz I. These trails formed during a brittle deformation stage that pre-dates ductile re-crystallization. (g) Pseudosecondary FIA associated with Qtz II-chlorite (FIA11). Enlarge—The enlargement shows the phase ratio details. (h) Small scale view of secondary FIAs crosscutting Qtz II. (i) Detail of secondary trails crosscutting euhedral Qtz II (FIA 13). In all photographs north points up.

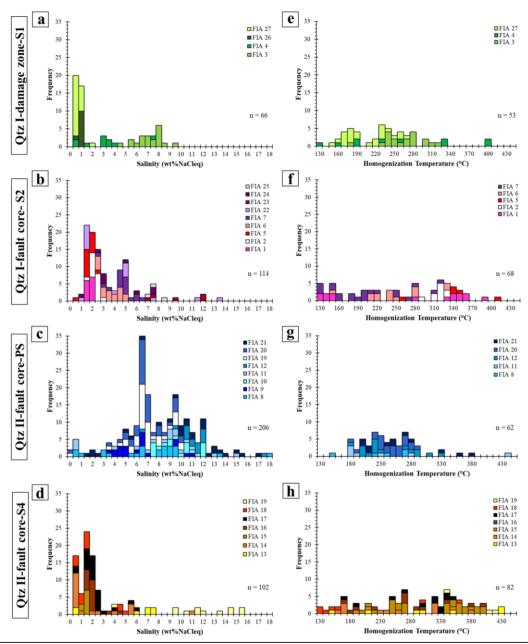


Figure 10. Microthermometric data of the studied FIAs. Panels a-d show the bulk salinities of individual FIAs calculated from the Tmice data, while panels e-h refer to the temperatures of final homogenization of the same assemblages. Notice that the data report the properties of individual FIAs according to their occurrence within Qtz I of the damage zone, Qtz I from the fault core, and Qtz II from the fault core. Notice that pseudosecondary (PS) and secondary (S) FIAs identify progressive later stages of fluid entrapment, and can be used to constrain the fluid properties in the fault zone. Notice also that the measured ranges of Thtot spread across T intervals that are too large to represent entrapment at equilibrium (e.g., FIA7 of Qtz I from fault core: 130-320 °C), which suggests post-entrapment re-equilibration of the inclusions. Fluid bulk composition is expressed as salinity, which is conventionally reported as weight percent of NaCl equivalents (wt%NaCleq, Roedder, 1984).

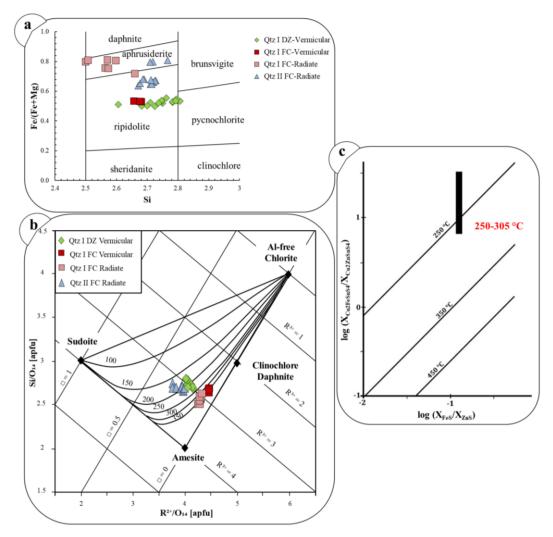


Figure 11. Chlorite chemical composition diagram and mMineral-pair geothermometry applied to the assemblages of the Qtz I- and Qtz II veins.

(a) Chlorite compositional diagram based on Hey (1954). The classification diagram shows a wide compositional range for chlorite across the BFZ300. Green, red, pink and light blue symbols indicate distinct chlorite textures in association towith Qtz I and Qtz II veins. (ba) Chlorite-quartz formation temperature estimated using the method of Bourdelle and Cathelineau (2015). Green, red, and light blue symbols indicate the distinct textural types of chlorite in Qtz I and II, respectively. The maximum temperature is from the Qtz I-chlorite pair from the fault core. The other groups of chlorites in the 150 250 °C range, indicate a second stage of quartz chlorite precipitation in the fault core and damage zone, in line with microthermometric constraints. (cb) Estimated tTemperature of formation of sphalerite-stannite in association with Qtz II vein (based on formation estimate We used thed with the method of Shimizu & Shikazono (1985), that uses Fe and Zn partitioning between stannite and sphalerite. The region of the plot that was calibrated with this geothermometer lies between the 250 and 450 °C isotherms. Hence, compositions corresponding to T<250 °C should be interpreted with caution.

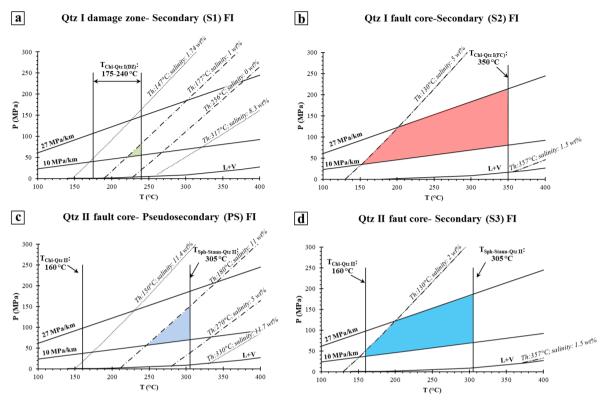


Figure 122. P-T diagrams showing the ranges of PT trapping conditions of the analysed fluid inclusions: P-T ranges have been estimated Estimated fluid pressure for the various typologies of FI petrographically discriminated petrographically and on the basis of identified in each structural domains. Fluid pressures are related to (a) secondary inclusions in Qtz I from the fault damage zone Qtz I, (b) secondary inclusions from Qtz I in the fault core Qtz I; (c-d) ppseudosecondary inclusions trapped in Qtz II in the fault core and (d) secondary inclusions in the Qtz II—Qtz II. Thin dashed lines indicate maximum and minimum isochores of FIAs in each structural domain. The light coloured areas are defined by the uppermost and lowermost sets of fluid inclusion isochores I, while the dark coloured areas identify the probable PT trapping ranges are defined by: i) the slope and position of the fluid inclusion isochores as determined by the most representative salinity and homogenization temperature range (Supplementary Material for details); ii) related to the pressure range calculated for isochores computed for the most probable composition of the pristine fluid (salinity between 0 and 5 wt%NaCleq, see text for more details). mThe pressure temperature areas are also defined by the mineral pairs geothermometry and iii) hydrostatic and lithostatic pore-fluid pressures computed assuming a regional geothermal gradient of c. 43 °C/km (assuming retrograde conditions of P c. 4 kbar and T c. 650 °C, from Kärki and Paulamäki, 2006). The IL (dotted vertical lines), and by the liquid-vapour equilibrium curves for the H₂O-NaCl modeled fluid is also indicated. The 240 °C vertical line represents the equilibrium temperature between chlorite and damage zone Qtz I. The 350 °C vertical line is the equilibrium temperature with Qtz II in the BFZ300 fault core. The thick lower curve marks the bundle of liquid vapour curves for a 1.5 wt% NaCl fault fluid.

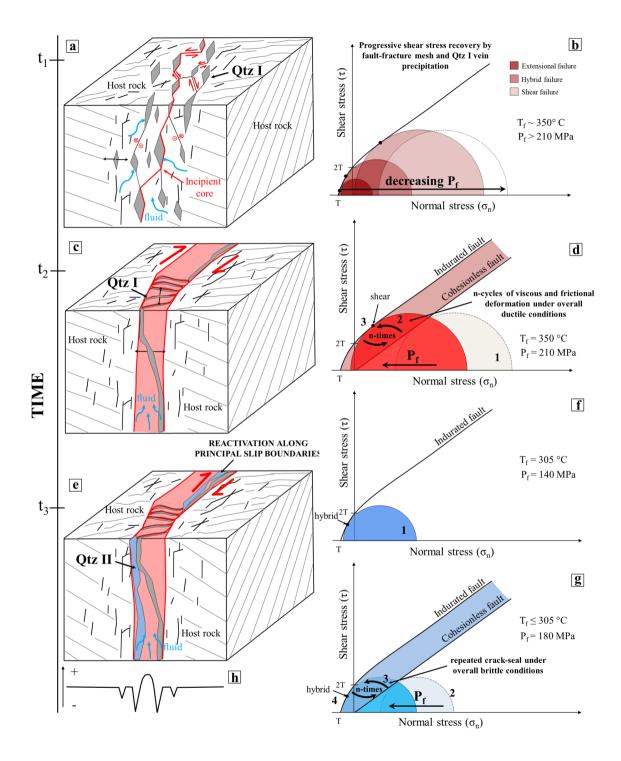


Figure 13. Conceptual model of the temporal and mechanical evolution of the BFZ300 fault zone (see text for more details). Grey lines: traces of metamorphic foliation. Black lines: fractures related to the BFZ300 structural development. (a) Initial embrittlement of the migmatitic basement occurred by fracture coalescence (red line) under (b) initial lower differential stress conditions and high fluid pressure and followed by a transient increase of differential stress. A first generation of quartz veins (Qtz I) precipitated inside the diffuse network of joints and hydrid/shear fractures which formed during this first deformation stage. (c) Progressive strain localization and fluid channeling within the fault core occurred by (d) episodically renewed fluid-pressure build-up driven by cycles of brittle and ductile deformation. (e-g) Progressive exhumation and cooling of the fault system occurred concomitant with several brittle reactivation episodes of the fault zone under hybrid conditions and fluid pressure lower than during the previous deformational stages. Lastly, a second generation of quartz veins (Qtz II) was emplaced, mainly along the principal slip boundaries of the fault core, following the Qtz I vein as shown by (h) the strength profile across the fault architecture, that suggests lower tensile strength values (and hence higher reactivation potential) along the Qtz I vein / host rock walls.

Table 1: Schematic summary of main microstructures, fluid properties, and PT deformation conditions in the quartz veins of the BFZ300 fault.

Structural zone and	Qtz type	Deformation type	Microstructures	Microthermometric properties	Fluid pressure (P_p) and mineral pair thermometry			
Sample Damage zone (PH-21)	Qtz I	Brittle/Ductile	Chl. Si FI MIL trail Migmatite	T_{mice} S1: -0.1 to -5.9 °C T_{htot} S1: 150-400 °C	$T_{\text{Chl-QtzI (DZ)}}$: 175-240 °C P_{f} (S1): 50-80 MPa			
Fault core (TPH120-4A)	Qtz I	Cyclic Brittle/Ductile	Intracrystalline healed fracture S2 FI trail	T _{mice} S2: -0.4 to -8.2 °C T _{htot} S2: 130-410 °C	$T_{Chl\text{-QtzI (FC)}}$: 350 °C P_f (S2) 30-210 MPa			
Fault core (TPH120-6) (TPH120-4)	Qtz II	Brittle	Growth plane S4 FI trail trail	T_{mice} PS: -0.1 to -13.6 °C T_{htot} PS: 150-440 °C T_{mice} S4: 0 to -11 °C T_{htot} S4: 130-430 °C	$T_{\text{Chl-QtzII}}$: 160-220 °C $T_{\text{Sph-Stann-Qtz II}}$: 250-305 °C $P_{f}(PS)$: 50-140 MPa $P_{f}(S4)$: 40-180 MPa			

Note: microstructures are coupled with the corresponding FI types and PT constraints derived from the collected dataset. See text for more explanations. Notice that we combine structural and geochemical data to constrain the relationships between stages of mineral-scale deformation and fluid circulation, which in turn defines the relative chronology of stages of fluid flow during faulting.

ML: median line; Blg: bulging.

1313 Table 2: Chlorite EPMA from various structural zones of BFZ300

Sample	4A	4A	4A	4A	4A	4A	PH21	PH21	PH21	2	2	2	6	6	6	6
Structural zon		FC	FC	FC	FC	FC	DZ	DZ	DZ	DΖ	DZ	DZ	FC	FC	FC	FC
Quartz type	Otz I	Otz I	Otz I	Otz I	Otz I	Otz I	Otz I	Otz I	Otz I	Otz I	Otz I	Otz I	Qtz II	Otz II	Otz II	Otz II
Textural type		Verm	Verm	Rad	Rad	Rad	Verm	Verm	Verm	Verm	Verm	Verm	Rad	Rad	Rad	Rad
Na ₂ O	0.04	0.07	0.00	0.08	0.08	0.03	0.05	0.02	0.04	0.03	0.01	0.05	0.04	0.06	0.01	0.01
TiO ₂	0.02	0.01	0.00	0.00	0.03	0.01	0.09	0.04	0.01	0.01	0.01	0.03	0.03	0.03	0.04	0.13
MnO	0.59	0.65	0.62	0.53	0.56	0.48	0.24	0.24	0.30	0.48	0.37	0.43	0.64	0.57	0.71	0.60
K_2O	0.06	0.02	0.04	0.07	0.06	0.04	0.01	0.01	0.03	0.10	0.05	0.07	0.03	0.02	0.05	0.01
MgO	13.66	13.79	13.74	6.61	5.13	6.75	13.95	14.06	13.29	12.85	12.57	12.59	4.85	4.87	8.73	8.05
SiO ₂	25.49	26.00	25.83	23.62	22.89	23.91	27.24	27.02	27.49	27.43	27.88	27.79	25.63	25.64	26.5	26.13
Cr_2O_3	0.00	0.01	0.04	0.00	0.00	0.06	0.04	0.03	0.06	0	0.06	0.01	0	0.02	0.01	0
FeO	27.86	27.74	27.87	36.61	38.49	36.75	24.68	25.21	26.07	25.97	26.06	25.77	34.26	33.84	30.08	30.47
CaO	0.03	0.04	0.05	0.00	0.06	0.03	0.01	0.02	0	0.05	0.05	0.03	0.01	0.04	0.04	0.02
Al_2O_3	22.04	22.13	22.00	22.89	23.35	22.98	24.13	24.75	24.91	24.02	23.48	23.21	24.23	24.64	24.49	25.02
Cl	0.00	0.00	0.01	0.03	0.02	0.04	0.01	0	0	0.01	0	0	0.03	0.02	0.02	0.01
Total	89.78	90.45	90.20	90.44	90.67	91.09	90.69	91.42	92.23	91.12	90.81	90.08	89.82	89.94	90.78	90.48
No. ions in																
formula																
Based on																
28 (O,OH)																
Na	0.02	0.03	0	0.03	0.03	0.01	0.02	0.01	0.01	0.01	0.01	0.02	0.02	0.03	0	0.01
Ti	0	0	0	0	0	0	0.01	0.01	0	0	0.01	0	0.01	0.0	0.01	0.02
Mn	0.10	0.11	0.11	0.10	0.10	0.09	0.04	0.04	0.05	0.08	0.06	0.07	0.12	0.10	0.12	0.11
K	0.01	0	0.01	0.02	0.02	0.01	0	0	0.01	0.02	0.01	0.02	0.01	0	0.01	0
Mg	4.25	4.25	4.25	2.14	1.68	2.17	4.18	4.19	3.93	3.86	3.79	3.82	1.55	1.55	2.69	2.49
Si	5.32	5.37	5.36	5.14	5.02	5.15	5.48	5.40	5.46	5.53	5.64	5.66	5.49	5.47	5.48	5.43
Cr	0	0	0.01	0	0	0.01	0.01	0	0.01	0	0.01	0	0	0	0	0
Fe ²⁺	4.86	4.79	4.83	6.66	7.06	6.62	4.15	4.21	4.33	4.38	4.40	4.39	6.14	6.04	5.20	5.29
Ca	0.01	0.01	0.01	0	0.01	0.01	0	0	0	0.01	0.01	0.01	0	0.01	0.01	0
Al	5.42	5.39	5.38	5.86	6.04	5.84	5.72	5.83	5.83	5.71	5.59	5.57	6.12	6.20	5.97	6.13
Cl	0	0	0	0.01	0.01	0.01	0	0	0	0	0	0	0.01	0.01	0.01	0
Fe	4.86	4.79	4.83	6.66	7.06	6.62	4.15	4.21	4.33	4.38	4.40	4.39	6.14	6.04	5.20	5.29
Al Tetr	2.68	2.63	2.64	2.86	2.98	2.85	2.52	2.60	2.54	2.47	2.37	2.34	2.51	2.53	2.52	2.57
Al Oct	2.73	2.76	2.73	3.00	3.06	2.99	3.20	3.22	3.29	3.24	3.23	3.23	3.61	3.67	3.45	3.56
Fe/(Fe+Mg)	0.53	0.53	0.53	<u>0.76</u>	0.81	0.75	0.50	0.50	0.52	0.53	0.54	0.53	0.80	0.79	0.66	0.68
Based on																
28 (O,OH) R ²⁺	9.11	9.04	9.08	8.80	8.74	8.79	8.33	8.40	8.26	8.24	8.19	8.21	7.69	7.59	7.90	7.79
Si	5.32	5.37	5.36	5.14	5.02	5.15	5.48	5.40	5.46	5.53	5.64	5.66	7.69 5.49	7.39 5.47	7.90 5.48	5.43
Based on	3.34	5.57	5.50	3.14	3.02	3.13	3.40	3.40	3.40	5.55	3.04	5.00	3.49	J.4/	3.40	5.45
14 (O,OH)																
R ²⁺	4.55	4.52	4.54	4.40	4.37	4.40	4.17	4.20	4.13	4.12	4.10	4.10	3.84	3.79	3.95	3.89
Si	4.55 2.66	2.68	2.68	2.57	2.51	2.58	2.74	2.70	2.73	2.77	2.82	2.83	2.75	3.79 2.74	3.95 2.74	2.71
31	2.00	2.08	2.08	2.31	2.31	2.38	2./4	2.70	2.13	2.11	2.82	2.83	2.13	2.74	2.74	2./1

Table 3: Representative EPMA of sulphides associated with Qtz II

	Structural	Qtz	-	-	-	-	=		-	-	<u> </u>		
Analysis	zone	type	Mineral	S	Fe	Cu	As	Pb	Ni	Zn	Ti	Sn	Total
TPH120-6-14	Core	II	pyrite	55.02	47.50	0.01	0.00	0.00	0.02	0.00	0.00		102.55
TPH120-6-17	Core	II	pyrite	54.08	47.19	0.00	0.01	0.00	0.00	0.00	0.00		101.28
TPH120-6-18	Core	II	sphalerite	34.46	6.46	0.09	0.01	0.00	0.03	59.62	0.02		100.69
TPH120-6-19	Core	II	sphalerite	34.48	6.24	0.08	0.06	0.00	0.04	59.61	0.02		100.53
TPH120-4A-34	Core	II	pyrite	54.49	47.40	0.05	0.00	0.00	0.00	0.00	0.00		101.94
TPH120-4A-35	Core	II	pyrite	54.13	47.26	0.02	0.04	0.00	0.00	0.01	0.55		102.01
TPH120-4A-38	Core	II	galena	13.40	0.00	0.00	0.00	86.63	0.00	0.32	0.01		100.36
TPH120-4A-59	Core	II	galena	13.50	0.06	0.00	0.01	87.04	0.00	0.10	0.01		100.72
TPH120-4A-40	Core*	II	sphalerite	35.06	9.46	0.05	0.00	0.00	0.00	56.74	0.01		101.32
TPH120-4A-43	Core*	II	sphalerite	34.69	9.04	0.01	0.03	0.00	0.00	57.51	0.01		101.28
TPH120-4A-41	Core	II	chalcopyrite	35.40	30.53	33.51	0.00	0.00	0.00	1.32	0.00		100.76
TPH120-4A-42	Core	II	chalcopyrite	35.78	30.78	33.59	0.03	0.00	0.01	1.22	0.01		101.42
TPH120-4A-19	Core **	II	stannite	29.79	12.53	28.41	0.07	0.08	0.00	0.92	0.000	27.86	99.66
TPH120-4A-22	Core **	II	sphalerite	33.82	8.15	0.06	0.00	0.03	0.02	57.27	0.006	0.00	99.36

Note: * - located within cataclastic band and close to stylolite. ** - located along stylolite
Sphalerite and stannite compositions from locations indicated by ** have been used to calculate the temperatures of sphalerite-stannite equilibrium following the
geothermometer of Shimizu and Shikazono (1985). See text for more explanations.