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# 1 The Tertiary structuration of the Western Subalpine foreland

# 2 deciphered by calcite-filled faults and veins

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- 17

### 18 Abstract:

19 The age of brittle deformation in the superficial part of orogens is generally constrained by relative, 20 cross-cutting structural relationships. However, it becomes possible to decipher the timing of fault 21 activity by a combination of methods based on U-Pb dating and stable isotope composition of 22 calcite-filled fault and veins. This methodology is applied to constrain the timing of deformation of 23 the frontal part of an orogenic system, through the example of the Tertiary development of 24 subalpine massifs fold and thrust belt (composed by Bauges, Chartreuse, Vercors massifs). The 25 architecture of the massifs is well constrained, but the chronology and the nature of involved fluids 26 is unknown. Clumped isotope  $\Delta_{47}$  analysis shows a significant variation of fluid temperature from 27 54°C to the west up to 149°C for the internal Vercors thrust to the east. These temperature estimates 28 highlight a deep underthrusting with a significant exhumation of the eastern Vercors (4-6 km), 29 while a shallow underthrusting at a depth of around 2 km is estimated for the central Vercors. 30 Carbon and oxygen stable isotope analyses, reveal three fluid signatures corresponding to heated 31 fluids from meteoric or basin-derived origin, which interacted at various extents with the host-rock. 32 These data are in agreement with a low altitude frontal belt, and with a progressive reequilibration 33 of downwards infiltrating fluids with the composition of the host-rocks. The successfully dated 34 calcites are those exhibiting the highest fluid-rock ratios, which allowed U enrichment in a mainly 35 uranium-poor rock environment. In-situ U-Pb calcite dating was performed on fault mirrors of main 36 thrusts and on fractured pebbles of the underthrusted molasses of eight major thrusts. Preserved 37 Oligocene ages of extensional veins related to pre-Alpine rifting demonstrate that U-Pb calcite is a 38 robust method to characterize multiple superimposed events of a whole mountain building history. 39 Furthermore, the presence of multiple deformation events in the same location point out the 40 importance of inherited structures on the strain localization processes. Thrust related U-Pb on 41 calcite ages on an E-W section between the Vercors and Chartreuse massifs record a major Miocene 42 shortening phase, and the timing of strain propagation towards the foreland. The oldest thrust dated 43 at 15 Ma, corresponds to the activation of the most internal preserved thrust mobilizing 149°C 44 fluids. In the central subalpine massifs, ages range from 14 Ma (Chartreuse) to 12 Ma (Vercors) 45 mobilizing 54°C fluids. On the western part, the initiation of the most external thrust has been dated 46 between 10 Ma and 7 Ma. These geochronological constraints are consistent with an 'in-sequence' 47 westward propagation of the deformation between 15 Ma and 7 Ma and contemporaneous with the 48 exhumation of the Belledonne External Crystalline Massif and the development of flexural basin 49 sedimentation. Paleostress calculations indicate a three-stage evolution of the subalpine nappe stack. Reverse and strike-slip deformations are interpreted as a diffuse tectonic reactivation in an
'hors-sequence' mode from 10 Ma to present in a broadly similar stress field.

52

### 53 1. Introduction

54 Absolute dating of brittle deformation has become possible thanks to recent developments of the U-55 Pb dating method on calcite (e.g., Roberts et al., 2020). This method has been applied to the 56 reconstruction of superficial mountain belts tectonic history (e.g., Smeraglia et al., 2019; Carminati 57 et al., 2020; Cruset et al., 2020; Curzi et al., 2021; Looser et al., 2021; Parizot et al., 2022). 58 However, its applicability may still suffer from a lack of understanding of calcite recrystallization 59 processes in the case of multiple stages of deformation and complex fluid history (Roberts et al., 60 2021). Several problems have hindered the development of this method, especially detection limits 61 of U and Pb in calcite, which remain a major obstacle (Lanzirotti and Hanson, 1995; Chew et al., 62 2014). For this reason, this method was initially limited in the 1990's to calcites with high uranium 63 contents (1-9 ppm), (e.g., Rasbury et al., 1997, 1998, 2000). A major issue has been to disentangle 64 crystallization histories within a heterogeneous mineral at the micron scale, as the method was 65 formerly undertaken by acid digestion and isotopic dilution (e.g., Smith and Farquhar, 1989). With 66 the development of in-situ Laser-Ablation ICP-MS techniques (Li et al., 2014), allowing micron 67 scale elemental mapping (Drost et al., 2018), it became possible to overcome this problem. 68 Nowadays, the low detection limit of U and Pb (~0.1ppm) combined to a high spatial resolution 69 (50-150µm spot diameter) make LA-ICP-MS dating on calcite a widely and useful method from 70 various contexts (e.g., Roberts et al., 2021), in particular for the development and chronology of the 71 fault network (Roberts and Walker, 2016; Ring and Gerdes, 2016; Beaudoin et al., 2018; Hansman 72 et al., 2018; Pagel et al., 2018; Parrish et al., 2018; Hoareau et al., 2021; Parizot et al., 2021; 73 Roberts et al., 2021; Smeraglia et al., 2021; Roberts and Holdsworth, 2022). Direct calcite dating 74 applied to main faults along transects allows to analyse the along-belt variations of deformation 75 ages, like the propagation of fold and thrust belts (Looser et al., 2021) and the polyphased evolution 76 of major faults (Bilau et al., 2021; Parizot et al., 2022). The use of methods performed on calcite 77 can brings up information on the physical and chemical parameters associated to the fluid 78 circulation during the deformation responsible for the calcite crystallization (Goodfellow et al., 79 2017; Bilau et al., 2021; Hoareau et al., 2021; Looser et al., 2021) but can also be used for structural 80 and micro-tectonic analysis to characterize the paleo-stresses recorded during the development of 81 brittle deformation . Still, only few studies combine all these techniques. Further, it remains 82 challenging to link these deformations and those rooted within the underlying crystalline basement 83 which can be highlighted by thermochronological data (e.g., Girault et al., 2022).

84 In this paper, we focus on the Western part of the Alps, where a good geological knowledge exists for the development of the Subalpine fold and thrust belt, by paleontological indirect dating of 85 foreland sediments (<sup>86</sup>Sr/<sup>87</sup>Sr on shells by Kalifi et al., 2021). Therefore, this is a suitable area for 86 87 testing the U-Pb method on calcite to establish an absolute chronology and infer the duration of deformation on major thrusts. Although the sedimentary sequence is well characterized by Kalifi et 88 89 al., (2021), the hypothesis of the tectonic control of sedimentation in these basins by the subsidence 90 driven by local faults instead of a more regional subsidence, driven by European plate flexure at a 91 larger scale, is subject to debate. Further, the possibility of testing the U-Pb calcite dating method 92 on various objects, such as veins and fault planes related to main thrusts activity and on deformed 93 pebbles of the underthrust conglomeratic molasse allows to test the suitability of this method. The combined geochronological (U-Pb calcite) and geochemical study (fluid proxies,  $\delta^{18}O_{calcite}$  and 94 95  $\delta^{13}$ C<sub>calcite</sub> stable isotopes,  $\Delta_{47}$  clumped isotopes) of Tertiary thrusts and molasses in the subalpine 96 massifs, may bring absolute ages on the brittle deformation and on the conditions of calcite 97 crystallization associated to the main tectonic events. These data are further used to reconstruct the 98 kinematics of the Western Alps fold-and-thrust belt propagation through the (re)activation of major 99 thrusts in the Bauges, Chartreuse and Vercors subalpine massifs, and link them to the exhumation 100 of the External Crystalline basements.

## 102 2. Geological Context

## 103 2.1. Overall structure of the Subalpine massifs

The Subalpine massifs (Bauges, Chartreuse and Vercors massifs) are part of the Alpine foreland, which correspond to the deformed Mesozoic and Tertiary sedimentary cover of the European margin in front of the exhumed External Crystalline Massifs (ECMs: Mont Blanc, Belledonne and Pelvoux). This external domain is bounded to the east by the Penninic Front, which marks the limit with metamorphic Internal Alps, representing remnants of the subducted Alpine Ocean and related extended continental margin (Fig. 1).



111 Fig. 1. (a) Geological map of Western Alps, with the location of the Subalpine fold and thrust belt massifs 112 (Vercors, Chartreuse and Bauges) and the External crystalline massifs (Pelvoux and Belledonne). Numbers 113 correspond to sampled locations detailed in Supplementary material 1, roman numbers and capital letters 114 correspond to major subalpine faults and thrusts. A: Eastern Bauges fault; B: Central Bauge Thrust. I Eastern 115 Vercors thrust; II: Eastern Chartreuse thrust; IIIa: Central Vercors thrust; IIIb: Central Chartreuse thrust; IV: 116 West Vercors thrust; V: Western Subalpine thrust. To construct this map, the ALOS World 3D-30m DEM 117 (AW3D30) from the Japan Aerospace Exploration Agency (©JAXA) was used. In addition, the geo-118 referenced database of BRGM's 1:50,000 vectorised and harmonised geological maps (Bd Charm-50) was 119 also used. The faults and the geological cross-section (b) are drawn from the Bd Charm-50 database and 120 from Kalifi et al. (2021).

121 The foreland basin (Fig. 1) is located in front of Subalpine massifs, and consists of Tertiary 122 sedimentary deposits over a large area. This domain shows minor deformations, such as large wave-123 length folds and strata tilting. By contrast, the Subalpine massifs represent a classical fold and thrust 124 belt, which accommodated the Tertiary alpine shortening. In this context, the Tertiary sedimentary 125 basins were locally underthrusted below the main thrusts of the Subalpine Massifs (e.g., Deville, 126 2021). Additionally to this thrust-controlled deformation, a mainly NE-SW dextral strike-slip fault 127 system is present at the scale of the Chartreuse massif (Fig. 1), and appears to crosscut the main 128 thrusts (Gidon, 1981, 1990). Despite this cross-cutting relationship, it is generally admitted that 129 both structures were developed almost contemporaneously (Gidon, 1990). The deformation 130 corresponds to the propagation of flat decoupling layers, mainly along the Triassic evaporites 131 above the Paleozoic crystalline basement, resulting in a thin-skin tectonic style (e.g., Mugnier et al., 132 1987; Butler, 1992; Philippe et al., 1998; Bellahsen et al., 2014; Deville, 2021). To the east, the flat 133 basal decollement roots within the basement at the western boundary of ECMs (e.g., Barfety and 134 Gidon, 1996; Nouibat et al., 2022). In this context, the exhumation of ECMs is in part controlled by 135 the activity of these basement thrusts combined with erosion (Bellahsen et al., 2014; Schwartz et al., 136 2017; Girault et al., 2022). However, the timing and geometrical relationships between exhumation 137 of ECMs and the propagation of the frontal fold and thrust belt still suffer from a lack of direct 138 dating constraints. The position of some of these structures seems to be highly controlled by 139 inherited N-S extensional faults related to the Oligocene Bresse-Rhône or Jurassic Tethyan (Butler, 140 1992).

141

### 142 2.2. Timing of deformation

143 In the Western Alps, direct constraints of the timing of deformation are mostly gathered using syn-144 kinematic minerals within ductile shear zones in the basement (e.g., Rolland et al., 2008; Rosenberg 145 et al., 2021). Timing of deformation within the sedimentary cover is constrained by cross-cutting 146 relationships of the brittle structures (thrust and fault) and stratigraphic constraints on detrital 147 material deposed within flexural basins (e.g., Kalifi et al., 2021). Underthrusting of ECMs below 148 the Penninic Front is constrained by Ar-Ar on phengite and U-Pb on allanite in shear zones that 149 developed within the basement between 29 and 34 Ma (Simon-Labric et al., 2009; Cenki-Tok et al., 150 2014; Bellanger et al., 2015). However, the timing of deformation of the Subalpine massifs, and the 151 propagation of the frontal fold-and-thrust belt remain unconstrained by absolute dating. The relative 152 timing of the deformation sequence is based on the deposition ages of the Neogene molasse 153 sediments obtained by Sr isotopic ratios on bioclasts compared to the global seawater curve (Kalifi 154 et al., 2021). These ages range between ~21.3 and 12.7 Ma (Kalifi et al., 2021) and show a 155 westward migration of the depocenters interpreted to result from footwall tectonic subsidence in 156 response to the activation of major thrusts (e.g., Philippe et al., 1998). However, the interpretations 157 of the Sr isotopic ratios remain difficult to decipher between local subsidence related to thrust 158 activity and a more regional tectonic subsidence, or even some eustatic variations. Further, due to 159 the narrow size of intra-continental marine basins, isotopic influence of the continental source may 160 play a significant role of the resulting sediment Sr isotopic ratios and thus resulting on significant 161 errors on the estimated ages, which must be considered with caution. Further north, the timing of 162 deformations in the Jura fold-and-thrust belt has been studied by in-situ U-Pb calcite dating (Looser 163 et al., 2021; Smeraglia et al., 2021). Looser et al. (2021) constrained two successive stages in the 164 development of the Jura between 14.3 and 4.5 Ma: (1) deformation on the basal cover-basement 165 décollement at 14.3±0.5 Ma, (2) a second phase 11.3 and 4.5 Ma, which they related to the 166 propagation of the northern part of the Jura fold and thrust belt. Smeraglia et al. (2021) obtained 167 similar ages within this second phase in central and southern Jura, with ages from thrusts (11.4±1.1 168 Ma to  $7.5\pm1.1$  Ma) and from tear fault and thrust reactivations ( $10.5\pm0.4$  Ma to  $3.9\pm2.9$  Ma).

169

## 170 **3. Sampling strategy**

171 Different tectonic structures related to the structuration of the subalpine foreland were targeted for172 this study (Fig. 2). These structures correspond to fault planes and calcite-filled fractures associated

173 with the activation of main thrusts in the Mesozoic cover and Tertiary molasses (Fig. 2). This 174 fracturing is recorded by centimetre scale calcite filled-veins within pebbles from the Neogene 175 conglomeratic molasses. The post-deposition nature of the deformation is certified by an observable 176 offset between each side of the fracture calcite filling-vein, see section 4.2. Each sample location 177 has been numbered in the Fig. 1. GPS data, names and other information are included in the 178 Supplementary material 1. Field photographs of outcrops are displayed in Supplementary material 179 2.



180

Fig. 2. Sketch illustrating the different types of studied structures and their relative position to the thrust.
Four types of structures have been studied and sampled: (1) main thrusts in the Mesozoic sedimentary cover,
(2) Strike-slip faults and (3) flexural slip fault in the hanging wall of the thrust, associated to the ramp
anticline development, (4) fractured pebbles with calcite filling in the molasses.

### 186 *3.1 Striated fault planes and veins*

187 We sampled slickenfibres and calcite filled-veins kinematically linked to major thrust and strike-188 slip faults. Field structural analyses have been performed on 16 locations from the three Subalpine 189 massifs (Fig. 1). In the Vercors massif central part, at Rencurel (location 13 in Fig. 1), the so-called 190 "Urgonian" (i.e., Aptian-Barremian) limestone is thrusted toward the NW above unknown age dark 191 marls and Miocene molasses (Fig. 3). The fault zone is marked by a meter-scale fault gouge. The 192 hanging wall of the thrust is marked by a main striated plane oriented ~N10°E. The dark marls are 193 highly deformed with C-S fabrics that locally encloses fragment of calcite veins (Fig. 3). These 194 veins are related to early deformation stage of central Vercors thrust (Roberts, 1994). The fault 195 gouge (sample B, Fig. 3a), a fragment of calcite filled-vein in C-S fabric in the dark marls (sample 196 A) and an undeformed sample taken as reference of the Urgonian hanging wall were sampled.



197

Fig. 3. (a) Field photographs of central Vercors thrust (Rencurel, location 13 in Fig. 1) with Urgonian, main fault plane in red and related cleavage in slates of the high-strain zone in blue. (b) Zoom on a calcite sigmoid close to the fault plane and C-S fabrics. (c) Stereogram of the fault planes, Urgonian bedding and dark marls cleavage, colours refer to (a). (d) Photography of calcite fault gouge sample B with sampling spots of  $\Delta 47$ and stable isotopes (s.i.) with slickensides underlined in red.

In the Bauges massif (location 1, Thrust A), fault gouge was collected in a reverse fault N146°E crosscutting Urgonian limestones (Supplementary material 2). No clear tectoglyphs indicate weither normal or reverse movement occur but in spite of the low inclination (~20°NE) it has been considered as a reverse fault. At location 2 thrust B (Fig. 1) a sample was collected in Upper Valanginian carbonates on a metre-scale top-to-the-west reverse fault plane (oriented N145°E). In the continuation of the fault from location 2, at location 3, the sample was collected in Urgonian limestones on a sinistral strike-slip fault (oriented N64°E). The sample from location 4 corresponds to a fault gouge developed in Urgonian limestones with sigmoidal structure exhibiting reverse topto-the-west kinematics related to thrust II (oriented N132°E). At location 11, the sample was
collected on a N19°E dextral strike slip fault plane also in Urgonian limestones (Supplementary
material 2).

215 In the Chartreuse massif the sample from location 12 was collected in Valanginian carbonates along 216 a reverse fault plane (N11°E) related to thrust IIIa. At location 14 the sample was collected 217 (location 14 in Fig. 1) in massive Portlandian carbonates, along reverse top-to-the-west fault plane 218 (N170°E) associated to a pop-up structure combined to ramp anticline of thrust IIIb. Further south, 219 on the same thrust, at location 15, decametre fold in Urgonian have been observed and a decimetre 220 reverse fault plane (N43°E) have been sampled. In the south-eastern part of the Chartreuse massif, 221 at location 7, the sample was collected in lower Valanginian carbonates along N111°E sinistral 222 strike-slip faults (Supplementary material 2).

223 In the NE of the Vercors massifs, at location 10, the sample was collected on a N85°E dextral 224 strike-slip fault plane in limestone from the hanging-wall of the top-to-the west eastern Vercors 225 thrust I. At the western most location 16 in Urgonian limestones, sample A on normal fault plane 226 (N16°E) and sample B from a fault gouge (N37°E) resulting from flexural slip were selected. These 227 structures are interpreted as related to a top-to-the-west thrust fault V and related to the ramp 228 anticline development during the NW-SE compression. The last location (17) with Upper 229 Valanginian limestones shows three types of structures, (1) vertical large extensional decimetre-230 scale calcite veins (N28°E); (2) strike-slip oblique fault planes (N104°E); (3) reverse fault plane 231 (N169°E). Type (1) and (2) where sampled, but only (1) was dated. In this outcrop, the cross-232 cutting relationships is evidenced by reverse fault planes displacing the vertical veins, which are 233 clearly older (see Supplementary material 3).

234

235 *3.2 Fractured pebbles* 

236 The conglomeratic Miocene molasses are located on the footwall of several thrusts. In footwall 237 basins bedding can be tilted due to the flexuration relative to the above thrust, see local geologic 238 cross-section in Supplementary material 4. The polygenic pebbles of the molasse include a variety 239 of limestones and ophiolite blocks (meta-basalt, gabbro, serpentinite) from a more internal origin. 240 Highly deformed pebbles were observed, especially in the eastern side of the Vercors-Chartreuse 241 massifs (Fig. 4). The tectonic structures include calcite filled-fractures with clear offsets, highly 242 elongated pebble with a thin cleavage and dissolving cups exhibiting pressure-dissolution processes 243 (Fig. 4c). The limited number of locations where the tectonic analysis is possible is due to (1) 244 sandstone dominance versus conglomeratic lithology in the molasses and (2) an overall low density 245 of fracturing within the pebbles, mainly concentrated at the proximity of faults.



246

Fig. 4. (a) East Vercors location 9 Miocene conglomeratic molasse with (b) zoom and correspondingstereoplot of fractures (in black) and bedding (in green, 'S0'). (c) Examples of strained pebbles figures

249	exhibiting brittle-ductile deformation: 1, Visible fracture with mm displacement highlighted by slickensides;
250	2, Dissolution cups with linked radial fracturing and offset; 3, Small calcite filled-veins with observable
251	offsets; 4, Widespread fracturation and fibrous crystallization without any visible offset.

Fractured pebbles were sampled at location 9 associated to Thrust I and at three locations in southeastern Chartreuse associated to Thrust II at locations 5, 6 and 8 (Supplementary material 2).

At location 8, bedding is sub-horizontal and three distinct fractured pebbles were dated. At location 6, close to the thrust, pebbles are less deformed than at location 9, but the molassic bedding is vertical. Location 5 also exhibits highly deformed molassic pebbles.

258

#### 259 4. Methods

#### 260 *4.1 Stable isotopes analyses*

261 In this study,  $\delta^{13}$ C and  $\delta^{18}$ O ratios were acquired using two methods: (1) by isotope ratio mass 262 spectrometer (IRMS) on calcite powders sampled with a dental micro-drill with a diameter of 1 mm 263 (host-rock and calcite cement independently) and (2) by secondary ion mass spectrometry (SIMS) 264 on polished thin-sections, using the Ion Mass Spectrometer (IMS) 1280 (CAMECA) at the Centre 265 de Recherches Pétrographiques et Géochimiques (CRPG) in Nancy, France. Four types of calcites 266 have been sampled: calcite filled-fractures within pebbles (fractured pebbles from location 6, 8 and 267 9), fault gouge calcite filling of thrust and calcite filled-veins cross-cutting the fault gouge (from 268 location 1, 13, 14, 15) and mineralized fault plane (location 3, 7, 10, 12). For the IRMS, 11 269 carbonate powders were analysed using a dual-inlet Isoprime 100 spectrometer (Elementar) coupled 270 to a multiCarb system to determine O and C isotope compositions of calcite at the Laboratoire des 271 Sciences du Climat et de l'Environnement (Université Paris-Saclay). Data were standardized to Pee 272 Dee Belemnite (PDB) based on repeated measurements of international reference materials NBS19 and NBS18, with respective values of -2.20% and -23.01% in PDB for  $\delta^{18}O$  and 1.95% and -273

274 5.01% in PDB for  $\delta^{13}$ C. The reported uncertainties are based on the external reproducibility of an 275 in-laboratory carbonate standard (MARGO) with 1SD of 0.05% for  $\delta^{18}$ O and 0.03% for  $\delta^{13}$ C. For the SIMS analysis, 5 thin sections were analysis for a total of 17  $\delta^{13}$ C and  $\delta^{18}$ O ratios on calcite and 276 277 dolomite according to the methodology described by Andrieu et al. (2018) and Peyrotty et al. 278 (2020). Thin sections were cut in order to match the 30x23 mm sample-holder and were coated with 279 gold before the analysis. The sample is exposed to a primary beam of Cs<sup>+</sup> ions under a vacuum 280 down to few mbar. The primary ion beam diameter varied from 20 µm for oxygen isotope measurements to 10 µm for  $\delta^{13}$ C carbon isotope measurements, and the depth of the resulting crater 281 282 was  $<1 \,\mu$ m. The  $2\sigma$  error range varies from 0.15% to 0.35% for oxygen isotopes, and from 0.2% to 283 1.3% for carbon isotopes. The calibration standard used was a calcite crystal (CCigA) with a  $\delta^{18}$ O value of 18.94% SMOW (Standard Mean Ocean Water) and a value of 1.04% PDB for  $\delta^{13}C$  and a 284 dolomite crystal with a  $\delta^{18}$ O value of 20.04% SMOW (Standard Mean Ocean Water) and a value of 285 286 3.56% PDB for  $\delta^{13}$ C. For a complete description of the materials and protocol, refer to Rollion-287 Bard et al. (2007). All results are presented in per mil (%) deviation from the PDB standard (%) 288 PDB).

289

### 290 *4.2 Clumped isotope analyses*

Clumped isotope  $\Delta_{47}$  analyses were performed to infer temperature and  $\delta^{18}$ O of the fluid during 291 292 calcite crystallization (Ghosh et al., 2006; Eiler, 2007; Huntington et al., 2009; Pagel et al., 2018; 293 Mangenot et al., 2019; Brigaud et al., 2020). Thirty mg of calcite was sampled using a dental micro-294 drill with a diameter of 1 mm. Clumped isotope  $\Delta_{47}$  measurements were performed at the California 295 Institute of Technology (Caltech, USA) with an automated acid digestion and gas purification 296 device coupled to a dual inlet Thermo MAT253, as described in Passey et al. (2010). Samples were 297 weighed into silver capsules (~ 8 mg) and reacted in a common phosphoric acid bath (~ 103 %) for 298 20 minutes at 90 °C under static vacuum. The resulting CO<sub>2</sub> was passed through an ethanol/dry ice 299 U-trap (~ -80 °C) before being collected on a liquid nitrogen temperature (-196 °C) U-trap.

300 Following the 20 minutes reaction period, the collected  $CO_2$  was thawed, entrained in helium, and 301 carried through a Porapak Q 120/80 mesh gas column held at -20 °C using He as the carrier gas. 302 The purified CO2 was analysed using a Thermo Scientific MAT 253 Mass Spectrometer set to collect masses 44-49. Mass 48 was only monitored to detect any hydrocarbon contaminant.  $\delta^{18}O$ 303 304 and  $\delta^{13}C$  data was also acquired as part of each  $\Delta_{47}$  analysis and calculated using the parameters 305 reported relative to the PDB reference frame based on the calibrated composition of the laboratory 306 working gas and the correction scheme and constants from Brand et al. (2010). In order to account 307 for the temperature dependence of oxygen isotope fractionation between CO<sub>2</sub> gas and carbonate 308 resulting from the reaction with phosphoric acid at 90 °C, a fractionation factor of 1.00811 was 309 used for calcite following Swart et al. (1991). The raw  $\Delta_{47}$  data was corrected for instrument non-310 linearity and scale compression (Dennis et al., 2011) using several heated (at 1000°) and 311 equilibrated gases (at 25°C) of various bulk isotopic compositions that were run during each 312 session. These gases were used to convert measurements into the inter-laboratory absolute reference 313 frame (Dennis et al., 2011). To guarantee accuracy of the  $\Delta_{47}$  data, we routinely analysed two 314 carbonate reference materials (Carrara marble and TV04). One of these two carbonate standards 315 was analysed once for every five analyses of the unknown samples in order to check for procedural 316 analytical stability and accuracy, and to determine the long-term external reproducibility of our 317 measurements. The  $\Delta_{47}$  values obtained for these carbonates over the course of this study (April to 318 July 2021) are:  $\Delta_{47-\text{CDES25}} = 0.409 \pm 0.016\%$  (1 $\sigma$ , n = 10) for Carrara;  $\Delta_{47-\text{CDES25}} = 0.666 \pm 0.011\%$ 319  $(1\sigma, n = 8)$  for TV04, i.e., within accepted  $\Delta_{47}$  values for TV04 ( $\Delta_{47-\text{CDES25}} = 0.655\%$ ) and Carrara 320  $(\Delta_{47-\text{CDES25}} = 0.405\%)$ . Finally, the corrected  $\Delta_{47}$  values were converted into temperatures using the 321 composite  $\Delta_{47-T}$  calibration of Bonifacie et al. (2017), which has been shown to be appropriate for 322 calcite and dolomite between 0 and 300 °C, and which has been shown to be consistent with 323 measurements made at Caltech. The oxygen isotopic compositions of the fluid ( $\delta^{18}O_{Fluid}$ ) from which the carbonates precipitated were calculated for each estimated  $T\Delta_{47}$  using the bulk  $\delta^{18}O_{\text{calcite}}$ 324 325 values and the calcite-water fractionation equation from Kim and O'Neil (1997).

### 327 4.3 Paleotensors

328 Three types of calcite-filled fractures have been characterized in the field: (1) striated fault plane, 329 (2) fault gouge cement with sigmoidal shear structures, (3) calcite filled-veins of fractured pebbles 330 from Neogene conglomeratic molasses. Standard azimuth and plunge measurements have been 331 performed on fault planes of structures (1) and (2). For the structures (3) measurements were done 332 on endured fractured pebble with observable offset along the fractures. In rare cases, slickensides 333 are observed so, standard measurements were made. Otherwise, slickenside direction is regarded as 334 equivalent as the maximal displacement line between the two sides of the pebble and perpendicular 335 to the pebble surface. The sense of the movement is indicated by the relative displacement between 336 the two sides. Structural data are sorted in population based on field identified generation and 337 conjugated families (Pascal, 2021). Inversions have been done with TectonicsFP (Ortner et al., 338 2002) using NDA method (Numerical method for Dynamic Analysis described in Spang, 1972). 339 The NDA results are plotted as right dihedral diagrams (Angelier and Mechler, 1977) with a 340 compressive (black) and extensive (white) quadrant containing respectively  $\sigma$ 3 and  $\sigma$ 1 principal stress axis. The P-B-T kinematic axis calculation (Turner, 1953; Marrett and Allmendinger, 1990) 341 342 allows to obtain the principal stress axis orientation (Ortner et al., 2002). The P-B-T axis of each 343 fault plane are plotted in a lower hemisphere with an equal-area stereographic projection. For each 344 location, inversion have been done with a 30° frictional internal angle "theta" (mean value for 345 undifferentiated type of rocks) and then with the best-fit calculated angle. TectonicsFP also 346 calculate fluctuation histograms of the differential angles between the direction of the measured striae and the direction of the calculated shear stress related to the data inversion. The larger the part 347 348 of the dataset close to 0 is, with a rapid decrease, the better it is, but no secondary maximum should 349 be recorded. In the following results, based on Mohr circle visualization, the result with the most 350 significant mechanical consistency is selected. This verification step is required in order to eliminate physically aberrant cases possibly deducted from the calculated best-fit of frictionalinternal angle.

353

### 354 *4.4 U-Pb calcite dating*

355 In-situ U-Pb calcite analyses were carried out at the CEREGE (Centre Européen de Recherche et 356 d'Enseignement des Géosciences de l'Environnement), Aix-en-Provence, France. On each thin-357 section, petrography using optical microscope, cathodoluminescence and SEM were done in order 358 to identify multiple calcite generations. In some samples, variations of calcite texture have been 359 recorded (coexistence of fibrous and blocky calcite filled-veins, Fig. 10 and Supplementary material 5). Analyses were carried out using the parameters given in Supplementary material 6 (adapted 360 361 from Ganade et al., 2022). For data processing, raw intensities and baseline correction were made with Iolite 3 (Paton et al., 2011). Instrumental drift based on NIST614 analyses (Woodhead and 362 Hergt, 2001), Pb isotopes composition and <sup>206</sup>Pb/<sup>238</sup>U are calculated using an in-house Python code. 363 Spots with mean <sup>207</sup>Pb intensities below 3 times the baseline intensity were excluded. Tera-364 365 Wasserburg plots were made using IsoplotR (model-1), intercept ages and initial Pb composition 366 (Vermeesch, 2018). Ages are quoted at  $2\sigma$  absolute with propagation of WC1 2.51% age error by 367 quadratic addition (Roberts et al., 2017). Excess variance of reference material is propagated into 368 sample data. Systematic uncertainties include age uncertainty of reference material.

369

### 370 **5. Results**

### 371 *5.1 Stable Isotopes*

372 46 samples have been analysed (32 in dissolution and 14 in SIMS). Corresponding  $\delta^{18}O_{calcite}$  and 373  $\delta^{13}C_{calcite}$  results are displayed in Supplementary material 7 and have been plotted in Fig. 5.



374

Fig. 5.  $\delta^{13}C_{\text{calcite (VPDB)}}$  versus  $\delta^{18}O_{\text{calcite (VPDB)}}$  plot. Dotted frames are identified domains based on our data. Fror bars (at  $2\sigma$ ) for conventional isotopic analysis are smaller than symbols and those for SIMS analysis appear to be much larger. Letters and roman numbers correspond to main thrusts (Fig. 1).

#### 379 5.1.1 Host-rock compositions

380 A total of eleven analyses has been made on pebbles and carbonate host-rocks. On Fig. 5, these data

appear to plot in a restricted field, close to previously published data from the Jura (Smeraglia et al.,

382 2020; Looser et al., 2021). This field ranges from -6 and -3%  $\delta^{18}$ O ratios and from -1.7 to 2%  $\delta^{13}$ C

383 ratios, which is thus considered as the 'host-rock buffered domain'.

384

#### 385 5.1.2. Host-rock buffered veins

386 A minority of calcite vein samples from our dataset (8/35 values) show a composition close to the

387 host-rock isotopic signature (Fig. 5). These values are obtained in veins from strained pebbles. This

388 'host-rock buffered' domain comprises different types of calcite veins, large calcite filled-veins 389 (few cm in width) with ~100  $\mu$ m blocky calcite, medium-size calcite veins (mm in width) with 390 fibrous calcite and small size calcite filled-veins (100-1000  $\mu$ m width) with either fibres or 391 elongated to blocky calcite (Supplementary material 5).

392

#### **393** *5.1.2 Unbuffered calcites*

394 A majority of calcite vein samples from our dataset (23/35 values), in either fault zones or strained 395 pebbles, show a composition distinct to the host-rock isotopic signature (Fig. 5). These veins show  $\delta^{18}$ O ratios that are significantly lower (-12.5 to -8.6 %) and  $\delta^{13}$ C ratios slightly lower (up to -396 397 2.5%) than their host-rocks. This trend of values is consistent with previously obtained vein values 398 from the Jura Massif (Smeraglia et al., 2020; Looser et al., 2021). In detail, some calcites, like 399 location 8 in the Chartreuse massif, plot at the boundary of the host-rock buffered domain showing a slight  $\delta^{18}$ O depletion of -6.9% while bearing a  $\delta^{13}$ C signature similar to the host-rock (-1.9%). 400 Similarly, at location 9, a calcite vein from a fractured pebble presents a  $\delta^{18}$ O ratio of -6.2% and 401  $\delta^{13}$ C ratio of 2.5%, slightly distinct from its host-rock. Similarly, in the Bauges massif, samples 402 403 from location 1 and 3 plot at the boundary of the 'unbuffered domain' and exhibit depleted  $\delta^{18}$ O values (-7.7% and -7.1%, respectively) and similar  $\delta^{13}$ C values (1.1% and 0.2%) compared to their 404 405 host-rocks.

406

## 407 *5.2. ∆*<sub>47</sub> *Analysis*

Two  $\Delta_{47}$  analyses were made in the Vercors massif: east Vercors (location 9) and central Vercors (location 13) (Table 1). The associated error of 0.014 is the 1 $\sigma$  analytical error obtained on the standard replicates (n=18). At location 9, a  $\Delta_{47CDES90}$  value of 0.363 ± 0.014 has been obtained for a large fibrous calcite filled-vein (Supplementary material 5) from a pebble sample corresponding to a temperature of 149 ± 12 °C. At central Vercors (location 13), a  $\Delta_{47CDES90}$  value of 0.521 ± 0.014

has been obtained for a fault gouge cement sample corresponding to a crystallization temperature of 54 ± 6 °C (Table 1). Combining calcite  $\delta^{18}$ O signature and crystallization temperature allows the calculation of  $\delta^{18}$ O<sub>fluid</sub> signature using Kim and O'Neil (1997) fractionation equation. By using this equation, we obtained a value of 15.9‰  $\delta^{18}$ O<sub>fluid</sub> (SMOW) ratio for east Vercors, while a value of 5.0‰  $\delta^{18}$ O<sub>fluid</sub> (SMOW) is obtained for the central Vercors sample (Table 1). The nature of circulating fluids using these data will be discussed in section 6.3.

### 419 Table 1

420 Clumped isotopes results.  $\delta^{18}$ Ofluid are calculated with Kim and O'Neil (1997) equation.

Location	$\delta^{18}O_{\text{calcite}}$	$\delta^{13}C_{\text{calcite}}$	$\Delta^{47}$	1σ	$\Delta^{47}$	$\Delta^{47}$	T°C	1σ	T°C	T°C	$\delta^{18}O_{fluid}$
	(VPDB)	(VPDB)	CDES90		min	max			min	max	(SMOW)
Location 9	-4.4	2.0	0.363	0.014	0.349	0.376	149	12	137	161	15.9
Location 13	-2.7	1.0	0.521	0.014	0.508	0.535	54	6	48	60	5.0

421

#### 422 *5.3. Paleo-stress tensors*

Fault data inversion results are shown on (Fig. 6) and Supplementary material 8. Over the whole study area, reverse and strike-slip faults have been measured in the vicinity of the main thrusts of the subalpine massifs (Bauges, Chartreuse, and Vercors). In the field, the relative chronology established between these two fault types shows the anteriority of the reverse compared to the strike-slip tectonics (see Supplementary material 3 relating to the description of location 17 where a relative chronology is established). In total, the fault data inversions yielded 18 reduced stress tensors and their respective histograms (Fig. 6).

430

### 431 5.3.1 Fractured pebbles in the molasse sediments

In the conglomeratic molasse formations, three reduced tensors were obtained at locations 6, 8, and 9. The orientations of the measured faults are inhomogeneous (in orientation and movement). This can be seen in the Angelier plots, in the weak groupings of the P-B-T axes, and in the histograms obtained (several maxima are observed in the histograms, and errors greater than 45° are visible). In these molasses, inverse regimes with E-W to NW-SE compressional axes (P-axes varying approximately between N87°E and N120°E) were found. The dip of the S0 planes measured at these locations varies between 19 and 82° and no data tilting was performed.

439

#### 440 5.3.2 Striated fault planes in thrusts crosscutting the Mesozoic sediment cover

441 Seven tensors with compressive regimes were obtained (vertical  $\sigma$ 3 axis; see locations 2, 12, 13, 14, 442 15, 16 and 17). Fault plane dip azimuth are from N330°E to N100°E (mainly N90°E) and dip of 443 ~30 to 75° (Fig. 6 and Supplementary material 8). E-W compressional axes were obtained for 444 almost all results (orientation of the P-axes oscillating globally between N89°E and N105°E). Only 445 locations 14 and 15 show an NW-SE compressional axis (P-axis orientation ~N130°E). The P-B-T 446 axes have low dispersion and the histograms obtained also show errors less than 30°. As for the 447 strike-slip tensors (see section 5.3.4 below), low inhomogeneity of the data sets is visible in the histograms of locations 14 and 17. It is worth noting that at locations 3, 4, 12, 13, 14, 15, and 17 448 449 described above, a monotonic attitude of the fault planes is observed in the Angelier plots. On the 450 contrary, the results of the inversions carried out on locations 1, 2, 7, 10, 16, and 17 are constrained 451 by the presence of conjugate faults or by distinct fault orientations.

452

### 453 *5.3.3 Strike-slip faults*

454 The faults measured in Jurassic and Cretaceous limestones in the study area yielded eight tensors 455 with strike-slip regimes (horizontal  $\sigma$ 1 and  $\sigma$ 3 axes; see locations 1, 2', 3, 4, 7, 10, 11 and 17). In 456 the case of strike-slip faults, E-W to NW-SE compressional axes (P-axis oriented N88°E to 457 N148°E) are observed at locations 1, 2', 7, and 10 computed with both dextral and sinistral fault 458 planes (Fig. 6, Supplementary material 8). The tensors at locations 3, 4, 11 and 17' are consistent 459 with a NE-SW compressional axis (~N36°E to ~N80°E) computed with both dextral and sinistral 460 fault planes. However, it should be noted that small inhomogeneities of the data are observed in the 461 histograms of locations 1, 4, and 11. At location 17, vertical extensional veins were sampled and the 462 structural measurements of these veins are not taken into account in the data inversion, so no tensor 463 can be related to the dated sample. At the latter location, only a relative chronology shows that the 464 extensional veins predate the reverse faults, which also predate the identified strike-slip tectonics. 465 Furthermore, the two inversions at location 17 are constrained by a small number of data (less than 466 8 data) whereas at the other study locations, between 8 and 30 data per location could be used for 467 the inversions.



468

469 Fig. 6. Calculated tensors based on fault orientations, displacement and striae data. Black and white 470 quadrants correspond to compression and extension quadrant, respectively. The P, B and T axes are 471 represented by red circles, white squares and blue triangles respectively. Numbers correspond to the location 472 of measurements (Fig. 7). For each study location, the orientation of the fault planes and their striations as

473 well as the stratification planes (S0) are given in the Angelier plots. In addition, the orientation and name of 474 the samples taken for U-Pb dating are also shown in the latter (coloured in red and orange for samples A and 475 B). The dotted planes are not included in the data inversions (such as the S0 planes and the samples from 476 location 17 which correspond to extensional vein orientations). For each location, the results of the 477 inversions are displayed as right dihedral diagrams (NDA method) and P-B-T axes (P-T method). In 478 addition, histograms related to the NDA method are given.

479

#### 480 *5.4. U-Pb dating*

In this study, we present 27 in-situ U-Pb ages on calcite from 17 different locations spread in
Bauges, Chartreuse and Vercors massifs, ordered from east to west (Fig. 7, data are available in
Supplementary material 9). In Miocene pebbles, a total of 10 ages were obtained from 4 locations
while for fault plane a total of 17 ages have been obtained from 13 locations (Fig. 7).

- 485 For each of the three sample types (mineralized striated fault plane, fault gouge cement and filling-
- 486 veins) success rate was about 60% (27 ages out of ~45 screened thin-section) with the highest rate
- 487 for in filling veins of fractured pebbles (65% with 9 samples over 14 gave at least 1 age).



489 Fig. 7. Simplified map with main thrusts and minor faults and sampled location, see Fig. 1 caption. Roman490 numbers refer to the thrust relative chronology.

492 The following ages are presented with a  $2\sigma$  error bar (including error propagation of WC-1 493 uncertainties). All the obtained ages presented below have satisfactory MSWD values mostly 494 around 1, with several ages showing a slightly higher value (up to 4.5).

495

### 496 *5.4.1. Dating the extension*

497 In south-western Vercors massif, at location 17, large (decimetre scale) N-S trending vertical veins

498 related to an E-W extensional stress field (Supplementary material 3), gave a maximum age of 32.3

499 ± 0.8 (MSWD=1.3), significantly older than for the rest of the fault planes. However, dispersion of 500 the U-Pb results is interpreted as resulting from variable U and Pb mobilization - open-system 501 behaviour - suggesting calcite recrystallization (Roberts et al., 2021). It is noted that a second group 502 of U-Pb results intercept the concordia at 23.8 Ma. Consequently, we interpret these two ages as 503 related to two stages of crystallization during the Oligocene extensional phase at 32 and 24 Ma.



Fig. 8. Compilation of Tera-Wasserburg plot from reverse, normal and strike-slip fault planes. Numberscorrespond to sample locations and colours refer to the sampled thrusts (Fig. 7).

510 East of the Bauges massif, at location 1 (Thrust A; Fig. 8), a calcite-filled fracture within the largest 511 reverse fault has been dated at  $5.7 \pm 1.6$  Ma (MSWD = 1.5). The next thrust to the west (B in Fig. 8) 512 has been dated in locations 2 and 3 (Fig. 8). At location 2, a reverse fault plane returned an age of 513  $6.5 \pm 1.2$  Ma (MSWD = 3.5). The relatively large error obtained on the age and on individual spots 514 is due to their low Pb contents. Although the MSWD is relatively high, the robustness of this 515 isochrone is provided by an important spread in  ${}^{238}U/{}^{206}Pb$  ratio over 0 to 450. At location 3, the 516 collected sample corresponds to strike-slip fault plane along the same Thrust B structure. It has been 517 dated by at  $12.2 \pm 0.8$  Ma (MSWD = 4.5). This relatively high MSWD is due to a significant variability of <sup>207</sup>Pb/<sup>206</sup>Pb ratios for a given range of <sup>238</sup>U/<sup>206</sup>Pb ratios, but can be also explained by 518 519 the high number of spots (n=89), thus the reliability of this age remains high. The robustness of this isochrone is provided by the spread in  $^{238}$ U/ $^{206}$ Pb ratio over 0 to 350. To the north of Chambery, in 520 521 the central part of the Bauges massif (location 4, Thrust II), an age of  $7.5 \pm 1.9$  Ma (MSWD=5.8) 522 has been obtained on a gouge calcite cement within a reverse fault zone. A lot of spots were discarded after <sup>207</sup>Pb cut-off leaving only 10 spots and a relatively important error bar. Although, 523 the remaining spots shown a sufficient spread in <sup>238</sup>U/<sup>206</sup>Pb ratios from 100 to 600, this age is thus 524 525 taken as indicative (Fig. 8). At the eastern limit of the Bauges massif, at location 11, an age of 16.5 526  $\pm$  0.5 Ma (MSWD=1.7) has been obtained from a N34° vertical (dextral) strike-slip fault plane. This 527 fault plane does not seem to be related to any major thrust (Fig. 7).

528

529 5.4.2.1. Chartreuse massif

To the south-east of the Chartreuse massif, three locations associated to Thrust II have been dated (Fig. 7, Fig. 9). A consistent age has been measured at locations 5 and 6, of  $12.7 \pm 0.8$  Ma (MSWD = 1.8) and  $13.9 \pm 2.1$  Ma (MSWD = 2.3), respectively. At location 8, three fractured pebbles gave

- 533 ages: (A) of  $4.7 \pm 1.0$  Ma, (B) of  $3.4 \pm 0.8$  Ma and (C) of  $2.1 \pm 1.8$  Ma (with MSDWs of 2.0, 1.8
- and 1.5, respectively). Since uncertainties on the age overlap these events likely occurred in a single
- tectonic phase around 3 Ma (Fig. 9).



537 Fig. 9. Compilation of Tera-Wasserburg plots from fractured pebbles. Numbers correspond to sample538 locations, roman numbers and colours refer to the sampled thrusts (Fig. 5).

540 To the south-east of the Chartreuse massif, a strike-slip fault plane from location 7 has been dated at 541  $6.5 \pm 0.6$  Ma with a MSWD = 1.5 (Fig. 8). To the north of the Chartreuse massif, at location 12, a 542 sample of the main reverse thrust, corresponding to the central Vercors thrust IIIa, has been dated to 543  $13.6 \pm 0.9$  Ma with a MSWD = 2.0 (Fig. 8). To the north-western part of the Chartreuse massif, at 544 location 14, related to thrust IIIb, a hundred-metre-long major thrust vein from a reverse fault plane 545 has been sampled and dated at  $11.1 \pm 0.6$  Ma (A1) and  $10.5 \pm 0.7$  Ma (A2) with MSWD values of 546 1.5 and 1.9, respectively. The overlapping error bars of these two ages likely suggest a single 547 fracturation event, but because they are from two different thin-sections, spots are not mutualized. 548 To the south in the same major thrust, a reverse fault plane has been dated at location 15 (Fig. 8). 549 Two ages have been obtained on this fault plane from the same thin-section: (A1)  $16.6 \pm 5.3$  Ma 550 (MSWD = 1.8) and  $(A2) 8.4 \pm 1.5$  Ma (MSWD = 0.99). Even though MSWD of A1 is acceptable, 551 <sup>238</sup>U/<sup>206</sup>Pb ratio only varies up to 25, which is reflected by the error bar but this age is taken as 552 indicative. The "A2" age shows an up to 150 <sup>238</sup>U/<sup>206</sup>Pb ratio with a younger age that is still 553 coherent with other location 14 ages (Fig. 8).

554

#### 555 5.4.2.1. Vercors massif

To the NE of the Vercors massif, in the footwall of the eastern Vercors thrust I (location 9, Fig. 7), five ages were obtained out of four strained pebbles thin-sections (Fig. 9). The oldest vein generation in location 9 (Fig. 9) is highlighted by 3 similar ages obtained in 3 different samples A ( $15.4 \pm 1.1$  Ma), B ( $15.4 \pm 1.2$  Ma) and 'C Rims' ( $15.2 \pm 0.6$  Ma) with good MSWDs (2.7, 1.6 and 1.3, respectively).

In order to decipher the relationship between vein opening and calcite U-Pb ages, a LA-ICPMS age
map (Fig. 10c-d) was undertaken in one of the largest calcite filled-vein of blocky-calcite of sample
C. This map exhibits two stages of calcite precipitation associated to opening of the 'generation 2'

- 564 vein (C 'Rims': age of  $15.2\pm0.6$  Ma and 'Core': age of  $8.9\pm0.7$  Ma with a MSWD of 1.3 and 1.2,
- 565 respectively).



Fig. 10. (a) Thin-section of fractured pebble C from location 9 with cross relationships of calcite filled-veins.
(b) Zoom on age cartography made on generation 2 filled veins, each spot corresponds to a U-Pb analysis.
Red numbers correspond to straddles ousted spots. (c) Cathodoluminescence photography evidencing the
dark borders of the generation 2 filled-veins dated at ~15 Ma and the lighter core dated at ~9 Ma (samples C
in Fig. 9). (d) <sup>208</sup>Pb interpolation from age spots highlighting the 2 different generations inside the generation
2 filled veins.

574 On Fig. 10b, each spot of the age map has been numbered, boundaries have been traced based on 575 LPNA and cathodoluminescence image (Fig. 10c), all data are available in Supplementary material 576 9. Thereafter, a <sup>208</sup>Pb cartography built by the interpolation of each analysed spot allows to delimit 577 the boundary between the two compositional domains (Fig. 10d). While the edge presents dark shades in cathodoluminescence and high <sup>208</sup>Pb contents, the central part presents bright 578 cathodoluminescence shades and low corresponding <sup>208</sup>Pb contents. Just below the dated area, an 579 elemental cartography has been made (Fig. 11a). For <sup>208</sup>Pb and <sup>57</sup>Fe the sharp limit of the two 580 generations calcite filled-vein is visible between the borders and the core. Whether it is for <sup>208</sup>Pb or 581 for  ${}^{57}$ Fe, the younger core generation is depleted (0-5 x10<sup>3</sup> and 1-2 x10<sup>6</sup> CPS respectively) 582 compared to the older rims' generation (10-20  $\times 10^3$  and 2-3  $\times 10^6$  CPS respectively). 583



Fig. 11. (a) Zoom on elemental map zone. (b) Elemental mapping of the generation 2 calcite filled veins
evidencing differences in various elemental contents in count per second: <sup>208</sup>Pb, <sup>57</sup>Fe, <sup>238</sup>U and <sup>88</sup>Sr.

Concerning <sup>238</sup>U and <sup>88</sup>Sr, the distribution in these elements is heterogeneous with a core more
depleted (0-80 x10<sup>3</sup> and 10-40 x10<sup>6</sup> CPS respectively) than the rims (0-160 x10<sup>3</sup> and 10-40 x10<sup>6</sup>
CPS respectively) (Fig. 11b). The contents in <sup>238</sup>U and <sup>88</sup>Sr show anti-correlated patterns.

- 591 In addition, in the same location 9, another younger age  $(3.8 \pm 0.2 \text{ Ma}, \text{MSWD} = 1.2)$  was obtained
- 592 from another sample (D). Further, five kilometers to the south, in the hanging-wall of eastern

593 Vercors thrust I, at location 10, a strike-slip fault has been dated at  $13.9 \pm 0.8$  Ma with a MSWD = 594 2.7 (Fig. 8). The MSWD value is highly improved (from 2.7 to 1.9) by removing the two spots close 595 to 100 value of  ${}^{238}\text{U}/{}^{206}\text{Pb}$  ratio, while the age and associated uncertainty only vary slightly (by -0.3 596 Ma and -0.1 Ma, respectively).

597 In the southern part of the Vercors massif, at location 13, the major thrust IIIa was sampled (Fig. 7). 598 The thrust gouge cement (sample B) and sigmoidal structures in the footwall (sample A, 20 cm 599 close to the fault plane) have been dated (Fig. 8). The fault plane veins (sample B) returned an age 600 of 7.7  $\pm$  3.4 Ma and sigmoid veins (sample A) of 12.1  $\pm$  0.4 Ma with a MSWD of 7.7 and 1.6, 601 respectively. For the first age, the low number of spots (14) and the high MSWD are explained by 602 the small size of dated calcite filled-veins (even with the reduced spot size) and the observation of 603 clast contamination for some spots. This age is thus taken as an indicative estimate of the younger 604 calcite generation. In contrast, sample A shows a well-constrained age with a good spread of <sup>238</sup>U/<sup>206</sup>Pb ratios over 0 to 400 (Fig. 8). 605

At the western border of the Vercors massif, in the Bas-Dauphiné basin, at location 16, a fault plane related to Thrust V flexural slip (Fig. 7) gave two ages on different samples of (A)  $8.9 \pm 1.4$  Ma and (B)  $7.9 \pm 2.4$  Ma with MSWDs of 1.9 and 1.6, respectively (Fig. 8). Sample A shows a robust isochrone while sample B age has to be taken with caution because the correlation relies mainly on 2 points with relatively low <sup>238</sup>U/<sup>206</sup>Pb ratios (25 and 70). The overlapping error bars of the two ages suggest a single fracturation event or tectonic phase.

612

### 613 6. Discussion

## 6.1. *Limits for the application of U-Pb on calcite method, and sample selection*

615 Calcite U-Pb geochronology is limited by its overall low U and Pb contents (~  $<10 \mu g/g$ ). Recent

616 technical advances of LASER ablation coupled with mass spectrometers with improved sensitivity

617 have allowed U-Pb dating of <1cm calcite veins and to identify multiple generations (Roberts et al.,

618 2021). Carbonate rocks usually show low uranium contents, typically 10 ppb to 10 ppm (e.g., 619 Roberts et al., 2020). Calcites with an isotopic signature ( $\delta^{13}$ C and  $\delta^{18}$ O) close to their host-rock are 620 thus interpreted as resulting from crystallization in a fluid at chemical equilibrium with the host-621 rock in a relatively closed system (small scale fluid flow) and thus generally resulting in a uranium-622 poor calcite (Fig. 5). It follows that, in our sample dataset, the majority of calcites falling within the 623 "host-rock buffered" stable isotopic compositional domain (Fig. 5) does not have a sufficient 624 uranium content to be datable. Conversely, the successfully dated calcites present a  $\delta^{18}$ O signature different from their host-rock ( $\pm 4\%$  and more) with similar  $\delta^{13}$ C signature. The origin of the 625 626 uranium is thus supposed to result from the interaction of fluids with the underlying crystalline 627 rocks, clay formations or minerals present in terrigenous formations like sandstones (e.g., Tartese et 628 al., 2013). Thus, the supply of uranium to the calcite system is often facilitated by a relatively large-629 scale fluid flow with significant interactions with various kinds of host-rocks (Bilau et al., 2021). 630 Uranium incorporation in calcite is not only related to parental fluid uranium contents but also relies 631 on calcite crystallization rates, which are a function of the opening fracture mode (Gabitov et al., 632 2014; Prajapati et al., 2018). Calcite texture can provide significant information to discriminate 633 between the several opening fracture modes (Bons et al., 2012; Prajapati et al., 2018). Fibrous or 634 elongated calcite crystals are observed in a continuous slow vein opening mode, whereas blocky 635 calcite is related to a more spontaneous opening mode with a larger crystallization space. This 636 consideration directly impacts the feasibility of U-Pb dating. Some progress on calcite U 637 incorporation knowledge has been made by the study of natural samples and synthesized calcite (Weremeichik et al., 2017). For a slow crystallization speed rate of 0.001 nm.s<sup>-1</sup> the sharing ratio of 638 639 U between fluid and calcite (K<sup>U</sup>) is around 0.02 (no entrapment) whereas, for a rapid crystallization speed of 1 nm.s<sup>-1</sup> the K<sup>U</sup> is 0.06 (100% entrapment) (Gabitov et al., 2014; Weremeichik et al., 640 641 2017). It follows that blocky calcite could entrap more U than fibrous calcite making it more 642 suitable for dating. Fibrous calcite crystallization related to syn-kinematic continuous slow opening 643 process and usually related to the beginning of the fracturation (Prajapati et al., 2018) seems to

644 incorporate less U. In our study, we confirm that, in terms of their stable isotopic composition, 645 fibrous calcite stands close to their host-rock signature. Although almost all the calcites (6/8) in the 646 host-rock domain were unsuccessful candidates for U-Pb dating, one of these was dated (SIMS analyse from sample D at location 9, with 1.3%  $\delta^{13}$ C and -5.6%  $\delta^{18}$ O, Fig. 5). It follows that nearly 647 648 all of the low  $\delta^{18}$ O<sub>calcite</sub> ratio (< -6%) calcites (18/19) appear to be suitable for U-Pb dating. 649 Furthermore, oxidation state of uranium is also important, oxidised uranium is mobile in fluid while 650 reduced uranium is easily incorporated in calcite structure (Gabitov et al., 2021). The following 651 process can be imagined, oxidating meteoric fluid deeply percolating, interacting with crystalline rocks and becomes enriched in  $\delta^{18}$ O and U then is mobilized by fault networks and become reduced 652 653 and crystallize calcite at the same time.

In conclusion, in a carbonate context similar to the subalpine massifs, a blocky calcite with a stable isotope signature different from the host-rock seems to be more suitable candidate for U-Pb calcite dating than those that equilibrated with the host-rock, and especially the fibrous calcite. This indication should thus be taken in consideration for future sample selection.

658

### 659 6.2. Significance of U-Pb calcite ages

660 The main question that we address here, following recent works on this theme (e.g., Roberts et al.,661 2021), is how can we interpret the obtained U-Pb calcite ages?

Based on either experimental or natural examples, it is shown that calcite crystallization is a rapid process. Natural example of fault-related veins dated by U-Th of calcite suggests a crystallization speed rate of ~0.03 to 0.80 mm.ka<sup>-1</sup> (Uysal et al., 2007, 2011; Williams et al., 2019). The calcite crystallization process can thus be considered as instantaneous in sight of the geological age and precision we are looking for to reconstruct the deformation sequence of a fold and thrust belt on millions of years' scale. In addition to the opening rate, the calcite texture is also a function of the fluid source and rugosity of the pore's wall (Nollet et al., 2009). Fibrous calcite is more likely to 669 develop in a local pore-fluid derived fluid with a slow opening rate whereas elongated blocky 670 calcite is more likely to grow with smooth vein walls, an external fluid source and a related higher 671 opening rate as in a single fracturation event (Nollet et al., 2009). At location 9, the age of 15 Ma is 672 observed on three different pebbles from Miocene conglomerate, which seems to indicate that the 673 porosity was filled within a period of time shorter than the uncertainty on the ages ( $\sim <0.5$  to 1 Ma). 674 With the assumption that the porosity is not left empty, the age of crystallization can be considered 675 as equivalent to the age of fracturing. Furthermore, pebble elongation and calcite cemented offset 676 are tectonic markers indicating a post-depositional deformation linked to thrust activity. An 677 important part of the field and petrographic analysis is thus to characterize each cogenetic calcite 678 generation in order to sample and date them independently. Within a single generation, even if a 679 calcite shows a growth zoning, the age between the edge and the core of a calcite grain (considered 680 as part of the same crystallization event) is the same at the sight of our 0.1 Ma analytical precision 681 (e.g., Bilau et al., 2021). In addition, it is not uncommon to observe calcite filled-veins with residual 682 porosity (partially-open, Prajapati et al., 2018) in the form of a void and the measured age is the 683 same independently of the proximity to the residual porosity.

684 Now that the calcite crystallization is considered as a fracturation age, is this age robust or may it be 685 reset by any further deformation or fluid flow events? The common idea we have about calcite is 686 that it is not a very resistant mineral, and that it is highly sensitive to fluid circulation (Lamy-687 Chappuis et al., 2016). As opposed to the (U-Th)/He method, uranium replaces calcium in the 688 calcite structure (Gabitov et al., 2021) and in the absence of any recrystallization evidence, that 689 could be observed in thin section, it has a stable lattice position in natural calcite, in uranyl form, 690 U(VI), (Reeder et al., 2000; Kelly et al., 2003; Ortega et al., 2005) but Gabitov et al. (2021) shows 691 that tetravalent U(IV) uptakes by calcite is four orders of magnitude higher that oxidised U(VI), 692 (Rasbury and Cole, 2009; Gabitov et al., 2021). In the absence of any dissolution or 693 recrystallization features, the U-Pb system in calcite seem to remain in a closed system. In calcite, 694 chemical zoning profiles alike zircon has not been observed. At the corresponding temperature 695 range to calcite growth, solid diffusion seems too slow and should be considered null. Therefore, a 696 reset of calcite age in response to diffusion is unlikely. However, the circulation of aggressive fluids 697 will lead to a complete dissolution of calcite and possible reprecipitation, anyway such features can 698 be observed in cathodoluminescence and are not present in our samples. Moreover, given this solid 699 behaviour of U-Pb system in calcite once, in order to obtain a robust isochrone from a Tera-700 Wasserburg plot, one has to recognize and date a single synchronous generation. Otherwise, it is not 701 possible to undertake any linear regression with all the dataset, as the dated spots will result in a 702 significant scattering. The assumption underlying the use of this diagram is the presence of a 703 cogenetic generation (e.g., Roberts and Holdsworth, 2022). In Fig. 10, the age map highlights that a 704 single age, reflected by a homogeneous composition for the whole calcite generation, is obtained. 705 The demarcation between the two age domains is sharp without any apparent mixing of ages at the 706 <10 µm scale. Furthermore, the older 15 Ma calcite is well preserved, even if the vein was re-707 opened under the same stress system (similar vein orientation). The fact that a 15 Ma age is also 708 obtained in two other pebbles confirms that the reopening of the vein did not alter it. It is thus likely 709 that a new porosity was created and was rapidly sealed by fluids and calcite precipitation while the 710 older calcite remained intact.

711

### 712 6.3 The nature of fluids during deformation, and insights for fluid-rock interaction and burial

Host-rock  $\delta^{18}$ O and  $\delta^{13}$ C signature leads to define a domain so-called «host-rock buffered domain» 713 714 to the analysed veins. Calcites from this domain are interpreted as resulting from the crystallization of a fluid with a low fluid/rock ratio in equilibrium with the host-rock. Calcites with a lower  $\delta^{18}O$ 715 716 value are suggested to fall in an 'unbuffered domain' relative to the host-rock composition. All 717 together, these analyses define the "unbuffered domain" of our vein dataset. Calcites from this 718 domain are interpreted as resulting from the crystallization of a fluid with a high fluid/rock ratio, 719 which did not allow it to fully equilibrate with the vein host-rock. Clumped isotopes may retain the 720 nature of percolating fluids and paleo-temperatures, which can be used to constrain burial dynamics

721 (Smeraglia et al., 2019; Curzi et al., 2021; Looser et al., 2021; Smeraglia et al., 2021). Moss (1992) 722 was the first to bring some burial temperature constrains using vitrinite reflectance ( $R_0$ ) in 723 Subalpine massifs. Cretaceous units have low  $R_0$  (<0.5%), which gave a burial temperature of 40-724 90°C (Fig. 12), according to Basin% $R_0$  and Easy% $R_0$  (vitrinite to °C conversion model, Nielsen et 725 al. 2017).

726 However, these estimates can be affected by the exposure time at a given temperature, so deduced 727 temperatures from Moss (1992) may be indicative. Jurassic units have higher R<sub>o</sub> from 0.8% to 728 1.25%, corresponding to significantly higher temperatures (~135°C to ~180°C). Furthermore, for 729 the same geologic units, higher R<sub>0</sub> values have been observed for the north Chartreuse than for the 730 south Vercors and also inside each Subalpine massif, with increasing values towards the east. The 731 N-S gradient has been interpreted as related to an earlier and more significant burial in the north. 732 while W-E gradient is related to a thicker tectonic load at the front of the ECM (e.g., Bellahsen et 733 al., 2014). Moss (1992) suggested that the Vercors and Chartreuse Subalpine massifs underwent a 734 rather limited burial (40-80°C for Hauterivian units and 100-160°C for Upper to Middle Jurassic units) for a short duration (<10 Ma). 735

These rather high temperatures obtained for the eastern part of Vercors compare well to Raman
Spectrometry of Carbonaceous Material (RSCM) conducted on ECM Mesozoic cover by Bellanger
et al. (2015) who obtained a uniform maximum temperature of ~330°C decreasing to <200°C in</li>
neighbouring east Vercors Jurassic limestones.





740

These recorded  $T_{max}$  at the base of the ECM Mesozoic sedimentary cover are consistent with a thickness of 8-11 km of tectonic nappes above ECMs (Philippe et al., 1998; Bellanger et al., 2015).  $\Delta_{47}$  temperatures related to syn-fracturation calcite obtained in this paper complement this rather significant increase in burial temperatures from west to east.  $\Delta_{47}$  temperatures increase from 54°C in the central Vercors thrust (location 13, Fig. 12) to 149°C in eastern Vercors thrust footwall (location 9, Fig. 12). The determination of the calcite crystallization temperature by an independent method combined to calcite  $\delta^{18}$ O signature allows the calculation of  $\delta^{18}$ O<sub>fluid</sub> signature with Kim and O'Neil (1997) equation (Table 1, Fig. 13). These two fluid isotopic signatures are interpreted as distinct fluid sources:

- Fluid composition in central Vercors corresponds to a colder fluid with a  $\delta^{18}O_{\text{fluid}}$  composition of 5% SMOW compatible with a heated meteoric fluid which slightly interacted with carbonate rocks (Hoefs, 2021).
- Fluid composition and paleotemperature in eastern Vercors are indicative of hydrothermal fluids
  which interacted with crystalline rocks and/or metamorphic fluids (e.g., Brigaud et al., 2020).



Fig. 13. Recalculated Subalpine and Jura  $\Delta_{47}$  temperature versus  $\delta^{18}O_{\text{Calcite (VPDB)}}$  and calculated  $\delta^{18}O_{\text{Fluid}}$ (SMOW) in dashed lines (Kim and O'Neil, 1997). Fields are based on literature: mollusc shells compiled in (Veizer et al., 1999) are supposed in equilibrium with marine water temperatures, following Mangenot et al. (2019); precipitation field relies on IAEA data of Wiser program, https://nucleus.iaea.org/wiser/index.aspx,  $\delta^{18}O_{\text{Fluid (SMOW)}}$  and Air temperatures from Malaussene, Carpentras, Draix and Thonon-les-Bains.

764

At central Vercors (location 13), the sealing calcite  $\delta^{18}$ O signature plots within the host-rock domain (Fig. 5) with a crystallization temperature of 54°C, the resulting parent fluid is about 5‰ SMOW. This positive value could result from an interaction (dissolution) with the Urgonian host rock. The fault gouge cross-cutting veins have a low  $\delta^{18}$ O<sub>calcite</sub> value (-9‰), highly depleted compared to the host-rock signature, which is interpreted to result from a cold (<60°C) meteoric fluid.

771 Based on these arguments, the recorded crystallization temperature of 54°C (Table 1, Fig. 13) is 772 interpreted as resulting from a surface or basin-derived fluid that equilibrated with surrounding 773 rocks at shallow ~1-2km depth. This shallow burial depth can be explained by a moderate 774 underthrusting of the molasse. In eastern Vercors, the estimated temperature is of 149°C. This 775 elevated temperature is coherent with the observation of a more ductile deformation style and with 776 similar temperature estimates obtained by independent methods (vitrinite reflectance and RSCM, 777 Fig. 12). Based on these data and taking into account a geothermal gradient of 25 to 35°C/km, the 778  $\sim$ 150°C temperature can be related to a maximum burial depth of 4-6km. The significant difference 779 in temperature ( $\sim 100^{\circ}$ C) recorded between eastern and central Vercors highlights a marked increase 780 of the tectonic thrust stack in front of the ECM. Because the fracturation and the vein opening are 781 supposed to occur at the burial peak, this high recorded temperature is interpreted as a 782 crystallization temperature with little influence of solid-state reordering (Henkes et al., 2014; 783 Stolper and Eiler, 2015; Mangenot et al., 2019). This temperature is close to the closure temperature 784 of the U-Pb system in calcite (150°-200°C, Lloyd et al., 2018) so the calcite could have been 785 exposed to higher temperatures (deeper burial) without being recorded. Consequently, the derived fluid signature can be interpreted as a maximum value of  $\delta^{18}O_{\text{fluid}}$ . The recalculation of the fluid signature at equilibrium at the  $\Delta_{47}$  temperature of 149°C leads to a minimum ratio of 15.9 ‰. This high value, which can only be associated with highly enriched fluids, may also be the result of diffusion in the solid state and would therefore be a maximum value.

Toward the north, the Bauges and Bornes massifs show a similar increase in maximum recorded temperatures estimated from RSCM data (Muirhead et al., 2020),  $\Delta_{47}$  on host-rock, and illite crystallinity (Mangenot et al., 2019). These data show a significant variation of maximum burial temperature (from <70 °C in the west to 300°C in the east of these massifs). The difference in maximum temperature between the Bauges-Bornes and Vercors massifs, and from west to east within these massifs, is interpreted as resulting from a variation in the structural nappe stack thickness.

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### 798 6.4. Relationship of the thrust propagation sequence with molassic basins emplacement

799 Sedimentation in the molassic basins in this zone is bracketed in the range ~20-15 Ma (Kalifi et al., 800 2021). These authors suggested that subsidence occurred in relation to the activation of the thrust 801 sequence (i.e., 'tectonic subsidence'). Along our dated transect within the Subalpine massifs, the 802 age of sedimentation suggested by Sr isotopic ratios clusters mostly between 18 and 16 Ma (Fig. 14; 803 Kalifi et al., 2021). The comparison of thrust U-Pb calcite ages shows a good fit with the basin 804 sediment ages, and suggests that thrust activation shortly followed the end of sedimentation in each 805 sector. However, The U-Pb calcite ages obtained in this study are mostly younger (15 Ma 806 maximum) than the molasse sedimentation ages in the Subalpine massifs, which suggests that the 807 timing of fault development is decoupled from that of sedimentation. It follows that the mechanism 808 of frontal basin opening is dominated either by a more eastern fault (located to the east of the 809 Belledonne ECM), or by eustatic variation and/or regional subsidence instead of local parameters 810 like in-sequence thrust propagation within the Subalpine domain. It seems that there is a link 811 between the activation of the central Vercors and eastern Chartreuse thrusts (dated at ~12 Ma in this

812 study) and molasse deposition at the front of Subalpine massifs in the Bresse basin, where 813 sedimentation is restricted to 12 to  $\sim$ 8.2 Ma (Kalifi et al., 2021). The perfect match of activity of the 814 Bresse foreland basin with the central Vercors thrust is suggestive of a tectonic control of this phase 815 of basin subsidence.



Fig. 14. (a) Timeline of the development of the thrust sequence on a NW-SE profile and comparison with the
sedimentation ages of molassic basins. (b) W-E Cross-section, modified from Kalifi et al., (2021) with major
thrust activation. Fault names related to roman numerals are described in Fig. 1 caption. See Supplementary
material 4 for a zoomed cross-section of the eastern Vercors side.

816

### 822 6.5 Evolution of the stress field and proposed tectonic model

823 The combination of paleo-stress reconstructions and U-Pb dating undertaken on the same calcite

sample allows to decipher the evolution of stress through time in a given geological area. All the

825 dated tensors that were obtained in Fig. 6 (i.e., all locations except 17) are shown in the compilation 826 (Fig. 15) featuring the tectonic evolution of the Alpine foreland constrained by U-Pb ages on 827 calcite. The fault data inversions carried out in this study show a good consistency of results at the 828 regional scale. Two main trends are observed for the compressional axes: E-W to NW-SE and NW-829 SE. In the overthrust limestone deformed cover (locations 1 to 17), homogeneous datasets, low 830 dispersion of the P-B-T axes and internal friction angles close to the average value accepted in the 831 rocks (~30°; except for locations 2', 4, 11 and 17 where internal friction angles lower than 20° are 832 obtained) show a good reliability of results, even in case of a small number of data (<8 data, 833 location 17). These results appear to be consistent with the geometry of the main tectonic faults that 834 were studied. On Fig. 15, paleostress results displayed for the oldest recorded calcite U-Pb calcite 835 ages, can be depicted in three stages corresponding to three distinct states of stress:

836 (1) During stage 1, ranging between 17-14 Ma the stress regimes of Bauges and Vercors 837 massifs were different. In the Bauges massif, a NE-SW compressive axis is recorded and 838 interpreted as resulting from a transpressional side effect of deformation during early stages 839 of deformation of the Bornes-Bauges and Jura massifs, in response to exhumation of 840 Aiguilles Rouges and Mont Blanc ECMs (e.g., Boutoux et al., 2016; Rolland and Rossi, 841 2016), while ECM exhumation remained low at ~17 Ma in the Belledonne massif (Girault et 842 al., 2022). In the meantime, NW-SE compression is recorded in the Vercors-Chartreuse 843 massif and is attributed to the first signs of Alpine compressional tectonic activity in these 844 subalpine massifs.

(2) During stage 2, from 14 Ma to 10 Ma, Bauges and northern Chartreuse show NE-SW and
NW-SE compressional axes, respectively. The influence of more significant displacements
in the Jura massif compared to the subalpine massifs could induce strain accumulation in the
limit of these massifs accommodated as local strike-slip deformation regime observed in
southern Bauges. In contrast, the northern Chartreuse massif underwent a more E-W
compression in relation to the thrust geometry, and the absence of evaporites inducing less

851 basal decoupling. At the same time, a similar pure E-W orientation is observed in Vercors 852 and southern Chartreuse massifs. We propose that the rapid activation of numerous frontal 853 thrusts in a short time period (~1 Ma) at ~12 Ma is related to a more rapid exhumation of the 854 Belledonne MCE indicated by ZHe ages (Girault et al., 2022). We suggest that the rapid 855 propagation of the deformation in the Subalpine Massifs is controlled by the activation of a 856 blind crustal-scale ramp located at the front of Belledonne MCE (Fig. 14). This change in 857 stress direction could result from the structural anisotropy of the region, and is especially 858 ascribed to the reactivation of N-S inherited crustal structures. The N-S sub-vertical 859 anisotropy corresponds to the East Variscan shear zone (e.g., Corsini and Rolland, 2009; 860 Simonetti et al., 2020) in the basement along the MCE, the Jurassic extensional faults (e.g., 861 Lemoine et al., 1986) and the Oligocene extensional faults like the one dated at 32 and 24 862 Ma (location 17, Fig. 8). These inherited faults have clearly controlled the localization of 863 major thrust as highlighted by the eastern Vercors structure (e.g., Fig. 14b, Schori et al., 864 2021).

(3) Since 10 Ma, ECMs have been uplifted at a somehow constant rate, as suggested by a similar magnitude of exhumation rates (about 1 mm.yr<sup>-1</sup>) deduced from thermochronology
(Girault et al., 2022) and vertical motions recorded by GPS data (Sternai et al., 2019). A fairly uniform E-W stress field is recorded from Bauges to south Vercors with the activation of the Western Subalpine thrust and multiple "hors-sequence" reactivation of the thrusts in a dominant strike-slip context until present (Rolland et al., 2022).





Fig. 15. Sketch depicting the evolution of strain context through time, the involved faults and related ECM
exhumation. Activation of the different thrusts is inferred by the first age recorded for each fault. Small grey
arrows are local data derived stress orientation while big grey arrows are regional interpreted stress
orientation. Black cross in a grey circle are exhuming zones.

### 877 6.6. *Timing of fold-and-thrust belt propagation*

The new U-Pb calcite ages obtained in this study allow to constrain the timing of the thrust propagation sequence in the Subalpine massifs. Based on the distribution of the different ages obtained from the different thrusts (Fig. 14), and the structural relationships (based on Deville, 2021; Kalifi et al., 2021 and references therein), it appears that four major thrusts have been activated from east to west:

- 883 (1) The most eastern thrust in the Vercors massif was activated around 15 Ma in a NW-SE884 compressional context.
- 885 (2) To the west, in-sequence thrust activation propagation occurred with the activation of the 886 central Vercors thrust and E-W shortening around 12 Ma. It appears from structural 887 relationships that this fault system is in continuity with the eastern Chartreuse thrust, and 888 that its southward propagation in Vercors is split into two main different faults (the II 889 and IIIa thrusts, as shown on Fig. 14). At this stage, based on the basement offset in the 890 rooted trust we assume that the deformation is mostly in a thick-skin mode as the 891 deformation front is not significantly transferred into the evaporitic layer at the base of 892 the sediment cover. In contrast, this phase correlates with an acceleration of the 893 Belledonne ECM, through the activation of a crustal ramp, rooted directly west of the 894 ECM.
- (3) A transfer of this motion occurred around 10 Ma on the western Vercors-Chartreuse
  thrusts (e.g., west Vercors thrust, Fig. 15). This rapid propagation of thrusts is supposed
  to be related to the transmission of stress by the Triassic decoupling evaporitic layer
  which marks a transition from thick-skin tectonics to a more thin-skin tectonic mode.
  However, based on geological cross-section (Fig. 14) it seems that the flat decoupling
  layer always branch on the same transcrustal fault at the base of Belledonne ECM.

901 (4) A final in-sequence activation of the mostly blind Western Subalpine thrust is 902 constrained at 7-9 Ma, still in agreement with a propagation of the decoupling layer on 903 basal evaporites. However, at massif scale, it appears that this front progressed more to 904 the west of Vercors than of Chartreuse, probably due to the lack of evaporites below this 905 later massif and a location of the basal décollement in Lias-Dogger marly shales 906 (Deville, 2021). As a result, we interpret the development of NE-SW dextral faults of 907 southern Chartreuse as a result of this differential propagation of the active front 908 between the two massifs. Indeed, the dextral lineaments clearly offset the 10-12 Ma 909 thrusts, and thus could be correlated with this ~7-9 Ma final propagation event. The 910 propagation of the deformations towards the foreland is likely to have induced an uplift 911 of the most internal subalpine massif and could therefore explain a more significant 912 erosion and exhumation (about 4-6 km) of these domains.

913 After this fold-and-thrust propagation that occurred between 15 and 7 Ma, widespread and diffuse 914 "out-of-sequence" reactivation occurred in the whole subalpine massifs. Widespread reactivations 915 of internal thrusts may result from the locking of thrust propagation to the west. This phase is 916 highlighted by the re-opening of veins in the eastern Vercors thrust around 9 Ma (Fig. 9). Younger 917 deformation (e.g.,  $6.5 \pm 0.6$  Ma at location 7) is recorded by a widespread strike-slip activity always 918 in an E-W ( $\sigma$ 1) compressional context which indicates a permutation of  $\sigma$ 2 and  $\sigma$ 3 stress axes. Since 919 ~6 Ma, this evolution towards a more strike-slip dominated regime in a similar context as nowadays 920 may be caused by a modification of the regional strain-field that could be ascribed to the Apulian 921 late rotation (e.g., Collombet et al., 2002; Rolland et al., 2012; Walpersdorf et al., 2018).

922

### 923 7. Conclusion

924 Through the example of the subalpine fold-and-thrust belt, this study establishes a multi-technique 925 methodology based on fault data inversion, conventional and clumped isotopes and U-Pb calcite 926 dating in order to provide a detailed comprehension of the Tertiary tectonic structuration of the 927 frontal part of the western Subalpine foreland. These combined approaches on calcite have allowed 928 to constrain the timing of the fold and thrust belt propagation, which occurred in an 'in-sequence' 929 mode between 15 Ma and 7 Ma. This propagation is dominated by a phase of thick-skin tectonics, 930 highlighted by the activation of central Vercors and eastern Chartreuse thrusts at 12 Ma. This 931 activation is correlated to the acceleration of Belledonne ECM exhumation, and it follows, based on 932 structural relationships, that the foreland thrusts were rooted in a crustal-scale ramp below the 933 ECM. The reorientation of the stress field from NW-SE to E-W compression since 12 Ma is 934 ascribed to the N-S orientation of the crustal anisotropies, which originate from the Variscan, 935 Tethyan, and early Alpine tectonic stages. In this work, the U-Pb dating of preserved Oligocene 936 calcites at 32 Ma and ~24 Ma agree with the reactivation of N-S faults within the Subalpine 937 massifs. After 10 Ma, and until 7 Ma, the most frontal (western) thrusts have been activated due to 938 the propagation of a décollement on the evaporites of the basement – cover interface. This phase 939 induced a differential motion of the Vercors with respect to the (evaporites-poor) Chartreuse 940 massifs, which explains the development of cross-cutting dextral strike-slip faults in the latter 941 massif. Finally, a further out-of-sequence reactivation is recorded from 6 Ma to 2 Ma, and is still 942 ongoing from the seismic records, that is related to a more regional transition probably related to 943 Apulian rotation. In this well-constrained tectonic context, stable isotope analysis brings some 944 complementary information about the origin of fluids and the fluid-rock interaction history, 945 providing independent constraints on the depth and connectivity of the fault systems. Most of the successfully dated calcites show a depleted  $\delta^{18}O_{calcite}$  signature compared to their host-rock, which 946 947 indicates an external fluid source, explaining a slight enrichment in U. This fluid source, 948 recalculated with clumped isotopes, is similar to surface meteoric fluids that interacted with the 949 host-rock during deformation. It is also observed that most successful dates are obtained in a blocky 950 calcite, which results from higher fluid/rock ratios, while the fibrous calcite exhibits lower U 951 contents and is isotopically close to the host-rock. Clumped isotope analysis highlights a strong 952 temperature gradient from west (~50°C) to east ( $\geq$ 150°C), in agreement with vitrinite and RSCM data, coherent with an increase of burial from 2 to 4-6 km. This burial is related to an increase of theunderthrusting below the Alpine frontal thrusts.

955

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