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1 2	Oligocene-Miocene burial and exhumation of the Southern Pyrenean foreland quantified by low-temperature thermochronology
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#### 15 Abstract

16 The central Pyrenees experienced an episode of rapid exhumation in Late Eocene-Early Oligocene 17 times. Erosional products shed from the range during this time were deposited in large paleovalleys of 18 the southern flank of the Axial Zone, leading to significant sediment accumulation. A recent numerical 19 modelling study of the post-orogenic exhumation and relief history of the central Axial Zone allowed 20 us to constrain this valley-filling episode in terms of timing and thickness of conglomeratic deposits. 21 This paper aims to test these results for the southern fold-and-thrust belt using apatite fission-track and 22 (U-Th)/He analysis on detrital samples from the Tremp-Graus and Ager basins. Inverse thermal-23 history modelling of the low-temperature thermochronology data indicates that the fold-and-thrust belt 24 was covered during the Late-Eocene to Miocene by 0.7 to 1.6 km of sediments and confirms the 25 timing of re-excavation of the valleys during the Miocene. A detailed analysis of the apatite (U-Th)/He 26 results shows that the significant scatter in grain ages can be explained by the influence of alpha-recoil 27 damage with varying eU content together with distinct pre-depositional thermal histories; the age 28 scatter is consistent with initial exhumation of the sediment sources during the Triassic and Early 29 Cretaceous.

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#### 31 1. Introduction

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The southern central Pyrenees have figured prominently in studies of orogenic wedge building, thrusting sequences and interactions between tectonics and surface processes, in part because of the exceptional exposure of syn-tectonic strata (Puigdefàbregas and Souquet 1986; Vergés and Muñoz 1990) and the quality of the ECORS seismic profile (ECORS Pyrenees Team 1988) shot through this area.

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39 Low-temperature thermochronology (mainly apatite fission-track; AFT) data (Fitzgerald et al. 1999; 40 Sinclair et al. 2005) provide evidence for a phase of rapid exhumation of the Axial Zone of the central 41 Pyrenees during Late Eocene – Early Oligocene times, accompanied by the development of a crustal-42 scale duplex (Muñoz 1992; Beaumont et al. 2000). The conglomeratic erosional products of this phase 43 of rapid exhumation were hypothesized to have buried the southern flank of the Axial Zone in a late 44 orogenic stage of its evolution, and to have been removed  $\sim 20$  Myr later as post-orogenic incision of 45 the Ebro drainage system reached the area (Coney et al. 1996). Recent magnetostratigraphic studies 46 (Beamud et al. 2003; 2011) have confirmed the depositional ages of these conglomerates, while the 47 link to exhumation of the Axial Zone is clearly established by their sedimentology (Vincent 2001). 48 However, the amount, extent and timing of excavation of these conglomerates have yet to be 49 quantified in the foreland basin where low-temperature thermochronological data are scarce. The only

50 currently available data are AFT ages and track-length distributions from the conglomeratic deposits 51 themselves (Beamud *et al.* 2011; Rahl *et al.* 2011), which confirm the timing of rapid erosional 52 unroofing of the Axial Zone and suggest that the deposits were sufficiently thick to result in partial 53 AFT annealing in samples from their base.

54

55 Two of us have recently constrained the thickness of the conglomerate deposits and the timing of their 56 re-incision, using thermo-kinematic modelling constrained by the existing thermochronology dataset 57 in the Southern Axial Zone (Fillon and van der Beek 2012). These models predict a thickness of the 58 deposits of  $\sim 2$  km on the southern flank of the Axial Zone, which was reached shortly after 30 Ma. 59 Valley incision is predicted to have occurred from Late Miocene times (9 Ma) onward, which we link 60 to the base-level drop resulting from opening of the Ebro Basin to the Mediterranean (Garcia-61 Castellanos et al. 2003; Urgelés et al. 2011). We thus have proposed a scenario of topographic 62 evolution from Axial Zone data that we extrapolated to the southern Pyrenean foreland, but no data 63 were available to test our model predictions.

64

Low temperature apatite fission-track (AFT) and (U-Th)/He (AHe) thermochronology provides a powerful tool to constrain the thermal history of the foreland. The AFT and AHe thermochronometers are sensitive to the 50-120 °C and 40-110 °C temperature ranges respectively (Farley 2000; Shuster and Farley 2009), therefore their combination can be used to constrain burial and exhumation in the fold-and-thrust belt from depths between ~1.5 and 3.5 km, depending on the local geothermal gradient.

71

72 We present new AFT and AHe data from Late Cretaceous-Palaeocene "Garumnian" sandstones 73 collected in the Tremp-Graus and Ager basins of the southern central Pyrenees (Fig. 1), in order to 74 provide direct estimates of the thickness and extent of the overlying conglomerate deposits, as well as 75 to further constrain the timing of excavation of the basin. The AHe thermochronometer is very 76 sensitive and requires high-quality samples, rendering the application of this method challenging in 77 sedimentary samples. However, we will show that despite limited and relatively scattered AHe ages, 78 we are able to obtain consistent Mesozoic to Cenozoic exhumation histories for three samples located 79 in the north, centre and south of the fold-and-thrust belt. We use a new thermal-history inversion code 80 (Gallagher 2012) that allows incorporation of variable AFT-annealing and AHe-diffusion kinetics.

81

## 82 2. Tectono-sedimentary evolution of the South Central Pyrenees

83

The south-central Pyrenean fold-and-thrust belt (also called South Central Unit, SCU) developed with the collision of the Iberian and European plates in the Late Cretaceous. Underthrusting of the Iberian plate below the European plate created a doubly-vergent Pyrenean wedge. The southern Pyrenees

87 evolved as the pro-wedge by inversion of Cretaceous extensional structures (Puigdefàbregas and 88 Souquet 1986; Bond and McClay 1995) followed by in-sequence thrust propagation towards the south 89 (Vergés and Muñoz 1990; Muñoz 1992; Vergés et al. 1995; Beaumont et al. 2000). The fold-and-90 thrust belt propagated further in the SCU, compared to the regions east and west of it, by sliding on a 91 thick Triassic evaporite layer (Fig. 1c). Thrusting is in-sequence at first order, with activation of the 92 Boixols thrust initiated in the Late Cretaceous, followed by the Montsec thrust, which transported the 93 Tremp-Graus Basin in a piggy-back manner from Paleocene to Late Eocene times (Puigdefabregas et 94 al. 1992), and finally the frontal Sierras Marginales thrust sheet, active from Middle Eocene to Late 95 Oligocene times (see Fig. 1 for locations).

96

97 From the ECORS seismic profile (ECORS Pyrenees Team 1988; Choukroune and ECORS Team 98 1989), as well as modelling and thermochronological studies of the Southern Axial Zone (Beaumont et 99 al. 2000; Sinclair et al. 2005; Fitzgerald et al. 2006; Gibson et al. 2007; Metcalf et al. 2009), rapid 100 exhumation of the internal units (Nogueres, Orri and Rialp, Fig. 1c) is known to have occurred by 101 vertical stacking (Muñoz 1992; Vergés et al. 1995) during Middle Eocene to Early Oligocene times. 102 Thermo-kinematic modelling of the *in-situ* low-temperature thermochronology (AFT and AHe) dataset predicts exhumation rates of 2.8±0.3 km Myr<sup>-1</sup> between 37 and 30 Ma in that area (Fillon and 103 104 van der Beek 2012). This rapid exhumation was associated with strong erosion of the southern Axial 105 Zone, the products of which were deposited in pre-existing palaeo-valleys as a thick pile of 106 conglomerates currently mainly exposed in the Sis, Gurp and La Pobla massifs (Fig. 1b). These 107 palaeo-valleys probably represented sediment transfer zones, supplying the Huesca fan system 108 (Vincent 2001), one of the main drainage systems of the Southern Central Pyrenees (with the Luna fan 109 system to the West) during Oligocene-Miocene times. The link between exhumation of the internal 110 massifs and deposition of the conglomerates has been confirmed by provenance studies (Vincent 111 2001) and by AFT analysis of pebbles from the Sis conglomerates (Beamud et al. 2011; Rahl et al. 112 2011), which both reveal unroofing of the Axial Zone and deposition of its erosional products in the 113 basin.

114

115 Present-day remnants of these extensive syn- to post-tectonic conglomerate deposits include the 116 Senterada and La Pobla basins (Beamud et al. 2003; Beamud et al. 2011), the Sis conglomerates 117 (Vincent, 2001), the Oliana fan (to the east of the CSU; Burbank et al. 1992b) and the Huesca fan 118 (southwest of our study area; Friend et al. 1996); they reach a maximum thickness of ~1000 m. 119 Magnetostratigraphic studies constrain deposition of the Sis conglomerates at ~40 to 27 Ma (Beamud 120 et al. 2003; Beamud et al. 2011), deposition of similar conglomerates to the east of the CSU, in the 121 Oliana area, at ~40 to 36 Ma (Burbank et al. 1992a), and in the Sierras Marginales thrust sheet 122 between 36.5 and 24.7 Ma (Meigs et al. 1996). The period of strong exhumation/erosion/deposition 123 was synchronous with closure of the Atlantic connection of the Ebro foreland basin at 36 Ma (Costa et 124 *al.* 2009). The basin became endorheic at that time and remained so until Late Miocene times (Arenas

125 and Pardo 1999; Garcia-Castellanos et al. 2003; Arche et al. 2010; Urgelés et al. 2011). During its

126 endorheic phase, the basin was progressively filled by conglomeratic deposits at its borders, grading to

127 lacustrine sediments in its centre, and developed into a large overfilled foreland basin.

128

#### 129 **3.** Mesozoic pre-depositional history

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131 Recently, two independent studies have reported zircon fission-track (ZFT), U-Pb and (U-Th)/He 132 (ZHe) ages of samples from the SCU (Filleaudeau et al. 2011; Whitchurch et al. 2011). ZHe analysis 133 of Garumnian sandstones of the Tremp Basin (sample ORC2; Filleaudeau et al. 2011) shows three 134 main age populations (Triassic, Early Cretaceous and Late Cretaceous), interpreted to reflect distinct 135 Mesozoic exhumation events affecting the source area(s). Whitchurch et al. (2011) applied detrital 136 ZFT dating to Garumnian sandstones of the Ager basin and found the same Early and Late Cretaceous 137 exhumation events, together with Carboniferous (Variscan) ZFT ages. A sample collected from the 138 Aren formation (the stratigraphic unit just below the Garumnian in the Tremp Basin) presents Early 139 Cretaceous and Carboniferous ZFT age peaks, lacking Late Cretaceous ages. The combination of these 140 two datasets shows an important phase of Early Cretaceous source-area exhumation at 134±15 Ma (all 141 uncertainties combined), with less well expressed events at ~80 Ma, ~225 Ma and 300-330 Ma. We 142 will discuss later the imprint of these pre-depositional exhumation phases on our sample ages.

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- 144

#### 4. Low-temperature thermochronology

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Five samples (AN01 to AN05) were collected along a N-S profile in Garumnian facies sandstones. These continental deposits of Maastrichtian-Middle Palaeocene (65-60 Ma) age crop out extensively throughout the fold-and-thrust belt (Fig. 1b). Other lithologies were also sampled, including Eocene sandstones, Santonian turbidites and volcanic rocks of the Nogueres zone, but none of these provided sufficient high-quality apatite grains for thermochronological analysis.

151

#### 152 **4.1. Apatite Fission-Track thermochronology**

Samples were prepared and analyzed at ISTerre (Grenoble, France). Apatite grains were separated from fine- to medium-grained sandstone samples using standard heavy-liquid and magnetic separation techniques. Apatite aliquots were mounted in epoxy, polished to expose internal crystal surfaces, and etched with 5.5 M HNO<sub>3</sub> for 20 s at 21 °C. Low-U muscovite sheets were fixed to the mounts, to be used as external detectors, and samples were irradiated in the FRM II Research Reactor at the Technische Universität München (Germany). Apatite samples were irradiated together with IRMM 540R dosimeter glasses and Durango and Fish Canyon Tuff age standards. After irradiation, the mica 160 detectors of all samples and standards were etched for 18 minutes at 21 °C in 48% HF. The samples 161 and standards were counted dry at 1250× magnification, using an Olympus BH2 optical microscope 162 and the FTStage 4.04 system of Dumitru (1993). Due to the low yield of apatite grains, all datable 163 grains were counted for each sample. Fission-track ages were calculated using the zeta-calibration 164 method and the standard fission-track age equation (Hurford and Green 1983). The  $\chi^2$ -test and age 165 dispersion (Galbraith and Green 1990; Galbraith and Laslett 1993) were used to assess the 166 homogeneity of AFT ages. We were able to measure horizontal confined track lengths and the lengths 167 of the track etch pits that outcrop on the etched internal surface (D<sub>nar</sub>) in four samples.

168

## 169 4.2. Apatite (U-Th)/He analysis

170 Apatite grains were carefully selected according to their morphology at ISTerre. Each grain was 171 placed into a platinum basket for He-extraction at Paris-sud University (Orsay, France). Two to five 172 replicates were analyzed per sample. The platinum baskets were heated using a diode laser to 1030±50 173 °C during 5 minutes, allowing total He degassing; a reheat under the same conditions allowed checking for the presence of He trapped in small inclusions. The <sup>4</sup>He content was determined by 174 comparison with a  $1-2 \times 10^{-7}$  ccSTP <sup>3</sup>He spike. After He extraction, platinum baskets were placed into 175 single-use polypropylene vials. Apatite grains were dissolved one hour at 90 °C in a 50 µl HNO3 176 solution containing a known concentration of <sup>235</sup>U and <sup>230</sup>Th, and then filled with 1 ml of ultrapure 177 MQ water. U and Th measurements followed a procedure similar to Evans et al. (2005). The final 178 solution was measured for U and Th concentrations by quadrupole ICP-MS (series<sup>II</sup> CCT Thermo-179 180 Electron at LSCE, Gif-sur-Yvette, France). Analytical accuracy and reproducibility for U-Th isotope 181 measurements by using ICP-QMS were better than  $\pm 0.5\%$  at  $2\sigma$  (Douville *et al.* 2010). The analysis 182 was calibrated using internal and external age standards, including Limberg Tuff, Durango and FOR3, 183 which provided mean AHe ages of  $16.8\pm0.7$  Ma,  $31.8\pm0.5$  Ma, and  $110.7\pm7.0$  Ma respectively. These 184 values are in agreement with literature data, i.e. 16.8±1.1 Ma for the Limberg Tuff (Kraml et al. 2006), 185 31.02±0.22 Ma for Durango (McDowell et al. 2005) and 112±10 Ma for the internal FOR3 standard. Individual ages were corrected by each grain's ejection factor  $F_T$ , determined using the Monte Carlo 186 187 simulation technique of Ketcham et al. (2011); the equivalent-sphere radius has been calculated using 188 the procedure of Gautheron and Tassan-Got (2010). The  $1\sigma$  error on AHe age should be considered at 189 8%, reflecting the sum of errors in the ejection-factor correction and age dispersion of the standards.

190

#### 191 **5. Results**

192

Apatite fission tracks were counted and measured in 4 samples located in the north (AN04), centre
(AN03, AN02) and south (AN01) of the fold-and-thrust belt (see Fig. 1 for location); the results are
reported in Fig. 2 and Table 1. Only sample AN01, collected south of the Montsec thrust (Fig. 1),

196 close to the Ager basin sample of (2011), shows a single age component (144±11 Ma). The other samples, for which more grains were counted, fail the  $\chi^2$ -test and have age dispersions >20%, with 197 198 central ages between 64±5 and 77±6 Ma. Individual samples contained insufficient dated grains to 199 deconvolve the sample ages into age components. However, since the samples were collected in the 200 same stratigraphic unit and show similar age structures, we calculated age components from the 201 combined data, using the radial-plotter software of Vermeesch (2009). Three age peaks are found 202 when pooling all single-grain ages: the major age component is Late Cretaceous in age (76.1±3.7 Ma), 203 the second peak age is Late Paleocene (i.e., younger than the depositional age;  $51.7\pm3.2$  Ma), and a 204 minor Early Cretaceous peak age is found at 142±10 Ma, with Early Cretaceous single-grain ages 205 encountered in all samples (Fig.2).

206

The youngest age population is clearly younger than the depositional age and single-grain ages <60 Ma are found in all samples except AN01, suggesting that they have been partially reset by postdepositional burial. Mean track lengths (MTL) are very short, ranging from 10.1  $\mu$ m in AN01 to 12.2  $\mu$ m in AN03 with standard deviations between 1.0 and 2.2  $\mu$ m, indicating relatively slow long-term exhumation rates. Note, however, that only between 10 and 27 track lengths could be measured in these samples due to the low apatite yield.

213

214 Apatite (U-Th)/He analyses were also performed on four samples: AN01, 02, 03 and AN05 (Fig. 3, 215 Table 2). A striking feature of the AHe ages is their large scatter; single-grain ages vary from 3.9 to 169 Ma. There appears to be some correlation between AHe ages and the effective uranium content 216 217  $(eU = [U]+0.24\times[Th])$ , expressed in ppm) of the grains, which varies between 4 and 25 ppm in samples 218 AN01, 02 and 03 (Fig. 3a). The relationship between single-grain AHe ages and eU content reflects 219 the increase of closure temperature as a function of the amount of  $\alpha$ -recoil damage, for which eU 220 content is a convenient proxy (Shuster et al. 2006; Shuster and Farley 2009). Moreover, the rapidly 221 increasing ages for eU > 15 ppm, with less variation below this value (Fig. 3a), are consistent with a 222 model in which He-retention increases non-linearly with the amount of  $\alpha$ -recoil damage (Shuster et 223 al., 2006; Flowers et al. 2009). Only sample AN05 does not fit this correlation; we suspect that this 224 may be related to its very high eU content and, in particular, quite extreme Th concentration compared 225 to the other samples. Thus, the AHe ages cannot be interpreted directly, but the Late Miocene-Pliocene 226 ages measured in samples AN01, AN02 and AN03 imply post-depositional burial and exhumation of 227 the basin. We will test the timing and amount of burial / exhumation through numerical inversion of 228 the data in the next section.

#### 230 6. Thermal-history modelling

231

232 In order to test what constraints the data provide on the post-depositional thermal history of the 233 samples, we use an inversion model (Gallagher et al. 2009, Gallagher 2012) that allows extracting 234 both optimal thermal histories and AFT annealing/AHe diffusion parameters by a Markov chain 235 Monte Carlo (MCMC) sampling method. The inversion code incorporates recent kinetic models of He 236 diffusion proposed by Flowers et al. (2009) and Gautheron et al. (2009), as well as the most recent 237 multi-kinetic AFT annealing model of Ketcham et al. (2007). The modelling proceeds from an initial 238 randomly chosen time-temperature path and set of kinetic parameters, for which a probability that the 239 model fits the data is calculated. Then the parameters are slightly perturbed, the probability of fitting 240 the data is recalculated and compared to the initial model. The model with the highest probability is 241 retained. This procedure is repeated a large number of times (the number of iterations being chosen by 242 the user), providing a large collection of models with their associated probabilities that allow 243 calculating model statistics (probability distributions of model parameters, etc.). A full explanation of 244 the modelling procedure is provided in Gallagher (2012).

245

#### 246 **6.1. Model set-up**

247 The parameter space (time, t and temperature, T) has been subdivided into three T-t boxes: from 300 248 Ma (beginning of the model run) to 70 Ma with temperatures between 140 °C and 0 °C, from 70 to 60 249 Ma with a temperature of  $10\pm10$  °C (e.g. surface conditions), and finally from 60 Ma to present, with a 250 temperature range of 140 °C to 0 °C. By doing this, the only constraint we impose on the model is the 251 deposition time. The results are the product of 200,000 iterations, which is a sufficient amount to 252 obtain a stable and robust solution (see discussion in Gallagher 2012). We present results of inversions 253 for samples AN01, 02 and 03, which were analysed with both AFT and AHe thermochronology; 254 samples AN04 and AN05, which come from the same structural position but for which only AFT 255 (AN04) or AHe (AN05) measurements were available, were combined for a test model but do not 256 provide sufficient thermal constraints to obtain a well-defined *T*-*t* path.

257

As we use a multikinetic AFT annealing algorithm (Ketcham *et al.* 2007) in this study, the single grain-age counting data and track-length measurements were incorporated in the model and combined with the average  $D_{par}$  (i.e. we do not take potential intra-sample kinetic variation into account). We acknowledge that the number of track lengths measured in the samples is less than optimal for inverse thermal history modelling; however, this limitation is offset by the fact that we have both AFT and AHe data and exploit variable annealing and diffusion kinetics.

264

265 The He-diffusion models of Gautheron *et al.* (2009) and Flowers *et al.* (2009) were used to reproduce 266 He-diffusion behaviour in apatite. These two models include the influence of  $\alpha$ -recoil damage and its 267 annealing on He-diffusion kinetics; in particular, they predict He retentivity to increase either linearly 268 (Gautheron et al. 2009) or non-linearly (Flowers et al. 2009) with the amount of  $\alpha$ -recoil damage. 269 Predicted AHe ages by the two models mainly differ for low eU contents (15-25 ppm), because this is 270 the range where the amount of  $\alpha$ -recoil damage corresponds to the slope change in the diffusion law 271 defined by natural and experimental data (Shuster et al. 2006; Shuster and Farley 2009). The 272 simulations therefore predict slightly different thermal histories depending on which He-diffusion 273 model is used; we will only present models using the Flowers et al. (2009) diffusion algorithm here; 274 modelling results obtained using the Gautheron et al. (2009) diffusion algorithm are presented in 275 Supplementary Figure 1.

276

The inversion procedure used here implicitly assumes that all grains share a single pre-depositional thermal history, whereas the zircon data discussed in Section 3 show that these sediments were derived from multiple sources with different pre-depositional histories. We will assess the effect of this simplification in the discussion (Section 7.2).

281

#### 282 6.2. Inferred thermal histories

The models predict very similar *T-t* paths for all three samples (Fig. 4), in accordance with their similar stratigraphic and structural position, showing burial from their time of deposition until Miocene-Pliocene times, followed by exhumation until the present-day. The pre-deposition *T-t* path is poorly constrained for samples AN01 and AN02, in contrast to the model for sample AN03, which predicts a well-constrained pre-depositional history with linear cooling from the Early Cretaceous (130 °C at 100 Ma) to the time of Garumnian sandstone deposition. For this sample, the model predicts burial to a temperature of ~70 °C reached between 11 and 5 Ma, before final exhumation.

290

291 Sample AN01 produced the widest range of single-grain AHe ages (7 Ma and 77 Ma); although the 292 AFT age (144±11 Ma) suggests a single source age. The sample is also furthest from the preserved 293 conglomeratic massifs. Nevertheless, the model predicts significant burial, to a maximum temperature 294 of 80 °C, and an onset of Miocene exhumation between 20 and 10 Ma. The T-t path of sample AN02 295 is quite similar, although this sample appears to have been buried most deeply, reaching a temperature 296 of up to 95 °C. The predicted onset of final exhumation is well constrained at 7 Ma. Finally, we 297 observe that the maximum Cenozoic burial temperatures predicted by the models are consistent with 298 their corresponding AFT ages; maximum predicted burial temperatures are higher for samples with 299 younger central AFT ages.

#### 301 7. Discussion

302

## 303 7.1. Sensitivity to eU variations

304 To test the thermal histories provided by inverse modelling and the match with our AHe ages, the 305 "expected" thermal histories (i.e. the weighted-average *T*-*t* path for the probability distributions shown 306 in Fig. 4; cf. Gallagher 2012) for the three samples have been introduced in a forward model (Ketcham 307 et al. 2005) to predict AHe ages for a grain size equal to the average size of our analysed crystals 308 (equivalent-sphere radius of  $57 \,\mu\text{m}$ ) and eU contents varying from 5 to 30 ppm, using the He-diffusion 309 model of Flowers et al. (2009). A comparison of predicted ages with our AHe data (Fig. 5) shows that 310 both single-grain ages of sample AN02, as well as most single-grain ages of sample AN03 are in good 311 agreement with the modelled path. Therefore, this test shows that the age scatter of these samples can 312 be largely explained by their varying eU content. Conversely, some grains of samples AN01 and 313 AN03 do not fit the modelled relationship between age and eU; therefore the measured ages could 314 have been influenced by another parameter, such as a different pre-depositional history. We will test 315 that possibility in the following section.

316

## 317 **7.2. Influence of the pre-depositional history**

The inverse modelling procedure we used cannot take into account variable pre-depositional histories of these detrital grains. However, the detrital ZFT and ZHe data of Whitchurch *et al.* (2011) and Filleaudeau *et al.* (2012) provide evidence for variability in timing and rates of source-area exhumation, as discussed in Section 3. We thus test to what degree such variable pre-depositional exhumation histories could explain the encountered dispersion of AHe ages.

323

324 The ZFT and ZHe data presented in Section 3 show pre-depositional exhumation ages between ~80 325 and ~300 Ma. From this starting point, we tested the influence of the pre-depositional history and eU 326 content by performing forward modelling (Ketcham 2005), using the AFT annealing model of 327 Ketcham et al. (2007) and the AHe-diffusion model of Flowers et al. (2009), with the same parameter 328 values as in Section 7.1. We imposed the post-depositional scenario from the inversion results 329 presented previously and tested how the final AHe age varies as a function of eU, maximum post-330 depositional temperature ( $T_{max}$ ) and onset time of initial (pre-depositional) exhumation (Fig. 6a). The 331 results are presented as contours of predicted AHe ages as a function of eU and onset time in Fig. 6b 332 and 6c, for T<sub>max</sub> of 70 and 80 °C respectively.

333

This forward modelling differs from the inverse modelling presented previously in two important aspects, therefore inducing somewhat different results: first, the forward models use fixed values for the diffusion parameters, whereas the inverse models allow varying these within the uncertainties propagated from the original experiments (Gallagher 2012). Second, we fix  $T_{max}$  as well as its timing and the overall shape of the thermal history path in the forward models, whereas these have more degrees of freedom in the inversions and show, for instance, clear tradeoffs between  $T_{max}$  and timing of initial (pre-depositional) cooling. We use these forward models to illustrate the effects of variable  $T_{max}$ , eU and pre-depositional history on the measured AHe ages rather than to provide additional constraints on these parameters.

343

344 For the two burial temperatures tested, the modelling reveals the same patterns, in agreement with the 345 relationship we expect between AHe age, eU content and T<sub>max</sub>. The modelled ages are older when 346 initial exhumation starts earlier and for higher eU contents. The predicted ages are younger when T<sub>max</sub> 347 increases; the maximum modelled AHe ages are 155 and 120 Ma for Tmax of 70 and 80 °C, 348 respectively. Moreover, for the same eU, the age scatter decreases with increasing T<sub>max</sub>. For example, 349 for an eU of 16 ppm (the average eU value of our grains), modelled ages vary from 10 to 105 Ma as a 350 function of onset time of initial exhumation for  $T_{max} = 70$  °C, but only from 10 to 40 Ma for  $T_{max} = 80$ 351 °C.

352

353 These forward models confirm the hypothesis that the pre-depositional history of the detrital grains 354 can have a major effect on AHe age variations. With these results, we can also identify what 355 variability in timing of pre-depositional exhumation would be required to explain the scatter in our 356 AHe ages, by plotting the ages and eU values of our samples on the contour plots (Fig.7b). First of all, 357 we note that the forward model with  $T_{max} = 70$  °C provides the best fit to the combined data; this value 358 is within the range of predictions of all inverse models. Secondly, when looking at the  $T_{max} = 70$  °C 359 plot, we can conclude that all grains except AN011 fit with a pre-depositional exhumation phase 360 starting between 150 and 90 Ma, which is in good agreement with the ZHe and ZFT age distributions 361 of Filleaudeau et al. (2012) and Whitchurch et al. (2011), which record a major Early Cretaceous 362 (~120-150 Ma) exhumation phase and a less well-expressed phase in the Late Cretaceous (~80 Ma). 363 These results are also in agreement with the expected model from the thermal-history inversion of 364 AN02 and AN03, predicting an onset of initial exhumation before the Late-Cretaceous (i.e. >100 Ma). 365 Only the age of AN011 is outside of this range and suggests Triassic initial cooling, consistent with 366 the occurrence of Triassic ZHe ages, which may result from a major volcanic episode associated with 367 the emplacement of the Central Atlantic Magmatic Province (Marzoli et al. 1999). Our previous 368 observations showed that the age dispersion between the two grains of this sample could not be 369 explained by their respective eU contents alone. We thus propose that variable pre-depositional 370 exhumation histories, with at least one grain recording Triassic initial cooling, constitute another 371 factor to explain the variable AHe grain ages.

To conclude, our forward modelling shows that the pre-depositional history of a detrital sample can considerably influence the measured AHe ages as well as the inferred temperature of burial. In our case, it seems that a combination of variable eU and variable timing of initial exhumation, consistent with higher-temperature detrital ZHe and ZFT data, can explain the scatter in AHe ages. Moreover, the forward models suggest that most apatite grains record an Early Cretaceous pre-depositional exhumation phase.

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#### 380 7.3. Cenozoic Burial and exhumation scenario

The inverse modelling results provide values for the amount and timing of Cenozoic burial heating that range from 70 to 95 °C, with final exhumation starting between 20 and 5 Ma. Even though the modelling predicts a variable range of maximum burial temperatures and associated timing, the firstorder pattern of the *T-t* paths in all cases shows significant burial during post-Eocene times, with subsequent exhumation starting in the Late Miocene–Early Pliocene.

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387 We interpret the burial recorded by the samples as being due to deposition of the Late Eocene-388 Oligocene conglomerates produced by rapid erosion of the Axial Zone, and explain the subsequent 389 exhumation by excavation of the modern southern Pyrenean river valleys. In this scenario, we do not 390 take into account any tectonic activity that could be responsible for late-stage exhumation of the SCU 391 (*i.e.*, during Miocene times). The main reason for this choice is that there is no clear evidence for such 392 tectonic activity in the SCU, mainly because no Miocene sediments are exposed in that area. 393 Moreover, as shown in Fig. 7, our AFT and AHe ages and the corresponding burial estimates 394 predicted by the thermal modelling are homogeneous for all samples, despite their provenance from 395 different thrust sheets. A few very young (Middle Miocene) AFT and AHe ages were reported further 396 west, in the Bielsa massif (Jolivet et al. 2007) and to the north of the study area in the Barruera massif 397 (Gibson et al. 2007), which have been interpreted as recording Miocene out-of-sequence thrusting. 398 However, it is unclear how these results could be extrapolated to the SCU.

399

400 Estimating the thickness of the conglomerate cover requires knowledge of the local geothermal 401 gradient. A previous interpretation of AFT data from the SCU (Beamud et al. 2011) used a geothermal 402 gradient of 30 °C km<sup>-1</sup>, as constrained by numerical modelling (Zeyen and Fernandez 1994) supposing a value for the thermal conductivity of sediments of 2.5 W m<sup>-1</sup> K<sup>-1</sup>. According to Fernandez *et al.* 403 404 (1998), however, the average present-day geothermal gradient for the SCU is 22±4 °C km<sup>-1</sup>, 405 significantly lower than the value for the Pyrenean Axial Zone ( $\sim$ 33 °C km<sup>-1</sup>); this discrepancy is apparently due to the high thermal conductivity (2.47 to 3.22 W  $m^{-1}$  K<sup>-1</sup>) measured in the sediments. 406 407 As there is no consensus on the subject, we will use in the following a 30 °C km<sup>-1</sup> geothermal gradient 408 to estimate the sediment thicknesses, thus providing a conservative estimate of the amount of burial.

410 Using this value for the geothermal gradient and a surface temperature of 20 °C, the minimum amount 411 of burial required to reach the T<sub>max</sub> recorded by our Garumnian samples can be estimated at 2.3 km for 412 sample AN03, 2.6 km for sample AN01 and to 3.2 km for sample AN02, with final exhumation rates 413 of 0.13 to 0.5 km Myr<sup>-1</sup>. As the thickness of the Palaeocene-Middle Eocene sediments deposited in the 414 Tremp-Graus Basin ranges from 1.2 km in the Montsec footwall area (sample AN01) to 1.6 km in the 415 Tremp area (samples AN02 and AN03), our results imply a minimum thickness of post-Middle 416 Eocene sediments of 700 m for sample AN03, 1.6 km for the sample AN02 and 1.4 km for sample 417 AN01, as represented in Fig. 7. Note that, because the Late Eocene-Oligocene conglomerates are 418 discordant on the Middle-Eocene and older sediments, it is difficult to estimate the exact thickness of 419 the latter. Note also that for a lower geothermal gradient of 22 °C km<sup>-1</sup> (see paragraph above), inferred 420 burial depths are significantly higher and require 1.6, 2.4 and 2.7 km of post-Middle Eocene 421 sediments, respectively, for samples AN03, AN01 and AN02.

422

423 Thermo-kinematic modelling of the *in-situ* AFT and AHe dataset in the Axial Zone of the central 424 Pyrenees (Fillon and van der Beek 2012) suggested that  $\sim 2$  km of conglomeratic deposits once 425 covered the southern flank of the central Axial Zone. The model also predicted re-incision and erosion 426 of the conglomerates during the Tortonian (9.2±0.5 Ma), implying late-stage exhumation rates of  $\sim 0.3$ 427 km Myr<sup>-1</sup> in the valley bottoms. The AHe ages confirm this interpretation and show continuous 428 excavation of the basin from the Late Miocene-Pliocene to the present.

429

430 Finally, it is interesting to note that our inverse models do not reproduce an Eocene signal of 431 exhumation linked to tectonic activity of the fold-and-thrust belt. However, the Late Eocene 432 conglomerates rest unconformably on the earlier syn-tectonic sediments, implying an initial period of 433 burial and unroofing between the Late Cretaceous and Late Eocene. We tested for this by forcing the 434 thermal history models to reach near-surface temperatures in the Eocene but found that this did not 435 affect the models to any significant extent. Therefore, if there was exhumation in the Eocene it could 436 not have been very large (>1 km) otherwise the AFT and AHe systems would have recorded this 437 event. Our modelling results thus support the idea of a strongly overfilled Ebro basin in the Oligocene-438 Miocene, characterised by up to 1.6 km of sediment aggradation at its northern edge.

439

### 440 **7.4. Extent of late orogenic conglomerate deposits**

The inferred amounts of burial are consistent with what we expected for samples AN02 and AN03, as these were collected close to the remnant conglomeratic massifs and the Axial Zone. A more surprising result is obtained for sample AN01, which is located in another structural unit, south of the Montsec thrust, and quite far from the Axial Zone. The closest conglomeratic outcrops to this sample are located further east and have been dated by magnetostratigraphy at 40 to 36 Ma (Oliana conglomerates; Burbank *et al.* 1992a). Nevertheless, the inversion results for sample AN01 suggest 447 2.6 km of burial before an onset of final exhumation between 10 and 20 Ma. These values are 448 comparable to those obtained for the northern samples. Thus, the Ager Basin, south of the Montsec 449 thrust (Fig. 1), seems to record the same post-Eocene burial/exhumation history as the Tremp Basin, 450 implying that infilling by the Huesca fan sediments extended to that basin. The burial of the Ager 451 Basin, which is not drawn in most paleo-geographic reconstructions (see Jones 2004 for example), 452 could result from an extension of the Huesca fan further to the east, or from extension of the Oliana 453 fan towards the west.

454

## 455 8. Conclusions

456

457 We have succeeded in modelling our dataset of AFT and AHe ages of detrital apatites to obtain 458 consistent T-t paths using He-diffusion kinetics that are a function of radiation damage and its 459 annealing. Our models show that the significant AHe-age scatter can be explained by variable eU 460 content together with variable pre-depositional exhumation histories, consistent with inferences from 461 published higher-temperature ZFT and ZHe data. Modelling of three samples predicts burial from the 462 Late-Cretaceous to the Miocene-Pliocene to reach maximum temperatures of 70 to 95 °C, requiring at 463 least 0.7 to 1.6 km of Late Eocene-Oligocene overburden. The inferred timing for the onset of 464 exhumation ranges from 20 to 5 Ma, with exhumation rates of 0.13 to 0.5 km Myr<sup>-1</sup>. These modelling 465 results thus show similar post-orogenic burial and exhumation in the southern Pyrenean foreland as 466 previously inferred for the southern Axial Zone and suggest a Late Miocene (pre-Messinian) onset of 467 Ebro Basin incision. They confirm that several km of sediments covered the southern Pyrenean flank 468 and filled the Ebro Basin, consequently to the onset of endorheism in the basin.

469

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Sample	position (lat/long)	Elevation (m)	no of grains	ρs (Ns) (10 <sup>6</sup> cm <sup>-2</sup> )	ρi (Ni) (10 <sup>6</sup> cm <sup>-2</sup> )	ρd (Nd) (10 <sup>6</sup> cm <sup>-2</sup> )	Ρχ² (%)	D	Central age (± 1σ, Ma)	MTL (µm)	sd (µm)	no tracks	Dpar (µm)	sd (μm)	no Dpar
AN01	41.96/00.85	624	8	1.133 (489)	0.632 (273)	0.74 (4262)	32.34	0.02	$144 \pm 11$	10.1	1.0	10	2.2	0.4	24
AN02	42.16/00.89	436	20	0.7641 (999)	0.8513 (1113)	0.75 (4262)	<< 1	0.26	$76.9 \pm 6.2$	11.3	1.4	19	2.1	0.2	36
AN03	42.18/00.89	504	22	1.264 (1639)	1.493 (1936)	0.75 (4262)	<< 1	0.26	$64.2 \pm 4.6$	12.2	1.6	27	2.2	0.2	61
AN04	42.22/00.84	902	13	1.0715 (810)	1.205 (911)	0.75 (4262)	<< 1	0.20	74.1 ± 7.6	11.3	2.2	19	2.1	0.1	18

Table 1 Apatite Fission-Track results.  $\rho_s$  and  $\rho_i$  are the spontaneous and induced track densities measured;  $\rho_d$  is the induced track density in the external detector, the number of tracks counted (Ns, Ni, Nd) are in brackets;  $P(\chi^2)$  is  $\chi^2$ -probability (in %), D is the single-grain age dispersion. A  $P(\chi^2) < 5$  and/or D > 100

0.15 indicates that the data distribution contains multiple age populations; MTL is the mean track length, with sd its standard deviation; Dpar is the average

etch-pit width measured parallel to the C-axis. Measurements were performed by CF with a  $\zeta$ -factor = 217.9±3.5 for IRMM-540 dosimeter glasses.

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Lab ID	Sample	$F_T$	Weight (ng)	Length/ Width/ Thickness (µm)		<sup>4</sup> He×10 <sup>-6</sup> (cc/g)	U (ppm)	Th (ppm)	Th/U	eU (ppm)	Age (Ma)	Age corr. ± 1σ (Ma)	
2486	AN011	0.76	2.9	128	101	95	122	12.9	19.2	1.5	17.5	58.0	76.9 ± 7.1
2488	AN012	0.80	5.7	181	123	108	4.1	2.8	6.3	2.3	4.3	5.2	$6.5 \pm 0.6$
2493	AN022	0.76	3.4	146	106	92	9.4	9.9	11	1.1	12.5	6.2	8.1±0.7
2495	AN023	0.84	6.7	156	139	132	5.4	5.8	33.6	5.8	13.9	3.2	$3.9 \pm 0.3$
2499	AN031	0.76	3.9	187	100	85	48.5	9.8	39.6	4	19.3	20.9	$27.6 \pm 2.5$
2501	AN032	0.87	5.3	177	135	117	117	22.3	20.5	0.9	27.2	35.7	41.0± 3.8
2548	AN033	0.69	10	151	77	67	120	12	54.9	4.6	25.2	39.6	57.4± 5.3
1641	AN034	0.84	11.5	200	150	187	20.6	15.9	8.9	0.56	18	9.5	11.3±1.0
1643	AN035	0.80	5	150	125	112	6.2	4	14.2	3.55	7.4	6.9	$8.7 \pm 0.8$
2552	AN051	0.80	4.6	190	113	97	326	49.2	616.3	12.5	197.1	13.8	17.2±1.6
2554	AN052	0.80	5	147	120	112	224	27.9	362.4	13	114.9	16.3	20.3±1.9
1878	AN053	0.85	12.6	275	175	125	53.2	6.4	29.9	4.67	13.6	32.6	38.5±3.5
1880	AN054	0.80	12.8	262	162	162	335	10.6	36.5	3.43	19.4	144.0	$169.1 \pm 15.6$

714 **Table 2.** Apatite (U-Th)/He results. Lab ID is the Orsay sample identification number, which is the second replicate <sup>3</sup>He spike pipette number,  $F_T$  is the 715 geometric correction factor for age calculation; age corr. is the age corrected by the ejection factor  $F_T$ , the uncertainty  $1\sigma$  was fixed at 8 % of the age. eU is 716 the effective uranium concentration.

- 717 Figure captions
- 718

Figure 1. a) Regional map of the Pyrenean range and its two foreland basins, the black box represents the study area shown in map b. NPZ: North Pyrenean Zone; SCU: South Central Unit. b) Simplified geological map of the southern central Pyrenees (modified from ICC 2002), with sample positions and location of cross-section shown in c. c) Geological cross–section drawn from the ECORS seismic profile (Muñoz 1992); sample positions have been projected on the section.

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Figure 2. Radial-plot and histogram representations (Vermeesch 2009) of the AFT counting data of samples AN01, 02, 03, and 04 combined. The radial plot is centred on the central age of the combined samples (79 Ma); grey lines indicate peak ages and the black dashed line represents the depositional age (65 Ma). Samples are individualised by different grey-shading of the data points.

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Figure 3. a) AHe ages plotted as a function of effective uranium content ( $eU = [U]+0.24\times[Th]$ ) for the four samples analysed; gray dashed line in a) represents exponential fit to the data. b) Sample AN05 has been separated from the others due to its high eU content and lack of correlation between eU and AHe ages, which contrasts with the other samples.

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Figure 4. Modelled *T-t* paths from joint AFT and AHe data for samples AN01, 02 and 03 (note variable timescales for these plots, constrained by the oldest single-grain AFT age). The black curves represent the expected model (weighted mean model) and its 95% confidence interval for the *T-t* paths; the yellow curve is the maximum likelihood model (best data fit). *T-t* paths are coloured according to their probability (scale on the right-hand side, which varies between samples). The gray boxes represent the prior constraints on the explored parameter space (see text for discussion) and dotted gray lines refer to the values of  $T_{max}$  and burial time discussed in the text.

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Figure 5. Sensitivity of AHe ages to eU variations. The curves represent predicted AHe ages as a function of eU for the *T-t* paths of the expected models (cf. Fig. 4), and for eU varying from 5 to 30 ppm. The dots represent the AHe data shown in Fig. 3 and Table 2.

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Figure 6. Effect of pre-depositional exhumation history on AHe ages. a) Forward model set up: AHe ages are predicted using the He-diffusion model of Flowers *et al.* (2009) for varying eU contents,  $T_{max}$ and starting times of pre-depositional exhumation ("exhumation 1"). b) AHe ages modelled as a function of eU content and starting time of the pre-depositional phase of exhumation, for two different maximum burial temperatures ( $T_{max} = 70$  and 80 °C respectively). Measured ages of our samples are plotted as black squares, or represented as dashed lines when they do not resolve a starting time.

- 754 Figure 7. Topographic profile along the same transect as A-A' in Figure 1b, showing the inferred
- palaeo-elevation of the Oligocene-Miocene deposits inferred from thermal-history modelling of the
- AHe and AFT data for samples AN02, AN03 and AN01 (dashed line is best estimate, orange band is
- 757 uncertainty), compared to the inferred maximum elevation over the southern Axial Zone from
- 758 inversion of *in-situ* thermochronology data (Fillon & van der Beek 2012). Yellow shading in Sierra de
- 759 Gurp represents current exposure of Late Eocene–Oligocene conglomerates. Locations of major thrust
- faults in the SCU fold-and-thrust belt are indicated for comparison to Fig. 1c.
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