

# Contribution of magmatism, partial melting buffering and localized crustal thinning on the late Variscan thermal structure of the Agly massif (French Pyrenees)

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2 late-variscan thermal structure of the Agly massif (French Pyrenees)

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#### 15 Abstract

Low pressure-high temperature (LP-HT) metamorphism, with geothermal gradients in the order of 16 50 to 100°C/km, is a common feature of the late evolution of collisional orogens. These abnormal 17 thermal conditions may be the results of complex interactions between magmatism, metamorphism 18 and deformation. The Agly massif, in the French Pyrenees, preserves the metamorphic footprints of 19 the late Variscan thermal structure of an almost continuous section from the upper and middle 20continental crust. The upper crust is characterized by a very high geothermal gradient of ~55 21 °C/km, evolving from greenschist to amphibolite facies, while the middle crust, exposed in a 22 gneissic core, exhibits granulite facies conditions with a near isothermal geothermal gradient 23  $(<8^{\circ}C/km)$  between 740 and 790°C. 24

The abnormal and discontinuous crustal geothermal gradient, dated at c. 305 Ma on syn-granulitic 25 monazite by LA-ICPMS, is interpreted to be the result of magmatic intrusions at different structural 26 levels in the crust: the Ansignan charnockite (c. 305 Ma) in the deepest part of the gneissic core, the 27 Tournefort granodiorite (c. 308 Ma) at the interface between the gneissic core and the upper crust 28 and the Saint-Arnac granite (c. 304 Ma) in the upper section of the massif. The heat input from 29 these magmas combined with the thermal buffering effect of the biotite dehydration-melting 30 reaction resulted in the near-isothermal geothermal gradient in the gneissic core (melt-enhanced 31 geotherm). The higher geothermal gradient (> 50°C/km) in the upper crust is only due to 32 conduction between the hot middle crust and the Earth's surface. 33

The estimated maximum finite pressure range suggests that about 10 to 12 km of crust are exposed in the Agly massif while the present day thickness does not exceed 5 to 6 km. This pressure / depth 36 gap is consistent with the presence of several normal mylonitic shear zones that could have 37 contributed to the subtraction of about 5 km of the rock pile. Monazite U-Th-Pb ages carried out on 38 monazite overgrowths from a highly mylonitized sample suggest that this vertical thinning of the 39 massif occurred at c. 296-300 Ma.

This later Variscan extension might have slightly perturbed the 305 Ma geothermal gradient, resulting in an apparent higher conductive geothermal gradient in the upper crust. Although the Agly massif has been affected by Cretaceous extension and Eocene Alpine compression, we suggest that most of the present-day thickness of the column rock was acquired by the end of the Paleozoic.

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**INTRODUCTION** 

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Low pressure-high temperature metamorphism (LP-HT) in the continental crust is defined by high 50 geothermal gradient (60-150°C/km) and temperature reaching about 600-800 °C, at pressure below 51 0.4 GPa. These conditions are usually reached in contact metamorphism settings, resulting in 52 hornfels and possible partial melting of pelitic rocks close to the contact with the intrusive rocks. 53 Nevertheless, numerous examples for LP-HT metamorphic terranes have been ascribed to regional 54 metamorphic environments (De Yoreo, Lux, Guidotti, Decker, & Osberg, 1989; Kim & Cho, 2003 55 Gibson, 1991; Okay et al., 2013; Yardley, Barber, & Grey, 1987). Such high geothermal gradients, 56 much higher than steady state gradients for thickened crust, require a considerable heat flux to reach 57 such thermal state in the continental crust and are more common in Archean cratons (Brown, 2007; 58 2009). They have been commonly ascribed to crustal underplating of mantle-derived basic magma 59 (Thompson & England, 1984), intrusion of large felsic magma in the middle and upper crust (De 60 Yoreo et al., 1989; Lux, De Yoreo, Guidotti, & Decker, 1986) or Metamorphic Core Complexes 61 (Coney, 1980). 62

These regional LP-HT metamorphic gradients are commonly reported in gneissic domes and often associated with magmatism and partial melting (De Yoreo et al., 1989; Kim & Cho, 2003). Over the past decades, numerous studies have highlighted the important role of partial melting, thermal buffering and advective heat transfer due to melt migration towards the structurally upper levels to produce this particular thermal structure of the crust (Depine, Andronicos, & Phipps-Morgan, 2008; Riel, Mercier, & Weinberg, 2016). One of the key feature of these studies is that the combined effect of radioactive heat production, high mantle heat source, heat diffusion, latent heat of melting

and heat advection by melt transfer result in discontinuous thermal structure of the continental crust. 70 The upper crust is characterized by high to very high LP-HT geothermal gradient. In contrast the 71 middle to lower crust is partially molten and characterized by a low to very low geothermal 72 gradient. This thermal structure has been modeled and defined by Depine, Andronicos, & Phipps-73 Morgan (2008) as a melt-enhanced geotherm. This discontinuous crustal thermal structure has been 74 described in various crustal settings including collisional setting, in the French Massif Central 75 (Trap, Roger, Cenki-Tok, & Paquette, 2016) and Himalaya (Groppo, Rolfo, & Indares, 2012), and 76 continental arc settings such as the Andean active margin in Ecuador (Riel et al., 2016). 77

Deciphering the thermal structure of the crust at a specific geological time (i.e. the geothermal 78 gradient) relies on the estimation of pressure and temperature conditions as well as dating peak 79 metamorphism for different rocks along a metamorphic field gradient-related cross-section. 80 Unfortunately, this task is not straightforward due to potential kinetics effect for amphibolite facies 81 rocks that makes part of the upper crust. Indeed, estimated peak temperature from equilibrium 82 thermodynamic modeling of phase relation may not reflect the "real" temperature because of kinetic 83 factors that delay the onset of nucleation and growth of metamorphic phases (Carlson, Pattison, & 84 Caddick, 2015; Pattison & Tinkham, 2009; Waters & Lovegrove, 2002). Additionally, field 85 metamorphic gradient may not be only the result of heat transport but could also be modified by 86 deformation processes. Heterogeneous deformation leads to low strain domains preserving relict 87 and metastable assemblages partially re-equilibrated while high strain domains are fully re-88 equilibrated. Localized crustal shear zones can also modify metamorphic gradient by juxtaposing 89 terrains of different grades, disrupting the observed metamorphic gradient and leading to transient 90 geotherms. 91

LP-HT metamorphism is a common feature in the Variscan belt, especially in Pyrenean massifs: St 92 Barthélémy (Saint Blanquat, Lardeaux, & Brunel, 1990; Saint Blanquat, 1993), Canigou (Gibson, 93 1991), Aston-Hospitalet (Barnolas & Chiron, 1996; Denèle, Olivier, Gleizes, & Barbey, 2007). This 94 LP-HT metamorphism is usually associated with the development of gneissic domes and a 95 widespread late Variscan magmatism between 300-320 Ma (Cochelin et al. 2017, ref therein). 96 Different geodynamic models have been proposed that might be responsible for the LP-HT 97 gradients: (1) early extension with thinning during continental rifting associated with mantle-98 derived magmas (Wickham & Oxburgh, 1985; Wickham, Oxburgh, Reading, & Vissers, 1987), (2) 99 late extension after crustal thickening (Van den Eeckhout & Zwart, 1988) or (3) compressional 100 setting with thickening associated with numerous intrusions in the crust and partial melting (De 101 Yoreo et al., 1989; Lux et al., 1986). 102

In the easternmost part of the French Pyrenees, the Agly massif is one of these gneiss domes that preserves a complex HT-LP late Variscan tectono-metamorphic evolution (figure 1a). However, its

present-day geometry is not only due to Variscan tectonics but also results from the superposition of 105 late Cretaceous extension and Eocene Alpine compression (Clerc & Lagabrielle, 2014; Vauchez et 106 al., 2013). The late Variscan metamorphism event is characterized by a high geothermal gradient in 107 the upper crust of about 80 to 100°C/km (Fonteilles, 1976; Guitard, Vielzeuf, & Martinez, 1996; 108 Vielzeuf 1984) and up to 150°C/km is some localities (Delay, 1989), partial melting in the middle 109 crust and different generations of magmatic intrusions (Delay, 1990; Fonteilles, 1976; Guitard et al., 110 1996). A detailed review of the bibliography reveals that quantitative thermobarometry has never 111 been published on the metamorphic gradient of the whole Neoproterozoic-Paleozoic sequence. 112 Furthermore, the available P-T estimations are restricted to the gneissic core, and mostly to the 113 deepest part near the Ansignan charnockite. 114

This paper is aimed at defining quantitatively the P-T conditions along the metamorphic gradient, 115 the age and thermal structure of the Agly massif during the Late-Variscan orogeny. A suite of 116 samples located at different structural levels were collected along an E-W cross section to constrain 117 peak P-T conditions via phase relation forward modeling. Two distinct lithologies have been used: 118 Ordovician pelites from the upper crustal level and Proterozoic pelites (kinzigites) from the genissic 119 core, in order to minimize uncertainties on thermobarometric estimates and highlight possible 120 metamorphic discontinuities along the metamorphic gradient. In-situ LA-ICPMS U-Th-Pb monazite 121 dating has been used to constrain the timing of peak metamorphism and compared to previously 122 documented magmatic ages. Finally, we will discuss and propose a conceptual model taking into 123 account the role of partial melting, heat advection during magmatism and localized deformation 124 responsible for the finite shape of the HT-LP discontinuous Variscan geotherm. 125

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# 128 **2 GEOLOGICAL SETTING**

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#### 130 2.1 The Agly massif in the Pyrenees

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The Pyrenees mountain belt was formed during the Tertiary collision of the Iberian microplate and the Eurasian plate (e.g. Choukroune, 1992). The Axial and Northern Pyrenean zones are characterized by a suite of exhumed Variscan massifs (e.g. Aston, Hospitalet, Canigou, Trois-Seigneurs, Agly), that consist of metasediments, para- and orthogneisses, migmatites and plutonic rocks, thrusted and juxtaposed to Mesozoic and Cenozoic covers.

137 The Agly massif is the easternmost Variscan massif of the Northern Pyrenean Zone in France 138 (Figure 1a). The southern boundary of the massif is an alpine NW-SE-trending and vertical fault

(Bélesta-Trilla fault) that brings into contact Variscan magmatic and metamorphic rocks with the 139 Boucheville syncline, consisting of Mesozoic sedimentary rocks (up to Albian) metamorphosed at 140 high temperature during Cretaceous time (~95 Ma) (Delay, 1990 and references therein). On its 141 northwestern edge, the Variscan Agly massif is locally thrusted northward over the Mesozoic cover 142 (Triassic formation) of the Saint-Paul de Fenouillet syncline (Figure 1). In contrast, the same 143 contact towards the northeast has been recently interpreted as a detachment fault that juxtaposed the 144 Agly massif with the Mesozoic Agly syncline that has been metamorphosed during Cretaceous time 145 (Vauchez et al., 2013). Although it is well acknowledged that both northern contacts have been 146 tectonically reactivated during the Tertiary orogeny, there is a controversy about the age of this 147 extensional tectonic history that could be either Paleozoic or Cretaceous (Vauchez et al., 2013; 148 Olivier, 2013). This controversy has been rejuvenated by two recent studies using (U-Th)/He 149 (Ternois et al., 2019) and U-Pb (Odlum & Stockli, 2019) in both zircon and apatite. The first study 150 concluded that the Agly massif acquired most of its present-day geometry during Paleozoic times 151 while the second argued that most of the thinning occurred during pre-Alpine hyper-extension. 152

Distinct geodynamic models have been proposed to account for these structural, metamorphic and 153 magmatic features. Bouhallier, Choukroune, and Ballèvre (1991) proposed a late Variscan 154 extensional setting after crustal thickening, while Paquet and Mansy (1991) rather suggested that 155 crustal thinning may have occurred during Cretaceous time. On the other hand, Delay (1990) argued 156 for a late Variscan compressional setting with heat provided by a charnockitic intrusion in the lower 157 part of the massif. More recently, a bulk compression tectonics associated with a transpressive 158 setting has been proposed to explain the structural evolution of the Agly massif (Olivier, Gleizes, & 159 Paquette, 2004; Olivier, Gleizes, Paquette, & Muñoz Saez, 2008). It appears from this review that 160 there is no consensus on the geodynamic model, and this situation may have arisen partly because 161 the geothermal gradient (thermal structure) and the age of metamorphic and deformation events 162 have not been well constrained. 163

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#### 165 2.2 Lithological units

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Since the pioneer work of Fonteilles and Guitard (1964) and Fonteilles (1970; 1976), the Variscan Agly massif has been subdivided into two main units: (1) A metamorphosed Paleozoic sequence, mostly exposed in the northeastern part of the massif with a thickness of ~2.5 to 3 km, that structurally overlies (2) a high grade gneissic core of presumed late Proterozoic age in the southwestern part of this area. Both units are intruded by Variscan to late Variscan plutonic bodies (Ansignan charnockite, Tournefort diorite and the Saint-Arnac granite). The two main units were 173 classically interpreted as a sedimentary cover on top of an older basement, respectively (Fonteilles174 & Guitard, 1964), but this interpretation is revised in this contribution.

The upper part of the Paleozoic sequence consists of Devonian limestones with minor and 175 discontinuous Silurian black phyllites and locally Late Ordovician (Caradoc) volcano-clastic rocks. 176 The lower part of the sequence is a monotonous and thick succession composed of an alternation of 177 siltstone and sandstone layers (Jujols group of Laumonier (1998)). Recent biostratigraphic age data 178 obtained on comparable rocks, on the southern slope of the Canigou massif (in the Pyrenean Axial 179 Zone, south of the Agly massif), suggest that the Jujols group may be of Late Cambrian - Early 180 Ordovician age and that most of the Early to Middle Ordovician sequence was eroded prior to the 181 deposition of the Late Ordovician volcano-clastic rocks during the Sarde tectonic event (Casas & 182 Palacios, 2012). The entire Paleozoic sequence has been affected by a low pressure-high 183 temperature (LP-HT) metamorphism that increases downward from lower greenschist facies 184 conditions at the top, leading to the transformation of Devonian and Silurian rocks into marbles and 185 calc-schists, respectively, to upper amphibolite facies conditions with partial melting involved, at 186 the bottom of the Jujols group (Fonteilles, 1970). 187

The "high grade gneissic core" is heterogeneous and consists of partially-melted metapelites 188 (locally garnet-cordierite-sillimanite-bearing gneiss) and metagreywackes interbedded with 189 190 discontinuous and highly deformed layers of calc-silicate gneisses and marbles. These metasedimentary are inter-layered with orthogneisses of variable thickness (on the order of meters) 191 which were emplaced as granitic sills during Early Cambrian at c. 530-540 Ma (Tournaire Guille, 192 Olivier, Paquette, Bosse, & Guillaume, 2019). In the western part of the massif, the uppermost part 193 of the gneissic core is intruded by the kilometer thick Riverolles orthogneiss. The "gneissic core" is 194 separated from the Paleozoic Jujols group by a discontinuous intercalation of white metacarbonate 195 (the so-called "base marble"), which in some cases is present few meters inside the gneisses. All 196 these observations suggest that the metasediments of the "gneissic core" of the Agly massif are a 197 highly metamorphosed equivalent to the Late Neoproterozoic sediments of the Canaveilles group, 198 observed for instance south of the Agly massif, in the Canigou dome (e.g. Cocherie et al., 2005; 199 Laumonier, 1998). The Late Neoproterozoic age of the sediments was first suggested by Autran, 200 Fonteilles, & Guitard (1966) and Vitrac-Michard and Allegre (1975), but they argued for a cover 201 and pre-Variscan basement relationships (see review in Padel, 2016). 202

The Paleozoic sequence (Jujols group) and the gneissic core (Canaveilles group) were intruded by late Variscan plutons. In the southwestern part of the massif, the deepest part of the gneissic core was intruded by the Ansignan charnockite. This intrusion is a 500 to 800 m thick laccolith parallel to slightly discordant to the main foliation and consists of an orthopyroxene-bearing granodiorite with megacrysts of K-feldspar defining an internal magmatic foliation. The age of magmatic

emplacement of this intrusion was initially constrained at  $314 \pm 6$  Ma by Respat and Lancelot 208 (1983) on monazite grains, and more recently re-evaluated at  $307 \pm 3$  Ma (Tournaire Guille et al., 209 2019) on zircons. The upper part of the gneissic core was intruded by the meter-thick Cassagnes 210 sills at the same time ( $308 \pm 3$  Ma, Tournaire Guille et al., 2019), and may be equivalent to the 211 Ansignan intrusion except that it is orthopyroxene-free and was emplaced in an upper structural 212 level. In the northwestern part of the Agly massif, the Paleozoic sequence was intruded by the late 213 Variscan Tournefort Diorite and the Saint-Arnac granite at  $308 \pm 1$  Ma and  $304 \pm 5$  Ma, 214respectively (Olivier et al., 2008). While the Tournefort diorite was emplaced at the base of the 215 Jujols Group sequence, the Saint-Arnac granite intruded the entire Jujols group. Emplacement of 216 the late Variscan Tournefort diorite and Saint-Arnac granite induced a local contact metamorphism 217 with the development of partial melting around the diorite and late muscovite crystallization around 218 the granite contact and in the metasediment septas present in the granite. 219

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#### 221 2.3 Tectono-metamorphic evolution

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The Paleozoic sequence shows a progressive downward increase of metamorphic conditions, with the following succession of isograds: chlorite out, biotite in, cordierite in, and alousite in, sillimanite in, muscovite out and finally water saturated partial melting. Based on semi-quantitative phase diagram projections and the estimation of the thickness of the Paleozoic sequence, the late-Variscan geothermal gradient has been estimated between 130 and 150 °C·km<sup>-1</sup> (Fonteilles, 1970) or at 80 °C·km<sup>-1</sup> (Delay, 1990).

The gneissic core recorded a more homogenous upper amphibolite to granulite facies conditions of 229 metamorphism. Nevertheless, the gneissic core is classically divided into two structural levels based 230 on their metamorphic assemblages (Fonteilles, 1970): the Belesta gneisses forming the upper part 231 with K-feldspar-sillimanite-bearing assemblages, and the Caramany gneisses in the deepest part 232 with orthopyroxene-bearing assemblages. Metamorphic conditions estimated on the Ansignan 233 charnockite or nearby lithologies have been estimated at  $0.6 \pm 0.1$  GPa and  $750 \pm 50$  °C (Andrieux, 234 1982),  $0.5 \pm 0.05$  GPa and  $800 \pm 100$  °C (Vielzeuf, 1984) or  $0.4 \pm 0.05$  GPa and 600-650 °C 235 (Delay, 1990). 236

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#### 238 2.4 Deformation

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240 The metamorphic rocks of the upper Paleozoic sequence and the lower gneissic core share a 241 predominant strain fabric observed in the entire Agly massif that corresponds to a moderately to shallowly dipping HT/LP foliation, holding a conspicuous N20 trending mineral and stretching lineation (e.g. Bouhallier et al., 1991; Olivier et al., 2004). Similarly, the Ansignan charnockite and the Cassagnes intrusives show a well-developed magmatic foliation, with a minor component of solid-state deformation. This magmatic foliation is parallel to the main LP/HT foliation of the surrounding gneisses (Delay, 1989; Olivier et al., 2004).

In addition to the main foliation, the gneissic core is characterized by the development of low angle 247 extensional mylonitic shear zones, mainly visible in the eastern part of the massif, dipping either 248 toward the north or toward the south and associated with a strong stretching lineation that trends 249 N20 (Delay, 1989). The main foliation and the mylonitic shear zones show a similar normal and 250 top-to-the north kinematics attributed to a same tectono-metamorphic event responsible for the 251 thinning of the Variscan crust (Vanardois et al. in press). Olivier et al. (2004; 2008) suggest a local 252 thinning during a late Variscan transpressive regime with the mylonitic normal shear zones that 253 developed in response to late doming. In contrast, Bouhallier et al. (1991) documented a major 254 detachment zone located between the Paleozoic sequence and the gneissic core (the Caladroy-255 Latour shear zone) that would be responsible for the subtraction of about 10 km of crustal material 256 and consequently the juxtaposition of the low pressure-medium temperature Paleozoic sequence 257 with the moderate pressure-high temperature gneissic core. The age of mylonitic shearing (either in 258 a context of crustal thinning or transpression) is still a matter of debate. For Delay (1990) and 259 Bouhallier et al. (1991), the extension is contemporaneous with the HT-LP metamorphism and 260 therefore would be late Variscan (c. 315 Ma). On the contrary, Olivier et al. (2008) argued that the 261 root of the ~304 Ma Saint-Arnac and Tournefort plutonic body underwent northward normal 262 shearing suggesting that the development of mylonitic shear zones took place just after 304 Ma. In 263 contrast, Paquet & Mansy (1991) suggested that the thinning responsible for the loss of ~ 10 km of 264 crustal material occurred during Mid-Cretaceous time (110-100 Ma). 265

The E-W trending fault that bounds the southern limb of the Mesozoic Agly syncline (north-eastern 266 part of the Agly massif) has recently been interpreted as an ancient Mid-Cretaceous detachment 267 fault (~100 Ma), with a top-to-the-north shearing sense (Vauchez et al., 2013). This contact may 268 have contributed to the exhumation of the Agly massif during the Iberian microplate rotation 269 relative to Eurasia and opening of the Bay of Biscay during or after Albian (Vauchez et al., 2013). 270 Vauchez et al. (2013) also suggested that some of the extensional mylonitic shear zones observed in 271 the Variscan basement, and described above, may also be related to this Mid-Cretaceous 272 extensional event. Furthermore, this Mid-Cretaceous deformation is coeval with an intense episode 273 of fluid circulation and fluid-rock interactions that are responsible for the local albitization of the 274 Saint-Arnac granite, the paragneisses of the gneissic core and, locally, the Paleozoic sequence 275 (Poujol, Boulvais, & Košler, 2010). 276

Most of the tectonic contacts bounding the Agly massif to the north and the south have been reworked during the Tertiary orogeny corresponding to the N-S convergence between Iberia and Eurasia during Eocene time. For instance, the reactivation of the western part of the upper Cretaceous detachment, described above, corresponds to the northward thrusting of the Agly massif over the Mesozoic cover of the Saint-Paul de Fenouillet syncline (Figure 1). Alpine deformation is also visible within the Agly massif where Mesozoic carbonate cover and Early Eocene Breccias are pinched into the high-grade rocks between steeply dipping N110 trending Alpine faults (Figure 1b).

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# **3 METAMORPHIC GRADIENT ESTIMATION**

287 As introduced in the previous section, the N20 pervasive foliation of the main metamorphic event 288 (LP/HT) defines a single tectono-metamorphic event throughout the entire massif that can be linked 289 to the Charnockite emplacement at 305 Ma. This lineation is similar to the stretching lineation of all 290 mylonitic shear zones. Thus, sampling along the metamorphic gradient should be in a N-S direction 291 in order to highlight the possible role of mylonitic shear zones on the finite metamorphic gradient 292 shape. However, the occurrence of major East-West trending post-Mesozoic steep faults and 293 Pyrenean thrusts that may have juxtaposed blocks of different structural level precludes a simple 294 north-south sampling. Furthermore, in the central part of the Agly massif, where the deepest part 295 located around the Ansignan charnockite is exposed, the metamorphic gradient in the Paleozoic 296 sequence is obliterated by the St Arnac granitic intrusion. For these reasons, we have chosen to 297 sample the different structural levels along an E-W cross section (Figure 2), from Força Real on the 298 Eastern side of the massif, towards the lowest structural levels near the Ansignan charnockite on the 299 western side. The location of this cross section is represented on the map (Figure 1b). The GPS 300 coordinates for each sample can be found in Table 1. 301

# 302 3.1 Petrology

# 303 3.1.1 Metamorphic evolution of the Late Cambrian - Early Ordovician 304 sequence (Jujol group)

The Agly massif preserves a typical LP-HT metamorphic gradient in the cover as suggested in previous studies (Fonteilles, 1970; Guitard et al., 1996; Vielzeuf, 1984) and referred to here as the upper Paleozoic sequence (Jujol group). The sequence of mineral isograds is biotite-in, closely followed by chlorite-out, cordierite-in, andalusite-in, sillimanite-in, muscovite-out/K-feldspar-in and onset of partial melting. We describe here the different reactions that could lead to the appearance and disappearance of these index minerals based on the petrography and available petrogenetic grids (Pattison & Tracy, 1991; Spear & Cheney, 1989). For clarity, all reactions are represented in the simplified KFMASH system. Consequently, the role of plagioclase and other Cabearing phases is not taken into account to describe the main phase transitions.

The first observable isograd of the LP-HT metamorphic gradient is the biotite-in. This reaction results in the appearance of small biotite flakes in the fine-grained chlorite-muscovite bearing schists. Biotite content continuously increases with increasing metamorphic grade. The low-grade chlorite-muscovite schists preserve evidences of a bedding that corresponds to an alternation of millimeter to centimeter thick phyllosilicates-rich and quartz-rich layers (Figure 3a).

The very fine-grained nature of the chlorite schists makes difficult the petrographic observation, and although we have not observed K-feldspar, we suggest that the first appearance of biotite is the result of the simplified reaction:

322

323

$$Chl + Kfs = Ms + Bt + Qtz + H_2O$$
 (1)

324

This reaction is consistent with the observation of micro-lithons bearing chlorite and surrounding preferred orientation of muscovite and biotite parallel to the HT/LP foliation (caption Figure 3a, see S cleavage).

In the field, the continuous increase of biotite content results in a striking change in color and grain size from a greenish and very fine-grained chlorite-muscovite schist to a darker and coarser biotitemuscovite schist (Figure 3b). The darker color is mostly due to the presence of graphite derived from organic matter, present until partial melting.

The next isograd corresponds to the appearance of cordierite porphyroblasts together with the complete disappearance of primary chlorite (Figure 3c). This isograd is immediately followed by the appearance of andalusite in the stability field of cordierite. Preferred orientation of cordierite and andalusite porphyroblasts underline the orientation of the main N20 stretching lineation. In high-aluminum pelites, several chlorite dehydration reactions can be responsible for the crystallization of cordierite at low pressure. All of them involve muscovite, chlorite and quartz as reactant phases and biotite and  $H_2O$  as products:

339

340	$Chl + Ms + Otz = Bt + Crd + H_2O$	(2)
540	$C_{III} + III_{III} + Q_{III} - D_{II} + C_{III} + II_{III} - D_{III} + C_{III} + II_{III} - D_{III} + C_{III} + C$	(2)

$$Chl + Ms + Qtz = Bt + Crd + And + H_2O$$
 (3)

$$342 Chl + Ms + Qtz = Bt + And + H_2O (4)$$

343

Based on field observations, cordierite appears before andalusite. This together with the pattern of 344 inclusions inside andalusite visible in figure 3d (i.e. only biotite and quartz without core to rim 345 differences) suggest that cordierite grew outside the stability field of andalusite due to reaction (2). 346 Nevertheless, this interpretation would require a more Mg-rich bulk composition. Several factors 347 can explain this compositional discrepancy. Measured bulk  $X_{Mg}$  composition varies from ~ 0.4 to 348 0.5 (see AFM diagram figure 4), which is very close to both chlorite and biotite that are close to 0.5 349 in X<sub>Mg</sub>. Thus, small retrograde changes in X<sub>Mg</sub> of these phases can potentially override the prograde 350 compositions and make interpretations based on AFM more difficult. Additionally, iron-sulfide is 351 usually present in such rocks, but bulk rock composition was not projected since no major element 352 of oxides were constrained. This would change the  $X_{Mg}$  of the equilibrium volume considered, and 353 most likely would be a function of temperature since iron sulfides usually vary along the pyrite-354 355 pyrrhotite join.

The pattern of inclusions inside and alusite together with the progressive disappearance of cordierite in the stability field of and alusite is interpreted as the consequence of the reaction:

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$$Ms + Crd = Bt + And + Qtz + H_2O$$
(5)

360

The next encountered isograd is the appearance of fibrolitic sillimanite (Figure 3e), that forms millimeter-size nodules elongated within the main stretching direction. In one sample, we found evidence of polymorphic breakdown of andalusite into sillimanite (reaction 6), having the two minerals coexisting but never in direct contact as observed by many authors (e.g. Carmichael, 1969). However, the sillimanite isograd mapped in the field rather corresponds to the muscovite breakdown reaction (reaction 7) that produced large cluster of fibrolite, which are easier to observe in the field.

368

369

$$And = Sil$$

(6)

$$370 Ms + Qtz = Kfs + Sil + H_2O (7)$$

371

The last observed isograd in the upper Paleozoic sequence corresponds to the onset of partial melting that is characterized by the formation of quartzo-feldspatic leucosomes parallel to the HT/LP main foliation (Figure 3f). In the field, the melt-in isograd is always located a few tens of meter away from the muscovite-out isograd that corresponds to reaction (7). Onset of partial melting is assumed to be water saturated or near water saturation. Partially molten micashists host tourmaline-bearing pegmatitic bodies that range in thickness from a few centimeters up to a several decameters. In the vicinity of these pegmatites, the micaschists are characterized by the 379 crystallization of late euhedral muscovite that cross-cut the main foliation (Ms2 in figure 3e).

In the field, the thickness of the partially molten micaschist zone represents only a few decameters 380 to a hundred meters. Then the molten micaschists evolve into a biotite-rich quartzo-feldpathic 381 gneiss characterized by minute porphyroblast of K-feldspar. This lithology has been referred by 382 Delay (1990) as the "pearled gneiss". Partial melting evidences in this lithology are rare and when 383 present consist of discrete dismembered leucosomes. We suggest that the onset of partial melting 384 resulted in a strong fractionation of water into the melt leading to local fluid-undersaturation and 385 therefore subsolidus conditions. Furthermore, in most locations, including Caladroy, the "pearled 386 gneiss" is strongly deformed and consists in mylonitized partial melted micaschists. Bouhallier et. 387 al (1991) described a high strain zone in Caladroy having an abrupt transition between the 388 micaschists and the gneisses. The transition between micaschists and pearled gneisses is also 389 characterized by a discontinuous intercalation of a marble layer, the so-called "base marble", that 390 can be located either at the interface between the partially molten micaschists and the gneiss or 391 within the gneiss itself. To conclude, we propose that the "pearled gneiss" is derived from the same 392 Early Ordovician protolith as the overlying micaschist and schist, and is produced by concomitant 393 water-undersaturated metamorphism and strain localization. 394

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396

#### 397 **3.1.2 Kinzigites of the gneissic core**

The gneissic core, located structurally below the metamorphosed upper Paleozoïc sequence (Jujol 398 group), is mainly composed of paragneisses, orthogneisses and metacarbonates, the latter being 399 more abundantly present in the deeper part of the massif (Figure 2). In this study, we have chosen to 400 focus our petrological analysis on kinzigites (bt-sil-crd-grt bearing gneisses) that occur as 401 discontinuous meter-scale layers at different structural levels in the gneissic core (Figure 2). These 402 rocks are good candidates for calibrating P-T peak conditions across the entire gneissic core using 403 P-T pseudosections. The studied samples show the same peak metamorphic mineralogical 404 assemblage that consists of cordierite + garnet + sillimanite + biotite + alkali feldspar + quartz  $\pm$ 405 plagioclase (see samples 10Ag04, 10Ag11 and 11Ag03, Figure 4). Sillimanite occurs either as 406 elongated prismatic grains, which define, with biotite, the main foliation wrapping around garnet 407 and cordierite porphyroblasts (Figure 4a), or small needle inclusions in garnet, together with quartz 408 and biotite; sillimanite commonly exhibits both textural positions in the same sample. Although 409 evidences of partial melting are unclear at the thin-section scale due to post-peak deformation, field 410 observations clearly suggest that these rocks were partially molten. All these minerals are stable at 411 peak metamorphic conditions, and are consistent with biotite dehydration melting reaction: 412

413

414

$$Bt + Sil + Qtz = Grt + Crd + Kfs + H_2O \text{ or } L$$
(8)

415

416 Some studied kinzigites also show late shear zones where a high-temperature retrogression 417 assemblage of Bi + Sill is observed. Samples 10Ag04, located near the Trilla bridge, shows that the 418 peak metamorphic assemblage is partially overprinted by brittle deformation that results in the 419 cataclasis of garnet porphyroblasts (Figure 4b). Similarly, sample 11Ag03, near the Cuxous castle, 420 is affected by numerous extensive ductile to brittle-ductile shear zones without low-temperature 421 retrogression, except for a strong retrogression of cordierite and K-feldspar (Figure 4c).

422

423

## 424 **4 THERMOBAROMETRY**

425

To constrain the P-T conditions along the metamorphic gradient, pseudosections were computed using two thermodynamic modelling programs, Perple\_X (Connolly & Petrini, 2002, Connolly, 2005) and Domino/Theriak (De Capitani & Petrakakis, 2010). Whole rock analyses are from XRF analyses and are presented in table 1. Since no mineral zoning and very limited retrogression was observed, the bulk rock XRF analyses represent a good estimate of the equilibrium volume for both the HT-LP rocks and the rocks from the gneissic core.

# 433 4.1.1 Late Cambrian – Early Ordovician LP-HT gradient

In the case of the Late Cambrian - Early Ordovician sequence, the bulk chemistry is rather 434 homogeneous (Figure 5). The effect of bulk composition variability on phase relations and the 435 location of major phase boundaries are thus very limited (Figure 5b). Therefore, phase relations 436 were computed with the bulk composition of sample 10Ag16, in the middle of the sample 437 composition cluster (white dot, Figure 4). The P-T phase diagram section produced in 438 NCKFMASHT (Figure 6a) shares similarities with the P-T projection (petrogenetic grid) of 439 Pattison & Tracy (1991) that was used by Guitard et al. (1996) to constrain semi-quantitatively the 440 LP-HT metamorphic gradient in the Agly massif for the first time. In this contribution we 441 rejuvenate this approach quantitatively with more petrological constraints and using recent 442 thermodynamic databases. 443

Figures 6b, c and 7 show that the metamorphic P-T conditions are bracketed in a temperature range from 500-520°C (chlorite breakdown, reaction (2) outside the stability field of andalusite) to 660446  $680^{\circ}$ C (muscovite breakdown, reaction (7) and the onset of partial melting at ~ 0.35 GPa.

Defining the exact temperature (and pressure) of intermediate samples characterized by the 447 appearance of andalusite and cordierite is more challenging because of the controversy regarding 448 the exact P-T slope and location of reaction (5) that involves both andalusite and cordierite. In the 449 computed NCKFMASHT phase diagram section (Figure 6a), the positive slope of the reaction (5), 450 here represented by the field Ms + Crd + Bt + And + Qtz + Pl + Ilm (labelled "Ms Crd Bt And" in 451 figure 6a), is in contradiction with field observations made in many low pressure contact aureoles as 452 well as the Agly massif, where cordierite appears just before andalusite (Pattison, Spear, Debuhr, 453 Cheney, & Guidotti, 2002; Pattison & Vogl, 2005; Pattison & Debuhr, 2015). As can be seen in 454 figure 6b, this inconsistency is not related to bulk compositional variations, although the position of 455 this equilibrium varies with X<sub>Mg</sub> bulk rock composition. 456

The reaction 5 has low entropy change, thus it is very sensitive to the quality of the thermodynamic 457 data. To overcome this known problem in low pressure metapelites, Pattison & Debuhr (2015) used 458 a modified Berman (1988) thermodynamic database (Pattison et al. 2002; Pattison & Debuhr, 2015; 459 Spear & Cheney, 1989; Spear & Pyle, 2010) instead of the Holland and Powell (1998) or (2011) 460 version; this database is referred to here as SPaC14 as in Pattison & Debuhr (2015). Their approach 461 was (1) to simplify the chemical system (excluding non-essential components like Na<sub>2</sub>O, CaO and 462 TiO<sub>2</sub>) to minimize uncertainties related to poorly constrained thermodynamic constant of pure end-463 member, and (2) to use simplified ideal site mixing solid-solutions. Following this approach, we 464 have computed the phase diagram section in a simplified KFMASH system shown in figures 6c and 465 6d using the 1998 version of the Holland and Powell thermodynamic database, updated in 2002, 466 and the SPaC14 database, respectively. The bulk composition used is the same as in figure 6a, 467 except that Na is projected through albite, Ca through anorthite and Ti through ilmenite, resulting in 468 a small variation in the molar amount of Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and FeO contents of the equilibrium volume 469 considered in the KFMASH system. As shown by Pattison & Debuhr (2015), the use of the SPaC14 470 database results in a phase diagram with a negative slope for the Crd + And + Bt + Ms equilibria 471 (5), that is more consistent with the appearance of cordierite and then and alusite along a prograde 472 path at ~ 0.3 GPa. In figures 6c and 6d, it is interesting to note that and alusite is predicted to be 473 stable at low temperature in equilibrium with chlorite and chloritoid (i.e. before the appearance of 474 cordierite). This feature is commonly described in contact aureoles, for example in the Bugaboo 475 aureole (Pattison & Debuhr, 2015) or the Little Cottonwood (Wohlers & Baumgartner, 2012), but is 476 in contradiction with field observations in the Agly massif. In the case of the Agly massif, we 477 suggest that the stabilization of andalusite and chloritoid is the result of excess aluminum in the 478 calculated bulk composition after albite and/or anorthite projection. At these P-T conditions, 479 paragonite is very likely to be stable but it is not taking into account in this modeling, resulting in a 480

481 higher Al<sub>2</sub>O<sub>3</sub> content of the effective bulk composition.

The main petrological constrains for the metamorphic gradient of the Paleozoic sequence are: 1) 482 chlorite breakdown and crystallization of biotite and cordierite, 2) crystallization of andalusite with 483 or without cordierite present, 3) and alusite/sillimanite polymorphic transformation, 4) muscovite 484 breakdown without cordierite stable, and 5) melting outside the stability field of muscovite. 485 Minimum pressure in the low-temperature side of the gradient (~550 °C) cannot be constrain using 486 solely the phase diagram. However, the thickness of rocks above the Paleozoic schists is inferred to 487 be approximatively 10 km (Olivier et al. 2004). Assuming a density of the crust of 2.7 g/cm<sup>3</sup> would 488 give a pressure around 0.27 GPa. The transition from andalusite to sillimanite, breakdown of 489 muscovite and partial melting occur on a very short distance in the field and a very limited 490 temperature range (620 to 670 °C) suggest that the maximum pressure is limited by the location of 491 the invariant point (i) at 0.35 GPa. Assuming a linear gradient, these two values (0.27 GPa at 550 492 °C and 0.35 GPa at 670 °C, allow us estimating the gradient at 55°C/km (2076 °C/GPa with 2.7 493  $g/cm^{3}$ ). Such value is in very good agreement with a conductive gradient between partially molten 494 schists at 670 °C and 0.35 GPa (i.e. a 13 km depth), and the surface since the resulting gradient 495 would be ~52 °C/km. This high value is however lower than the previous qualitative estimates of 80 496 to 100 °C/km (Fonteilles, 1976; Delay, 1989; Guitard et al., 1996; Vielzeuf 1984). 497

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#### 500 4.1.2 Kinzigites from the gneissic core

To constrain the P-T conditions in the gneissic core, we have selected 10 kinzigite layers 501 from various structural levels. The pristine kinzigite samples preserve the same peak mineral 502 assemblage produced by biotite dehydration-melting reaction, which has a steep slope in P-T space 503 (Figure 8a). In addition, divalent cations composition of garnet was used to better constrain P-T 504 conditions and highlight continuous or discontinuous changes in pressure and temperature along the 505 cross-section. A phase diagram section was computed for each sample with Perple\_X using whole 506 rock XRF composition. Garnet compositions were determined using electron microprobe with 507 wavelength dispersive spectrometry (WDS) during 3 sessions. The first two sessions were 508 conducted with a JEOL Superprobe 8200 at the Institute of Earth Sciences at the University of 509 Lausanne, the third session was on a Cameca SX100 at the University of Lille 1. Synthetic and 510 natural minerals were used for standardization. Average compositions and variations for the flat 511 core of garnets for each sample can be found in table 2, full analyses can be download with the 512 supplementary materials. 513

514

There is no discernable zonation at the level of precision of EPMA analyses (Figure 7),

except for small increase in Fe/(Fe + Mg) at the rim, due to retrograde interdiffusion with biotite. 515 Spessartine content can locally increase at the rim when garnet is retrogressed by biotite due to a 516 very high preference for incorporation of Mn in garnet compare to biotite (Kohn & Spear 2000). 517 The assumed equilibrium composition used to constrain P-T conditions were always coming from 518 garnet cores. Figure 8a is an example of pseudosection obtained for the sample 11Ag03. Almandine 519 and pyrope content in garnet are suitable to constrain P-T conditions because the slope of their 520 isopleths is either temperature or pressure dependent in the quadrivariant garnet – melt – bearing 521 stability fields located on both sides of the main equilibrium (8). There is generally a very good 522 agreement between almandine and pyrope compositions for all samples, with isopleths usually 523 located close from each other (Figure 8a). Spessartine and grossular content are very low, i.e. below 524 5 % of the X site of garnet, except for 4 samples (Table 2), thus they do not represent a good 525 constraint for P-T condition due to a limited accuracy of the thermodynamic database for these low 526 contents. 527

A synthesis of estimated P-T conditions is shown in figure 8c and shows a range in P-T 528 conditions from 0.49 GPa, 730°C to 0.65 GPa, 790°C. However, there is good agreement between 529 the pressure estimates from thermodynamic modelling and the relative structural position (i.e. 530 depth) of samples in the gneissic core (Figure 8c). Samples located in the upper part of the gneissic 531 core above the Ansignan charnockite intrusion have pressures ranging between 0.49 and 0.55 GPa 532 (Figure 8c). The minimum pressures of 0.50 to 0.52 GPa were obtained on samples 10Ag03, 533 10Ag09, 11Ag03 and 11Ag04 which are located a few tens to hundreds of meters below the sheared 534 contact with the Paleozoic sequence. The difference in pressure with the deepest sample form the 535 upper Paleozoic sequence (partially melted micaschist: 0.35 GPa at 670°C) is about 0.15 GPa. 536

Sample 10Ag11 located in the western central part of the massif and structurally in the 537 middle of the gneissic core gave P-T conditions of 0.57 GPa and 750°C. In contrast, samples 538 10Ag01 and 10Ag02 that are the deepest samples, below the Ansignan charnockite, are 539 characterized by pressures estimates of 0.66 and 0.62 GPa for temperatures of 770-800°C, 540 respectively. However, sample 10Ag04, that is also located below the charnockite and may belong 541 to the same kinzigite layer as 10Ag01 and 10Ag02, gave inconsistent pressure estimate of 0.52 GPa. 542 If we disregard sample 10Ag04, then the difference in pressure between samples located in the 543 deepest level and 10Ag11 is around 0.1 GPa (~2.7 km), which contrasts significantly with the 544 observed thickness on cross-section that does not exceed 200 m. 545

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# 5 U-Th-Pb MONAZITE GEOCHRONOLOGY

#### 551 5.1 Studied samples

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Three kinzigites (10Ag02, 10Ag04 and 11Ag03) have been collected in order to constrain the 553 554 timing of metamorphism. These samples are located at different structural levels and show various degree of deformation and retrogression. The deepest samples, 10Ag02 (Agly lake) and 10Ag04 555 (near the Trilla bridge), come from the same structural level (Figure 1 and 2). They are pristine 556 kinzigites that consist of garnet, cordierite, sillimanite, quartz and feldspar with minor biotite. 557 Sample 10Ag04 shows a more penetrative foliation with localized thin shear zones where garnet is 558 fractured and partially dismembered (Figure 9b). Sample 11Ag03 comes from the uppermost level 559 of the gneissic core, near the castle of Cuxous. It is characterized by the development of a mylonitic 560 foliation, normal shear sense ductile shear zones and brittle fractures that affect sillimanite and K-561 feldspar porphyroclasts (Figure 9c). Peak metamorphic phases like K-feldspar and cordierite are 562 almost completely pinnitised in this last sample. 563

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## 565 5.2 Analytical methods

566

In situ U-Th-Pb isotopic data were obtained by laser ablation inductively coupled plasma 567 spectrometry (LA-ICP-MS) at Laboratoire Magmas & Volcans (Clermont-Ferrand, France). The 568 analyses involved the ablation of minerals with a Resonetics Resolution M-50 powered by an ATL 569 Atlex Excimer laser system operating at a wavelength of 193 nm. For monazite, spot diameters of 9 570  $\mu$ m were used, associated with repetition rate of 1 Hz and laser fluence of 9 /cm<sup>2</sup>. The ablated 571 material was carried by helium and then mixed with nitrogen and argon before injection into the 572 plasma source of an Agilent 7500 cs ICP-MS equipped with a dual pumping system to enhance 573 sensitivity (Paquette et al., 2014). The alignment of the instrument and mass calibration were 574 performed before every analytical session using the NIST SRM 612 reference glass, by inspecting 575 the signal of  $^{238}$ U and by minimising the ThO+/Th+ ratio (< 1%). The analytical method for isotope 576 dating is similar to that developed and reported in Paquette and Tiepolo (2007) and detailed in 577 Hurai et al. (2010) and Gasquet (2010). The signals of <sup>204</sup>(Pb+Hg), <sup>206</sup>Pb, <sup>207</sup>Pb, <sup>208</sup>Pb, <sup>232</sup>Th and 578 <sup>238</sup>U masses were acquired. The occurrence of common Pb in the sample was monitored by the 579 evolution of the <sup>204</sup>(Pb+Hg) signal intensity, but no common Pb correction was applied owing to the 580 large isobaric interference from Hg. The <sup>235</sup>U signal was calculated from <sup>238</sup>U on the basis of the 581 ratio  ${}^{238}U/{}^{235}U= 137.88$ . Single analyses consisted of 30 seconds of background integration with the 582

laser off, followed by 60 seconds integration with the laser firing and a 20 second delay to wash outthe previous sample and prepare for the next analysis.

Data were corrected for U-Pb fractionation occurring during laser sampling and for 585 instrumental mass discrimination (mass bias) by standard bracketing with repeated measurements of 586 the Trebilcock monazite (Tomascak et al., 1996) reference material. Repeated analyses of the 587 monazites Itambé and Bananeira (Gonçalves et al., 2016) reference materials, treated as an 588 unknown independently control the reproducibility and accuracy of the corrections. Data reduction 589 was carried out with the software package GLITTER® from Macquarie Research Ltd (Jackson et 590 al., 2004; van Achterbergh et al., 2001). For each analysis, the time resolved signals of single 591 isotopes and isotope ratios were monitored and carefully inspected to verify the presence of 592 perturbations related to inclusions, fractures, mixing of different age domains or common Pb. 593 Calculated ratios were exported and reported in <sup>208</sup>Pb/<sup>232</sup>Th vs. <sup>206</sup>Pb/<sup>238</sup>U "Concordia" diagrams 594 generated using the Isoplot/Ex v. 2.49 software package of Ludwig (2001). Owing to the lack of 595 correlation between Th/Pb and U/Pb uncertainties, a low correlation coefficient value of 0.1 was 596 considered in the calculations. The concentrations of U-Th-Pb were calibrated relative to the values 597 of the GJ-1 zircon (Jackson et al. 2004) and Trebilcock (Tomascak et al., 1996) reference materials. 598 599

#### 600 5.3 **Results**

#### 601 5.3.1 Pristine kinzigite from the gneissic core (10Ag02 and 10Ag 04)

Monazites in sample 10Ag02 (Agly lake) are located in the matrix and range in size from 20 to 100 602 µm. Two distinct compositional domains are defined based on Y distribution: a Y rich core and Y 603 poor rim (Figure 10a). Th and U content is constant between both domains. Six monazite grains 604 were analyzed and most of the analyses are concordant in a <sup>232</sup>Th/<sup>208</sup>Pb vs <sup>238</sup>U/<sup>206</sup>Pb concordia 605 diagram (table 3, Figures 10a and 11a). Y-rich cores give the oldest ages, with <sup>232</sup>Th/<sup>208</sup>Pb ages 606 ranging from 240  $\pm$  7 Ma to 311  $\pm$  9 Ma. The four oldest analyses were selected to calculate a 607 concordant age at  $305 \pm 4$  Ma (MSWD<sub>(C+E)</sub> = 1.5). Y-poor rims show a spread of concordant ages 608 that range for c. 307 to 232 Ma, which precludes the calculation of concordant age (Figure 11a, red 609 610 circles).

Monazites in sample 10Ag04 are either present as inclusions in garnet or grains in the matrix. They range in size from 50 to 150  $\mu$ m (Figure 10b). Two compositional domains are defined based on Y and Th/U compositions. Monazite included in garnet (m2 and m5) and the core of monazite in the matrix (m3, m6, m8, m9) are characterized by high and variable Y content with a very constant Th/U of 6.5 ± 1 (Table 3, Figure 10b). Monazite in the matrix are all characterized by small overgrowths (10 to 20  $\mu$ m) with very low Y content (m1, m3, m4, m6, m8, m9) and a very high and

variable Th/U, ranging from 10 to 34 (Table 3, Figure 10b). Grain m4 is homogeneous in 617 composition, with low Y content and high Th/U ratio, it is interpreted to belong to the same 618 generation as the overgrowths based on its composition. Monazite included in garnet do not show 619 low Y and high Th/U overgrowth suggesting that the formation of the overgrowths is post-garnet 620 crystallization and monazite entrapment. Eight monazite grains were dated. Seventeen isotopic 621 analyses belonging to the five monazites with high Y content and low Th/U ratio were performed. 622 On a <sup>232</sup>Th/<sup>208</sup>Pb vs <sup>238</sup>U/<sup>206</sup>Pb concordia diagram, all sixteen analytical points are concordant and 623 allow to calculate a concordia age of  $306 \pm 2$  Ma (MSWD<sub>(C+E)</sub> = 1.05), which is consistent with the 624 concordia age obtained on 10Ag02 (Figure 11a and b). The constant Th/U ratio and the occurrence 625 of monazite included in garnet suggest that this age correspond to the age of the peak 626 metamorphism. The second compositional domain corresponding to the low Y and high Th/U 627 overgrowth, yield a second set of twelve concordant dates with a concordia age of  $300 \pm 2$  Ma 628  $(MSWD_{(C+E)} = 0.71)$  (Figure 11d). 629

#### 630

# 631 5.3.2 Deformed kinzigite of the upper gneissic core (11Ag03)

Five monazites located in the matrix were mapped. They range in size from 50 to 120 µm. Most of 632 the grains show homogeneous, large, Y-depleted core surrounded by an Y rich overgrowth that do 633 not exceed 10 µm in size (Figure 10c). Thorium content and distribution are homogeneous at the 634 grain scale. Monazite 1 differs from the others because it preserves an irregular very low Th core 635 (table 3 and Figure 10c). Three of the monazite grains analyzed (m1, m2 and m3, Figure 9c) show a 636 cluster of nine concordant dates that allow calculating a concordia age of  $296 \pm 2$  Ma (MSWD<sub>(C+E)</sub> 637 = 1.3) (Figure 11c). A single analysis has been obtained on the Th poor core of monazite 1 (#16d: 638  $^{232}$ Th/ $^{208}$ Pb age of 319 ± 10 Ma, table 3) and was not considered for the calculation of the concordia 639 age. The 296 Ma age is consistent within errors with the age obtained on the high Th/U monazite of 640 sample 10Ag04 (300  $\pm$  2 Ma) and can either be interpreted as the age of fluid percolation during the 641 mylonitic deformation or fluid exsolution from crystalizing melt of migmatites. Although monazite 642 m3 has the same compositional features as the other grains, it shows a spread of concordant dates in 643 the <sup>232</sup>Th/<sup>208</sup>Pb vs <sup>238</sup>U/<sup>206</sup>Pb concordia diagram. This spread in age along the concordia has been 644 observed in all analyzed samples, especially 10Ag02 and 11Ag03 (Figure 11a and c), it could 645 reflect a younger partial lead loss. We speculate that this isotopic disturbance could be the result of 646 localized fluid percolation during a Cretaceous event (ca. 100Ma) that affects the Pyrenees range 647 (Vitrac-Michard & Allègre, 1975, Boulvais et al., 2007, Poujol et al., 2010). 648

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Our new geochronological data combined with phase relation modeling suggest that peak metamorphism in the gneissic core was achieved at *c*. 305 Ma and that the massif was affected by deformation between 300 Ma and 296 Ma. The age of metamorphism in the upper Paleozoic sequence was not directly and quantitatively constrained by geochronology but we assume it is related to the same late Variscan event. In the following, we will see that the finite metamorphic field gradient observed in the Agly massif results from the superposition of a thermal (magmatic and metamorphic) event (305 Ma) and a later tectonic (296-300 Ma) event.

#### 661 6.1 Geothermal gradient during the main metamorphic event (c. 305 Ma)

Figure 12 shows the reconstructed metamorphic gradient for the entire rock column of the Agly 662 massif. It can be clearly seen that the geothermal gradient of the Agly massif is not linear and does 663 not result from a purely conductive heat transfer. The middle crust preserved in the gneissic core of 664 the Agly massif shows a very low to near isothermal gradient ( $\sim < 8^{\circ}C/km$ ), while the low pressure 665 assemblages in the upper crust (i.e. and alusite-sillimanite transition, cordierite stable) defines a 666 significantly higher geothermal gradient (> 50°C/km) in the Late Cambrian – Early Ordovician 667 sequence. This thermal structure of the continental crust has been previously described in high-668 temperature and mid-crustal granulite terranes (Hollister, 1982; Patiño-Douce et al. 1990; De Yoreo 669 et al. 1991; Thompson & England, 1984; Thompson, 1999). Two different tectonic settings have 670 been called upon to explain such abnormal geothermal gradients. First, rifting resulting in an 671 extremely thin continental crust, with a possible mantle derived basaltic sill intrusion in the lower 672 crust providing sufficient heat to reach such high temperatures. This has been proposed in the 673 Pyrenean belt by Wickham and Oxburgh (1985; 1987), partly based on stable isotopes signature in 674 the uppermost crust (equivalent of the Late Cambrian - Early Ordovician sequence in the Agly 675 massif). The second alternative model is referred as the "melt-enhanced geotherm" (Depine et al. 676 2008) for which the combined effect of (1) upward heat advection associated with melt transfer and 677 (2) thermal buffering in the middle crust due to dehydration-melting reactions (either biotite or 678 hornblende breakdown reaction). In the case of the Agly massif several arguments are in favor of a 679 melt-enhanced thermal structure at c. 305 Ma: 680

681

682 1) Contemporaneous magmatism and metamorphism at ca. 305 Ma.

683 The main metamorphic event responsible for the abnormal geothermal gradient at 305 Ma is coeval

with a widespread magmatic event between 308 and 304 Ma (Figures 13a and b). In the deepest part of the Agly crustal section, the Ansignan charnockite and the Cassagnes sills were emplaced at 307  $\pm$  3 Ma and 308  $\pm$  3 Ma, as a 500 to 800 m laccolith and decameter sills, respectively, parallel with the main gently dipping foliation developed at peak metamorphic conditions. The upper part of the gneissic core and the micaschist belonging to the Paleozoic sequence are intruded by the kilometer thick Tournefort diorite at 308  $\pm$  1 Ma and St Arnac granite at 304  $\pm$  5 Ma that is exposed on a 25 km<sup>2</sup> surface (Olivier et al., 2008).

691

#### 692 2) Melt source and upward advection of heat by melt

693 The Ansignan charnockite is a composite laccolith intrusive that consists of a massive  $\sim 500$  to 800 m sill of orthopyroxene ± garnet and K-feldspar megacrysts granite (i.e. the charnockite) associated 694 with norite bodies up to 300 m in its core and garnet-bearing leucocratic granites in its upper 695 margins. The source of the charnockite and associated magmatic rocks has been a matter of debate 696 in the 1980's (Fonteilles, 1981; Fourcade, 1981; Pin, 1989). All these studies agreed that the 697 charnockite was related to the partial melting of continental crust and more particularly 698 metasediments. Pin (1989) who conducted the most extensive isotopic analysis on the different 699 lithologies, suggested that the charnockitic magma without garnet (the more widespread facies of 700 the charnockitic complex), characterized by  $\epsilon Nd_{300Ma},$  ranging from -6.4 and -4.4 and  $^{87}Sr/^{86}Sr_i=$ 701 0.7079 was produced by partial melting of metasediments due to the intrusion of contaminated 702 mafic magmas, corresponding to the norite bodies ( $\epsilon Nd_{300Ma} = -2.5$  to -3.4 and  ${}^{87}Sr/{}^{86}Sr_i = 0.7062$ -703 0.7067). These charnockitic magmas are characterized by a strong HREE fractionation (Fourcade, 704 1981; Tournaire-Guille et al., 2019, Figure 11b) suggesting that melts, produced by a high degree of 705 partial melting, were in equilibrium with a garnet-rich restite at a pressure greater than 6 kbar (see 706 below and Figure 13a). Chemical exchange between the mantle-derived mafic magmas with lower 707 crust-derived magmas resulted in a moderate Nd ans Sr isotopic hybridization. Pin (1989) invocated 708 a second stage corresponding to the ascent and emplacement of this hybridized magma (norite and 709 charnockite) into upper crustal levels, corresponding to the present day gneissic core of the Agly 710 massif, at conditions estimated at  $0.5 \pm 0.05$  GPa (~18 to 22 km) at  $750 \pm 100^{\circ}$ C (Vielzeuf, 1984; 711 1986 and Figures 13a and b). The latent heat of crystallizing magmas induced partial melting of the 712 surrounding metasedimentary host-rock and the formation of the garnet-bearing charnockite and 713 leucogranites commonly observed at the margins of the intrusions. These garnet-bearing magmas 714 are characterized by a stronger crustal signature with the lowest  $\epsilon Nd_{300Ma} = -6.4$  and -7.3 and the 715 highest- ${}^{87}$ Sr/ ${}^{86}$ Sr<sub>i</sub> = 0.7109 – 0.7127 recorded in the Ansignan intrusive complex. 716

The Cassagnes granodiorite, that consists of K-feldspar-bearing megacrystic sills of about 10 meters thick, has a close age at  $308 \pm 3$  Ma and very similar major / trace element composition to the Ansignan charnockite (see Figures 10 and 11 in Tournaire-Guille et al., 2019). Therefore, we suggest that both magmas have the same origin and source depth. The main difference between them is that Cassagnes sills do not contain orthopyroxene nor garnet probably because they were emplaced in a much shallower crustal level than the Ansignan granodiorite. Based on P-T constrains on kinzigite located near the Cassagnes sills (10Ag06 and 11Ag03) on the eastern part of the gneissic core, we suggest that they were emplaced at 0.5 GPa at 740°C corresponding to a depth of ~ 18 km (Figures 13a and b).

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The Tournefort diorite and Saint-Arnac granite are calc-alkaline magmas, similar to the other major late-Variscan intrusions in the Pyrenees like the granite of Millas, Quérigut, Saint Laurent, Maladetta or Mont-Louis. They are interpreted as the result of the emplacement of magmas in the upper crust issued from partial melting of a deep old magmatic continental crust or hybridization of deep seated crustal magmas with mantle-derived basalt intrusive and related to continental strike slip and exhumation tectonics during the late Variscan collision (Lagarde et al., 1992).

It is clear from field observations that the Saint-Arnac granite and Tournefort diorite were not 733 produced in-situ but rather transferred from an unknown depth in the deep crust to the upper 734 Paleozoic sequence. Because the Tournefort diorite intruded andalusite-bearing micaschist, we 735 estimate the maximum depth of emplacement at about 11 km based on the 0.3 GPa pressure 736 estimated on the host rock (Figures 12 and 13a). Field evidence, like surrounding rocks xenoliths, 737 magma mingling and diorite enclaves in the granite, suggests that the Saint Arnac pluton is located 738 739 at the same depth in the micaschist. The granite intruded all the Ordovician micaschist and schist series up to the Devonian marbles found as decameter xenolith in the wall of the granite. We 740 therefore suggest that the Saint Arnac granite has a minimum thickness of ~2500 m, corresponding 741 to the estimated thickness of the Ordovician series. The Saint-Arnac granite would have been 742 emplaced in the depth bracket of 9.5 and 12 km (Figure 13a). This estimation differs significantly 743 with Olivier et al. (2008), who suggested that the granite is 5 km thick with a roof located at a 744 shallow depth of 2.7 to 4 km (0.1 to 0.15 GPa). The emplacement of both intrusions is associated 745 with a local contact metamorphism that is characterized by partial melting around the diorite and 746 crystallization of large muscovite flakes in the surrounding micaschists around the granite and 747 micaschist xenoliths. 748

At the scale of the Agly massif, all these magmatic intrusions represent a significant volume that is difficult to establish with confidence but may be greater than 30% of the basement (Figure 1). Their migration from a deep source, currently not accessible in the exposed Agly massif, towards upper crustal levels constitutes a very efficient and dynamic heat advection process. The crystallization of late muscovite in micaschists around the Saint-Arnac granite and partial melting surrounding the

Tournefort diorite are consistent with magmatic heat advection in the previously LT Paleozoic 754 sequence. In the gneissic core, the upward heat advection during melt transfer and heat released 755 during the crystallization of magmas would increase temperature. This is in good agreement with 756 the observation of high temperatures ranging from 730 °C to 790 °C for pressure from 0.49 to 0.65 757 GPa (Figure 13b) for the entire gneissic core. At these depths, with a steady state-geotherm, the 758 temperatures would only be about 400-600 °C (figure 13a). The juxtaposition of high-grade rocks 759 from the lower crust as the explanation of the metamorphic gradient observed in the Agly is thus a 760 very unlikely explanation. These high temperatures cannot be achieved without the additional heat 761 source from magmas, even with a high mantle heat flux (Figure 13a). We suggest that the low 762 gradient at about 750 °C in the gneissic core is the combination of melt migration, plutons 763 formation and thermal buffering by the biotite-dehydration melting reaction that produced garnet 764 +/- cordierite. In contrast, the geothermal gradient in the upper crust, above the shallower intrusion 765 to the surface, is significantly higher (~ 55 °C/km) and correspond to a diffusive geothermal 766 gradient (compare Figures 13a and b). The orthogneisses of the gneissic core might not have been 767 as affected as the metapelites by the thermal buffering since they are less fertile in terms of melt 768 production (Schorn, Diener, Powell, & Stüwe, 2018). Nevertheless, they represent only a marginal 769 portion of the gneissic core (Figure 1). Thus, the temperatures computed from metapelites, buffered 770 by the biotite dehydration-melting reaction, can be interpreted as representative of the thermal state 771 of the gneissic core. 772

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#### 774 6.2 Thinning of the massif

Evidences for a major thinning of the Agly massif have been well accepted for decades now, 775 especially based on the difference between depth inferred from previous geothermobarometric 776 studies and thickness of the rock column of the massif in its present geometry. This thickness has 777 been inferred with paleo-pressure calibrations to be of about 11 km between the top of the Paleozoic 778 Jujol group and the bottom of the Charnockite and represents now a lithologic pile of only 5-6 km 779 of exposed rocks. Nevertheless, the timing and importance of this thinning is still a matter of 780 debate, as recently outlined by the succession of publications (Olivier at al. 2004; 2008; Olivier 781 2013; Vauchez et al. 2013a; 2013b). Even more recently, two studies have rejuvenated this debate 782 with new data, using (U-Th)/He (Ternois et al., 2019) and U-Pb (Odlum & Stockli, 2019) in both 783 zircons and apatite. The first study founded that the Agly massif never experienced temperature 784 higher than 250 °C during Cretaceous rifting while the second study argued that the gneissic core 785 was still at temperatures higher than 450 °C before rifting. In terms of the thinning itself and 786 concerning the late Variscan tectonics only, two different deformation settings have been proposed. 787

First, the presence of one major subtracting zone responsible for the main thinning of the massif 788 was proposed by Bouhaillier et al. (1991) and Delay (1990). This detachment zone would act as a 789 major normal fault located at the boundary between the Late Cambrian - Early Ordovician 790 sequence (Jujol group) and the gneissic core. It has been referred to the Latour-Caladroy mylonitic 791 zone (Figure 1 and labelled "Caladroy SZ" on the cross-section, figure 2) since many mylonites 792 with normal shear behavior have been observed between the city of Latour-de-France and the castle 793 of Caladroy. In contrast, Olivier et al. (2008) argued that the thinning of the massif was 794 accommodated by numerous normal shear zones over the entire massif. 795

The reconstruction of the geothermal gradient in the Jujol group and in the gneissic core from the 796 present study clearly shows a pressure gap of 0.15 GPa between the bottom of the Late Cambrian – 797 Early Ordovician sequence and the uppermost level of kinzigite (Figure 12). A simple pressure 798 conversion into depth (using an average density of 2.7), indicates that about 5.5 km of rocks may 799 have been substracted by the Latour-Caladroy mylonites (Figure 12c). The fact that most of the 800 deformation and subtraction was localized at the boundary between the partially molten micaschists 801 802 and the gneissic core is not surprising since partial melting induces a major strength drop when melt proportion reaches 7 vol% (Rosenberg & Handy, 2005). The onset of partial melting triggered by 803 muscovite dehydration reaction is known to produce under 10% of melt (White, Powell, & Holland, 804 2001), such value would then be in good agreement with deformation experiments. 805

We also suggest that thinning and associated rock subtraction is not restricted to the Latour-806 Caladroy mylonite but also occur within the gneissic core. Pressures estimated for the deepest 807 kinzigite level close to the Charnockite are 0.62-0.66 GPa (10Ag01 and 10Ag02, Figure 12). This 808 would give a pressure difference between the shallowest and the deepest kinzigite levels for the 809 gneissic core of about 0.10 to 0.15 GPa, or a thickness of about 3.7 to 5.5 km. Since the present-day 810 gneissic core thickness do not exceed 2.5-3 km, we suggest that the numerous mylonites observed 811 throughout the entire gneissic core might be responsible for the remaining thinning of the massif (as 812 quoted Ansignan and gneissic core mylonites on Figure 12c). 813

Monazite overgrowths with highly variable Th/U ratio from mylonitized kinzigites yield ages of 814 about 296-300 Ma (samples 11Ag03 and 11Ag04; Figures 11c and 11d). These overgrowths may 815 have been produced by fluid-assisted dissolution/ precipitation during localized fluid percolation in 816 shear zones pathways. Furthermore, the highly variable Th/U ratio is also a good evidence that 817 monazite rims were not in equilibrium rock-wide but rather locally equilibrated with a fluid. The 818 origin of the fluid responsible for the dissolution/precipitation of the overgrowth could be the result 819 of crystalizing melt from migmatites. This would require to be near the solidus at significantly 820 lower temperature and pressure than 0.6 GPa and 750-790 °C determined at 305 Ma. Such drop in 821 pressure and temperature would likely be the result of tectonic assisted exhumation, such that the 822

age at 296-300 Ma would be the age of the thinning of the massif. To conclude, we suggest that the 823 thinning of the Agly massif occurred between 300 and 296 Ma after peak metamorphism and the 824 main magmatic event at c. 305 Ma. The Cretaceous thinning during rifting prior to the Pyrenean 825 orogen would not have significantly disturbed the geometry acquired at the end of the Variscan 826 orogeny, in agreement with the (U-Pb)/He on zircons and apatite data from Ternois et al. (2019). 827 Nevertheless, blocs might have slightly moved along the E-W Pyrenean fault that crosscut the 828 massif, especially the southernmost bloc where the Ansignan charnockite is located, that most 829 probably moved to slightly upper levels along the E-W fault located in the Caramany lake. 830 831

- 0.51
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- 833 7 CONCLUSION
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The detailed petrological and geothermobarometric data from the present study shows that 835 the Agly massif has experienced an abnormal thermal state during late-Variscan orogeny. This has 836 resulted in a very high geothermal gradient of 55 °C/km in the upper crust (above 13 km depth) and 837 a near isothermal gradient in the middle crust. New monazite U-Th-Pb geochronology show that 838 839 this metamorphic event took place at c. 305 Ma. This abnormal geothermal gradient is the consequence of the coeval emplacement of several intrusions at different structural level during a 840 limited time period c. 304-308 Ma. The heat provided by these intrusions allowed the middle crust 841 to reach partial melting temperatures at depth below 13 km, and very high temperatures, i.e. 842 ~750°C, between 18 and 25 km depth and be responsible of a "regional contact metamorphism". 843 The biotite dehydration-melting reaction, strongly endothermic, buffered temperature in the middle 844 crust, resulting in the observed near isothermal geothermal gradient. The high geothermal gradient 845 in the upper crust is mostly due to heat conduction between the anomalously hot middle crust and 846 the cold surface. 847

This gradient was latter disturbed by normal mylonitic shear zones, resulting in the thinning 848 of the massif from a thickness of 15 km to a final 5-6 km thickness. Most of this thinning was 849 concentrated within a major detachment zone located at the onset of partial melting, between the 850 Late Cambrian - Early Ordovician sequence and the gneissic core. This detachment zone was 851 responsible for the subtraction of 5-6 km of rocks thickness. The remaining thinning most likely 852 took place at several levels inside the gneissic core, resulting in the abundance of small mylonitic 853 zones. The tectonic juxtaposition of a very hot middle crust (i.e. ~750 °C) and a colder upper crust 854 probably increased the temperature of the later. Nevertheless, due to the already existing thermal 855 structure resulting of the metamorphic event at c. 305 Ma, temperatures in the upper crust were 856 already higher than normal, close to 660 °C at the contact with the gneissic core. The temperature 857

increase due to thinning was only marginal and well below the precision of any present geothermobarometric method. This thinning event could be linked to the second age observed in monazite overgrowths and determined to be at c. 296-300 Ma. Nevertheless, this should be the focus of a detailed geochronologic study, aiming specific deformed monazites inside strongly deformed samples to directly date the thinning event.

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## 1081 APPENDIX

1082 Chemical system, software and activity-composition relationship model used for the 1083 thermodynamic modelling.

Fig	Chemical	Software	Thermod	Solid	a-X models	Reference
ure	system		ynamic	Solution	abbreviation	
			database			
Fig	NCKEM	Dorplo V	Holland	V foldener	Son	Waldhaum and
Fig		Perpie_A	попапа	K-leidspar	Sall	
. 6a	ASHT		and	Plagioclase	Pl(h)	Thompson (1968)
and			Powell	Melt	melt(HP)	Newton et al. (1980)
60			(2002)	White Mica	Pheng(HP)	White et al. (2001)
				Garnet	Gt(WPPH)	Holland & Powell
				Chlorite	Chl(HP)	(1998)
				Cordierite	hCrd	White et al. (2005)
				Biotite	Bio(HP)	Holland & Powell
				Staurotide	St(HP)	(1998)
Fig	KFMAS	Perple_X	Holland	K-feldspar	San	Waldbaum and
. 6c	Н		and	Plagioclase	Pl(h)	Thompson (1968)
			Powell	Melt	melt(HP)	Newton et al.
			(2002)	White Mica	Mica(CHA)	(1980)White et al. $(2001)$
				Garnet	Gt(WPPH)	Coggon & Holland
						coggon a monana
				Chlorite	Chl(HP)	(2002)
				Chlorite Cordierite	Chl(HP) hCrd	(2002) White al. (2005)
				Chlorite Cordierite Biotite	Chl(HP) hCrd Bio(HP)	(2002) White al. (2005) Holland et al. (1998)
				Chlorite Cordierite Biotite Orthopyroxn	Chl(HP) hCrd Bio(HP) Opx(HP)	(2002) White al. (2005) Holland et al. (1998) ideal mixing
				Chlorite Cordierite Biotite Orthopyroxn e	Chl(HP) hCrd Bio(HP) Opx(HP) St(HP)	<ul> <li>(2002)</li> <li>White al. (2005)</li> <li>Holland et al. (1998)</li> <li>ideal mixing</li> <li>Holland et al. (1998)</li> </ul>
				Chlorite Cordierite Biotite Orthopyroxn e Staurotide	Chl(HP) hCrd Bio(HP) Opx(HP) St(HP)	<ul> <li>(2002)</li> <li>White al. (2005)</li> <li>Holland et al. (1998)</li> <li>ideal mixing</li> <li>Holland et al. (1998)</li> <li>Powell and Holland</li> </ul>
				Chlorite Cordierite Biotite Orthopyroxn e Staurotide	Chl(HP) hCrd Bio(HP) Opx(HP) St(HP)	(2002) White al. (2005) Holland et al. (1998) ideal mixing Holland et al. (1998) Powell and Holland (1999)

						ideal mixing
Fig	KFMAS	Domino/	SPac14	same as Fig	Ideal site	
. 6d	Н	Theriak		бс	mixing	
Fig	NCKFM	Perple_X	Holland	K-feldspar	San	Waldbaum and
. 8a	ASHT		and	Plagioclase	Pl(h)	Thompson (1968)
and 8b	and 8b	Powell (2002)	Melt	melt(HP)	Newton et al. (1980)	
00			(2002)	White Mica	Mica(CHA	White et al. (2001)
				Garnet	Gt(WPPH)	Coggon & Holland
				Chlorite	Chl(HP)	(2002)
				Cordierite	hCrd	White al. (2005)
				Biotite	Bio(HP)	Holland et al. (1998)
				Staurotide	St(HP)	ideal mixing
						Holland et al. (1998)
						Ieal mixing

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# 1093 Figure captions:

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Figure 1: Geographic and geologic setting of the Agly massif; a) situation of the Agly massif in the Pyrenees in respect to all Variscan massifs, modified after Olivier et al. (2004); b) geological map of the massif produced during this study; the E-W cross section is discontinuous to take into account the displacement along the Planèzes fault, which was most likely active during very late Variscan stage.

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Figure 2: Schematic E-W cross-section located on figure 1. The different shear zones (SZ) havebeen highlighted in dark black.

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Figure 3: Petrography of the Late Cambrian - Early Ordovican samples, with increasing 1105 metamorphic grade from left to right and top to bottom. a) Polarized photomicrograph of a 1106 greenschist facies sample with sedimentary bedding and main schistosity; S<sub>n-1</sub> microlithon shows 1107 the transition between chlorite grade and biotite grade. b) Polarized photomicrograph for a typical 1108 1109 biotite grade sample, with biotite (Bt) and muscovite (Ms) in the main schistosity. c) Polarized photomicrograph of a cordierite grade sample, with a slightly rotated large poikilitic cordierite (Crd) 1110 porphyroblast with biotite, quartz and oxides inclusions. Biotite is also present in the matrix, in the 1111 main schistosity, as well as muscovite. d) Polarized photomicrograph of a cordierite-andalusite 1112 grade sample, with large poikilitic and alusite (And) porphyroblasts with biotite, quartz and oxides 1113 in inclusions. Biotite and muscovite are mainly present in the matrix aligned in the main schistosity, 1114

interbedded with quartz (Qtz) and plagioclase (Pl). e) Polarized photomicrograph of a sillimanite grade sample, with typical fibrolite (Sil) patches, aligned in the main foliation. Muscovite (Ms2) is only present as secondary mineral, discordant to the main foliation. f) Polarized photomicrograph of a partially melted sample, with small aggregates of quartz (Qtz), alkali feldspar (Kfs) and plagioclase (Pl), representing the remaining melt pockets. Biotite (Bt) and sillimanite (Sil) are aligned in the main foliation.

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Figure 4: Petrography for three kinzigite samples from different structural levels of the gneissic 1123 core, 10Ag11 located slightly above the Ansignan charnockite; 10Ag04 represents the deepest level, 1124 below the charnockite; 10Ag03 is the shallowest level, near the Cuxous castle at about 500m from 1125 the contact with the Late Cambrian - Early Ordovicain sequence. a) polarized photomicrograph for 1126 10Ag11, with large garnet porphyroblasts and prismatic sillimanite (Sil) and biotite (Bt) aligned in 1127 the main foliation. Note the fresh cordierite grain on the lower right corner of the microphotograph. 1128 b) Polarized photomicrograph for sample 10Ag04, showing fractured garnet, fresh cordierite and 1129 monazite (Mnz) grains used for the geochronolical study. c) Polarized photomicrograph of the 1130 sample 11Ag03, showing highly deformed K-feldspars (Kfs), grain size reduction for prismatic 1131 1132 sillimanite (Sil) and retrogressed cordierite ("Crd"). Note the grain size reduction in figure b) and c) in the shear bands. 1133

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Figure 5: Thompson-like AFM diagram projected from muscovite end-member. Black dots
represent the XRF-analyses for the different samples of the Late Cambrian – Early Ordovician
sequence. White dot corresponds to sample 10Ag16 considered as a representative composition (see
text for explanations).

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Figure 6: Thermodynamic modelling for the Late Cambrian – Early Ordovician sequence. Sample 1144 10Ag16 was select since it is representative of the composition for the different samples of this 1145 sequence (white dot on figure 4). a) Pseudosection computed with Holland and Powell 1146 thermodynamic database (version 1998, updated in 2002) for an average composition of the schists 1147 in the NCKFMASHT system. Outlined on this pseudosection are: stability field of staurolite (light 1148 red area), upper limit of chlorite stability (green line), andalusite (And) to sillimanite (Sil) transition 1149 and muscovite breakdown (black lines), and hydrated granite solidus (red line) b) Compilation of

constrains for all samples in the system NCKFMASHT, the green lines represent the breakdown of 1150 chlorite, the yellow lines are the bounds for the stability field of staurolite, the black lines are the 1151 upper limit for the stability of muscovite, the blue lines represent the upper limit (in terms of 1152 pressure) of stability for cordierite, and red lines are the wet solidus curves; c) Pseudosection 1153 computed with Holland and Powell thermodynamic database (version 1998, updated in 2002) in the 1154 KFMASH system, with TiO2 projected through ilmenite, CaO projected through anorthite and NaO 1155 projected through albite. d) Pseudosection computed with Domino/Theriak and the SPaC145 1156 thermodynamic database in the KFMASH system (using the same composition than for 6c). See 1157 text for more explanations. 1158

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Figure 7: a) BSE image of a garnet from sample 10Ag03 analyzed with EPMA for major elements determination. The bold black line represents the position of the transect analyzed. b) Garnet endmembers composition along the transect shown on figure 8a. The garnet analyses highlight the absence of zonation for major elements composition, even near the rim of the grain. Note the discontinuous scale due to very high almandine content, above 80 % here and commonly above 65% for all garnet compositions.

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Figure 8: thermodynamic modelling for the gneissic core, using Holland and Powell 1169 thermodynamic database (version 1998, updated in 2002) and NCKFMASHT system of 1170 components. a) Pseudosection for sample 11AgG03, located near the Castle of Cuxous, close to the 1171 Late Cambrian - Early Ordovician sequence. The shaded area in blue represent the small divariant 1172 field for the dehydration-melting reaction of biotite. Red and green lines are isopleths of garnet 1173 composition, almandine and pyrope contents, respectively. The bold lines for both contents 1174 represent the garnet composition for this sample (see table 2). The brown area between the two bold 1175 lines for isopleths represent the P-T conditions determined for this sample based on the assemblage 1176 and the garnet composition. b) Summary of all kinzigite samples P-T conditions determinations. As 1177 for figure 9a, red lines represent almandine contents of garnets for each sample, and green lines are 1178 pyrope contents, black lines are the upper and lower limits for the biotite dehydration-melting 1179 reaction. c) Cross section with the location and P-T conditions for all kinzigites samples. Those who 1180 are not strictly on the cross section (10Ag03 and 10Ag05) are projected based on their distance to 1181 the boundary between the gneissic core and the late Cambrian – early Ordovician sequence. 1182

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Figure 9: Large polarized photomicrographs for the 3 samples used for the geochronological study: a) 10Ag02; b) 10Ag04; c) 11Ag03. The position of the monazite grains analyzed for U-Th-Pb by LA ICP-MS are highlighted with yellow circles, with their associated names next to it. The limits of the field of view (dashed rectangles) used for the figure 6 for the petrological study for samples 10Ag04 (figure 5b) and 11Ag03 (figure 5c) are shown with dashed lines. Normal shear zones are highlighted for the highly deformed sample 11Ag03 using asymmetric arrows.

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Figure 10: Yttrium and Thorium X-ray maps of the different monazite grains analyzed for the U-Th-Pb geochronology for samples 10Ag02 (A), 10Ag04 (B) and 11Ag03 (C). The white dots on Y maps represent the location of LA ICP-MS analyses, with the label number. For sample 10Ag04, monazite grains in inclusion in garnet are labelled with "grt" on top.

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Figure 11: Concordia diagrams for all U-Th-Pb analyses of monazite grains. A) Analyses for 1199 sample 10Ag02, close to the Ansignan charnockite, all Y-rich cores define a statistical meaningful 1200 population with an age of  $305 \pm 4$  Ma ( $2\sigma$ ). B) Analyses of monazite for the sample 10Ag04, also 1201 1202 close to the charnockite, taking only the spot representing Th/U ratio of  $6.5 \pm 1$ , they define a single population of ages with a mean of  $306 \pm 2$  Ma  $(2\sigma)$ , one very low concordant age has been 1203 excluded. C) Analyses of monazite for sample 11Ag03, the most deformed and retrogressed 1204 kinzigite sample, located in the upper part of the gneissic core. Nine of the analyses successfully 1205 represent a single population of ages, with a mean at 296  $\pm$  2 Ma (2 $\sigma$ ). The analyses of m3 have 1206 been excluded as well as the low Th core in m1 (figure 11c). Monazite grain 3 seems to be more 1207 affected by the Cretaceous deformations, with tailing of the ages toward younger values. D) 1208 1209 Analyses of high Th/U zones of monazite grains from sample 10Ag04. 12 of these analyses clearly 1210 define a single population of ages with a mean value of  $300 \pm 2$  Ma  $(2\sigma)$ .

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Figure 12: Reconstruction of the metamorphic gradient after the main Variscan metamorphic event
with both the gneissic core samples and the late Cambrian – early Ordovician sequence.

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Figure 13: reconstruction of the evolution of the geothermal gradient of the Agly massif during the Late-Variscan HT-LP metamorphic event. a) Intrusion of the magmatic bodies (grey boxes) at different structural levels of the Agly massif between 308 and 304 Ma, with a lower crustal source for these magmas, see text. b) At the time of the main metamorphic event (c. 305 Ma), geothermal gradient is directly inferred from the P-T conditions determined from thermodynamic modelling for the gneissic core (red symbols) c) Reconstruction of the geothermal gradient after the main Late-Variscan thinning event, at c. 296-300 Ma. The late Cambrian – early Ordovician sequence is slightly heated due to the juxtaposition of the hot gneissic core. Blue areas represent the two main subtractive zones responsible for the thinning of the massif.

1226 Mineral abbreviations around the alumino-silicate triple point are kyanite (ky), sillimanite (sil), 1227 andalusite (and). The two main dehydration reactions described in the text are highlighted, 1228 muscovite dehydration (ms -) and biotite dehydration melting (bt sil q = grt crd L).

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1231 Table 1: XRF analyses of all samples used in this study. GPS location are in the French system1232 Lambert 2 étendu.

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Table 2: Average garnet core analyses from EPMA for kinzigites samples of the gneissic core with their respective variations, computed as 1 standard deviation (1s). Structural formulae are computed based on 12 oxygens, the relative proportion of almandine (Alm), pyrope (Py), grossular (Gr) and spessartine (Sps).

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Table 3: Summary of all U-Th-Pb data for monazite used in this study with their respective texturalpositions.

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1242 Table 4: Summary of P-T conditions for all kinzigite samples, determined from pseudosections (see1243 figure 8b).

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