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1 Tectono-geomorphological evolution of the Eastern

2 Pyrenees: insights from thermo-kinematic modeling

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17 ABSTRACT

Constraining the tectono-geomorphological evolution of the Pyrenees is still a major challenge, especially in the Eastern Pyrenees where late Neogene exhumation history and topographic evolution appear contrasted and have been debated. In this study, we performed thermo-kinematic (Pecube) modeling using a relatively dense spatial compilation of previously-published low-temperature thermochronological data for the

23 Eastern Pyrenees, and quantified the late-Neogene exhumation history based on different 24 topographic evolution models reconstructed from literature constraints. Our modeling 25 outcomes suggest a major decrease in the regional exhumation rate between 37-35 Ma, 26 associated to an early-end contractional tectonics for the Eastern Pyrenees. Two main 27 periods of the Têt fault normal activity have been characterized, with the existence of 28 pluri-kilometric (2-3 km) post- Oligocene-Miocene displacement along the western 29 segment of the Têt fault. The different topographic evolution models show relatively 30 similar results for the regional exhumation since ca. 35 Ma and for the most recent (mid 31 to late-Miocene) tectonic event recorded along the Têt fault. Conversely, inversion results 32 obtained for pre-35 Ma regional exhumation rates, and more importantly for the 33 Oligocene-Miocene activity of the Têt fault appear contrasted between the investigated 34 topographic scenarios. Overall, this study confirmed a mid to late Miocene significant 35 extensional tectonic event for the Eastern Pyrenees, which may have had a non-negligible 36 role in late-stage relief evolution for this part of the orogen. However, deciphering the 37 long-term evolution of topographic relief through thermo-kinematic modeling appears 38 still challenging for slowly-exhuming mountain ranges given the limited resolution of 39 low-temperature thermochronological data to topographic changes.

40

41 KEYWORDS

42 Eastern Pyrenees, Low-temperature thermochronology, Tectonics, Thermo-kinematic

43 models, Topographic evolution, Exhumation

44 **1. INTRODUCTION**

45 The Earth's surface evolution is controlled by the combination of tectonic, erosion and climate processes (e.g. Beaumont 1981; Willett, 1999; Dadson et al., 2003; Braun, 46 47 2010; Herman et al., 2013; Jamieson and Beaumont, 2013; Thieulot et al., 2014). The 48 existence of feedbacks amongst these mechanisms (Whipple, 2009) constitutes one of the 49 major difficulties to deconvolute the impact of each individual forcing on landscape 50 dynamics (Braun, 2012). In the upper crust, isotherm shapes and wavelengths are 51 especially controlled by the overlying topography and its evolution (Stüwe et al., 1994; 52 Mancktelow and Grasemann, 1997; Braun, 2002; Foeken et al., 2007; Valla et al., 2010), 53 therefore the upper-crust thermal evolution is influenced by rock exhumation and can be 54 impacted by (i) major topographic changes or (ii) fault motion and associated isotherm 55 advection (Batt and Braun, 1997; Ehlers and Chapman, 1999; Ehlers and Farley, 2003). 56 Low-temperature (low-T) thermochronology coupled with 3D thermo-kinematic 57 modeling provides a quantitative way to assess the crustal long-term thermal evolution 58 and has the main advantage to investigate the respective contributions of the regional 59 topographic evolution and/or the tectonic component(s) in rock exhumation (e.g. Farley, 60 2002; Ehlers and Farley, 2003; Glotzbach et al., 2011; Braun et al., 2012).

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Thermo-kinematic 3D modeling based on low-T thermochronology data has been widely used in active mountain ranges to constrain the topographic evolution and faultactivity history, especially in mountain ranges with rapid changes in either topographic relief or tectonic forcing like for instance the Himalayas (e.g. Coutand et al., 2014; Zhang et al., 2015; Wang et al., 2016; Grujic et al., 2020; Pitard et al., 2021; Shen et al., 2022), 67 the Andean Cordillera (e.g. Schildgen et al., 2009, 2022; Avdievitch et al., 2018; 68 Bermùdez et al., 2021; Gérard et al., 2021; Zapata et al., 2021) or the European Alps (e.g. 69 Vernon et al., 2009; van der Beek et al., 2010; Valla et al., 2011, 2012; Beucher et al., 70 2012). However, only few 3D thermo-kinematic modeling studies have been devoted to 71 the evolution of mountain ranges with limited or slow long-term topographic relief and 72 tectonic evolution like the Pyrenees (i.e. Gibson et al., 2007; Fillon and van der Beek, 73 2012; Erdös et al., 2014; Curry et al., 2021; Capaldi et al., 2022). This approach is 74 particularly challenging in mountain ranges formed by long tectonic histories with 75 multiple phases since (1) overall exhumation rates can be relatively low between the 76 different tectonic phases (<0.1 km/Ma; e.g. Braun and Robert, 2005), and (2) the 77 topographic and tectonic settings and their long-term evolutions are complex. Therefore, 78 it remains difficult to disentangle these two components in rock exhumation through low-79 T thermochronology in slowly-exhuming mountain range settings.

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81 In the Pyrenees, regional 3D thermo-kinematic modeling has been focused on the 82 central (Gibson et al., 2007; Erdos et al., 2014; Capaldi et al., 2022) and southern (Fillon 83 and van der Beek, 2012) parts of the orogen. Recent study of Curry et al. (2021) has 84 provided new quantitative constraints for the western and central part of the Axial Zone 85 to evaluate thrust sheet-controlled exhumation as well as along-strike exhumation 86 differences in the Pyrenees. Moreover, these previous studies have been particularly 87 focused on the Paleogene Pyrenean contractional phase. The Eastern Pyrenees have a 88 specific evolution, reflected by a complex tectonic history with an Eocene contractional 89 phase (Laumonier, 2015; Ternois et al., 2019; Calvet et al., 2021; Ford et al., 2022)

90 followed by 2 significant post-orogenic periods: i) a transtensionnal phase associated 91 with the opening of the West European Rift (Angrand and Mouthereau, 2021; Milesi et 92 al., 2022), and ii) an Oligocene-Miocene extensional phase associated to the Gulf of Lion 93 opening (Calvet et al., 2021; Jolivet et al.; 2020, 2021; Romagny et al., 2020; Séranne et 94 al., 2021; Taillefer et al., 2021; Milesi et al., 2022). The Têt normal fault is the most 95 prominent tectonic structure in the Eastern Pyrenees, and localizes high-relief massifs in 96 such as the Mont-Louis (hanging wall), and the Canigou and Caranca (footwall) massifs. 97 The development of these high topographic reliefs has been related to normal faulting 98 during the Oligocene-Miocene period (Maurel et al., 2008, Milesi et al., 2022). However, 99 the topographic evolution of the area is still highly debated (e.g. Augusti et al., 2006; 100 Gunnell et al., 2008, 2009; Suc and Fauquette, 2012; Calvet et al., 2014; Huyghe et al., 101 2018, 2020). The potential links between the topographic and tectonic evolutions of the 102 Eastern Pyrenees are also not entirely clear due to their location between the 103 contractional Pyrenean domain and the extensional area of the Gulf of Lion. Finally, the 104 long-term topographic evolution of the Eastern Pyrenees (Huyghe et al., 2012, 2020) 105 appears to differ significantly from the topographic evolution model proposed for the 106 western and central parts of the Pyrenees (Curry et al., 2019; 2021).

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In this study, we present the first 3D thermo-kinematic modeling outcomes for the Eastern Pyrenes, based on a large synthesis of previously-published (U-Th)/He on apatite (AHe) and zircon (ZHe), and apatite fission-track (AFT) data (Gunnell et al., 2009; Maurel et al., 2002, 2008; Milesi, et al., 2019, 2020a, 2020b, 2022). We also investigate 3 different models for the topographic evolution of the Eastern Pyrenees, with the objective

to integrate topographic evolution in 3D thermo-kinematic modeling and quantification of rock exhumation. By independently prescribing the long-term topographic evolution, we aim to provide constraints on the timing and rates of the Cenozoic to modern regional rock uplift as well as on the tectonic deformation along the Têt fault. Based on these results, we discuss the onset and timing of the contractional tectonic record in the Mont-Louis massif, and the late-Cenozoic extensional period(s) along the Têt fault in the Canigou and Carança massifs.

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2. GEOLOGICAL SETTING

2.1 Topographic and tectonic evolution of the Pyrenees

123 In the Pyrenees, collision and orogen building occurred from the Late Cretaceous 124 to the Middle Miocene (e.g. Roure et al., 1989; Muñoz, 1992; Vergés et al., 2002; 125 Mouthereau et al., 2014; Ford et al., 2022), and formed a double-wedged mountain range. 126 The Axial Zone (Fig. 1) is composed of a stack of crustal nappes formed during the main 127 Eocene-early Oligocene orogenic build up and linked to the maximum of shortening and 128 exhumation (Sinclair et al., 2005; Bosch et al., 2016a; Jolivet et al., 2007; Labaume et al., 129 2016; Mouthereau et al., 2014; Vacherat et al., 2016; Waldner et al., 2021). During the 130 Lutetian (lower Eocene), the paleo-elevation of the Pyrenees has been estimated to 131 around 2 ± 0.5 km (Huyghe et al., 2012), which is associated with the main phase of the 132 Pyrenean orogen starting around 50 Ma (Fitzgerald et al., 1999; Sinclair et al., 2005; 133 Vacherat et al., 2014, 2016; Ternois et al., 2019, 2021). Crustal flexural models (Curry et 134 al., 2019) suggest topographic growth from 56 to 23 Ma and a sharp increase in topography at 38 Ma that is in contradiction with the more progressive topographic 135

growth deduced from stable isotope data (Huyghe et al., 2012). Flexural models showed a
minor decay of overall Pyrenean topography from 23 Ma to present-day. On the contrary,
the topographic model for the Eastern Pyrenees deduced from stable isotope analyses
(mammal teeth from the Cerdanya basin; Huyghe et al., 2020) suggests a slow relief
development since the Miocene (Fig. 2).

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142 The exhumation history of the Pyrenees is marked by a southward propagation of 143 the orogenic-wedge deformation (Fitzgerald et al., 1999; Sinclair et al., 2005; 144 Mouthereau et al., 2014). The Pyrenean orogen has developed diachronously, from East 145 to West due to spatially-variable pre-orogenic crustal extension (e.g., Puigdefäbregas et 146 al., 1992; Whitchurch et al., 2011), some authors also proposed a role of major base-level 147 changes on the timing and rates of post-orogenic exhumation (Conev et al., 1996; Fillon 148 and van der Beek, 2012). Recent thermo-kinematic study from Curry et al. (2021) has 149 focused on the western and central part of the Pyrenees (Fig. 1) and, based on a large 150 compilation of low-T thermochronological data, their outcomes confirm a first-order 151 westward propagation of rock uplift and exhumation since the Eocene. This observed 152 propagation of tectonic deformation and exhumation may be explained by the inherited 153 crustal configuration rather than differential onset of shortening along strike (Teixell et 154 al., 2018; Muñoz, 2019). On the other hand, southward propagation of the deformation (Fitzgerald et al., 1999; Sinclair et al., 2005; Metcalf et al., 2009) has been confirmed by 155 156 low-T thermochronology in the central part of the Pyrenees, while only geological 157 evidences support southward propagation of the deformation in the Western Pyrenees 158 (Teixell et al., 2016).

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160 The post-orogenic tectonic evolution in the western and central Axial Zone has 161 not been well constrained. AHe ages from middle to late Miocene (15-6 Ma) are 162 interpreted in many ways, such as: (i) the result of a distinct tectonic event (Jolivet et al., 163 2007; Bosch et al., 2016a; Fillon et al., 2021), (ii) the erosional response to major 164 drainage changes with the connection of the foreland Ebro Basin to the Mediterranean 165 Sea (Coney et al., 1996; Fitzgerald et al., 1999; Fillon and van der Beek, 2012), (iii) 166 large-scale erosion resulting from Pliocene climate changes, as proposed for the 167 European Alps (Cederborn et al., 2004), or (iv) a lithological control on post-orogenic 168 erosion with lower erosion rates for Variscan metamorphic lithologies in the eastern part 169 of the Axial Zone compared to unmetamorphosed lithologies in the western part (Bernard 170 et al., 2019). A recent thermochronological study in the Ebro basin (South Pyrenean 171 Zone) evidenced also a potential long-wavelength tectonic event at ~10 Ma, linked to the 172 cessation of slab retreat in the Western Mediterranean domain (Rat et al., 2022).

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2.2 Topographic and tectonic evolution of the Eastern Pyrenees

The eastern part of the Axial Zone is characterized by the emplacement of southverging nappes rooted in the northern part (Laumonier et al., 2015; Sibuet et al., 2004; Vergès et al., 1995; Teixell et al., 2016). Balanced cross-sections (Ternois et al., 2019) suggest an Eocene thrusting of the Aspres-Mont-Louis massifs onto the Canigou massif as already proposed by Maurel et al., (2008) based on low-T thermochronology. In addition, the eastern part of the Pyrenees (east of $\sim 1^{\circ}$ E) underwent post-orogenic crustal thinning (Chevrot et al., 2018; Diaz et al., 2018). This crustal-thinning phase has been

182 explained by regional extensional episodes associated to the (re-)activation of major 183 orogenic structures as normal faults in response to the Gulf of Lion opening from the end 184 of the Priabonian to the Quaternary (e.g. Calvet et al., 2021; Taillefer et al., 2021, Milesi 185 et al., 2022). In the study area (Fig. 1b), two main crustal tectonic structures are 186 identified: the Têt and Py faults. The most prominent tectonic structure, the Têt fault, is a 187 crustal-scale NE-SW fault with pluri-kilometric (~3 km) displacement associated to the 188 main opening stage (25-20 Ma) of the Gulf of Lion during the Oligocene-Miocene 189 (Maurel et al., 2008; Milesi et al., 2022). The Py fault delimits the Canigou and Caranca 190 blocks in the footwall of the Têt fault (Fig. 1b), and recent thermochronological data 191 suggest a late Priabonian to early Rupelian (35-32 Ma) activity of this fault associated to 192 at least 2 km displacement (Milesi et al., 2022).

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194 A major debate exists on the geomorphological evolution of the Eastern Pyrenees, 195 related to the formation timing and relative elevation of Oligocene-early Miocene high-196 elevation low-relief surfaces (Bosch et al., 2016b; Monod et al., 2016; Calvet et al., 197 2021). Some authors proposed the formation of these low-relief surfaces at low 198 elevations, followed by a major uplift event ($\sim 2 \text{ km}$) since the early Miocene (Gunnell et 199 al., 2008; 2009). Another model proposed the development of these low-relief surfaces at 200 high elevations due to the smoothing of the relief before the Late Miocene, associated to 201 a limited surface uplift of around 400 m since then (Babault et al. 2005; Bosch et al., 202 2016b). Rock exhumation associated to late-Miocene normal faulting is considered to 203 have played a subordinate role, although significant exhumation events have been 204 proposed by (i) NW-SE fault activity (Taillefer et al., 2021) and (ii) thermal modeling in the Carança massif, with predicted rock cooling between 12 to 9 Ma, interpreted as the
normal motion of the western segment of the Têt fault (Milesi et al., 2022).

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208 A paleo-elevation reconstruction for the whole Eastern Pyrenees, based on stable-209 isotope data coupled with regional geological constraints, has been proposed by Huyghe 210 et al. (2020). This model suggests that the topographic uplift in the Eastern Pyrenees is a 211 long-term process initiated in the Late Burdigalian (~20 Ma, Fig. 2) in response to the 212 pre-breakup uplift in the Gulf of Lion. The Burdigalian period is considered as the end of 213 the main rifting period for the western part of the Gulf of Lion. During this period, the 214 topographic conditions in the Eastern Pyrenees were proposed to be close to sea level 215 (Bache et al., 2010; Huyghe et al., 2020). The post-Burdigalian period in the Eastern 216 Pyrenees is still debated, with a proposed uplift of ~ 1 km since the Tortonian to 217 Messinian (i.e. since around 10 to 6 Ma) based on palynological constraints (Agusti et al., 218 2006; Suc and Fauquette, 2012). Stable-isotope data obtained from the analyses of small 219 mammal teeth (Huyghe et al., 2020) complete these palynological data and suggest an 220 uplift estimate of around 500 m since the Messinian (ca. 6 Ma). On the contrary, Tosal et 221 al. (2021) recently proposed, from the study of leaf taxa, that the Cerdanya Basin was 222 located at an elevation comprised between 1100 and 1500 m already during the 223 Tortonian, although this estimate may reflect local conditions and should be confirmed 224 by additional taxa analyses.

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- **3. THERMO-KINEMATIC MODELING**

3.1 Low-temperature thermochronology data in the Eastern Pyrenees

228 We focused our 3D thermo-kinematic modeling on the South Mérens, Caranca and 229 Canigou blocks. The North Mérens block is not included due to the sparce low-T 230 thermochronological record in this area (Milesi et al., 2022). We compiled AHe, AFT, 231 ZHe ages and mean track lengths (MTL) from previously-published datasets (Gunnell et 232 al., 2009; Maurel et al., 2002, 2008; Milesi, et al., 2019, 2020a, 2020b, 2022). For our 233 study, we used only samples issued from crustal basement that record long-term rock 234 exhumation and were not affected by recent geothermal anomalies or hydrothermal 235 circulations (41 samples in total, Fig. 1b). In the Têt fault footwall, samples have been 236 selected in the first 5-km distance to the fault, since the cooling history of farther samples 237 may have been potentially impacted by other local faults. Details of the selected samples 238 and associated low-T thermochronological ages are available in the Supplementary Table 239 S1.

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3.2 Modeling approach

Inverse modeling has been performed using the thermo-kinematic modeling code Pecube v4.0, based on a finite-element method to solve the heat transport equation in 3D (Braun, 2003; Braun et al., 2012), and coupled with the neighborhood algorithm (NA) inversion method (Sambridge 1999a, 1999b). The detailed model setup is fully described with all modeling parameters and our inversion approach in the Supplementary Information.

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We explored the 3 crustal blocks separately to constrain their respective rock exhumation patterns: the South Mérens, Carança and Canigou blocks. No activity on the

251 Têt fault has been defined when modeling the South Mérens exhumation history (hanging 252 wall of the Têt fault), only exhumation in response to regional uplift has been considered 253 (Milesi et al., 2022). We modeled a two-stage regional exhumation model based on low-254 T thermochronological age-elevation relationships (Milesi et al., 2022) using AHe, ZHe 255 AFT and MTL data (Supplementary Table S1). The model runs for 60 Myr, with a 256 transition event between the two exhumation phases called "transition time" and ranging 257 from 60 to 0 Ma. Each phase is associated to exhumation rates during the first (V1) and 258 second (V2) phases, which range from 0 to 2 km/Ma and from 0 to 1 km/Ma, 259 respectively. The output exhumation scenario obtained for the South Mérens block is thus 260 considered as the regional exhumation scenario and used for subsequent modeling.

261 The Carança and Canigou blocks have been then investigated with prescribing an 262 activity on the Têt fault. For the Canigou block, a single activity phase for the Têt fault 263 has been tested following the work of Milesi et al. (2022). This approach is based on low-264 T thermochronological age-elevation relationships, which suggest a simple exhumation 265 history for this area using AHe, ZHe AFT and MTL data. To further test this hypothesis, 266 we have run an exhumation scenario with 2 activity phases for the Têt fault, however 267 output results show poor inversion convergence for the tested parameters on the Têt fault 268 activity (see Supplementary Figure S1). In the single-phase scenario, the model runs for 269 60 Myr, with a regional background exhumation fixed by best-fitting output results 270 obtained in Section 4.1 (South Mérens block). The Têt fault activity is allowed between 271 35 and 5 Ma, with the onset time ranging from 35 to 25 Ma, and the ending time from 25 272 to 5 Ma. The fault slip rate ranges from 0 to 6 km/Ma.

273 For the Caranca block, two potentially distinct activity phases for the Têt fault 274 have been tested following the proposed thermal history of Milesi et al. (2022) using 275 AHe, ZHe AFT and MTL data. The model runs for 60 Myr, with a regional background 276 exhumation fixed by best-fitting output results obtained in Section 4.1 (South Mérens 277 block). The first fault-activity phase is allowed between 35 and 15 Ma, with the onset 278 time ranging from 35 to 25 Ma, and the ending time from 25 to 15 Ma. The fault slip rate 279 during this first phase ranges from 0 to 6 km/Ma. The second fault-activity phase is 280 allowed between 15 and 5 Ma, with the onset time ranging from 15 to 11 Ma, and the 281 ending time from 11 to 5 Ma. The fault slip rate during the second phase ranges from 0 to 282 4 km/Ma.

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284 Three main topographic evolution models (TEMs, Fig. 4) have been tested for the 285 different crustal blocks: (1) a model without topographic change and a constant 286 topography since 56 Ma which is similar to the present-day topography (Fig. 1b); (2) a 287 Global Pyrenees model based on the topographic outcomes from Curry et al. (2019); and 288 (3) an Eastern Pyrenees model (Fig. 2) based on the mean paleo-elevation curve proposed 289 by Huyghe et al. (2020) and the maximum paleo-elevations defined by palynological 290 constraints (Agusti et al., 2006; Suc and Fauquette, 2012). Due to the lack of continuous 291 record for the maximum paleo-elevations, a conservative hypothesis has been established 292 when no data is available. At 56 Ma, topographic parameters for the Eastern Pyrenees 293 have been inferred from the literature, lower mean relief and topography have thus been 294 implemented based on the lack of detrital material in adjacent basins, and the absence of 295 alluvial cone/delta at that time (Calvet et al., 2021). All the parameters considered are

available in the Supplementary Table S2, and the mean paleo-elevation curves for the
three models, together with resulting topographies and associated topographic
amplification factor/offset, are illustrated in Figure 4.

299

300 4. RESULTS

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4.1 Hanging wall of the Têt fault: the South Mérens block

Inversion results (Table 1) demonstrate that regardless of the considered TEM, the 1D probability density functions (PDFs) show no statistical convergence for the output exhumation rates V1. However, we can note that all output values are above 0.2 km/Ma and that a maximum of probability is reached for exhumation rates between 0.3 and 0.6 km/Ma for the Eastern Pyrenees TEM (Fig. 5). For the Global Pyrenees and No topographic change TEMs, V1 is highly variable with a more pronounced probability towards high exhumation rates (>0.5 km/Ma, Fig. 5).

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The output transition time is well constrained and concordant for all three TEMs. Inversion results for the transition time show a continuous probability distribution (40-30 Ma range), with a maximum probability between 37 and 35 Ma (Fig. 5).

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The exhumation rate V2 is also well constrained for all three TEMs and varies only slightly between them. The output PDFs for V2 are close to a Gaussian distribution, with exhumation rates of 0.04 ± 0.03 km/Ma, 0.06 ± 0.01 km/Ma and 0.09 ± 0.03 km/Ma for the No topographic change, the Global Pyrenees and Eastern Pyrenees TEMs, respectively. Overall, the regional exhumation scenario for the different TEMs is very
well constrained between ~35 Ma and present day.

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321 The best-fitting parameters obtained for each TEM have subsequently been used 322 in a forward Pecube model to predict low-T thermochronological ages and time-323 temperature paths (Fig. 6). Low-T thermochronological ages modeled for the South 324 Mérens block are close to observed data, except for ZHe ages that are underestimated for 325 all considered TEMs (Fig. 6a). Modeled AHe ages with the Eastern Pyrenees TEM are 326 slightly younger for the top samples compared to the two other TEMs. Between 1600 and 327 2100 m, AHe ages are not well reproduced for all TEMs. However, it must be noted that 328 the observed AHe ages are older than AFT ages obtained at similar elevations. The 329 general trend of the AFT elevation profile is generally well reproduced with slightly older 330 modeled than observed ages. Predicted MTLs are relatively consistent between the used 331 TEMs, but overall overestimated the observed data except for mid-elevation samples 332 $(\sim 1600 \text{ m elevation}, \text{Fig. 6a}).$

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Best-fitting predicted thermal histories (Fig. 6b) show similar overall trends, with rapid cooling until ~35 Ma followed up by slow cooling until present day. Some differences are observed between the considered TEMs: (i) before 35 Ma, the output cooling rates are close to 17, 14 and 23 °C/Ma for the No topographic change, Eastern Pyrenees and Global Pyrenees TEMs, respectively. Output cooling rates for this period must be considered carefully when considering the large output PDFs and thus high uncertainties obtained for V1 (Fig. 5d), with the better statistical estimate being given by

the Eastern Pyrenees TEM. (ii) Cooling rates after 35 Ma are more impacted by the choice of the TEM. For the No topographic change TEM, the output cooling rate is constant (~1 °C/Ma). The Global Pyrenees TEM yields a very low cooling rate between 343 35 and 23 Ma (~0.3 °C/Ma) which increased towards present day (1 °C/Ma). Finally, the Eastern Pyrenees TEM yields a gradual decrease in cooling rate from 2.8 °C/Ma between 346 34 and 23 Ma to around 0.7 °C/Ma since 23 Ma.

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4.2 Eastern footwall of the Têt fault: the Canigou block

349 Inversion results for the Têt fault activity (Table 1) slightly differ according to the 350 used TEMs. For the No topographic change and Global Pyrenees TEMs, the Têt fault 351 activity is limited in time with maximum probabilities from 26-25 to 24.5 Ma (Fig. 7a) 352 and from 28-26 to 24.5 Ma (Fig. 7b), respectively. Inversions based on the Eastern 353 Pyrenees TEM do not well constrain the fault-activity onset time but suggest a longer 354 activity period between >28 and 23.5 ± 1.5 Ma. These output differences are also marked 355 for the output fault slip rates (Fig. 7d). This parameter is not well constrained for the No 356 topographic change and Global Pyrenees TEMs, all inversions yielding high slip rates 357 above 1.0 and 1.5 km/Ma, respectively. In contrast, inversions with the Eastern Pyrenees 358 TEM show a maximum probability for output slip rate at 1 ± 0.5 km/Ma, significantly lower than for other TEMs (Fig. 7d). 359

360

Predicted AHe ages are close for the No topographic change and Eastern Pyrenees
 TEMs (Fig. 8a), and consistent with observed ages for elevation >1400 m. AHe ages
 predicted with the Global Pyrenees TEM are younger than for the two other TEMs and

364 also compared to observed ages for elevation >1800 m. Observed AFT ages for top 365 samples (>2000 m) are well reproduced by numerical inversions with all the considered 366 TEMs (Fig. 8b). Only inversions with the Global Pyrenees TEM show predicted AFT 367 ages in agreement with observed ages for elevation <1600 m. Predicted MTLs differ 368 between the considered TEMs and samples: for most samples, the Global Pyrenees and 369 Eastern Pyrenees TEMs provided similar MTL predictions except for the low-elevation 370 sample for which predicted MTL differ for the Global Pyrenees TEM. The No 371 topographic change TEM predicted relatively short MTLs (<14 µm) and close to 372 observed data for most of samples, while the Global Pyrenees TEM produced shorter 373 MTL and close to observations for the low-elevation sample (Fig. 8a). The only ZHe age 374 in the Canigou block cannot be properly reproduced by numerical inversions whatever 375 the considered TEM, which all predict too young ZHe ages (Fig. 8a).

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Best-fitting output thermal histories (Fig. 8b) show differences essentially due to the contrasted timing and slip rate of the Têt fault activity (Fig. 7d). At around 25 Ma, high cooling rates (~130 °C/Ma) are observed for the No topographic change TEM, while slower cooling is predicted for the Global Pyrenees and Eastern Pyrenees TEMs (around 70 and 40°C/Ma, respectively). Posterior to this fault-activity phase, predicted cooling rates are lower and consistent between all TEMs (2-4 °C/Ma, Fig. 7d).

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4.3 Western footwall of the Têt fault: the Carança block

Inversion results for the first fault-activity phase (Table 1) differ according to the used TEM (Fig. 9). For the No topographic change TEM (Fig. 9a), the first phase is

387 similar but slightly shorter compared to the Canigou block, i.e. from 26-25 to 25-24 Ma, 388 and associated with a high slip rate (maximum of probability at 5.5-6 km/Ma). The 389 Global Pyrenees TEM (Fig. 9b) suggests a longer fault activity between 28 ± 2 and 390 around 25-24 Ma. The output slip rate is not well constrained. For inversions based on 391 these 2 TEMs (No topographic change and Global Pyrenees), the first phase of the 392 Carança block appears similar to the predicted fault activity of the Canigou block. 393 Inversions based on the Eastern Pyrenees TEM suggest a longer fault activity, with a 394 maximum probability for the onset time at 32 ± 1 Ma and ending time at around 15 Ma. 395 For this TEM, the end of first fault-activity phase is thus suggested later than for the 396 Canigou block. Inversions based on the Eastern Pyrenees TEM yield a well-constrained 397 slip rate of <1 km/Ma that is lower than for the other TEMs (Fig. 9d) but consistent with 398 output slip rate for the Canigou block (Fig. 7d).

399

The inversion outcomes for the second fault-activity phase (Table 1 and Fig. 10) are similar whatever the considered TEM. Inversion results show a maximum probability for the fault-activity duration between around 15 and 11 Ma. The PDFs for the slip rate are close to Gaussian distributions, with a maximum probability in the range of 0.7-0.8 km/Ma (Fig. 10d).

405

Predicted AHe ages are close for the 3 considered TEMs but they overall underestimate the observed AHe ages. Output AHe ages are rather consistent with observed low-elevation ages (1050-1250m, Fig. 11a). Predicted AFT ages are consistent or very close to the observed ages for all considered TEMs. Predicted ZHe ages are

410 different according to the used TEMs, being increasingly younger for inversions based on 411 the Global Pyrenees, Eastern Pyrenees and No topographic change TEMs, respectively. 412 Modeled ZHe ages with the Global Pyrenees TEM are close to observed ages for the base 413 of the elevation profile (900-1000 m) whereas predictions with the No topographic 414 change TEM are more coherent with observed ages (1200-1400 m). Predicted MTLs are 415 well reproducing observed data for the different TEMs (Fig. 11a). All TEM models fail to 416 explain observed ZHe ages (>1600 m), and predict rather young ZHe ages compared to 417 observations (Fig. 11a).

418

419 Best-fitting output thermal histories (Fig. 11b) show differences between TEMs 420 that are essentially due to the contrasted duration and intensity of the first fault-activity 421 phase (Fig. 9d). The main differences are similar to those obtained for the Canigou 422 inversions (Section 4.2). For the first phase, high cooling rates are observed for the No 423 topographic change (\sim 140 °C/Ma) and Global Pyrenees TEMs (\sim 70 °C/Ma), while 424 slower cooling is predicted for the Eastern Pyrenees TEM (~10 °C/Ma). This first cooling 425 phase is followed by a decrease in cooling rate regardless of the TEM (<5 °C/Ma). 426 Output thermal histories show a similar pattern between 15 Ma and present day between 427 the three TEMs: output cooling at ~20 °C/Ma associated to the second fault-activity 428 phase between 15 and 11 Ma, followed by slow cooling (1-2° C/Ma, Fig. 11b).

429

430 **5. DISCUSSION**

431 **5.1 Pyrenean contractional stage**

432 Our inversion results allow reconstructing a two-stage regional exhumation 433 history for the South Mérens block (hanging wall of the Têt faut, Fig. 1), with a well-434 constrained transition time between 37 and 35 Ma regardless of the considered TEM 435 (Fig. 5). Exhumation rates V1 are not well constrained, with a probability peak between 436 0.3 and 0.6 km/Ma for the Eastern Pyrenees TEM and >0.5 km/Ma for the global 437 Pyrenees and No topographic change TEMs. Exhumation rates V2 appear significantly 438 lower and are well constrained (close to Gaussian distributions) below 0.1 km/Ma for all 439 the TEMs. These results overall agree with thermal histories obtained with OTOt 440 modeling, showing rapid cooling between 40 and 35 Ma, followed by slow cooling since 441 35 Ma (Milesi et al., 2022, Fig. 12). The regional exhumation scenario for the different 442 TEMs is also very well constrained between \sim 35 Ma and present day, which is the main 443 period of interest for this study. More complex exhumation scenarios (e.g. considering a 444 three-phase exhumation history) have also been tested, without significant improvement 445 regarding low-T thermochronological age predictions (Milesi, 2020).

446 Best-fitting inversion models showed similar thermal-history predictions for the 3 447 TEMs, only slightly influenced by the topographic evolution after 35 Ma (Fig. 6). 448 Cooling rates associated to the first exhumation phase are between 15 and 20°C/Ma. 449 consistent at first order with QTQt outcomes. However, QTQt results (Milesi et al., 2022) 450 suggested an apparent two-stage cooling history before 35 Ma: a first phase between 50 451 and 38 Ma ($>5^{\circ}C/Ma$) followed by a second phase between 38 and 35 Ma ($\sim30^{\circ}C/Ma$). 452 This two-stage history is not constrained in Pecube-derived thermal histories (Fig. 12), 453 which can be explained by both the relative simplicity of our implemented Pecube 454 exhumation scenarios (i.e. only two exhumation phases considered) but also by the lack

of low-T thermochronological data for the 50-38 Ma period (Fig. 6 and Table S1).
Indeed, most of ZHe and AFT data from high-elevation samples are not well reproduced
whatever the considered TEMs (Fig. 6).

458

459 Predicted cooling rates associated to the second exhumation phase showed slight 460 differences between the considered TEMs, but are overall quite low (up to 1°C/Ma, 461 except for the Eastern Pyrenees TEM with 2.8 °C/Ma between 34 and 23 Ma, Fig. 6). 462 Observed AHe and AFT data are well reproduced by our best-fitting Pecube models, 463 except for samples characterized by ages inversion (i.e. AHe ages are older than AFT 464 ages). Such an ages inversion cannot be reproduced by Pecube due to the relatively 465 simple AHe diffusion model used in this code (Braun et al., 2012), since for example it 466 does not consider apatite chemistry nor size effects on He diffusion (see Section 3.2; e.g. 467 Gautheron et al., 2009).

468

469 Predicted regional exhumation rates prior to ca. 35 Ma are all above 0.2 km/Ma, 470 with PDF peaks for rates >0.3 km/Ma from our inversion results (Figs. 5 and 12) that can 471 be related to the activity of major regional thrusts located further South of the Mérens 472 block (Fig. 1a), like the Cadi-Canigou thrust (Ternois et al., 2019). Regional thrusting in 473 the Eastern Pyrenees has been linked to the contractional phase of the orogen, associated 474 to the convergence between the Iberian and European plates during the Eocene 475 (Fitzgerald et al., 1999; Whitchurch et al., 2011; Rushlow et al., 2013; Mouthereau et al., 476 2014; Bosch et al., 2016a; Labaume et al., 2016; Cruset el al., 2020). The end of this 477 contractional phase is well constrained in our inversion results between 37 and 35 Ma,

478	followed by a major decrease of exhumation rates (<0.1 km/Ma) for the eastern part of
479	the Axial Zone (Fig. 1), which contrasts with faster exhumation rates at 1.0 ± 0.1 km/Ma
480	proposed further West between 40 and 30 Ma (Curry et al., 2021). These exhumation
481	differences can first be explained by our model set-up, for which the transition time is an
482	inversion parameter and not constrained by an a priori prescribed time window (see
483	Section 4.1) as in Curry et al. (2021). In addition, we propose that this temporal
484	difference may reflect a first-order westward propagation of rock uplift and exhumation
485	starting in the eastern part of the Pyrenees (Whitchurch et al., 2011; Fillon et al., 2016;
486	Curry et al., 2021). The observed early end of the contractional event corresponds also to
487	the proposed period for the onset of extensional tectonics in the Eastern Pyrenees,
488	associated with the large-scale West European Rifting (Ziegler, 1992; Romagny et al.,
489	2020; Angrand and Mouthereau, 2021; Jolivet et al., 2021; Mouthereau et al., 2021;
490	Séranne et al., 2021; Milesi et al., 2022).

- 491
- 492
- 493 **5.2** Têt fault activity
- 494

5.2.1 The Canigou block

Inversion results for the Têt fault activity differ according to the used TEM, but overall our model outcomes reveal that the fault-activity duration is not well constrained (Fig. 7). For the No topographic change and Global Pyrenees TEMs, the Têt fault activity appears shorter in time (from 26-25 to 24.5 Ma and 28-26 to 24.5 Ma, respectively) compared to the Eastern Pyrenees TEM (between >28 Ma and 23.5 \pm 1.5 Ma). Consistently, predicted fault slip rates are also not well constrained for the different

501 TEMs (Figure 7e) but they appear relatively high and unconstrained for the No 502 topographic change (1.0-6.0 km/Ma) and Global Pyrenees (1.5-6.0 km/Ma) TEMs, while 503 they seem slightly lower and more constrained for the Eastern Pyrenees TEM (1 ± 0.5) 504 km/Ma). Output slip rates can be translated into vertical rock uplift/exhumation rates 505 using the implemented fault dip in Pecube models, with the following estimates: >0.85506 km/Ma, >1.3 km/Ma and 0.85 ± 0.40 km/Ma for the No topographic change, Global 507 Pyrenees and Eastern Pyrenees TEMs, respectively. All these estimates are well higher 508 than output exhumation rates of 0.33 to 0.56 km/Ma derived from age-elevation 509 relationships for the same period (Milesi et al., 2022). However, our estimated 510 exhumation rates associated to the Têt fault activity are in the same order of magnitude 511 than exhumation rates obtained by thermochronological studies along other normal faults 512 worldwide (e.g. Fletcher et al., 2000; Ehlers et al., 2001). These differences of 513 exhumation rates can be explained by the simple diffusion models used in Pecube code 514 and modeling assumptions (see Section 3.2).

515

Best-fitting inversion models show a good agreement between model predictions and 516 517 observed low-T thermochronological ages (Fig. 8), with noticeable differences between 518 the No topographic change/Eastern Pyrenees TEMs and the Global Pyrenees TEM. The 519 main differences are illustrated by AHe and low-elevation AFT ages predicted for the 520 Global Pyrenees TEM, which are younger than for the two other TEMs. These 521 differences are also visible in output thermal histories, with a more pronounced cooling 522 event (~120 °C/Ma) for the Global Pyrenees TEM than for the two other TEMs (~90 523 °C/Ma). In contrast with the QTQt output Oligocene-Miocene cooling rates (~30 °C/Ma,

524 Fig. 12), Pecube thermal histories reveal faster cooling during that period. It is 525 particularly visible for Global Pyrenees and Eastern Pyrenees TEMs (Fig. 8b), with 526 prescribed Oligocene-Miocene cooling from 350°C to 120°C and 260°C to 30°C, 527 respectively. These important differences can be due to the potential combination of 2 528 independent cooling phases in our Pecube approach: a Priabonian cooling phase, 529 followed by a second Oligocene-Miocene phase, which are predicted for QTQt modeling 530 (Fig. 12, Milesi et al., 2022). Furthermore, these output thermal histories are reflecting 531 the complexity to properly model the observed ZHe age for the Canigou block (Fig. 8a) 532 and the MTL data from top-profile samples of the Canigou block (Fig. 8b). Performed 533 inversion models with 2 fault-activity periods tend to refine fault slip rates during the 534 Oligocene-Miocene phase rather than highlighting a Priabonian tectonic event, most certainly due to the lack of data for this period (Supplementary Table S1 and Figure S1). 535 536 It is important to note that with the Eastern Pyrenees TEM, the output fault activity is 537 starting earlier (\sim 28 Ma) which is closer to the observed ZHe age. Additional ZHe and 538 ZFT data in the Canigou area would allow to better constrain this early tectonic phase. 539 Considering our inversion results and the relatively good AHe/AFT model predictions, no 540 post Oligocene-Miocene activity of the Têt fault is required to explain low-T 541 thermochronological data in the Canigou block.

542

543

5.2.2 The Carança block

Inversion results for the first fault-activity phase differ according to the considered TEM (Fig. 12). For the No topographic change TEM, this first phase appears limited in time (from 26-25 Ma to 25-24 Ma). The Global Pyrenees TEM suggests a 547 slightly earlier onset of fault activity (28 ± 2 Ma) but close ending time (25-24 Ma), 548 similarly to the Canigou block (Section 5.2.1). Inversions based on the Eastern Pyrenees 549 TEM suggest a longer fault activity, between around 32 Ma and 15 Ma. Estimated faut 550 slip rates are high (5.5-6 km/Ma) for the No topographic change TEM, these are much 551 lower for the Eastern Pyrenees TEM (<1 km/Ma) and not well constrained for the Global 552 Pyrenees model (Fig. 10d). These differences in quantitatively constraining fault slip 553 rates can be explained by the major differences in the low-T thermochronological dataset 554 for the Carança block compared to the Canigou block, with a complete ZHe age-elevation 555 profile and low-elevation AHe ages that record exhumation for the mid to late Miocene 556 period (Fig. 11a). Output Oligocene-Miocene cooling rates (Fig. 11) are relatively high 557 (>100 °C/Ma) for the No topographic change and Global Pyrenees TEMs, similar to the 558 Canigou block (Fig. 9), while slower cooling is predicted for the Eastern Pyrenees TEM 559 (~10 °C/Ma). However, the output cooling rate for the Global Pyrenees TEM has to be 560 considered cautiously and may not be representative due to the largely unconstrained 561 fault slip rate with this TEM (Fig. 10d). Finally, our results suggest the existence of 562 diachronism and magnitude differences for the Têt fault activity along its western 563 segment (i.e. Canigou vs. Carança block), confirming previous observations with QTQt 564 models (Fig. 12; Milesi et al., 2022).

565

Predictions for the second fault-activity phase are relatively similar whatever the considered TEM. Inversion results for the fault duration show a maximum probability between around 15 and 11 Ma, with an associated fault slip rate of 0.7-0.8 km/Ma (Fig. 10). These results indicate the existence of a major post Oligocene-Miocene activity of

570 the Têt fault along the Caranca block, with associated rock exhumation (estimated using 571 slip-rate predictions and Têt-fault dip) above 0.6 km/Ma that is 5-10 times higher than the 572 regional exhumation rate recorded in the South Mérens block (see Section 5.1) over the 573 same period. Independently of the considered TEM, the low-T thermochronological 574 dataset for the Carança block cannot be only explained by large-scale change(s) in 575 topographic relief (Gunnell et al., 2008; Huyghe et al., 2020; Calvet et al., 2021). Output 576 cooling at about 20 °C/Ma is associated with this late-stage tectonic exhumation phase 577 between 15 and 11 Ma, and is consistent to first order with the previously recognized 578 cooling phase, but of lower magnitude (around 10 °C/Ma) and duration (between 11 and 579 9 Ma), in QTQt thermal models (Milesi et al., 2022). Observed differences between the 580 two modeling approaches (Pecube vs. QTQt) can be explained by (i) the relatively simple 581 He diffusion model used in Pecube (see Section 3.2), and (ii) the lack of topographic 582 evolution in QTQt modeling approach. Overall, our results indicate a major tectonic stage 583 during the middle to late Miocene, with kilometric-scale (2-3 km) displacement inferred 584 on the Têt fault along the Carança block only, which cannot be reproduced for the 585 Canigou block (Supplementary Figure S1). This fault-displacement estimate is much 586 higher than the previously-proposed 150-500 m for this period and applied 587 homogeneously along the Têt fault (Pous et al., 1986; Réhault et al., 1987; Roca & 588 Desegaulx, 1992; Calvet, 1999; Carozza & Baize, 2004; Delcaillau et al., 2004; Agusti et 589 al., 2006). Our results are also consistent with a westward propagation of the deformation 590 along the Têt fault with a proposed more recent activity along the Carança block 591 compared to the Canigou block (Carozza & Delcaillau, 1999; Carozza & Baize, 2004; 592 Milesi et al., 2022), marked by the more recent activity along the Carança block 593 compared to the Canigou block, and the latest activity of the Têt fault further West, along

- the Cerdanya Basin, during late Miocene times (Pous et al., 1986; Agustì et al., 2006).
- 595

596 The main differences between observed and predicted low-T thermochronological 597 ages (Fig. 11a) are essentially marked for high-elevation ZHe and AHe ages. ZHe age 598 differences may be linked to lateral variations in timing and exhumation rates associated 599 to the contractional deformation phase in the Eastern Pyrenees as it has been observed at 600 the scale of the whole Pyrenees (Curry et al., 2021). In addition, AHe ages located below 601 1400 m elevation show an important dispersion between 15 and 20 Ma. This observed 602 dispersion can be explained by (i) differences in apatite U-Th content and grain size 603 (Milesi et al., 2022), and/or (ii) additional activity of secondary NW-SE fault(s) between 604 20 and 15 Ma (Fig. 1b). These NW-SE faults are particularly well expressed in the 605 western part of the study area (Taillefer et al., 2021) and can create an important 606 segmentation of the Caranca block, therefore resulting in slightly different cooling 607 histories within the different sub-blocks. The potential activity of such NW-SE faults 608 could also explain the local elevation increase of the Cerdanya basin documented by 609 Tosal et al. (2021) during late Miocene-Pliocene times.

610

611 **5.3 Tectono-geomorphological evolution of the Eastern Pyrenees**

The original approach of our study was to assess regional exhumation and tectonic histories from 3D Pecube inversions using contrasted TEMs over the last around 60 Myr (Fig. 4). Figure 13 summarizes the three tested topographic evolution scenarios along with the output key periods in exhumation/tectonic activity:

- (i) Prior to 37 Ma for the South Mérens block, inferred exhumation rates are
significantly higher for the Global Pyrenees or No topographic change TEMs (>0.5
km/Ma) compared to the Eastern Pyrenees TEM (0.3-0.6 km/Ma);

619 - (ii) since around 37-35 Ma, i.e. the end of the contractional phase, until present
620 day, inferred regional exhumation rates are very close (0.04-0.09 km/Ma, Fig. 12),
621 whatever the considered TEM;

622 - (iii) from 33 to 15 Ma, corresponding to the main Têt-fault activity period along 623 the Canigou and Carança blocks (phase 1), the Global Pyrenees and No topographic 624 change TEMs predict a shorter phase with much higher fault-slip rates compared to 625 inversions with the Eastern Pyrenees TEM (Fig. 12). As these differences are observed 626 for both the Canigou and Carança blocks, our results confirm that the topographic relief 627 evolution scenarios have had a significant impact on the thermochronology cooling ages 628 predicted by the model;

629 - (iv) between 15 and 11 Ma, a second fault-activity phase but only along the
630 Carança block is inferred from our Pecube inversions whatever the considered TEM.
631 Output fault-slip rates are also very consistent for the different TEMs, between 0.7 and
632 0.8 km/Ma (Fig. 10d).

633

Overall, these results show that it remains difficult to use low-T thermochronological data from the Eastern Pyrenees to quantitatively assess the longterm topographic evolution of the area, even if the proposed topographic evolution models since ca. 60 Ma are very contrasted (Fig. 13). We suggest the possible following reason for this limitation: topographic relief changes for the Eastern Pyrenees, even if

639 they appear important in amplitude, are spread out over tens of millions of years, limiting 640 their impact on the crustal thermal structure and rock exhumation compared to the 641 regional rock uplift and local fault activity (Valla et al., 2010; 2011).

642

643 Despite the lack of quantitative information on the robustness of the different 644 TEMs from low-T thermochronology, this approach allowed us to highlight and quantify 645 the main extensional tectonic events that have affected the Eastern Pyrenees. Our results 646 confirm a major Oligocene-Miocene displacement (pluri-kilometric) along the Têt fault, 647 confirming previously-proposed interpretations for the Canigou massif (Maurel et al., 648 2008; Milesi et al., 2022). This major tectonic phase may be related to the opening of the 649 Gulf of Lion (e.g. Séranne, 1999; Mauffret et al., 2001; Maillard et al., 2020; Romagny et 650 al., 2020; Jolivet et al., 2021). Our new results moreover highlight some differences in 651 fault timing and slip rates for this extensional phase between the Carança and Canigou 652 blocks, even though these results must be considered carefully, in particular for the 653 Canigou block, due to the potential contribution of an earlier fault-activity phase during 654 the Priabonian. One major outcome of this study is the characterization of a second, post Oligocene-Miocene, extensional tectonic event between 15 and 11 Ma, with kilometric 655 656 displacement associated along the Têt fault but restricted to its western segment (i.e. the 657 Caranca block). Even if this episode is less important in magnitude than the Oligocene-658 Miocene one, this has been triggering important displacement along the western part of 659 the Têt fault, and can have significantly contributed to the regional relief changes 660 recorded in the Eastern Pyrenees (Gunnell et al., 2008; Huyghe et al., 2020; Calvet et al., 661 2021). This tectonic event may have been recorded at the scale of the whole Pyrenees, as recent thermochronological evidence in the Ebro basin (South Pyrenean Zone) have highlighted a long-wavelength tectonic event at ~10 Ma (Rat et al., 2022) similar to what was also proposed for the western part of the Pyrenees (Fillon et al., 2021). Our study provides further evidence for large-scale lithospheric and sub-lithospheric processes, particularly well expressed in the Eastern Pyrenees, that coincide with the cessation of slab retreat in the West Mediterranean Sea (Jolivet et al., 2021).

668

669 **CONCLUSIONS**

670 3D Pecube thermo-kinematic inversions, using different topographic evolution 671 models for the Eastern Pyrenees, reveal a major decrease in regional exhumation rates 672 between 37-35 Ma. This change is associated to an early end of the contractional tectonic 673 period in the Eastern part of the orogen, while deformation and rock exhumation 674 propagated to the West. The detailed investigation of the Têt fault activity along the 675 Canigou and Carança blocks has been favored by the dense existing low-T 676 thermochronological datasets. Our inversion results highlight two main period of normal-677 fault activity with (i) a major Oligocene-Miocene phase recorded in the Canigou and 678 Caranca blocks, (ii) a secondary mid- to late-Miocene phase with, however, kilometric 679 displacement (2-3 km) restricted along the western segment of the Têt fault (Caranca 680 block), and (iii) no latest Miocene or Pliocene major phase that could have been recorded 681 by low-T thermochronometers. These outcomes confirmed the presence of significant 682 extensional tectonics in the Eastern Pyrenees that may have had a non-negligible role in 683 the late-stage construction of the topographic relief. Finally, our approach and results 684 highlight the benefits of integrating long-term topographic evolution models at a regional

685	scale	to	further	constrain	the	exhumation	and	tectonic	histories	from	low-T
686	thermochronological data and inverse 3D thermo-kinematic modeling.										

687

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697

698 Supplementary Information

699 Supplementary Information to this article can be found online at ...

FIGURES AND CAPTIONS



Figure 1. a) Geological map showing the main structural domains of the Pyrenees (modified after Taillefer et al., 2017). The main Neogene normal faults of the Eastern

Pyrenees are reported in red. The area delimited for 3D thermo-kinematic modeling from Curry et al. (2021) is outlined with a purple-dashed box, and our study area with an

orange-dashed box. b) Location of AHe ages and c) AFT and ZHe ages (literature data,

see text for details) projected on SRTM1s DEM under GMT (Wessel et al., 2019). The

different crustal blocks (South and North Mérens in the Mont Louis massif, Carança and

710 Canigou) are delimited by regional major faults (bold italics, modified after Milesi et al.,

711 2022), while secondary faults and main basins (Cerdanya, Conflent) are also indicated.

- 712 Location of palynological data discussed in the main text are indicated by green stars in
- 713 panels b-c.



715 Figure 2. Topographic relief evolution for the Eastern Pyrenees since Ypresian times 716 (modified after Huyghe et al., 2020). Mean paleo-elevation curve for the Eastern 717 Pyrenees is based on studies from (1) Huyghe et al. (2012); (2) Bache et al. (2010); (3) 718 Huyghe et al. (2020). Maximum paleo-elevation curves are issued from palynological 719 data from (4) Suc and Fauquette (2012) and (5) Agusti et al. (2006), Mean paleo-720 elevation curve for the whole Pyrenees is issued from (6) flexural model outputs from 721 Curry et al. (2019), and (7) low-T thermochronological studies from Fitzgerald et al. 722 (1999); Sinclair et al. (2005); Vacherat et al. (2014, 2016) and Ternois et al. (2019, 723 2021).



724

Figure 3. Topographic profile of the Canigou massif to schematically illustrate the impact
 of the amplification factor and offset parameters used to describe topographic changes in

Pecube modeling. Different scenarios are considered with (1) an amplification factor of 2
and offset of 1500 m (red line) and (2) an amplification factor of 0.5 and offset of -750 m

729 (orange line). The modern topography (amplification factor 1 and offset 0 m) is shown by

the black line.



732 Figure 4. Three topographic evolution models (TEMs) used for Pecube thermo-kinematic modeling. In blue is represented the TEM for the No topographic change. In green is 733 734 shown the TEM for the Global Pyrenees issued after Curry et al. (2019). Finally, in red 735 appears the TEM for the Eastern Pyrenees (after Huyghe et al., 2020; see Section 3.3 for 736 further details). The resulting DEM for each evolution step is presented (upper: Global 737 Pyrenees; lower: Eastern Pyrenees) with associated values of amplification factor (Amp.) 738 and offset (Off.) in meters (see Section 3.2 and Supplementary Table S2 for full details). The surface trace of the normal Têt fault implemented in Pecube models is represented in 739 740 the modern DEM (dashed black line).



Figure 5. 3D Pecube inversion results for the South Mérens block, using the 3 TEMs: (a)
No topographic change, (b) Global Pyrenees, (c) Eastern Pyrenees. In these model runs, 3
parameters are inverted: exhumation rates V1 and V2, and the transition time between the
two exhumation phases. Scatter diagrams show results of Pecube and NA inversion with

the probability density function (PDF) adjacent to the axes for each parameter. Each dot 746 747 corresponds to a forward model; its color is proportional to the value of the reduced misfit between model predictions and the observed low-T thermochronological data. 748 749 Note that violet corresponds to high misfits and yellow to low misfits. The yellow stars 750 are the best-fitting model (values given on top for each TEM). Red and black lines are 2D 751 posterior marginal probability density functions (PDFs) with 2σ (black) and 1σ (red) 752 confidence contours. d) Probability density function (PDF) results for the 3 inverted 753 parameters and three considered TEMs (blue: No topographic change, green: Global 754 Pyrenees, and red: Eastern Pyrenees).



Figure 6. Age-elevation profiles with best-fitting predicted vs. observed AHe, AFT and 758 ZHe ages and MTLs, and output thermal histories from the South Mérens block. a-b) 759 Black dots (AHe, a), squares (AFT, b), diamonds (ZHe, b) and triangle (MTL, subplot in b) are observed data and associated uncertainties. Colored dots, squares, diamonds and 760 761 triangles are predicted data for the 3 TEMs. c-d) Best-fitting output thermal histories for 762 top (c) and bottom (d) samples of the South Mérens block, considering the 3 different 763 TEMs. Inset plot shows the predicted cooling rates for the 3 TEMs.





Figure 7. 3D Pecube inversion results of the Têt fault activity for the Canigou block, using the 3 TEMs: (a) No topographic change, (b) Global Pyrenees, (c) Eastern Pyrenees. In these model runs, 3 parameters are inverted: the Têt fault start and end times, and the slip rate on the fault. Scatter diagrams show results of Pecube and NA inversion with the probability density function (PDF) adjacent to the axes for each parameter. Each dot

770 corresponds to a forward model; its color is proportional to the value of the reduced 771 misfit between model predictions and the observed low-T thermochronological data. 772 Note that violet corresponds to high misfits and yellow to low misfits. The yellow stars 773 are the best-fitting model (values given on top for each TEM). Red and black lines are 2D 774 posterior marginal probability density functions (PDFs) with 2σ (black) and 1σ (red) 775 confidence contours. d) Probability density function (PDF) results for the 3 inverted 776 parameters and three considered TEMs (blue: No topographic change, green: Global 777 Pyrenees, and red: Eastern Pyrenees).

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Figure 8. Age-elevation profile with best-fitting predicted vs. observed AHe, AFT and ZHe ages and MTLs, and output thermal histories from the Canigou block. a-b) Black dots (AHe, a), squares (AFT, b), diamonds (ZHe, b) and triangle (MTL, subplot in b) are observed data and associated uncertainties. Colored dots, squares, diamonds and triangles are predicted data for the 3 TEMs. c-d) Best-fitting output thermal histories for top (c) and bottom (d) samples of the Canigou block, considering the 3 different TEMs. Inset plot shows the predicted cooling rates for the 3 TEMs.



Figure 9. 3D Pecube inversion results of the first phase of the Têt fault activity for the 788 Carança block, using the 3 TEMs: (a) No topographic change, (b) Global Pyrenees, (c) 789 Eastern Pyrenees. In these model runs, 6 parameters are inverted: the Têt fault start and 790 end times, and the slip rate on the fault for the 2 phases of fault activity. Scatter diagrams 791 show results of Pecube and NA inversion with the probability density function (PDF)

792 adjacent to the axes for each parameter. Each dot corresponds to a forward model; its 793 color is proportional to the value of the reduced misfit between model predictions and the 794 observed low-T thermochronological data. Note that violet corresponds to high misfits 795 and yellow to low misfits. The yellow stars are the best-fitting model (values given on top for each TEM). Red and black lines are 2D posterior marginal probability density 796 functions (PDFs) with 2σ (black) and 1σ (red) confidence contours. d) Probability density 797 798 function (PDF) results for the 3 inverted parameters and three considered TEMs (blue: No topographic change, green: Global Pyrenees, and red: Eastern Pyrenees). 799



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Figure 10. 3D Pecube inversion results of the second phase of the Têt fault activity, using
the 3 TEMs for the Carança block: (a) No topographic change, (b) Global Pyrenees, and
(c) Eastern Pyrenees. In these model runs, 6 parameters are inverted: the Têt fault start

and end times, and the slip rate on the fault for the 2 phases of fault activity. Scatter diagrams show results of Pecube and NA inversion with the probability density function 806 (PDF) adjacent to the axes for each parameter. Each dot corresponds to a forward model; 807 its color is proportional to the value of the reduced misfit between model predictions and 808 the observed low-T thermochronological data. Note that violet corresponds to high 809 misfits and to low misfits. The yellow stars are the best-fitting model (values given on top 810 for each TEM). Red and black lines are 2D posterior marginal probability density 811 functions (PDFs) with 2σ (black) and 1σ (red) confidence contours. d) Probability density

812 function (PDF) results for the 3 inverted parameters and three considered TEMs (blue:

813 No topographic change, green: Global Pyrenees, and red: Eastern Pyrenees).

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Figure 11. Age-elevation profiles with best-fitting predicted vs. observed AHe, AFT and ZHe ages and MTLs, and output thermal histories from the Carança block. a-b) Black dots (AHe, a), squares (AFT, b), diamonds (ZHe, b) and triangle (MTL, subplot in b) are observed data and associated uncertainties. Colored dots, squares, diamonds and triangles are predicted data for the 3 TEMs. c-d) Best-fitting output thermal histories for top (c) and bottom (d) samples of the Carança block, considering the 3 different TEMs. Inset plot shows the predicted cooling rates for the 3 TEMs.

Parameters	Range	Pec	ube inversion result	s			
		South Merens block					
		No topographic change	Global Pyrenees	Eastern Pyrenees			
Transition time (Ma)	0-60	36.0 ± 1.5	37.0 ± 2.1	35.2 ± 1.8			
V1 (km/Ma)	0-2	>0.5	>0.5	0.3-0.6			
V2 (km/Ma)	0-1	0.04 ± 0.03	0.06 ± 0.01	0.09 ± 0.03			
		Canigou block					
		No topographic change	Global Pyrenees	Eastern Pyrenees			
Têt fault start (Ma)	25-35	26-25	28-26	>28			
Têt fault end (Ma)	25-5	24.5	24.5	23.5 ± 1.5			
Slip rate (km/Ma)	0-6	>1.0	>1.5	1.0 ± 0.5			
		Carança block					
		No topographic change	Global Pyrenees	Eastern Pyrenees			
Têt fault start phase 1 (Ma)	35-25	26-25	28.0 ± 2.0	32 ± 1.0			
Têt fault end phase 1 (Ma)	25-15	25-24	25-24	15			
Slip rate phase 1 (km/Ma)	0-6	5.5-6	-	<1.0			
Têt fault start phase 2 (Ma)	11-15	15	15	15			
Têt fault end phase 2 (Ma)	5-11	11	11	11			
Slip rate phase 2 (km/Ma)	0-4	0.7 ± 0.2	0.7 ± 0.3	0.8 ± 0.4			

823 Table 1. Synthesis of Pecube inversion results



826 Figure 12. Summary results of QTQt thermal modeling (modified after Milesi et al., 2022; red box indicates prescribed time-temperature constraint from ZHe data used in 827 828 QTQt modeling) and 3D Pecube inversions from this study, using 3 different TEMs 829 (blue: No topographic change, green: Global Pyrenees, and red: Eastern Pyrenees). Diamonds schematically (see legend for details) represent the probability density 830 functions (PDFs) for the timing of the different exhumation or fault-activity phases: PDF 831 832 values >0.2 are considered for South Mérens inversion, and >0.1 for Canigou and 833 Carança inversions. Output exhumation and fault-slip rates are indicated (bottom right).

- 834 Time ranges explored for the start and the end of the Têt fault activity are indicated in
- 835 light orange and light grey colors, respectively.



836

Figure 13. Summary information of the three topographic evolution models (TEMs) used
in Pecube thermo-kinematic modeling (blue: No topographic change; green: Global
Pyrenees; red: Eastern Pyrenees, see Section 3.3 for further details). The temporal
windows highlighted in light pink (i.e. 55-37 Ma for the regional exhumation and 33-15
Ma for the Têt fault activity) and light orange (i.e. 35-0 Ma for the regional exhumation
and 15-11 Ma for the Têt fault activity) represent divergent and convergent Pecube
inversion results (Fig. 12) between the different TEMs, respectively. See text for details.

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