Extensional reactivation of the Penninic Frontal Thrust 3 Ma ago as evidenced by U-Pb dating on calcite in fault zone cataclasite. 2

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15 Abstract

16 In the Western Alps, the Penninic Frontal Thrust (PFT) is the main crustal-scale tectonic structure of 17 the belt. This thrust transported the high-pressure metamorphosed internal units over the unmetamorphosed European margin during the Oligocene (34-29 Ma). Following the propagation of the 18 19 compression toward the European foreland, the PFT was later reactivated as an extensional 20 detachment associated with the development of the High-Durance extensional fault system (HDFS). 21 This inversion of tectonic displacement along a major tectonic structure has been widely emphasized 22 as an example of extensional collapse of a thickened collisional orogen. However, the inception age of 23 the extensional inversion remains unconstrained. Here, for the first time, we provide chronological 24 constraints on the extensional motion of an exhumed zone of the PFT by applying U-Pb dating on 25 secondary calcites from a fault zone cataclasite. The calcite cement/veins of the cataclasite, formed 26 after the main fault slip event, at 3.6±0.4-3.4±0.6 Ma. Cross-cutting calcite veins featuring the last fault activity are dated at 2.6±0.3-2.3±0.3 Ma. δ^{13} C and δ^{18} O fluid signatures derived from these 27 28 secondary calcites suggest fluid percolation from deep-seated reservoir at the scale of the Western 29 Alps. Our data evidence that the PFT extensional reactivation initiated at least ~3.5 Ma ago with a 30 reactivation phase at ~2.5 Ma. This reactivation may result from the westward propagation of the 31 compressional deformation toward the External Alps, combined to the exhumation of External 32 Crystalline Massifs. In this context, the exhumation of the dated normal faults is linked to the eastward 33 translation of the HDFS seismogenic zone in agreement with the present day seismic activity.

34 **1. Introduction**

35 Dating of major tectonic inversions in orogens is generally achieved by indirect and relative dating, 36 but rarely by the direct dating of fault-related minerals using absolute geochronometers. For instance, 37 tectonic cycles are defined worldwide by the sediment unconformities or by exhumation ages through 38 thermochronological investigation. However, the recent progress in U-Pb dating of carbonate using 39 high-resolution Laser Ablation analyses (Roberts et al., 2020) allows us to directly date minerals 40 formed during fault activity and thus to establish the age of tectonic phases by absolute radiometric 41 dates (Ring and Gerdes, 2016; Goodfellow et al., 2017; Beaudoin et al., 2018;). This method is 42 especially well suited to disentangle the successive tectonic motions along a given tectonic structure. 43 U-Pb dating can be coupled to stable isotopic analysis to infer the nature of fluids through time, which 44 may give insights of the scale of fluid circulations and thus the scale of the active tectonic structure and changes in the stress regime (e.g., Beaudoin et al., 2015; Rossi and Rolland, 2014). In the Western 45 46 Alps, the Penninic Frontal Thrust or PFT represents a major thrust structure at lithospheric scale (e.g., 47 Tardy et al., 1990; Mugnier et al., 1993; Zhao et al., 2015) that accommodated the main collisional phase during the Paleogene-Neogene (e.g., Ceriani et al., 2001; 2004). Later on, this thrust was 48 reactivated as a normal fault, and the extensional deformation is still ongoing (Sue and Tricart, 1999; 49 Tricart et al., 2006; Sue et al., 2007). This transition from compression to extension in a collisional 50 51 chain has been diversely interpreted to reflect slab breakoff, crustal overcompensation or post-glacial 52 and erosion-induced isostatic rebound (e.g., Champagnac et al., 2007; Sternai et al., 2019). However, 53 until now, no direct dating of the tectonic shift from compression to extension on the PFT has been 54 obtained, which leads to many possible geodynamic scenari. At the present day, a large range of ages 55 for this transition has been hypothesized from ~12 to 5 Ma (Tricart et al., 2006), to only few ten's ka 56 (Larroque et al., 2009) which shows the lack of direct dating of brittle deformation (Bertrand and Sue, 57 2017). In this study, we applied the Laser Ablation U-Pb dating method on secondary calcites from a 58 cataclasite fault zone that testify of the extensional deformation of an exhumed paleo-normal fault 59 during the PFT inversion.

The purpose of this study is (1) to provide absolute chronological constraints on the structural
inversion of the PFT, and (2) give insights into the scale and nature of fluid circulations along this
major fault using stable isotope analysis of carbon and oxygen.

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64 **2. Geological setting**

The western Alpine collisional belt results from the convergence and collision of the European and
Apulian plates, which culminated with top-to-the west displacement on the PFT acting as the major
Alpine tectonic structure in the Late Eocene to Oligocene times (e.g., Dumont et al., 2012; Bellahsen
et al., 2014). This lithospheric-scale structure accommodated westward thrusting of highly

69 metamorphosed "Internal zone" units over slightly metamorphosed "External zone" units (Fig. 1, 70 Schmid and Kissling 2000; Lardeaux et al., 2006; Simon-Labric et al., 2009; Malusà et al., 2017). The 71 External zone is composed of the European non-metamorphosed Mesozoic and Paleozoic sedimentary 72 cover and its Paleozoic basement corresponding to the External Crystalline Massifs.



Fig. 1. Geological map of Western Alps showing the location of the study area. External Crystalline Massifs: Ar, Argentera; B, Belledonne; MB, Mont Blanc; P, Pelvoux. Internal Crystalline Massifs: DM, Dora-Maira; GP, Grand Paradis; MR, Mont Rose. PFT: Penninic Frontal Thrust. Insert modified from Schwartz et al. (2017).

78 The Internal zone corresponds to a high-pressure metamorphic wedge formed by the stacking of the 79 paleo-distal European margin of the Briançonnais zone, comprising the Internal Crystalline Massifs 80 and their sedimentary cover, with the oceanic-derived units of the Piedmont zone. These units were 81 incorporated and juxtaposed in the subduction accretionary prism since the Early Late Cretaceous until 82 the Late Eocene (e.g., Agard et al., 2002; Schwartz et al., 2007). The timing of subduction and 83 collision is well constrained by numerous dates on metamorphic minerals (e.g., Duchêne et al., 1997; 84 Rubatto and Hermann, 2001; Lanari et al., 2012, 2014). Eclogite facies recrystallization records 85 subduction of the distal European margin at 32.8 ± 1.2 Ma in the Dora Maira massif, which was later 86 transported as a tectonic nappe during the collision (Duchêne et al., 1997). PFT activation and 87 underthrusting of External Crystalline Massifs are indicators of the transition from subduction to continental collision in the Internal zones, between 44 and 36 Ma (e.g., Beltrando et al., 2009). This 88 89 transition is marked by shear zone development at greenschist facies conditions and recrystallization 90 during burial of the Alpine External zone in the PFT footwall compartment (Rossi et al., 2005; 91 Sanchez et al., 2011; Bellahsen et al., 2014). The early ductile PFT activity is dated at 34-29 Ma by

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92 ⁴⁰Ar/³⁹Ar dating of syn-kinematic phengite from shear zones in the Pelvoux and Mont Blanc External Crystalline Massifs (Seward and Mancktelow, 1994; Rolland et al., 2008; Simon-Labric et al., 2009; 93 94 Bellanger et al., 2014; Bertrand and Sue, 2017) and by U-Pb on allanite (Cenki-Tok et al., 2014). The 95 age of the PFT hanging wall tectonic motion and joint erosion is highlighted by the exhumation of the 96 Briançonnais units constrained by apatite fission tracks (AFT) at 26-24 Ma (Tricart et al., 1984, 2001, 97 2007; Ceriani and Schmid, 2004). However, the PFT reactivation as a normal fault remains 98 unconstrained. The onset of PFT extensional activity has been proposed to the Late Miocene (~12 to 5 99 Ma), based on indirect AFT ages in the Pelvoux External Crystalline Massif (Tricart et al., 2001, 100 2007), that record a cooling episode related to relief creation and erosion. The current seismicity (e.g., 101 Rothé, 1941; Sue et al., 1999, 2007) and observed GPS motions (Walpersdorf et al., 2018; Mathey et 102 al., 2020), all along the so-called High-Durance Fault System (HDFS) highlight the fact that extensional and minor strike-slip deformations along the PFT are still ongoing. This seismicity mostly 103 occurs at shallow depths, less than 10 km, and mainly at 3 to 8 km, where the HDFS is structurally 104 105 connected to the PFT (Sue and Tricart 2003, Thouvenot and Fréchet, 2006; Sue et al., 2007).



Fig. 2. Study area of the Penninic Frontal Thrust, east of the Pelvoux External Crystalline Massif (ECM). HighDurance Fault System is represented in red from Tricart et al. (2001) and Sue et al. (2007). Location of sampled
sites is indicated. The location of the extensional fault dated by U-Pb on calcite (samples FP18-2 and FP18-3) is
marked by a star. Colour of site circle refers to the host rock age: red, Eocene sandstone flysch (grès du
Champsaur); green, Cretaceous carbonates; blue, Jurassic carbonates; purple, Triassic carbonates. Sample
descriptions are shown on Suppl. Mat. 1. © Google Earth for background relief map.

112 The study area is focused on a portion of the PFT located in the southeast of the Pelvoux External 113 Crystalline Massif in the Western Alps (France) (Figs. 1-2). Here, the PFT rests on Late Eocene 114 (Priabonian) autochtonous nummulitic flysch so-called the "Champsaur sandstone" (Fig. 2), which lies 115 unconformably on the Pelvoux crystalline basement. In the southern part, the PFT lies on the 116 Cretaceous Helmintoid flysch nappes, Fig. 2. These two flysch units are intensely deformed by top-to-117 the-west PFT compressional deformation. The PFT hanging wall corresponds to the Brianconnais zone 118 composed of Mesozoic and Paleozoic sedimentary units, which underwent high pressure metamorphism (Lanari et al., 2012; 2014). The Briançonnais zone is composed of the Briançonnais 119 120 Zone Houillère, which consists of Carboniferous sediments overlying a crystalline basement, 121 stratigraphically overlain by Middle Triassic to Cretaceous sediments (limestones and calcschists). The 122 PFT structure is well shown in the Tête d'Oréac section of the Fournel Valley transect (Fig. 3, Sue and 123 Tricart, 1999). Here, normal faults cross-cut the Brianconnais series and branch down on the PFT, 124 which was reactivated as a detachment (Tricart et al., 2001).



Fig. 3. a: General view and geological interpretation of the Fournel Valley southern slope with the studied site
of the Tête d'Oréac. b: Outcrop interpretation of the Tête d'Oréac with extensional features in late Cretaceous
calcschists in agreement with the High-Durance Fault System and Wulff stereogram, lower hemisphere. c:
Calcschist oriented sample FP18-1 evidencing multiple calcite vein generations. V1 is related to the main
compressional phase related to the Tête d'Oréac anticline formation and V2 are related to extensional
reactivation of the PFT during onset of the High-Durance Fault System. Cs: Late Cretaceous calcschists;
Js+Tm: Middle Triassic to Late Jurassic dolomitic to siliceous limestones; Tl: Lower Triassic sandstones.

The normal faults are tilted by a passive rotation of about 30 degrees towards the west during their exhumation in relation with the activity of the High-Durance Fault System (Sue et al., 2007).

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135 **3. Sampling strategy and analytical methods**

136 *3.1. Sampling strategy*

We collected key samples of each brittle-ductile deformation phase, both in the PFT footwall and hanging wall (Suppl. Mat. 1), to provide a petrographic and stable isotopic dataset which will allow discussing the nature of fluids throughout the PFT activity associated to the late compressional and extensional history. Field analysis is supported by petrographic observations on 28 samples, including 8 host rocks, 6 from compressive structures and 14 from extensive structures. Based on this dataset, we selected three fault breccia samples to date the PFT extensional reactivation.

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144 *3.2. Cathodoluminescence*

145 Cathodoluminescence (CL) analysis provides shades that are mainly representative of oxidation state of trace element and their contents, i.e. Mn²⁺ and Fe²⁺ (Barnaby and Rimstidt, 1989). These differences 146 in calcite chemical composition are an indicator of different mineral precipitations related to slight 147 variations in fluid composition (Goodfellow et al., 2017). CL can also highlight crystal growth patterns 148 149 or grain boundary interactions (Beaudoin et al., 2015). Using cross-cutting criteria as well as CL, a 150 relative chronology of the calcite generations and related microstructures has been made. Analyses were performed with a spot camera mounted-Cathodyne device (cold cathode) with the following 151 152 parameters: vacuum ~50mTorr; voltage 16-18 kv; electron beam ~200 µA. Used description 153 terminology is based on Bons et al. (2012).

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155 3.3. O and C stable isotope analysis

156 Stable isotope measurements were achieved on the different generations of microstructures identified 157 by thin section observations and CL images, at Geosciences Paris Sud (GEOPS) laboratory of the Paris-Saclay University, France. Results are presented in Table 1. The protocol is described in detail by 158 159 Andrieu et al. (2015). Several milligrams (~1mm³) of sample for each calcite generation were collected using a Dremel 4000 with a 3.2 mm head. Samples were then dissolved with pure 160 orthophosphoric acid (H₃PO₄): Sample tubes provided with two compartments (one for the sample and 161 one for the acid) were sealed under a pressure of 1.5×10^{-2} mbar. They were immersed in a water bath at 162 25°C before the acid was poured on the sample and let to react for 24 h. Complete reaction is 163 necessary to avoid any artificial isotopic fractionation. The produced CO₂ is collected using an 164 extraction line and a liquid nitrogen trap is used to ensure that only CO₂ is collected. Pure CO₂ is 165 166 analyzed on a VG Sira 10 dual inlet IRMS (Isotope Ratio Mass Spectrometer). Data validity is 167 supported by concurrent analysis of the international standard IAEA CO-1. δ^{13} C and δ^{18} O are 168 expressed in ‰ relative to V-PDB (Vienna Pee Dee Belemnite) by assigning a δ^{13} C value of +1.95‰ 169 and a δ^{18} O value of -2.20‰ to NBS19, (1).

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$$\delta^{13}C = \left[\frac{\binom{1^3}{C}}{\binom{1^3}{C}}\right]_{Sample}}{\binom{1^3}{C}} - 1 \times 1000 \quad (1).$$

171 For oxygen isotope measurements, switch from PDB values to SMOW (Standard Mean Oceanic172 Water) were made using the Kim et al. (2015) equation, (2).

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$$\delta^{18}O_{SMOW} = 1.03086 \times \delta^{18}O_{PDB} + 30.86$$
 (2).

174 The ratio of carbon and oxygen isotopes is related to the parental fluid of calcite and can be used as a
175 fluid tracer. Reproducibility was checked by replicate analysis of in-house standards and was ±0.2‰
176 for oxygen isotopes and ±0.1‰ for carbon isotopes.

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178 *3.4. U-Pb dating of calcite*

179 In-situ uranium and lead isotope analyses of carbonates were carried out at CEREGE (Centre 180 Européen de Recherche et d'Enseignement des Géosciences de l'Environnement), Aix-en-Provence, 181 France. Results are presented in Suppl. Mat. 2. Data were acquired on 150 µm thick thin sections. 182 Laser ablation analysis was performed with an ESI excimer Laser Ablation system with a 6 inches two 183 volume cell (ESI), coupled to an Element XR SF-ICP-MS (Sector Field Inductively Coupled Mass Spectrometer, Thermo-Scientific). Analyses were done at 10 Hz and 1.1-1.15 J.cm⁻². Samples were 184 first screened to check signal intensities and maximise the spread of ²³⁸U/²⁰⁶Pb ratios (e.g. map of 185 186 Suppl. Mat. 3) to obtain the highest U-Pb variability. A typical analysis consists of 3 seconds of preablation to clean the sample surface, followed by 20 seconds of gas blank and ~20 seconds of 187 188 measurement on a static circle spot of 150 µm diameter (approximately 8-9 acquisition cycles per 189 second). These parameters lead to approximately $\sim 20-25 \ \mu m$ depth hole ($\sim 1 \ \mu m/s$) on a carbonate 190 material. Ablated particles are carried out of the cell with a He gas flux of 1300 ml/min and then 191 mixed with Ar sample gas (typically 0.8-0.9 l/min). Unknown samples were corrected by standard 192 bracketing with synthetic NIST-614 glass for instrumental drift and lead isotope composition 193 (Woodhead et al. 2001) and a natural calcite spar WC-1 of 254.4 ± 6.4 Ma (Roberts et al., 2020) for inter-elemental fractionation effect, every 20 measurements. No downhole correction was applied 194 195 since no natural calcite standard with homogeneous U-Pb ratio allows such correction. However, the 196 large aspect ratio used in this set up is supposed to limit this effect. Unknown sample were first 197 processed with the Iolite software (Paton et al., 2011) for baseline correction. Raw ratios were then reduced for instrumental drift, lead isotope composition and inter-elemental fractionation using an in-198 199 house excel spreadsheet macro designed for carbonate samples. Ages are obtained using IsoplotR 200 software and plotted in a Tera-Wasserburg diagram using model (1) age (Vermeesch, 2018). An

additional error propagation of 2.51% in quadratic addition on the final age, tied to the WC-1 standard,
is expressed in brackets in the Tera-Wasserburg plot.

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4. Results

- 205 *4.1. Deformation phases and miscrostructures*
- 206 4.1.1. Brittle-ductile deformation features

During the westward thrus motion, the Tête d'Oréac cross-section passes through the PFT (Fig. 3) and preserves a succession of units that were stacked on each other during to the west thrust motion of the PFT. The main schistosity (S1) is parallel to the initial bedding (S0) in Cretaceous calcschists. S0-S1 is sub-horizontal and penetrative throughout the studied area. At the outcrop scale, S1 is clearly visible and shows dissolution surface with the development of stylolithic joints (Fig. 4).



Fig. 4. a: General sketch of sample FP18-1 evidencing cross-cutting relationships for two main vein generations
 fig. 3.c. b-d: Microscope and cathodoluminescence pictures showing the different vein calcite generations.

214 Quartz anisotropy is observable in LPA which indicate an important deformation syn-post V1. This 215 correlate a strong transposition of structures during PFT compressive motion or opened initially near 216 parallel to S1 either way a ductile deformation is recorded. These early compressional features are 217 cross-cut by numerous steeply dipping eastward normal faults linked to the extensional reactivation of 218 PFT. Early stages of extension are featured by centimetre scale "en-echelon" veins (V2) indicative of 219 an early brittle-ductile extensional deformation followed by dissolution on the horizontal composite 220 (S0-S1) cleavage. Larger V2 veins, expressed at centimetre scale, cross-cut the cleavage and show 221 elongated calcite fibres of ~1000 μm at the vein walls (Fig. 4). Similar shades for early V2 and fibrous 222 V2 are observed in CL. At vein cores, the fibrous calcite is then replaced by a blocky calcite that is 223 less luminescent in CL.

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4.1.2. Brittle deformation features

226 The internal structure of one major extensional fault is investigated in the Tournoux scarp (Fig. 5). The 227 fault zone is highlighted by a metre-scale cataclasite fault gouge with variable amounts of 228 deformations. The top-to-the East (N90°E) normal sense of shear is represented by sigmoids and down-dip slickenside. At thin-section scale, for sample FP18-2, the cataclasite is composed of 229 centimetre-scale host rock clasts with very small (<20 µm) limestone grains. Two types of calcite 230 231 fillings have been identified. The first one contains organic matter has a « dusty appearance » with 232 bright shades in CL (Fig. 5C). The second one shows large and clear crystals that grew in the cracks 233 and porosity, showing sector zoning patterns highlighted in CL and Laser Ablation-Inductively 234 Coupled Plasma-Mass Spectrometry (LA-ICP-MS) maps (Fig. 5D; Suppl. Mat. 3). ~700 µm large 235 hexagonal, clear and organic matter free, calcite crystals have been selected for U-Pb dating.

These calcite crystals represent the latest pervasive fluid circulation episode through the porosity and provide a minimum age for the cataclasite. In sample FP18-3, the matrix is cross-cut by calcite veins with variable diameters (300-1300 µm) and is free of any further deformation. On the basis of their homogeneity and their youngest relative age relationships, these late calcites have also been targeted for U-Pb calcite dating (see section 4.3). Samples FP19-12A-B (described in supplementary data) were collected in a west-dipping conjugate normal fault and exhibits similar deformation features.

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Fig. 5. a-b: Outcrop interpretation of the Tournoux scarp showing various degrees of cataclasis in Triassic
dolomitic limestone with Wulff stereogram lower hemisphere. Squares are sampled area, sample FP18-2 is a
highly cataclased sample, while sample FP18-3 is less intensely cataclased and is cross-cut by millimeter-scale
calcite veins. c, d, e: Microscope and cathodoluminescence pictures showing several calcite filling generations.
« clear calcite » shows zonings and seems to crystallize into a primary porosity left within the cataclasite. The
clear calcite and veins from the cataclasite are dated using the U-Pb dating on calcite method.



250 4.2. $\delta^{13}C$ and $\delta^{18}O$ stable isotope results

Stable isotopes analyses were performed in calcites from various host rocks samples belonging to the
different units highlighted in the studied PFT section (Fig. 3) and are supposed to be representative of
the different (compressional and extensional) key tectonic phases (Fig. 6).

For host rock analysis, upper Cretaceous planktonic calcschists from the Tête d'Oréac show the lowest δ^{18} O host rock value of 16.8-17.1 ‰ and of δ^{13} C of 2.1-2.2 ‰. Triassic carbonates show a range between 23.7-26.5 ‰ for δ^{18} O and between 1.9-2.3 ‰ for δ^{13} C (with a higher value of 3.4 ‰ for the Ponteil scarp). Upper Jurassic calcshists gave δ^{18} O ratio of 28.5 ‰ and δ^{13} C of 1.3 ‰. The western Late Eocene Flysch (Champsaur sandstone) gave lowest δ^{13} C ratio of -0.3 ‰ and a δ^{18} O ratio of 21.9 ‰. Analysed brittle-ductile veins either related to the compressional or to the onset of the extensional tectonic phases stand very close to their host rocks, near to the meteoric water field defined by Nardini et al. (2019) (Fig. 6). However, the V2 veins associated to the brittle normal fault development, clearly show lower δ^{18} O values (<15‰) compared to their host rocks, with a trend towards lower δ^{13} C values. These isotope signatures are similar to those measured in calcite from veins of the Mont Blanc External Crystalline Massifs (Rossi and Rolland, 2014).



Fig. 6. Stable isotopic data from samples indicated on Fig. 2. Domains represented by dashed red, black and
blue lines are from the literature (Nardini et al., 2019; Rossi and Rolland, 2014 and references therein). The
coloured green domain corresponds to veins associated to brittle-ductile structures. These veins show similar
isotopic compositions as their host rocks. The orange domain features the signature of cataclased normal fault
samples, which show a different isotopic composition as compared to their host rock, and are similar to deep
metamorphic fluids (e.g., Crespo-Blanc et al., 1995; Rossi and Rolland, 2014; Rolland and Rossi, 2016).

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272 4.3. Calcite LA-ICPMS U-Pb dating results

273 Petrographic analysis has been complemented by screening using LA-ICP-MS on 24 thin-sections 274 from samples of 7 locations around the PFT related to compressive and extensive structures. Among 275 these, 20 screened samples show high common lead contents, and sometimes higher lead to uranium intensity signals. U-Pb dating of such carbonates with high lead concentrations remains highly 276 277 challenging, especially for very young samples. However, four samples (samples FP18-2, 3 and FP19-278 12A&B described in section 4.1 and supplementary data) from the Tournoux normal fault site bear sufficient ²³⁸U (~0-8.5 ppm for FP18-3A&B and ~0-4.5 ppm for FP18-2B and FP19-12A&B), and 279 ²⁰⁶Pb, ²⁰⁷Pb (~0-1.9 ppm for FP18-3A&B and ~0-13.1 ppm for FP18-2B and FP19-12A&B). 280



Fig. 7. Tera-Wasserburg concordia plot of (a) Highly cataclased sample FP18-2 calcite filling (b) and (c)
sample FP19-12A veins and FP19-12A 'clean calcite' filling (d) and (e) sample FP18-3 veins, and
corresponding maps of sampled spots (150 μm). MSWD: Mean Square Weighted Deviation. An additional error
propagation tied to WC-1 standard uncertainty is taken into account.

- Lead contents are based on NIST614 intensities and uranium contents are based on WC-1 intensities
 (Jochum et al., 2011; Roberts et al., 2017; Woodhead et al., 2001), giving measurable and significant
 radiogenic signal. Five ages have been obtained on these four samples (Fig. 7).
- A first group of ages of ~3.5 Ma is represented by three samples. The cataclasite 'clean calcite' infill (sample FP18-2B; Fig. 5) gives age of 3.42±1.44 Ma (n=41, MSWD=0.93). This quite large uncertainty is due to a relatively moderate U/Pb variability and the resulting low radiogenic signal measurable in this sample. Samples FP19-12A&B give two similar within-error ages for the vein calcite and 'clean calcite' infill, of 3.55±0.38 (n=58, MSWD=1.5) and 3.43±0.57 (n=53, MSWD=0.83), respectively.
- 295 A second group of ages of ~2.5 Ma is obtained on different cross-cutting veins of the latest generation 296 of sample FP-18-3 (Fig. 5), represented by slightly younger, but distinct out of error margins, ages of 2.59±0.23 Ma (n=42, MSWD=1.2; FP-18-3B in Fig. 7b) and 2.34±0.19 Ma (n=53, MSWD=1.1; FP-297 298 18-3B in Fig. 7c). The higher spread in U/Pb ratios measured in these two latter ages results in more 299 precise and robust ages. These two age groups obtained on extensional faults connected to the PFT 300 highlight for the first time at least two phases of deformation constrained out of error bars: a first 301 phase of brittle deformation forming the cataclasite at 3.5±0.4 and one or two discrete brittle events at, 302 or comprised within, 2.6±0.2 and 2.3±0.2 Ma. These ages show that the sated conjugated faults have 303 beed active for at least 1 Myr, and are featured by only several datable events, representing co-seismic 304 motions on the faults.

306 5. Discussion

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307 Onset of extensional tectonics in the Alps has remained a topic of debate for the last 20 years. A 308 Miocene age has been proposed for the onset of the extensional activation of the PFT based on AFT 309 datings on both sides of this major fault, i.e. in the Pelvoux External Crystalline Massif and in the 310 Champsaur sandstones to the west and in the Brianconnais zone to the east (Tricart et al., 2001; 2007; Beucher et al., 2012). The Brianconnais zone corresponds to the east hanging wall compartment of the 311 312 PFT. In this compartment, AFT ages ranging from 30 Ma to 20 Ma are interpreted as the exhumation 313 age of this area related to the compressional activity of the PFT during the Alpine collision, which 314 motion is constrained by direct ⁴⁰Ar/³⁹Ar dating on phengite at 35-25 Ma (Simon-Labric et al., 2009; 315 Bellanger et al., 2015). To the west (footwall of the PFT), the AFT ages range from 13 Ma to 4 Ma in 316 the Pelvoux External Crystalline Massif (Beucher et al., 2012), and from 9 to 4 Ma in the Champsaur 317 sandstones (Tricart et al., 2007), and are interpreted as the extensional reactivation of the PFT by these 318 latter authors. As the AFT dates record an exhumation age associated with cooling below ~100°C (Ault et al., 2019), they may not correspond to an age of PFT activity but rather record an erosion 319 320 process that is related to both climatic and tectonic processes (e.g., Champagnac et al., 2007). Sternai 321 et al. (2019) suggest that vertical movement in the Western Alps may be mainly ascribed to erosion 322 and deglaciation (Nocquet et al., 2016) and may also include a significant mantle convection 323 component (Salimbeni et al., 2018). However, the External Crystalline Massifs exhumation was also 324 driven by frontal thrusting, activated during middle Miocene at the western front of these massifs 325 (Boutoux et al., 2015) and by strong erosional processes that enhanced exhumation since the Late Miocene (Cederbom et al., 2004). Along the PFT, younger AFT and phengite ⁴⁰Ar/³⁹Ar ages of ~10 Ma 326 327 were obtained on the Plan de Phasy (Guillestre) metagranite mylonites (Tricart et al., 2007; Lanari et 328 al., 2014). These ages have been interpreted as the result of hydrothermal fluid circulation, which may 329 be linked to tectonic activity of the High-Durance Fault System. However these fluid circulations may 330 be passive through the PFT network and may not correspond to extension onset. Therefore, the age of 331 PFT activity remains unconstrained and requires some direct datings. In the following discussion, we 332 show how absolute U-Pb dating of fracture infill calcite brings quantitative time constraints on PFT 333 fault movement.

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335 5.1. Deformation and scale of fluid flow in the brittle-ductile structures

The measured δ^{18} O and δ^{13} C isotope ratios of veins from brittle-ductile structures are close or similar 336 to their host rocks, and remain close to the field of carbonates precipitated from meteoric water 337 338 (section 4.2). Based on several studies in the frontal parts of Alpine orogens (Smeraglia et al., 2020; 339 Nardini et al., 2019), these isotope signatures are thought to be representative of meteoric water inflow from the most superficial domains. Three important parameters are involved to control this surface-340 341 derived fluid regime: (i) lack of large-scale structures (ii) pressure-solution microstructures (evidence 342 of local fluid) (iii) presence of a shallow impermeable clay-rich layers which isolate upper crust from 343 more deeply-rooted systems (section 4.1. and Fig. 3). Rossi and Rolland (2014) report similar stable 344 isotope signatures in the Mont Blanc External Crystalline Massif sedimentary cover (Helvetic schists). 345 There, the vein calcites bear similar stable isotope values as the host Helvetic schists, which is in 346 agreement with the fluids to have equilibrated with their host rocks in a closed system with low 347 fluid/rock ratios (Rolland and Rossi, 2016). In our study, observations of veins show that they were closely related to schistosity acting as a stylolithic dissolution surface (section 4.1). This observation is 348 349 consistent with local fluid interactions and equilibrium with the host rock, resulting from a pressure-350 dissolution-recrystallization transfer mode (e.g. Passchier and Throw, 2005). Based on this, we suggest 351 that the external fluid signature was buffered by the host rock signature. These fluid compositions show that 'en-echelon' veins are linked to an early deformation, where the porosity was still not 352 connected by the fault network (Fig. 3). In such a system, the veins kept the host rock signature and no 353 354 crustal-scale fluid flow circulation is evidenced.

355

356 5.2. Scale of fluid flow in the brittle extensional structures

- 357 Major (> metre-scale width) faults are related to shallower, or higher stress contexts (e.g. Passchier 358 and Throw, 2005). The isotopic composition of calcite that crystallised in these brittle extensional 359 faults is significantly different from their host rock (section 4.2; Fig. 6). Indeed, calcites related to these major faults have δ^{18} O lower than 10 ‰ from their host rock and a δ^{13} C ranging between -5 to 4 360 % PDB (while the δ^{13} C ratio of Trias host rock is of 2 %). This signature is similar to that of 361 362 exogenous metamorphic fluid origin (Crespo-Blanc et al., 1995; Rossi and Rolland, 2014). The observed CL pattern of calcites also argues for variations in the fluid composition, between the 363 different veins and progressively within a given vein. Similar signatures are recorded in the Mont 364 365 Blanc External Crystalline Massif shear zones and veins in a similar structural context (Rossi et al., 2014). There a similar spread of δ^{13} C- δ^{18} O values is observed in the marginal part of the crystalline 366 367 basement, at the contact with the Helvetic schists. This spread is interpreted as a mixing between 368 fluids flowing down through the sedimentary cover and upwards fluids originating from shear zones in 369 the Massif Central (Rolland and Rossi, 2016). The chemical signature of calcite veins in the Massif 370 Central shear zones is correlated to a Mg-K-rich metasomatism, both arguing for CO₂-bearing fluids 371 representative of a deep source, which is rooted in the mantle via vertical shear zones (Rossi et al., 372 2005). This deeply rooted fluid cell is also suggested by fluids significantly hotter (150-250 °C) than their host-rock at ca. 10 Ma along vertical faults in Belledonne Massif, which are in continuity with 373 374 the central Mont Blanc Massif shear zones (Janots et al., 2019). Indeed, deep metamorphic fluid 375 circulation is in good agreement with a crustal-scale fluid pathway which is activated during the 376 extensional motion of the PFT, connected to the Rhône-Simplon right-lateral fault (Bergemann et al., 377 2019; 2020). This crustal-scale network suggests that extensional faults are in-depth connected to the 378 PFT, when it was reactivated as a detachment. Deep connection with the PFT crustal scale structure (e.g. Sue et al., 2003) would allow fluid circulation from interface of European slab with the deep 379 380 subduction/collisional metamorphosed prism. In our study, the isotopic dataset shows a significant 381 difference between the deep fluids signature recorded by the Mont Blanc veins (Rossi and Rolland, 382 2014; Rolland and Rossi, 2016) and the compositions of the veins related to brittle-ductile structures 383 (Fig. 6). This variability suggests a mixing process between the local fluids trapped in the early 384 extensional (closed system) and these exogenous fluids from a deep crustal origin.
- 385

386 5.3. Timing of PFT extensional inversion

All ages obtained from the investigated Tournoux normal fault scarp, give direct time constraints on the final stages of extensional slip, and are interpreted as a minimum age for the extensional reactivation of the PFT. The oldest event is the formation of the highly deformed cataclasite calcite filling/veins ~3.5 Ma (3.4±1.5 Ma, 3.6±0.4 and 3.4±0.6). This calcitic cementation occurred directly 391 after the main cataclastic deformation event and before the late cross-cutting veins. Latter cross-392 cutting veins gave the same \sim 2.5 Ma age, with two within-error dates of 2.6±0.3 and 2.3±0.3 Ma. The 393 ~3.5 Ma and ~2.5 Ma do not represent the same slip event on the fault. It is noteworthy that all these 394 ages are calculated assuming secular equilibrium in the U-series decay chain. As fluids are generally characterized by an excess in ²³⁴U with respect to ²³⁸U, resulting in an excess of radiogenic ²⁰⁶Pb, the 395 calculated ages should be considered as maximum ages (see for example Walker et al., 2006). The 396 magnitude of the offset ages due to initial ²³⁴U/²³⁸U disequilibrium can be significant and the true age 397 398 could be younger by several hundreds of thousands of years. In the present case, it was not possible to 399 carry out classical isotopic analyses of uranium by isotopic dilution to measure any detectable residual 400 ²³⁴U/²³⁸U disequilibrium because of the size of the carbonate phases. It could be hazardous to speculate on the initial ²³⁴U/²³⁸U disequilibria of the fluids, but the quite high uranium concentrations (up to the 401 ppm level) observed in analysed minerals of samples FP-18-2 & 3 (Fig. 7) are likely indicative of an 402 oxidizing environment and thus of a moderate initial ²³⁴U excess (Walker et al., 2006). To assess the 403 impact of this excess on the final age, we have tested various initial ²³⁴U/²³⁸U activity ratios ranging 404 between 1 to 2 as illustrated in Figure 8. For an initial (²³⁴U/²³⁸U) activity ratio of 2, the true age is 405 lower by about ~370 ka. The obtained ages assuming an initial (²³⁴U/²³⁸U) ratio of 1 are thus regarded 406 407 as maximum ages.

As they remain undeformed, the latter veins are considered as the youngest tectonic slip along the fault. Furthermore, the geometry of the Tournoux normal fault reguarding the PFT position indicates that this normal fault was connected to the PFT, which acted as a detachment Zone (Fig. 9). Thus, it may represent the paleo-HDFS seismogenic zone, which was later exhumed in the footwall part of the active extensional fault. Main activity of this paleo-fault can be bracketed between 3.4-2.2 Ma based on the above results.



Fig. 8. Impact of the initial ²³⁴U excess on the final age estimation. Several initial ²³⁴U/²³⁸U activity
ratios have been tested ranging between 1 to 2. This spread in initial ²³⁴U/²³⁸U leads to an age
difference of 0.37 Ma. The obtained U/Pb age of 2.59 Ma, assuming equality of ²³⁴U and ²³⁸U contents
is thus a maximum age.

419 5.4. Evolution of PFT through time

420 The structural and dating results presented in this paper, combined with the literature on PFT footwall421 and hanging wall exhumation lead to the following reconstitution of its evolution (Fig. 9).

422 The investigated PFT paleoseismic zone is located 3 to 10 km west of the active HDFS seismogenic 423 zone. Nowadays, the extensional deformation is mainly localised on one active fault and mostly occurs 424 mostly at 3 to 8 km depth (Sue et al., 2007; Mathey et al., 2020). This study gives insights into the 425 uplift rate and lateral displacement of the High-Durance Fault System footwall and hanging wall since 426 the passage of the investigated paleo-PFT through the upper boundary of the seismogenic crust some 427 2-3.5 Ma ago. Since then, the PFT hanging wall, represented by the active extensional deformation 428 front of the HDFS was significantly shifted eastward, while its footwall was uplifted up to 3 km (Fig. 429 9). This leads to a mean vertical tectonic motion on the order of $> 1 \text{ mm.yr}^{-1}$ for the footwall 430 compartment of PFT on this period of time. This rate is consistent with the vertical GPS rates 431 measured for the Pelvoux External Crystalline Massif (Nocquet et al., 2016; Sternai et al., 2019).



Fig. 9. Evolutionary geological cross-section sketch of PFT (modified from Tricart et al., 2006). a,
Compressional activation of the PFT resulting in joint External Crystalline Massifs burial and Briançonnais
exhumation during the Oligocene. b, Extensional reactivation of the PFT and setting up of the High-Durance
Fault System during the Pliocene as evidenced in this study. At this point the dated extensional fault passes
through the upper boundary of the seismic zone at ca. 2-3 Ma. c, At present-day, compressional deformation has
migrated westward (frontal part of External Crystalline Massifs, since c. 15 Ma) and extensional seismic activity
of the High-Durance Fault System is recorded at shallow depth 3-10 km east of the studied paleoseismic zone.

440Our data support the hypothesis that the present HDFS is the result of eastward shifting of extensional441deformation, accommodated by successive jumps on several faults. Faults were likely active on a scale442of ≥ 1 Myr before becoming inactive. Calcite U-Pb ages obtained on the Tournoux scarp constrain co-443seismic motion on two conjugate faults. The two age groups obtained on these extensional faults444connected to the PFT highlight at least two phases of deformation, at 3.5 ± 0.4 Ma and one or two

- discrete brittle events at, or comprised within, 2.6±0.2 and 2.3±0.2 Ma, which gives insights into the long-term activity of the of at least 1 Myr, but with only several datable events, which argues for an apparent contradiction. Indeed, co-seismic displacement on the fault suggest a significant magnitude for the related earthquake (Wells and Coppersmith, 1994), which is apparently incompatible with the very few datable motions. This gives some weight to a deformation regime which may alternate phase of creeping on the fault plane, without any brittle deformation, with very rare phases of brittle deformation.
- The vertical uplift and exhumation of the Pelvoux External Massif since 3.5 Ma may thus mainly result from the cumulated fault motion on these several fault segments. These data are thus in agreement with a significant tectonic component in the measured uplift signal of External Crystalline Massifs, which is in agreement with a clear difference in uplift rates measured between ECMs and Internal Alps (Nocquet et al., 2016; Sternai et al., 2019).
- We support the hypothesis that the HDFS is the result of the eastward extensional deformation shiftand the successive formation of younger faults normal fault participated at the exhumation of thedirectly to the west block.
- 460

461 **6.** Conclusion

462 Significant constraints on the evolution of fault systems can be acquired by coupling stable isotopic 463 analysis and U-Pb dating on calcite. These methods have been successfully applied to unravel the tectonic reactivation of the PFT for the first time. Five U-Pb ages on calcite have been obtained on 464 465 extensional fault structures connected to the PFT, gave two distinct group of ages of 3.5±0.5 Ma for 466 the main deformation phase represented by the cataclasite calcite cement, cross-cut by later discrete 467 phases represented by mm-large veins dated from 2.6±0.3 to 2.3±0.3 Ma. The 3.5 Ma age represents a 468 minimum age for the onset of extensional brittle reactivation of the PFT. Earliest extensional ductile-469 brittle structures cannot be dated due to low uranium contents and low U/Pb ratios. Associated to those 470 two (ductile and brittle) deformation stages, stable isotopic ratios of carbon (δ^{13} C) and oxygen (δ^{18} O) 471 of calcite samples collected within the kilometre-scale extensional faults show an evolution from a 472 closed to an open fluid system. The isotopic signature of fluids related to the brittle deformation stage 473 corresponds an open system due to the activation of a crustal-scale fluid circulation cell when the 474 HDFS developed in connection with the deeper PFT deeper structure. The fluids associated to this 475 open system show a deep crustal/mantle signature similar to that measured along the PFT across the 476 Alpine arc. This deeply rooted upward fluid circulation occurred when extensional fault activity was 477 connected to the PFT reactivated as a detachment, which suggests a crustal-scale extensional 478 reactivation at this stage. These constraints on PFT fluid regime are the first direct evidence for a 479 transition towards a crustal-scale fluid regime at the onset of brittle extensional reactivation in the

- Alps. The direct ages of PFT motion give insights into the long-term incremental displacement of the
 HDFS footwall, and Pelvoux Massif exhumation, which corresponds to its passage through the upper
 part of the seismogenic zone, at a mean rate of > 1 mm.yr⁻¹ in the last 3 Ma.
- 483

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490 Author contributions

AB, YR and SS wrote the manuscript and all authors discussed the results and contributed to the final
article. TD supported AB for map creation and cross-sections. YR, SS, TD, CG and AB participated to
field trip sampling. AB did the sample petrographic characterization with optical microscope and
cathodoluminescence. NG, AG and PD led U-Pb dating with AB. BB and AN supervised AB for stable
isotopes analysis, for results interpretation and protocol application respectively.

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497 Supplementary Materials

- 498 Suppl. Mat. Table S1. Sample locations and descriptions.
- 499 Suppl. Mat. Fig. S1. Tournoux's scarp general view.
- 500 Suppl. Mat. Fig. S2. Field photographies FP19-12 site.
- 501 Suppl. Mat. Fig. S3. La-ICPMS elemental maps, FP18-2B.
- 502 Suppl. Mat. Fig. S4. FP19-12B thin section with map localisation.
- 503 Suppl. Mat. Fig. S5. La-ICPMS elemental map, FP19-12B.
- 504
- 505 Suppl. Mat. Table S2. U-Pb on calcite La-ICPMS data.
- 506

	N°Sample	¹³ δC PDB	¹⁸ δΟ SMOW
Whole Rock	FP18-1A	2,15	17,18
	FP18-1A	2,09	16,90
	FP18-4	1,92	24,83
	FP18-7	2,25	25,76
	FP18-9	-0,32	22,00
	FP18-10	1,26	28,59
	FP18-11	2,1	26,51
	FP18-13	3,43	23,73
Early veins (V1)	FP18-1B	2,59	19,89
- · · ·	FP18-1C	2,48	20,74
	FP18-1C	2,48	18,84
	FP18-9	-0,18	22,59
	FP18-10	1,29	28,99
	FP18-11	1,58	23,37
En-echelon veins (V2)	FP18-1A	2,12	17,65
	FP18-1A	2,18	18,15
	FP18-1B	1,96	17,71
	FP18-1D	1,88	17,98
	FP18-5	1,66	25,80
Cataclasite infill (V2)	FP18-2A	0,03	7,43
	FP18-2B	-0,09	7,86
	FP18-3B	-4,62	11,41
	FP18-3B	-0,75	12,10
	FP18-6	-3,04	10,23
	FP18-6	-0,95	12,68
	FP18-13	3,46	13,29
	FP18-13	2,53	14,38
	FP18-13	3,4	7,21

- 507 Table 1. Stable isotope data, first bloc for host rock, second bloc compressive veins, third early small scale
- *extensional features, fourth main large scale extensional features.*

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