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### Distribution and intensity of High-Temperature Low-Pressure metamorphism across the Pyrenean-Cantabrian belt: constraints on the thermal record of the pre-orogenic hyperextension rifting

Maxime Ducoux<sup>1,\*</sup>, Laurent Jolivet<sup>2</sup>, Emmanuel Masini<sup>1,3</sup>, Romain Augier<sup>4</sup>, Abdeltif Lahfid<sup>5</sup>, Matthias Bernet<sup>3</sup>, and Sylvain Calassou<sup>6</sup>

<sup>1</sup> M&U SAS, 3 rue des Abattoirs, 38120 Saint-Égrève, France

<sup>2</sup> Sorbonne Université, CNRS-INSU, ISTeP UMR 7193, 75005 Paris, France

<sup>3</sup> Institut des Sciences de la Terre (ISTerre), Université Grenoble-Alpes, Grenoble, France

<sup>5</sup> BRGM, ISTO, UMR 7327, Orléans, France

<sup>6</sup> TOTAL SE, CSTJF, avenue Larribau, 64000 Pau, France

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**Abstract** – Whereas a straightforward link between crustal thinning and geothermal gradients during rifting is now well established, the thermal structure of sedimentary basins within hyperextended domains remains poorly documented. For this purpose, we investigate the spatial distribution of rift-related High-Temperature Low-Pressure (HT/LP) metamorphism recorded in the preserved hyperextended rift basins inverted and integrated in the Pyrenean-Cantabrian belt. Based on Vitrinite Reflectance ( $R_0$ ) data measured in 169 boreholes and more than 200 peak-metamorphic temperatures  $(T_{max})$  data obtained by Raman Spectroscopy of Carbonaceous Material (RSCM) added to ~425 previously published  $T_{\text{max}}$  data, we propose a new map depicting the spatial distribution of the HT/LP metamorphism of the Pyrenean-Cantabrian belt. We also provide three regional-scale geological cross-sections associated with  $R_o$  and  $T_{max}$ data to constrain the distribution of paleo-isograds at depth. Based on these results, we show that the impact of rift-related metamorphism is restricted to the pre- and syn-rift sequence suggested by the depth profiles of  $R_0$ values measured in different tectonostratigraphic intervals (pre-, syn- and post-rift and syn-convergence sediments). However, a small strip of early orogenic sediments (Santonian in age) appears also affected by high temperatures along the North Pyrenean Frontal Thrust and above the Grand Rieu ridge, which we attribute to the percolation of hot hydrothermal fluids sourced from the dehydration of underthrust basement and/or sedimentary rocks at depth during the early orogenic stage. The map shows that the HT/LP metamorphism (reaching  $\sim 500$  °C) is recorded with similar intensity along the Pyrenean-Cantabrian belt from the west in the Basque-Cantabrian Basin to the east in the Boucheville and Bas-Agly basins, for similar burial and rift-related structural settings. This thermal peak is also recorded underneath the northern border of the Mauléon Basin (calibrated by wells). It suggests that the high temperatures were recorded at the basement-sediment interface underneath the most distal part of the hyperextended domain. At basin-scale, we observe in the Basque-Cantabrian, Mauléon-Arzacq and Tarascon rift segments an asymmetry of the thermal structure revealed by different horizontal thermal gradients, supporting an asymmetry of the former hyperextended rift system. Using our results, we compare the Pyrénées to the Alps that also recorded hyperextension but no HT/LP metamorphic event and suggest that the high-temperature record within the basins depends on high sedimentation rate promoting a thermal blanketing effect and circulation of hydrothermal fluids.

**Keywords:** thermal evolution / hyperextended rifted margin / Pyrenean-Cantabrian belt / H*T*/L*P* metamorphism / RSCM Method / Vitrinite Reflectance

Résumé – Distribution et intensité du métamorphisme haute-température basse-pression de la chaîne Pyrénéo-Cantabrique : des contraintes sur l'enregistrement thermique de l'hyperextension anté-orogénique. Bien que l'association de l'amincissement crustal et de gradients géothermiques élevés

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<sup>&</sup>lt;sup>4</sup> Université d'Orléans, ISTO, UMR 7327, 45071 Orléans, France

<sup>\*</sup>Corresponding author: maxime@mandu-geology.fr

lors du rifting continental soit largement reconnue, la structure thermique des bassins sédimentaires dans la partie distale des systèmes de rift reste mal documentée. Pour cela, nous étudions la distribution spatiale du métamorphisme Haute-Température/Basse Pression (HT/BP) enregistrée dans les bassins préservés du système de rift hyper-aminci, par la suite inversés et intégrés dans la chaîne Pyrénéo-Cantabrique. Basé sur la réflectance de la Vitrinite (R<sub>o</sub>) mesurée dans 169 puits et plus de 200 données de pic de température lié au métamorphisme ( $T_{max}$ ) obtenues avec la méthode de Spectroscopie Raman de la Matière Carbonée (RSCM) ainsi que plus de  $425 T_{\text{max}}$  provenant d'études précédentes, nous proposons une nouvelle carte de la distribution spatiale du métamorphisme HT/BP de la chaîne Pyrénéo-Cantabrique. Nous proposons également trois coupes géologiques regionales, sur lesquelles nous avons placé les données de  $T_{\rm max}$  et de R<sub>o</sub> afin de contraindre la distribution des paléo-isogrades en profondeur. Basé sur ces résultats, nous montrons que l'impact du métamorphisme lié au rifting est restreint aux sédiments pré- et syn-rift, ce qui est suggéré par la tendance des profils des valeurs de Ro en profondeur mesurées dans les différents intervalles tectonostratigraphiques (sédiments pré-, syn- et post-rift ainsi que syn-convergence). Cependant, une fine bande de sédiments syn-orogéniques (d'âge Santonien) est affectée par des températures relativement élevées au-dessus de la ride de Grand Rieu et le long du Chevauchement Frontal Nord Pyrénéen, que nous attribuons à la percolation de fluides hydrothermaux chauds provenant de la déshydratation du socle chevauché et/ou des sédiments profonds, lors du stade d'inversion précoce. La carte présentée montre que le métamorphisme (atteignant ~500 °C) est enregistré avec la même intensité du bassin Basque-Cantabrique à l'ouest, aux bassins de Boucheville et du Bas-Agly à l'est, pour un enfouissement et un positionnement lors du rifting équivalents. Le pic thermique est également enregistré sous la bordure nord du bassin de Mauléon (calibré par des puits). Cela suggère que les hautes températures ont été enregistrées à l'interface soclesédiments au niveau de la partie la plus distale du domaine hyper-aminci. À l'échelle des bassins, nous observons dans les segments Basque-Cantabrique, Mauléon-Arzacq et Tarascon une asymétrie de la structure thermique, révélée par différents gradients thermiques horizontaux, supportant une asymétrie de l'ancien système de rift hyper-aminci. En utilisant nos résultats, nous comparons les Pyrénées avec les Alpes qui ont également enregistré l'hyper-extension mais pas d'évènement métamorphique HT/BP, ce qui suggère que l'enregistrement des hautes températures dans les bassins dépend de taux de sédimentation élevés, favorisant un effet de couverture thermique et de circulations de fluides hydrothermaux.

**Mots clés :** évolution thermique / marge hyper-amincie / Pyrénées / métamorphisme H*T*/B*P* / méthode RSCM / reflectance de la Vitrinite

### **1** Introduction

One of the key characteristics of continental rifting is the development of hot geothermal gradients. At rift-scale, these transient thermal anomalies primarily result from the thinning of continental lithosphere under extensional tectonics (e.g. Buck et al., 1988; Brune et al., 2014; Elders et al., 1972; Royden et al., 1980). The observed structural variability between different rift systems is generally considered as mostly due to rifting intensity. localization and velocity with respect to the thermal conductivity of the rifted lithosphere (e.g. Allen and Allen, 2013; Hantschel and Kauerauf, 2009; Jolivet et al., 2020; Lavier and Manatschal, 2006; Manatschal, 2004; Mohn et al., 2010; Osmundsen et al., 2016; Péron-Pinvidic and Manatschal, 2009; Péron-Pinvidic and Osmundsen, 2016; Sutra et al., 2013). For incipient or early stages of rift systems, regional thermal predictions through time and space provided by basin modeling (e.g. Callies et al., 2018; Lescoutre et al., 2019; Nirrengarten et al., 2020; Ungerer et al., 1990), generally fit well the McKenzie-type pure-shear rifting models (McKenzie, 1978). Similar approaches were also applied for more mature rift cases corresponding to distal domains of hyperextended rift systems but failed to predict geological observations (Nirrengarten et al., 2020; Peace et al., 2017; Pross et al., 2007). Unfortunately, boreholes used for these studies penetrated only basement highs and young syn-rift sedimentary units that are not documenting the syn-rift thermal structure within deeper and thicker basins. It has been shown on both onshore and offshore records that hyperextended domains of rifts usually display much higher and spatially variable syn-rift peak temperatures  $(T_{max})$  in close association with indications for hydrothermal and/or magmatic processes (diagenetic, metasomatic or fluid inclusion datasets, spatial distribution of T<sub>max</sub>, e.g. Jagoutz et al., 2007; Larsen et al., 2018; Manatschal, 2004; Nirrengarten et al., 2020; Royden et al., 1980). These observations strongly support the importance of syn-rift heat advection into distal rift domains. In this respect, the sedimentary blanket (either pre-, syn- or post-kinematic) may also strongly influence the basin thermal record by impacting fluid dynamics and therefore the advected heat (e.g. Callies et al., 2018; Clerc et al., 2015; Wangen, 1995). This is due to the petrophysical characteristics (conductivity and permability) and spatial distribution of the sediment blanket. It is noteworthy that, so far, there is no available database describing the thermal record of a hyperextended rift system at a sufficiently large scale (i.e. lithospheric scale), covering different structural domains of rifts from proximal to distal, with variable syn-rift sedimentary thicknesses and not overprinted by subsequent burial and/or tectonic phases such as lithospheric break-up, thermal and magmatic climax, or orogen-related.

At present, the alternative way to measure and describe the thermal record of lithospheric thinning and subsequent lithospheric break-up is to use fossil analogues that are

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**Fig. 1.** Tectonic and geological framework of the Pyrenean-Cantabrian belt. (a) Main collision related geological features of the Western Mediterranean. (b) Geological map of the Pyrenean-Cantabrian collision belt (after 1 million-scale Geological Map of Spain and Geological Map of France, with RGF93 projection), with location of Figures 3 and 4.

currently cropping out in mountain belts (e.g. Taiwan: Conand et al., 2020; Alps: Decarlis et al., 2017; Gabalda et al., 2009; Pinto et al., 2015; Pyrénées: Clerc et al., 2015; Ducoux et al., 2019; Golberg and Levreloup, 1990; Lescoutre et al., 2019). This is what we intend to accomplish in this study, using the fossil Pyrenean rift record that was shown to preserve a riftrelated thermal anomaly. The Pyrenean belt is an ideal candidate to study the thermal imprint of continental rifting, because: (i) this orogen is one of the typical examples of a mountain belt derived from an inverted hyperextended rift system (Clerc et al., 2012, 2013; Jammes et al., 2009, 2010a; Lagabrielle and Bodinier, 2008; Lagabrielle et al., 2010, 2016; Masini et al., 2014; Teixell et al., 2016, 2018; Tugend et al., 2014) and (ii) pre-and syn-rift sediments affected by HT/LP metamorphism not affected by subduction and collision metamorphic overprints. Owing to decades of industry (SNEAP, Elf Aquitaine, and TOTAL) and academic researches (e.g. RGF or OROGEN projects), we provide in this study a new compilation of  $T_{\text{max}}$  data documenting the spatio-temporal thermal record of hyperextension rifting across the entire Pyrenean segment of the belt. As measurements were performed on carbonaceous material coming from both surface and drillholes (Vitrinite Reflectance values and Raman Spectroscopy of Carbonaceous Material [RSCM] data), this unique dataset of a sedimentary rift system further enables to contextualize the so-called "Pyrenean HT/LP event" as well as to investigate the role of sediment burial in both time and space. Considering these results, we compare the Pyrenean-Cantabrian record to the Alps that show a generalized intense serpentinisation and brecciation of peridotites without significant HT/LP metamorphism of the syn-rift sediments. These differences are probably explained by a diversity of hyperextension architectures with variable extension rates,

the width of mantle exhumation domain and syn-rift sediment thicknesses. These various examples are thus perfect sites to study the thermal evolution of the ocean-continent transition (OCT) during hyperextension, a domain rarely attainable on present-day continental margins.

### 2 Geological setting

The Pyrenean-Cantabrian belt is a roughly 1000 km long fold and thrust belt, striking E-W from the Cantabrian belt in NW Spain to its eastern termination in the Pyreneo-Provençal belt in the south-east of France at the junction with the western Alps (Fig. 1a). Even though the precise Iberia-Eurasia kinematics scenario remains strongly debated (e.g. Barnett-Moore et al., 2016; Jammes et al., 2009; Neres et al., 2013; Nirrengarten et al., 2017; Olivet, 1996; Sibuet et al., 2004; Srivastava et al., 2000; Vissers and Meijer, 2012; Vissers et al., 2016), it is well established that the Pyrénées first resulted from the Late Cretaceous to Miocene tectonic inversion of the former Cretaceous rifted domains (Clerc et al., 2012, 2013; Jammes et al., 2009, 2010a; Lagabrielle and Bodinier, 2008; Lagabrielle et al., 2010, 2016; Masini et al., 2014; Mouthereau et al., 2014; Muñoz, 1992; Teixell et al., 2016, 2018; Tugend et al., 2014). Prior to the formation of the Pyrenean-Cantabrian belt, this area recorded several phases of extension after the Variscan orogeny. Post-Variscan extension was first recorded during the Permian and Triassic in the whole Western Europe by the development of several rift basins filled by continental clastic and volcaniclastic red sediments (Arche and López-Gómez, Autran and Cogne, 1980; Boillot, 1996: 1984: García-Mondéjar, 1996; Rat, 1988; Vissers, 1992; Winnock,

1974; Ziegler and Dèzes, 2006). The second event corresponds to the main divergence phase between Europe and Iberia, which occurred during Late Jurassic-Cretaceous times. In detail, two event can be differenciated (Nirrengarten et al., 2018; Tavani et al., 2018; Tugend et al., 2015). A leftlateral transtensional rifting phase during the Late Jurassic-Early Cretaceous period mostly recorded in the Bay of Biscay and south of the Pyrénées into the Iberian Range (Boillot et al., 1979; Cadenas et al., 2018, 2020; Déregnaucourt and Boillot, 1982; Ferrer et al., 2008; García-Mondéjar, 1996; Jammes et al., 2009; Montadert et al., 1979; Tugend et al., 2014, 2015). Then, a second phase of rifting, from Aptian to Cenomanian times, lead to the formation of rapidly subsiding rift basins in our study area (Brunet, 1984). Scattered exposures of sub-continental ultramafic rocks mainly reworked in or associated with Upper Triassic evaporites and/or Cretaceous syn-rift sediments (e.g. Clerc et al., 2012, 2013; de Saint Blanquat et al., 2016; DeFelipe et al., 2017; Fabriès et al., 1991, 1998; Lagabrielle and Bodinier, 2008; Lagabrielle et al., 2016) demonstrate that this phase of rifting recorded hyperextension characterized by extremely thinned continental crust and local mantle exhumation (Asti et al., 2019; Jammes et al., 2009, 2010a; Lagabrielle and Bodinier, 2008; Lagabrielle et al., 2010, 2019a, 2019b; Masini et al., 2014; Pedrera et al., 2017; Tugend et al., 2014). Therefore, the Pyrenean rift systems contain pre-rift salt-bearing rocks (Mid-Upper Triassic), which are acting as a regional decoupling layer for Mesozoic extensional structures (Jammes et al., 2009, 2010b; Labaume and Teixell, 2020; Lagabrielle et al., 2010, 2019a, 2019b; Teixell et al., 2016, 2018), and promoting salt-tectonics as well (Canérot, 1988, 1989; Canérot and Lenoble, 1993; Canérot et al., 2005; Ducoux et al., 2019; García-Senz et al., 2019: Izquierdo-Llavall et al., 2020; James and Canérot, 1999; López-Mir et al., 2014; Saura et al., 2016).

At variance with the Bay of Biscay that recorded breakup and an incipient seafloor spreading stage, all other rifted domains aborted at or before reaching a hyperextension stage like the Parentis Basin (Bois and Gariel, 1994; Ferrer et al., 2009; Jammes et al., 2010a, 2010c; Pinet et al., 1987; Tomassino and Marillier, 1997), the Basque-Cantabrian Basin (DeFelipe et al., 2018; Ducoux et al., 2019; Lescoutre et al., 2019; Pedrera et al., 2017; Pedreira et al., 2007; Roca et al., 2011), the Cameros Basin (Casas-Sainz and Gil-Imaz, 1998; Rat et al., 2019), the Columbrets Basin (Etheve et al., 2018) as well as the North Pyrenean Zone (Clerc and Lagabrielle, 2014; Clerc et al., 2012; Jammes et al., 2009, 2010a; Lagabrielle and Bodinier, 2008; Lagabrielle et al., 2010; Masini et al., 2014; Tugend et al., 2014) (Fig. 1). Extreme lithospheric thinning is commonly associated with a high-temperature and lowpressure (HT/LP) metamorphism referred to in this case as the Pyrenean Metamorphism (Azambre and Rossy, 1976; Bernus-Maury, 1984; Clerc and Lagabrielle, 2014; Clerc et al., 2015; Dauteuil and Ricou, 1989; Ducoux et al., 2019; Golberg and Leyreloup, 1990; Ravier, 1959). This HT/LP metamorphism can be observed along E-W-striking narrow domains of the North Pyrenean Zone (NPZ) corresponding to the so-called Internal Metamorphic Zone (IMZ), in the Basque-Cantabrian Basin (BCB), and further to the south in the Cameros Basin (e.g. Rat et al., 2019). In these domains, Mesozoic sediments recorded metamorphic temperatures up to 600 °C at pressures

as low as 4 kbar for the NPZ (Bernus-Maury, 1984; Clerc et al., 2015; Golberg and Leyreloup, 1990; Vauchez et al., 2013), 580 °C associated with pressure around 3-5 kbar for the BCB (Ducoux et al., 2019; Martinez-Torres, 1989; Mendia and Gil Ibarguchi, 1991; Mendia, 1987), and up to 350 °C for the Cameros Basin (González-Acebrón et al., 2011; Mantilla Figueroa et al., 2002; Rat et al., 2019). This apparent gradual increase of peak-temperature along the IMZ, from west to east is currently explained by non-cylindircal extensional deformation processes along the rift domain (Clerc and Lagabrielle, 2014; Clerc et al., 2015). HT/LP metasediments of the IMZ also display an intense ductile foliation, first attributed to the Pyrenean collisional event (Choukroune 1972, 1976), subsequently related to the pre-orogenic Cretaceous rifting event (Clerc and Lagabrielle, 2014; Clerc et al., 2015; Golberg 1987; Golberg and Levreloup, 1990; Lagabrielle et al., 2010). The Variscan basement, cropping out in the Axial Zone and North Pyrenean Massifs was coevally affected by intense metasomatism and magmatic albititic activity (Boulvais, 2016; 2006, 2007; Fallourd et al., 2014; Pin et al., 2001, 2006; Poujol et al., 2010). Published geochronological data for this HT/LP metamorphic event, indicate ages ranging from Albian to Santonian (110-85 Ma) (Albarède and Michard-Vitrac, 1978a, 1978b; Casquet et al., 1992; Chelalou et al., 2016; Clerc et al., 2015; Golberg and Maluski, 1988; Golberg et al., 1986; Montigny et al., 1986) among which the younger ages are coeval with the onset of Pyrenean shortening. The early stages of the tectonic inversion of the hyperextended rift system rapidly followed the end of the rifting phase during the late Santonian, with the deposition of a syn-orogenic sequence (e.g. Ford et al., 2016; Gómez-Romeu et al., 2019; Mouthereau et al., 2014). This interpretation is supported by field observations in the South Pyrenean Zone (García-Senz 2002; Garrido-Megias and Rios 1972; McClay et al., 2004; Mouthereau et al., 2014; Muñoz, 1992; Teixell, 1998; Vergés and García-Senz, 2001; Vergés et al., 1995), by seismic reflection data in the NPZ (Biteau et al., 2006) and by kinematic reconstructions based on magnetic anomalies, (e.g. Macchiavelli et al., 2017; Nirrengarten et al., 2017; Olivet, 1996; Roest and Srivastava, 1991; Rosenbaum et al., 2002). After a short period of tectonic quiescence during the early Paleocene (Desegaulx and Brunet, 1990; Dielforder et al., 2019; Ford et al., 2016; Grool et al., 2018; Rougier et al., 2016; Ternois et al., 2019), the main collisional phase responsible for the present-day structure of the Pyrenean-Cantabrian belt occurred in Eocene-Oligocene times (Mouthereau et al., 2014; Muñoz, 1992, 2002; Teixell et al., 2018; Vergés et al., 2002) and ended during the Chattian (Ortiz et al., 2020). Thrust faults were however still active in the Southern Pyrénées until the early Miocene (Hogan and Burbank, 1996; Jolivet et al., 2007; Labaume et al., 2016; Millán Garrido et al., 2000; Millán Garrido, 2006; Muñoz, 1992; Oliva-Urcia et al., 2015; Roigé et al., 2019; Teixell, 1996). After the main collisional event, the Eastern Pyrénées were affected by extensional deformation associated with the opening of the Valencia Trough and Gulf of Lion since the middle Oligocene (e.g. Etheve et al., 2018; Gorini et al., 1993, 1994; Jolivet et al., 2020; Mauffret et al., 1995, 2001; Roca, 2001; Roca et al., 1999). The finite structure of the Pyrenean-Cantabrian belt shows an asymmetric double-verging tectonic wedge above the underthrusted Iberian continental lithosphere



Orthez-102 and Bellevue-1 boreholes. (b) Log of the Les Cassières-2 well with plotted Vitrinite Reflectance data along the depth profile. (c) Log of the Bellevue-1 well with plotted Vitrinite Reflectance and dip data along the depth profile. (d) Log of the Orthez-102 well with plotted Vitrinite Reflectance data along the depth profile. Each log is associated with graph showing the evolution of the thermal maturity of organic matter by Vitrinite Reflectance (Ro) versus depth. The brown curve represents the trend of Ro values related to normal statistical and steady Fig. 2. Vitrine Reflectance (R<sub>o</sub>) depth profiles along three wells located in the Western Pyrénées. (a) Geological map of the Western Pyrénées with the locations of the Les Cassières-2, geothermal gradient through time (Cardott and Lambert 1985). The orange line corresponds to a sharp shift of Ro data corresponding to a maturity break.



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increasingly reworked by extensional tectonics toward the east (Beaumont *et al.*, 2000; Chevrot *et al.*, 2018; Jolivet *et al.*, 2020; Mouthereau *et al.*, 2014; Muñoz, 1992; Roure *et al.*, 1989; Teixell, 1998; Teixell *et al.*, 2016, 2018; Vergés *et al.*, 1995).

### 3 Data and methods

In order to constrain the distribution of the thermal record of the Cretaceous H*T*/L*P* metamorphism, we used two analytical methods: (i) the Vitrinite Reflectance ( $R_o$ ) data as an indicator of the diagenetic thermal evolution of organic matter in the range of 50 to 400 °C that can be applied to areas with strong hydrothermalism and (ii) the Raman Spectroscopy of Carbonaceous Materials (RSCM) method as reliable indicator of peak metamorphic temperatures (from 200 °C until 640 °C). Except  $R_o$  data obtained in boreholes, all  $T_{max}$  values are measured on rock sampled on the surface. Using finite thermal maturity data has limits, especially to determine the paleo-geothermal gradient, because  $T_{max}$  cannot be calibrated in age and depth.

### 3.1 Vitrinite reflectance

We provide a large set of unpublished Vitrinite Reflectance data (courtesy of SNEAP, Elf Aquitaine, and TOTAL R&D) in the Mesozoic sedimentary rocks measured in 169 wells drilled in the Western Pyrénées, including the Basque-Cantabrian, Mauléon, Arzacq and Tarbes basins (Figs. 2-4). Vitrinite Reflectance  $(R_0)$  analysis is the most commonly used organic indicator of thermal maturity in low to very low grade metasediments. It is generally used for oil-exploration in order to determine source-rock thermal maturity and maximum temperature  $(T_{max})$  recorded in sedimentary rocks (Taylor et al., 1998). In this study, we used the maturity evolution of  $R_0$ values in boreholes along depth profiles to provide vertical constraints on the relative thermal maturity experienced by rocks from different tectono-stratigraphic levels. In order to fit with RSCM data calculated in this study, we provide a conversion of the Vitrinite Reflectance values to relative  $T_{max}$ based on the formulas published by Barker and Pawlewicz (1994) applied for hydrothermal metamorphism (Tab. S1–S4), but we also provide relative  $T_{max}$  calculated with formulas applied for classical burial heating. The formula for hydrothermal metamorphism is probably more appropriate for rift-related HT-metamorphism because heat seems not only produced by burial. Therefore, we used the formula for hydrothermal metamorphism in pre- and syn-rift sediments located in the NPZ, but we used the formula for burial heating in post-rift, syn- and post-convergence sediments. Evidence for intense metasomatism has been actually reported for both the sediments and basement rocks of the NPZ (e.g. Clerc et al., 2015). For the Aquitaine Basin, only the formula for burial heating is used. However, it should be acknowledged that this conversion is informative and does not represent absolute temperature values as the  $T_{\text{max}}$  obtained with RSCM. It should be further noticed that we will use the Vitrinite Reflectance data in a comparative way, both vertically and laterally to discuss the time and space distribution of the HT event across the Pyrénées. For the purpose the Vitrinite Reflectance value to temperaure conversion itself is not a major issue.

#### 3.2 Raman spectroscopy

Raman spectroscopy analyses were all performed using a Renishaw (Wotton-under-Edge, UK) InVIA Reflex microspectroscope at the BRGM (French Geological Survey) in Orléans equipped with a 514 nm Spectra Physics argon laser in circular polarization. The laser was focused on the sample by a DMLM Leica (Wetzlar, Germany) microscope with a  $100 \times$ objective (NAD 0.90). The Rayleigh diffusion was eliminated by edge filters and the signal was dispersed using a 1800 g/mm grating before being analysed by a Peltier-cooled RENCAM CCD detector. Measurements were performed in situ on polished thin sections cut normal to the main planar fabrics and parallel to the stretching lineation when present (XZ structural planes). To avoid polishing induced damage, CM particles were systematically analyzed below a transparent adjacent mineral, usually calcite or quartz (Beyssac et al., 2002b; Pasteris, 1989; Scharf et al., 2013). Ten to twenty-five points were measured for each sample with 10 to 15 accumulations of 10 seconds acquisition periods. The measured Raman spectra of the carbonaceous material were decomposed for all Raman peaks of carbon by using the PeakFit (v4.06) software (Systat Software Inc<sup>®</sup>).

This analytical method allows to characterize the structural evolution of carbonaceous material (CM), reflecting a transformation from disordered to well-ordered CM during a metamorphic event (Wopenka and Pasteris, 1993). The irreversible reorganization and polymerization of these materials is reflected in their Raman spectrum by the decreasing width of the graphite G band and the gradual disappearance of the defect bands, first D3 and D4, then D1 and D2. The Raman spectrum of well-ordered CM (perfect graphite) contains only the G band. The link between this increasing graphitization and temperature was quantified, leading to a tool to determine peak temperatures attained by metamorphic rocks (Beyssac et al., 2002a). Since graphitization is an irreversible process, the RSCM method gives the peak metamorphic temperatures (Beyssac et al., 2002a; Pasteris and Wopenka, 1991). This is the basis of the RSCM geothermometer, which was calibrated in the range between 330 and 640 °C by Beyssac et al. (2002a). Due to uncertainties related to petrological data used for calibration, the RSCM geothermometer has an absolute precision of  $\pm 50$  °C. Considering a range of measurements, the relative uncertainties on temperature are limited, around 10-15 °C (Beyssac et al., 2004), allowing accurate estimation of field thermal gradients (Bollinger et al., 2004). The RSCM calibration established by Beyssac et al. (2002a, 2002b) was extended towards low temperatures in the range of 200-330 °C with an absolute precision of ±25 °C (Lahfid et al., 2010).

This geothermometer has been applied on 208 thin sections, sampled in Paleozoic to Upper Cretaceous carbonates and pelitic rocks, in order to characterize the distribution of the recorded peak metamorphic temperatures ( $T_{\rm max}$ ) in the study area. All results are presented in Table S5 (see Supplementary material) and Figures 5–7. A systematic sampling was performed in the metamorphic units to complete the previous HT/LP metamorphism map provided by Clerc *et al.* (2015).

# 4 Thermal record of Cretaceous rifting in the Pyrenean-Cantabrian belt

In addition to  $425 T_{\text{max}}$  measurements from previous studies focused on the HT/LP metamorphism, we provide in this paper 208 new  $T_{\text{max}}$  across the Pyrenean-Cantabrian belt, from the Basque-Cantabrian Basin to the west to the Bas-Agly syncline to the east. Twenty were measured in Paleozoic rocks and 188 in Mesozoic metasediments. Syn- to post-rift outcropping rocks in the Western Pyrénées seems to be colder (Clerc *et al.*, 2015; Saspiturry *et al.*, 2020). As they are only discriminant for the rocks exposed today at the surface, we used 169 measurements of Vitrinite Reflectance from boreholes to get insights on the vertical  $T_{\text{max}}$  evolution in this area. Gathering both of the surface and subsurface data allow to propose a new isometamorphic map as well as sections related to former rift basins, which shows the imprint of the paleothermal regime.

# 4.1 Thermal maturity measured with vitrinite reflectance

In this study, we used a large data set of vitrinite reflectance values collected over several decades of petroleum exploration and provided by TOTAL R&D (Tabs. S1-S4). These data are from samples mainly collected in the north Pyrenean foreland, especially in the Aquitaine Basin and the north-western part of the North Pyrenean Zone. In order to describe the thermal maturity of the pre- and syn-rift sediments which are not oucroping in the Western Pyrénées, we used the depth evolution of thermal maturity depth profile of the area along three key boreholes: Bellevue-1, Les Cassières-2 and Orthez-102 wells (Figs. 3 and 4). The observations performed along these wells were then generalized to the Western Pyrénées with a series of maps of vitrinite reflectance values for each tectono-stratigraphic unit from rifting (pre-, syn- and post-rift) to the subsequent collision (early- and late-convergence sequences).

# 4.1.1 Thermal maturity of organic matter along depth profiles

Bellevue-1, Les Cassières-2 and Orthez-102 boreholes, used as examples for describing the thermal maturity of organic matter along depth profiles and are consistently reaching the pre-rift layers. It should be noticed that, considering the thickness and nature of the syn-rift strata they penetrated, they are also respectively representing a distal to proximal trend on the northern side of the Mauléon Basin until the Arzacq Basin further north (Fig. 2). From both surface and subsurface data, this area is described as the highest thermal maturity area of the former hyperextended rift system in the Mauléon Basin (Lescoutre *et al.*, 2019).

The Bellevue-1 well is located within the northern Mauléon Basin and corresponds to the more distal palaeogeographic position (Fig. 2a). The post-/syn-rift boundary is documented by this well at 210 m in depth and the well reached the pre-rift successions at about 2300 m (Fig. 2b). Average  $R_o$  values measured in the post-rift deposits are about ~1%. Using the two formulas for burial heating and hydrothermal metamorphism of Barker and Pawlewicz (1994), such a R<sub>o</sub> corresponds respectively to temperatures of ~135 °C eq (with burial heating formula) and of ~150 °C eq (with hydrothermal metamorphism formula, see Table S2). The maturity trend of organic matter shows a break across the post-rift and the top of syn-rift sediments with R<sub>o</sub> values that slightly increased from 1.0 to 1.49%. From this maturity break, Ro values increase in depth from 1.49 to 2.56% ( $\sim$ 200 to  $\sim$ 275 °C eq) until the base of the syn-rift sediments reached at 2320 m in depth at the exception of the central part of the well where a relative stability is recorded (likely due to a post-deposition deformation as it coincides with a change in the sedimentary bedding). Downsection, the pre-rift sediments starting with the Neocomian deposits were penetrated until the end of the borehole at 6909 m in depth. In the upper part, R<sub>o</sub> values rapidly increase from 2.99 to 3.64% (~290 to ~325 °C eq) between 2400 to 2700 m depth. After a lack of data with only two lower  $R_0$ values ( $\sim 3.30\%$ ) between 2875 to 4000 m depth, R<sub>o</sub> displays a roughly constant trend ranging between 4.46 to 5.26%, corresponding to equivalent temperatures between 343 and 364 °C eq (Table S2). This gradient shift between 3300 to 4000 m depth starts from the axial plane of the Bellevue anticline revealed by dip directions (Fig. 2b). Then, the constant R<sub>o</sub> trend in depth until the base of the well correspond to the steeply dipping limb of the Bellevue fold. In addition of the bedding, the fact that the same stratigraphic level (Barremian) was drilled along more than 2300 m in depth is further indicating this structural setting. As the R<sub>o</sub> trend is following the bedding, this well further reveals that the thermal imprint was recorded before the folding at this location. This vertical stratigraphic level is consistent with the constant value of R<sub>o</sub> along depth profile.

As for the Bellevue-1 borehole, Les Cassières-2 well is located within the northern Mauléon Basin corresponds to a less distal palaeogeographic position compared to the Bellevue-1 well (Fig. 2a). In details, the Les Cassières-2 borehole penetrated the early convergence, post- and syn-rift units and reached the Lower Cretaceous (Neocomian) pre-rift sediments at 5692 m depth (Fig. 2c). The thermal maturity is relatively high in this well, even in the earliest syn-convergence Santonian sediments with Ro values of 2.13%, corresponding to a temperature of  $\sim 200$  °C eq (with burial heating formula, Table S1). It is noteworthy that RSCM measurements and the burial heating formula yield the same evaluation as a  $T_{\text{max}}$  in this area (Saspiturry *et al.*, 2020). From a petroleum system point of view, this value further indicates that the gas window was reached at this stratigraphic level (Tissot and Welte, 1984). Going downsection, Ro values slightly increased from 2.55 to 2.6%. These R<sub>o</sub> recorded in the post-rift sequence are significantly higher than the equivalent measurements of the Bellevue-1 as the corresponding temperature (with burial heating formula) would be in the order of ~210 °C eq (Table S1). At 2200 m depth, maturity evolution rapidly increased, indicating an important break in the trend of maturity as Ro values rapidly increased from 3.05% (~300 °C eq) at 2400 m up to 5.30% (>350 °C eq) at 4200 m (Fig. 2c). Even though absolute values recorded in Les Cassières-2 well differ from the Bellevue-1 well, their vertical trends are similar and show the same maturity break at the top of the syn- to post-rift boundary (Cenomanian). From 4200 m downward, the lower part of the syn-rift sequence shows an

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inflection break in the maturity trend which is almost stable until reaching the pre-rift cover. Then, Ro values are increasing to values ranging from 5.15 to 5.83% (~360 to ~380 °C eq) across the drilled pre-rift successions. At the scale of the entire well, this vertical trend of R<sub>o</sub> values does show significant variations while the sedimentary bedding is constantly gently dipping southwards (Fig. 2c). It should be further noticed that it significantly diverges from a theoretical burial-related record as shown by the expected trend for a stable 30 °C/km geothermal gradient (brown curve in Fig. 2c, Cardott and Lambert 1985). We therefore conclude that the thermal record cannot be explained by the sedimentary burial only. Accounting for similar sedimentation rates between syn- and post-rift times ( $\sim 1 \text{ km}/10 \text{ My}$ ), this shift between our observations and a constant thermal model therefore suggests a high syn-rift paleo-thermal gradient until post-rift times in this area. As R<sub>o</sub> values within the syn-rift sediments systematically exceed the value of 3%, they indicate that the coalification of the organic matter reached the level of meta-anthracites and semi-graphite indicative for metamorphic conditions.

Compared to the above-described Les Cassières-2 and Bellevue-1 boreholes, the Orthez-102 borehole is located further north corresponding to a more proximal structural setting in respect of the rift palaeogeography. It actually penetrated a duplicated stratigraphy on each side of the southdipping North Pyrenean Frontal Thrust (Fig. 2d). The drilled upper unit (hanging-wall) corresponds to a thin postconvergence sequence overlying the syn-convergence, postand syn-rift sediments and terminated within the pre-rift strata. Paleo-geographically, this hanging-wall unit is derived from the northern margin of the Mauléon Basin. Beneath the North Pyrenean Frontal Thrust, the Orthez-102 well penetrated the autochthonous syn-rift sediments at ~4500 m depth until 5489 m deep (Fig. 2d). Despite a lack of data in most of the syn-rift strata of the upper unit, the vertical interpolation of the data at the top and the base of the syn-rift strata suggests a roughly similar vertical maturity trend as in the Les Cassières well. Indeed, the measured Ro values range between 0.92 and 1.27% in the post-rift strata (late Cenomanian, Fig. 2d), which correspond respectively to temperatures of ~130 and ~155 °C eq (Table S3) and the  $R_0$ values of the pre-rift strata ~1.3 km deeper are between 2.87 and 4.05% (287 to 331 °C eq). Most likely because of the lack of data, the thermal maturity break observed within the Les Cassières-2 and Bellevue-1 wells cannot be observed in the allochthonous syn-rift unit of the Orthez-102 well even though the underlying pre-rift sediments reached similar thermal maturities (Fig. 2d). In the footwall of the North Pyrenean Frontal Thrust, from 4350 m depth and deeper, in the maturity trend recorded by the autochthonous syn-rift units shows a signifantly steeper slope than within the hanging wall with values increasing downward from 0.62 to 1.03% (~95 to 140 °C eq). It should be further noticed that these values show the same trend as the brown curve which represents the normal statistical Ro gradient for a normal burial-related thermal gradient of 30°C/km. This indicates that the thermal maturity of organic matter in the autochthonous unit did not recorded a high-temperature event and therefore differs from the thermal record of the neighboring Mauléon Basin.

### 4.1.2 Mapping the thermal maturity of organic matter within each tectono-stratigraphic unit

Thermal maturity data obtained from the three previously described boreholes provide key information on the intensity, the vertical (*i.e.* timing) and spatial distribution of the HT Pyrenean event recorded mainly within the pre-rift and until the midle part of syn-rift sediments. With this information, we compare mean  $R_o$  values from 169 wells (Table S4) where thermal maturities were analyzed, for each tectonostratigraphic layer of the Western Pyrénées (pre-, syn-, and post-rift as well as early- and late convergence deposits). This comparison is illustrated in five maps showing the values for the different stratigraphic levels (Figs. 3 and 4).

Mean R<sub>o</sub> values for the pre-rift unit were measured in Upper Triassic-Barremian sediments (Fig. 3a). Pre-rift sediments were affected by the rift-related and the subsequent convergence-related tectono-thermal events. Their thermal maturities were abundantly documented by Ro measurements. From this dataset, distinctive tectono-thermal domains can be identified within the NPZ and further west in the Basque-Cantabrian Basin. The northern Mauléon Basin systematically shows high thermal maturity with a range of mean R<sub>o</sub> between 3 to 5.60%, corresponding to estimated temperatures between 300 and 370 °C eq, indicating that these rocks underwent metamorphic conditions (Tissot and Welte, 1984). Similar to the Mauleon Basin, the western part of the Basque-Cantabrian Basin shows very high thermal maturity of the organic matter for the pre-rift units located along its northern side. It should be however acknowledged that only a few measurements are available (Robert, 1971) with Ro values exceeding 6-7% corresponding to temperatures above 400 °C eq. Further east, along the Pamplona transfer zone, the thermal maturity remains significantly high with a R<sub>o</sub> of 3.5% (Fig. 3a). However, further east across the pamplona transfer zone (i.e. South Pyrenean Zone), the thermal maturity of the pre-rift units, significantly decreases with Ro values <1%, suggesting an estimated temperature lower than ~130 °C eq. Therefore, the high-temperature domain is restricted to the eastern Basque-Cantabrian Basin on the western side of the Pamplona transfer zone. Concerning the Aquitaine foreland Basin further to the NE, we define in its southern part an area of moderate to high thermal maturity with mean R<sub>o</sub> values ranging between 1.09 and 3.56%. This area concerns the Arzacq Basin which was intensively drilled for petroleum exploration and has also recorded the Cretaceous rifting (Biteau et al., 2006; Brunet, 1984). Further east, the Comminges Basin shows the same range of R<sub>o</sub> values. Farther north, the pre-rift strata of the rest of the Aquitaine foreland shows a contrasting lower thermal maturity with  $R_0 < 0.9\%$ .

As for the pre-rift, the map of mean  $R_o$  values for the synrift units (Aptian-Cenomanian) (Fig. 3b), shows the same spatial distribution of thermal maturities with slightly lower  $R_o$ values, except in the Comminges Basin where measured  $R_o$ values of the syn-rift unit are higher than in the pre-rift units.

The map of mean  $R_o$  values for the post-rift unit is restricted to few measurements in Turonian-Coniacian sediments acquired in the Mauléon and Comminges basins (Fig. 3c). These sediments were deposited after the end of rifting during the post-rift thermal relax. The northern part of the Mauléon Basin shows a moderate to high thermal maturity



**Fig. 3.** Maps displaying the spatial distribution of  $R_o$  data for each rift-related tectono-stratigraphic unit. (a) Average  $R_o$  values measured in prerift sediments (Triassic to Barremian). (b) Average  $R_o$  values measured in syn-rift sediments (Aptian to Cenomanian). (c) Average  $R_o$  values measured in post-rift sediments (Turonian to Coniacian).



**Fig. 4.** Maps displaying the spatial distribution of  $R_o$  data for each convergence-related tectono-stratigraphic unit. (a) average  $R_o$  values measured in early-convergence sediments (Santonian to Maastrichtian). (b) average  $R_o$  values measured in later-convergence sediments (Danian to Chattian).

of the post-rift unit with an average  $R_o$  of 1.30% (~155 °C eq) and a higher value of 2.58% (~210 °C eq).  $R_o$  is 0.71% in the western part of the Mauléon Basin, indicating a low thermal maturity of syn-rift sediments for this area that can be linked to a more limited sedimentary burial (Lescoutre and Manatschal, 2020). The Arzacq Basin located north of the North Pyrenean Frontal Thrust, and the Comminges Basin eastwards both show  $R_o$  values <0.62 which indicate a low thermal maturity in these areas for the post-rift event. Similar values are recorded east of the Basque-Cantabrian Basin with  $R_o$  of 0.7%. Mean  $R_o$  values for the early convergence unit is derived from measurements within the Santonian-Maastrichtian sediments (Fig. 4a). This period corresponds to the onset of inversion of the hyperextended rift system (Gómez-Romeu *et al.*, 2019). Two tectono-thermal domains can be distinguished based on the  $R_o$  data. The first one represents an area with very low-grade thermal maturity with values of  $R_o < 1\%$ indicating estimated temperatures  $< 150 \,^{\circ}\text{C}$  eq. This area is restricted to the Aquitaine foreland Basin and is limited in the south by the North Pyrenean Frontal Thrust. The second domain located further south, corresponding to the Mauléon Basin and the NPZ, shows relatively higher  $R_o$  values exceeding 1% with a maximum value of 2.29% (~260 °C eq). These values are roughly similar to the underlying post-rift mean  $R_o$  values.

Mean  $R_o$  values for the late convergence unit (main orogenic collisional phase) is derived from the measurements within Paleocene-Eocene deposits (Fig. 4b).  $R_o$  is relatively constant in the overall late-convergence unit with values ranging between 0.28 to 0.51%. This range of  $R_o$  indicates estimated temperatures <60 °C eq, corresponding to very low thermal maturities reaching during the main collisional phase of orogeny despite of the sedimentary and tectonic burial.

# 4.2 Distribution of HT/LP metamorphism-related peak temperature of the Pyrenean-Cantabrian belt

## 4.2.1 Mapping the peak-temperatures using RSCM geothermometry

Using the RSCM geothermometer, we provide a riftrelated maximum peak-metamorphic temperature map (Fig. 5). This map displaying different tectonostratigraphic units was built with 633  $T_{\text{max}}$  from our dataset from which 208 are unpublished so far, in addition of 425 from the literature (Angrand et al., 2021; Chelalou et al., 2016; Clerc et al., 2015; Cloix, 2017; Corre, 2017; Ducoux et al., 2019; Golberg and Leyreloup, 1990; Izquierdo-Llavall et al., 2020; Revelli, 2013; Saspiturry et al., 2020; Villard, 2016). Most of the measurements were obtained by the analysis of sedimentary successions of the IMZ from the Basque-Cantabrian Basin in the west until the Agly area in the east (Fig. 5a). Some additional measurements were also retrieved using the Paleozoic metasediments belonging to the upper crustal basement rocks in respect of the Mesozoic rift history. In previous studies (Clerc et al., 2015; Golberg and Leyreloup, 1990), the eastern part of the IMZ was considered to record the highest metamorphic conditions compared to the Central and Western Pyrénées. Actually, even though the Boucheville and Bas-Agly basins yielded a range of  $T_{\text{max}}$  values between 500 to 600 °C within the pre- and syn-rift sediments (Chelalou et al., 2016; Clerc et al., 2015; Golberg and Leyreloup, 1990). Such a range of  $T_{\text{max}}$  was also recorded farther west, close to the Bessède-de-Sault Paleozoic massif (>550 °C), within the southern margins of the Tarascon Basin (>600 °C), in the Aulus Basin where  $T_{\text{max}}$  exceed 600 °C and in the Montillet Basin where the mean  $T_{\text{max}}$  exceeds 550 °C (Fig. 5a). In more details, the Aulus and Tarascon basins from the Central Pyrénées show a range of  $T_{\text{max}}$  between 500 and 630 °C, within their pre- and syn-rift sediments. In this central part of the Pyrénées, the maximum values are generally obtained along the southern side of the North Pyrenean Zone within the Aulus Basin in close association with mantle rocks (e.g. Lherz, Clerc et al., 2012; Bestiac: de Saint Blanquat et al., 2016). A thermal break is observed in the post-rift sequence located along the southern edge of the North Pyrenean Fault (in red in Fig. 5a) with  $T_{\rm max}$  generally ~350 °C. Northwards, the northern margin of the Tarascon Basin lower moderate  $T_{\text{max}} \sim 350 \,^{\circ}\text{C}$  for the same stratigraphic levels at the interesting exception of the ~570 °C yielded by an Upper Triassic pre-rift outcrop. The Trois-Seigneurs Paleozoic massif located between the Aulus and Tarascon basins consists in a rift palaeo-high as its basement is directly covered by post-rift sediments. These

sediments yielded much lower temperatures <200 °C than the surrounding Aulus and Tarascon rift-related troughs (Fig. 5a). It was reported that the Camarade Basin located further north beyond the Arize massif did not recorded the H*T* metamorphic event (Clerc *et al.*, 2015). However, the analysis its pre- and syn-rift sediments contrasts with these interpretations as it provided  $T_{\text{max}}$  measurements ranging between 230 to 330 °C. Such a thermal record cannot be reported from the syn-convergence strata located north of the North Pyrenean Frontal Thrust where  $T_{\text{max}}$  were systematically lower than <200 °C.

Further west, there is a lack of data in most of the Ballongue Basin until its western border corresponding to Milhas-Arguenos area. There, measured  $T_{\text{max}}$  are similar to those observed in the Aulus Basin and are bracketed between 420 and 605 °C (Fig. 5a). As for the Aulus and eastern Tarascon basins this area of high  $T_{\text{max}}$  is characterized by several mantle outcrops (see Clerc *et al.*, 2012; de Saint Blanquat *et al.*, 2016; Lagabrielle *et al.*, 2010 for more information).

Some tens of kilometers to the west, our results show that the Montillet Basin located between the Axial Zone and the Barousse Massif does show  $T_{\text{max}}$  values among the highest reported from the Pyrénées (up to  $625 \pm 17$  °C, Fig. 5a). The pre- and syn-rift sediments from the entire basin shows a range of T<sub>max</sub> between 480 and 625 °C. South of the Montillet Basin beyond the North Pyrenean Fault, recorded T<sub>max</sub> from post-rift sequences abrubtly decrease under 330 °C, with values of  $321 \pm 6$  and  $317 \pm 4$  °C (Fig. 5a). North of the Montillet Basin within the Barronies Basin, the syn-rift sequence recorded a lower HT/LP metamorphism  $(314 \pm 20 \text{ to } 477 \pm 29 \degree \text{C}, \text{Fig. 5a})$ . From the western border of the Montillet and Baronnies basins westwards,  $T_{\text{max}}$  values from the outcropping pre- and syn-rift sequences of the Western Pyrénées are globally decreasing.  $T_{\rm max}$  measured in the Chaînons Béarnais area consistently range from 310 to 390 °C, except along the North Pyrenean Fault within pre-rift sediments where  $T_{max}$  values often exceed 430 °C and can even reach locally  $T_{\text{max}} > 500$  °C (Fig. 5a). It should be noticed that  $T_{\text{max}}$  are increasing in the vicinity of mantle outcrops with values exceeding 400 °C, except for the Saraillé and Urdach outcrops (Clerc et al., 2015; Corre, 2017).  $T_{\rm max}$  are also consistently higher within pre-rift sequence than in younger deposits as shown by the Tres Crouts area, located in the eastern Chaînons Béarnais (see Labaume and Teixell, 2020; Villard, 2016), with values ranging between 420 and 500 °C (Fig. 5a). Westwards, calculated  $T_{\text{max}}$  decrease under 300 °C in the central part of the Mauléon Basin even reaching values under 200 °C along its southern margin (Fig. 5a).

On the western side of the Basque Massifs, the pre- and syn-rift cover of the eastern Basque-Cantabrian Basin, corresponding to the Nappe des Marbres unit, is also affected by the HT/LP metamorphism (Fig. 5a). From this point of view, this basin constrasts with the neighbouring Mauléon Basin as it shows  $T_{\text{max}}$  ranging from 250 to 580 °C. Similarly to the Tarascon Basin located in the Central Pyrénées, the eastern Basque-Cantabrian Basin recorded a thermal N-S asymmetry. It should be however noticed that they are opposite in direction as higher  $T_{\text{max}}$  values are located along its northern border along the Leitza Fault and decrease southward until 250 °C. It is interesting to note that here again highest temperatures were likely associated with mantle rocks as reported by DeFelipe *et al.* (2017). See more details below.





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### 4.2.2 Metamorphic gaps and limits of the HT/LP metamorphic domain

All of the  $T_{\text{max}}$  dataset presented in this paper is used and interpolated to build a regional isometamorphic map to display the spatial distribution of the rift-related HT/LP metamorphism (Fig. 5b). One of the main striking information that can be inferred from this map is that the metamorphic domain is continuously recorded across the entire belt. Based on different N-S distributions, we will however distinguish the Eastern/ Central Pyrénées from the Western/Basque Cantabrian Pyrenean records. In the Eastern and Central Pyrénées first, we show that the highest  $T_{\text{max}}$  values (between 450 to 630 °C) were indifferentially collected along the IMZ from the Montillet Basin in the Central Pyrénées until the Boucheville Basin in the eastern termination of the belt. The southern part of the IMZ is consistently bearing the highest  $T_{\text{max}}$  values  $(>500 \,^{\circ}\text{C})$  and is sharply separated from the much colder late syn- to post-rift record of the Axial Zone (<350 °C) by the North Pyrenean Fault (red line in Fig. 5). Some higher  $T_{\text{max}}$ exceeding 350 °C were obtained from the Paleozoic basement rocks underneath the colder Mesozoic cover suggesting that it may be inherited from the Late Variscan regional metamorphism.

It is commonly assumed that the northern border of the IMZ is limited by the North Pyrenean massifs (Clerc et al., 2015). However, it arises from our dataset that the metamorphism is actually strongly recorded within the Bas-Agly syncline and more moderately within the Camarades and Baronnies basins. Within the Bas Agly syncline, the metamorphic imprint is of the same order than in the Boucheville Basin with  $T_{\text{max}}$  exceeding 450 °C (e.g. Ducoux et al., 2021a). This is the Bas-Agly north-directed basal thrust that marks the northern limit of the HT/LP metamorphism from which with  $T_{\text{max}}$  are lower than 250 °C. Even though the Camarades and Baronnies basins are in a similar structural situation than the Bas Agly syncline, the recovered Tmax values seem to show a less intense metamorphism. However, they are documenting the shallowest part of the syn-rift record from which the deeper levels are not described so far (Espurt et al., 2019). Consequently,  $T_{\text{max}}$  measured at the surface may not reflect metamorphic conditions at the base of this basin.

In the Western Pyrénées, the westward lateral extension of the metamorphic domain is bounded by the North Pyrenean Frontal Thrust. The Chaînons Béarnais area and its equivalent and less deformed Mauléon Basin are less pervasively affected by Pyrenean deformations. The eastern part of the Chaînons Béarnais only is limited from the Axial Zone by the westward continuation of the North Pyrenean Fault. This major structure is actually replaced by the south-vergent thrust Lakhora thrust in the rest of the area that marks the new structural and thermal boundary of the IMZ with the Pyrenean Axial Zone (e.g. Labaume and Teixell, 2020; Teixell et al., 2016). Towards the north, there is no outcropping equivalent of the North Pyrenean Paleozoic massif until the North Pyrenean Frontal Thrust that directly juxtaposes the Chaînons Béarnais and Mauléon Basin cover ontop of the Aquitaine Basin. A likely equivalent of the North Pyrenean Paleozoic massifs would be the buried Grand Rieu ridge (more details below). In this lateral extent of the IMZ, the metamorphic imprint seems less intense than in the Central and Eastern Pyrénées. However, as for the

Baronnies Basin, the deepest part of the rift basin corresponding to the pre- and early syn-rift layers are not exposed in this part of the Pyrénées and are buried beneath a thick pile of syn- and postrift sediments, except within the Chaînons Béarnais area (Fig. 5). This is where the highest  $T_{\rm max}$  values were recovered from the pre-rift strata (>450 °C). These values are comparable to those obtained from the Montillet and Aulus basins of the Central and Eastern Pyrénées. We will partially answer to the missing record of the base of the rift basin using the drillhole data in the following.

As for the Chaînons Béarnais and Tarascon Basin, the highest metamorphic record from the eastern Basque-Cantabrian Basin at the southwest border of the studied area is located in the northern border of the IMZ and is limited by a north-vergent thrust corresponding to the Leitza Fault. This limit corresponds to a severe thermal gap between the metamorphic domain and the late syn- to post-rift sequences located in its footwall, that recorded  $T_{\rm max} < 200 \,^{\circ}{\rm C}$  (Fig. 5). The metamorphic domain is not limited to the south by a major fault but shows a progressive decrease of the metamorphic temperatures instead (Ducoux *et al.*, 2019).

#### 4.2.3 Peak-temperatures distribution at basin-scale

According to our new results on the HT/LP metamorphism, we show that the IMZ has reached high  $T_{\text{max}}$  values >450 °C across the entire Pyrenean rift system, with numerous areas exceeding 550 °C. The IMZ is bounded by first order Pyrenean faults suggesting that they may have been important riftrelated limits as the HT/LP metamorphic event predates the Pyrenean orogenesis. Studying the  $T_{\text{max}}$  distribution in each rift basin is generally complex, because: (i) these rift basins were affected by pervasive orogenic deformations, (ii) the dataset is neither dense enough nor equally distributed to determine precisely the pre-orogenic thermal structure. Nevertheless, using surface data, two favorable areas can be used to describe the spatial record of the HT/LP metamorphism at basin scale (Figs. 6 and 7) as the Eastern Basque-Cantabrian Basin (Nappe des Marbres) and the Central Pyrénées. For the first one the pre- and syn-rift sequences crop out and a dense coverage of surface data  $T_{\text{max}}$  measurements as published by Ducoux et al. (2019). Aside from the folded structure of the basin documented by this previous study, the regional thermal structure clearly shows a complex 3D pattern. First of all, isograds are E-W-elongated but present-day surface  $T_{max}$ values decrease toward the east and the south (Fig. 6). The highest  $T_{\text{max}}$  values significantly exceed 500 °C and are recorded within the pre- and syn-rift sediments located south of the Leitza Fault. This north-directed thin-skinned thrust fault is sharply juxtaposing the highest  $T_{\text{max}}$  values of its hanging-wall with the lowest values of its footwall. It contrats with the more gradual decrease of  $T_{\text{max}}$  values southwards (values <250 °C 10 km away from the fault). Less obvious features can be observed in the Central Pyrénées within the Tarascon Basin. Restricted between the Axial Zone (North Pyrenean Fault) to the south, the Trois-Seigneurs Massif to the west and the Arize-Saint-Barthélémy massifs to the NW, the Tarascon Basin exposes syn-rift sequences and is significantly less deformed than the Aulus Basin located further to the SW (Fig. 7). The spatial record of the HT/LP metamorphism indicates that it was



**Fig. 6.** Close-up of the global map of the HT/LP metamorphism focused on the eastern part of the Basque-Cantabrian Basin (modified from Ducoux *et al.*, 2019). (a) map of the spatial distribution of the  $T_{max}$ . (b) isometamorphic map showing a N-S trending decrease of the HT/LP metamorphism.

more intensively recorded along the southern margin of the basin with  $T_{\text{max}}$  values above 600 °C and then  $T_{\text{max}}$  abruptly drops down to 350 °C toward the north. We also observe an eastward  $T_{\text{max}}$  decrease from  $604\pm20$  to  $410\pm16$  °C and a westward decrease to ~345 °C along the eastern margin of the Trois-Seigneurs massif and toward the eastern termination of the Aulus Basin. Even though the Tarascon Basin is less documented by  $T_{\text{max}}$  data, it also shows a roughly similar thermal asymmetry than the Basque-Cantabrian Basin but with an opposite direction.

# 5 Interpretations of the thermal structure *versus* architecture of the Pyrenean-Cantabrian belt

In order to discuss the record of the HT/LP metamorphism in depth along the Pyrenean-Cantabrian belt, we projected available  $T_{max}$  data from the RSCM method and  $R_o$  values along three N-S cross-sections located respectively in the Eastern Basque-Cantabrian Basin, in the Western Pyrénées and



Fig. 7. Close-up of the global map of the HT/LP metamorphism focused on the Tarascon Basin in the central Pyrénées. (a) map of the spatial distribution of the  $T_{max}$ . (b) isometamorphic map showing a S-N trending decrease of the HT/LP metamorphism.

in the Central Pyrénées (Figs. 8–10). It should be acknowledged that this section is dedicated to discuss the 3D spatial distribution of metamorphism at first order and not to make an exhaustive description of the structure of the Pyrenean-Cantabrian belt, already documented by previous studies (*e.g.* DeFelipe *et al.*, 2017; Labaume and Teixell, 2020; Lescoutre and Manatschal, 2020; Mouthereau *et al.*, 2014; Pedrera *et al.*, 2017; Roca *et al.*, 2011; Tugend *et al.*, 2014).

#### 5.1 The Basque Cantabrian Basin

The Basque-Cantabrian Basin, relatively well preserved from pervasive collisional deformations, results from the shortening of a former hyperextended rift system that recorded mantle exhumation (DeFelipe *et al.*, 2017, 2018; Lescoutre and Manatschal, 2020; Pedrera *et al.*, 2017; Roca *et al.*, 2011; Tugend *et al.*, 2014). The sedimentary cover of the basin consists in a bivergent thin-skinned fold and thrust belt,





(km)

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**Fig. 9.** N-S geological cross-section across the Western Pyrénées (see location on Fig. 1) associated with measured  $T_{\text{max}}$  and Vitrinite Reflectance data. The southern part of the section drawn from surface geology is modified from Labaume and Teixell (2020). According to the seismic reflection data, the vertical scale of the whole section is in two-way traveltime (second). To preserved coherency within the whole section, the southern part of the section constructed with surface geology was drawn in the prolongation of the seismic data based on TWT vertical-scale. (a) 100 km-long N-S stiking geological cross-section through the Chaînons Béarnais, Grand Rieu ridge, Arzacq Basin and the Aquitaine platform, built form a 2D reflection seismic composite line available in Figure S1. (b) close-up focused on the Chaînons Béarnais and the Grand Rieu ridge. (c) close-up focused on the Aquitaine platform.

thrusted southward above the Iberian continental crust along the South Pyrenean Frontal Thrust and northward on top of the European continental crust represented by the Cinco Villas massif along the Leitza Fault (Fig. 8a). The Upper Triassic evaporites act as the main décollement layer located at the interface between the Mesozoic sedimentary series and the underlying basement. As pointed out in the previous section, this basin is anequivaly and homogeneously affected by the HT/LP metamorphism (Fig. 8). Only specific areas expose metamorphic rocks, especially the Nappe des Marbres Unit located in the NE termination of the Basque-Cantabrian Basin (Ducoux et al., 2019). As paleo-isogrades crosscut the preorogenic geometry of the basin at large-scale (Fig. 8), these authors suggested that a significant part of the folding in the Nappe des Marbres Unit, and more widely of the Basque-Cantabrian Basin, predated the regional HT/LP peak metamorphism conditions and resulted from the interaction between salt tectonics and rifting processes. Paleo-isograds are well constrained in the northern part of the cross-section but are extrapolated for the southern part. At basin-scale only a small strip of the basin is affected by temperatures exceeding 500 °C along its northern boundary affecting both pre- and synrift sediments. It structurally corresponds to the basal and more deeply buried part of the basin today exposed along the Leitza Fault. It should be noticed that the highest  $T_{\text{max}}$  coincides with the reported occurence of mantle rocks (serpentinites and ophicalcites) observed along the north-eastern margin of the Nappe des Marbre Unit (DeFelipe et al., 2017; Mendia and Gil Ibarguchi, 1991) demonstrating that the rifting came to a mantle exhumation stage. Toward the south,  $T_{max}$  gently decreases below 250 °C, as shown by the distance-temperature diagram and the geometry of the isograds (Fig. 8b). Isograds are only deformed by collision-related folds and faults. The rest of the basin shows relatively low to moderate metamorphic grade with temperatures on the order of 300 °C, suggested at its base, much lower than the thermal peak recorded at the northern border of the Nappe des Marbres unit. The Leitza Fault abruptly juxtaposes two units of contrasted thermal

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Fig. 10. N-S-striking geological cross-section (see location on Fig. 1) across the Central Pyrénées from the Axial Zone to the south to the Aquitaine Foreland Basin to the north. (a) Geologic cross-section associated with the distribution of the HT/LP metamorphism and relevant graph ( $T_{max}$  versus distance). The isograds were plotted with a margin of error. (b) Close-up on the Mesozoic basins.

records; the lowest conditions being experienced by the socalled Central Depression mostly made of post-rift sediments (with thin and discontinuous syn-rift sequence) unconformably lying on the Paleozoic basement (Fig. 8b). This abrupt lateral decrease in intensity of the metamorphism is shown by the dip of isograds and plotted  $T_{\text{max}}$  in the isometamorphic map (Figs. 6 and 8).

### 5.2 The Western Pyrénées: the eastern Mauléon-Arzacq rift system across the Chaînons Béarnais area

By using 2D seismic reflection composite line (Fig. S1, courtesy of TOTAL R&D), we constructed a ca. 100 km-long geological cross-section through the western part of the Chaînons Bearnais area corresponding to the Eastern Mauléon Basin, going across the Arzacq/Aquitaine foreland basins in the north. All of the nearby available  $T_{max}$  and  $R_o$  values were then projected on this section considering their depth and stratigraphic levels (Fig. 9). As for the Basque-Cantabrian Basin, the Chaînons Béarnais area results from the thinskinned shortening of the pre- and syn-rift cover above the Upper Triassic main décollement. It was also paleogeographically corresponding to the former distalmost part of the hyperextended rift basin as shown by outcropping occurences

of deep basin syn-rift sedimentary facies associated with late syn-rift magmatic intrusions and mantle rocks exposures. Even thought it was more or less intensively deformed during orogenesis as a function of the still debated structural interpretations, it still preserves its first order proximal to distal relationships with its neighboring underthrusted Iberian and European crustal necking zones to the south and the north respectively (Izquierdo-Llavall *et al.*, 2020; Labaume and Teixell, 2020; Teixell *et al.*, 2016; Tugend *et al.*, 2014).

In the southern part of the section, the Chaînons Béarnais display an alternation of salt walls enclosing tightened and inverted minibasins during the collision (Izquierdo-Llavall *et al.*, 2020; Labaume and Teixell, 2020). As for the Nappe des Marbres Unit, salt tectonics may have therefore strongly influenced the pre-orogenic structure of the rift basin, mostly composed of pre- and syn-rift sediments lying above the Upper Triassic salt décollement level and hyperextended rift crust. Thus, a significant part of the folding should predate HT/LP metamorphism in this case too (Izquierdo-Llavall *et al.*, 2020; Labaume and Teixell, 2020). This observation is in agreement with  $T_{max}$  values mostly around 350 °C obtained from surface outcrops. Local higher temperatures within pre- and syn-rift metasediments are observed in the Tres Crouts area as well as in the central and southern parts of the

Chaînons Bearnais (Corre, 2017; Izquierdo-Llavall et al., 2020; Villard, 2016). Even though there is no direct calibration for the  $T_{\text{max}}$  reached at the bottom part of the Chaînons Béarnais, high  $T_{\text{max}}$  values up to  $480 \,^{\circ}\text{C}$  were measured in the vicinity of some of the mantle exposures, *i.e.* the basement underneath the basin. At a first order, it indicates that  $T_{\text{max}}$  may have been significantly higher in depth close to the basin floor where the syn-rift sedimentary burial may have been higher too. The restored geometry of paleo-isograds that we propose also use the  $T_{\rm max}$  estimate temperatures from R<sub>o</sub> values from the nearby Oloron-1 borehole that penetrated >5 km thick syn-rift sequence (Fig. 9a, b). This range of  $T_{\text{max}}$ is actually similar to those obtained from in the central and eastern part of the belt along the IMZ. According to the estimated temperatures obtained from Ro measurements at depth in the different wells located north of the Oloron-1 well, the metamorphism intensity shows a northward moderate to rapid decrease until the North Pyrenean Frontal Thrust that crosscut metamorphic isograds (Fig. 9b). The Chaînons Béarnais unit thrust the Paleozoic basement of the Axial Zone towards the south by reactivating a rift-related north-dipping detachment fault surface (Labaume and Teixell, 2020). It is noteworthy that the underlying basement of the Chaînons Béarnais is poorly exposed on our section as the deformation is essentially thin-skinned. However, two first order observations are providing insights on its variable nature and structural context spatially: (1) the central and northern parts of the Chaînons Béarnais unit bear mantle outcrops indicating that they were lying ontop of an extremely thinned continental crust and/or exhumed mantle rocks; (2) a shortcut of the southern Chaînons Béarnais basement is exposed on the eastern side of the Aspe valley a few kilometers east of our section. It is made of Paleozoic basement rocks covered by its pre-salt autochthonous Lower Triassic cover. Of course, the rift paleogeography should not have been cylindrical, but these observations regionally indicate the occurrence of preserved blocks of upper crust located in the hanging-wall of the reactivated detachment of Labaume and Teixell (2020) but located south of exhumed mantle domains, *i.e.* as the extended basement of the southern Chaînons Béarnais as well as the Mauléon Basin (e.g. Lescoutre and Manatschal, 2020; Masini et al., 2014; Saspiturry et al., 2020; Tugend et al., 2014). As direct constraints on the basement are not numerous in the area, we therefore choose to use structural observations in addition of  $T_{\text{max}}$  in our Western Pyrénées section (Fig. 9a, b). Whatever the detailed syn-rift structure, it is unconformably overlain the post-rift deposits showing moderate  $T_{\text{max}}$  of ~320 °C. This range of  $T_{\text{max}}$  indicates a persisting moderate to high geothermal gradient as recently proposed by recent study in the area (Caldera *et al.*, 2021).

Toward the north and underneath the North Pyrenean Frontal Thrust, the Grand Rieu ridge belonging to the European border of the inverted rift basin corresponds to a former palaeo-high limiting the Mauléon from the Arzacq Basin located farther north. This rift-related setting is indicated by the onlapping post-rift and syn-convergence deposits above a major erosional unconformity which truncates discontinuous remnants of syn- and pre-rift deposits if not directly the basement (Biteau *et al.*, 2006; Canérot *et al.*, 2005, see La Commande-101 borehole). In this area, the post-rift sequence show a moderate to high thermal maturity of organic matter with  $R_0$  of 1.21% until 2.7%, corresponding to temperatures between 180 to 280 °C eq, observed respectively in Cardesse-2 and Le Rouat-1 boreholes (Fig. 9a, b and Tab. S4). Underlying Permian-Lower Triassic sediments preserved on top of the Grand Rieu ridge recorded similar  $R_o$  values with 2.3 and 3.1%, whereas the basement shows higher  $R_o$  of 4.7% (~350 °C). The Pau anticline, penetrated by the Pont-d'As-5 well (Fig. 9a, b) is well imaged by the seismic data. As there is no evidence for a significant top-basement décollement south of this thin-skinned fold neither on seismic data nor in wells, we interpret this structure as the surface expression of the orogenic tightening of the Arzacq Basin located further north (i.e. frontal triangular-shape structure). At equivalent burial, recorded R<sub>o</sub> values within the post-rift sequence show a large lateral variation ranging from 2.5-2.7% for the Le Rouat-1 borehole in the south to 1.0 and 0.7% respectively for La Commande-101 and Pont d'As-5 boreholes (Fig. 9a, b). As for the post-rift sequence, syn-convergence deposits display values of  $R_o \sim 0.6-0.7\%$  in the La Commande-101 and ~0.5-0.6% in the Pont-d'As-5 boreholes, while it recorded 1.7% in the Le Rouat-1 borehole. These values characterize a lower thermal maturity of organic matter and are similar to R<sub>o</sub> documented by Pont-d'As-5 borehole in the younger postconvergence sediments with values of  $R_0 \sim 0.4\%$  (Fig. 9b). It is interesting to note that this decrease from post-rift to synconvergence deposits and from south to north corresponds, as a paradox, to the maximum syn-orogenic sedimentary burial underneath and along the North Pyrenean Frontal Thrust. On the northern margin of the Grand Rieu ridge, pre-rift sediments show moderate to high values of  $R_o \sim 2.5\%$  corresponding to a temperature of 270 °C eq. A thermal maturity break is observed within the overlying syn-rift sediments with  $R_0$  of 0.7%. This specific point is discussed below.

The northern slope of the Grand Rieu ridge is flanked by a north-dipping low-angle fault which accommodated the formation of the Arzacq Basin northwards (Ducoux et al., 2021b). This syncline-shaped basin highlighted by the pre-rift units above the salt décollement level is infilled by a thick sequence of syn-rift sediments (~2s TWT). By using estimated temperatures from Ro, we suggest that the deepest part of this basin filled by ~5 km of compacted pre- and syn-rift sediments, recorded temperatures exceeding 250 °C (Fig. 9a), at the depocenter that may explain the significant quantity of dry gas discovered in this oil and gas province (e.g. Biteau et al., 2006). Further north, the top basement and the pre-rift sequence get shallower along the gently rising northern margin of the Arzacq Basin until the Aquitaine platform. The northern edge of this section represents the Aquitaine Platform where the pre-rift units is covering a thin layer of Upper Triassic evaporitic level which accommodate its underlying slightly faulted basement. The syn-rift sequence appears thinner in this more proximal area (<1 s TWT) and is pinching out toward the north similarly than the pre-rift sequence (Fig. 9a, c). A succession of boreholes penetrating the Aquitaine Platform documents the thermal maturity of organic matter in the overall stratigraphic column (see above and Fig. 9c). Average Ro values in the pre-rift sediments are  $\sim 1\%$ . In the Theze-1 borehole R<sub>o</sub> values ranging between 0.8 to 1.27% contrast with  $R_{\rm o}$  values exceeding 2.5% on the southern side of the Arzacq Basin (e.g. Pont-d'As-5) with a similar burial (Fig. 9a). The rest of the drilled sediments on the northern side of the

Arzacq Basin (post-convergence to syn-rift) show low to very low thermal maturity with  $R_o$  values <0.5%, decreasing upsection to 0.3% (Fig. 9c). This range of values corresponds to estimated temperatures <60 °C.

#### 5.3 The Central Pyrénées

Unlike the Basque-Cantabrian Basin and the Western Pyrénées, the Central Pyrénées were intensively affected by a collisional overprint (*e.g.* Choukroune and the ECORS Team, 1989; Ford *et al.*, 2016; Mouthereau *et al.*, 2014). Only discontinuous remnants of the Mesozoic sediments belonging to the IMZ are preserved between the Axial Zone and the North Pyrenean Massifs. Further north, the Camarade basin, located at the northern margin of the Arize massif, overthrusted the Aquitaine Foreland Basin, which is mainly filled with synconvergence sediments (Fig. 10). Along this transect,  $T_{max}$  values were measured in all units from the Axial Zone in the south to the Camarade Basin in the north. This transcect documents both the  $T_{max}$  recorded within the Mesozoic sediments affected by the Cretaceous rift-related metamorphism only and Paleozoic rocks that were also affected by earlier Variscan metamorphism.

The Axial Zone records a gentle decrease of temperature from 495 °C in the south to  $347 \pm 2$  °C close to the North Pyrenean Fault, its northern limit (Fig. 10a). Pinched between the Axial Zone and the IMZ, deformed post-rift sediments show relatively high  $T_{\text{max}}$  of  $327 \pm 14$  °C. Further north across the North Pyrenean Fault, the IMZ records very high  $T_{\rm max}$  of  $549 \pm 35$  and  $562 \pm 30$  °C.  $T_{\text{max}}$  observed in the post-rift sediments directly overlying the Trois-Seigneurs massif contrast with the IMZ, with values <200 °C. This massif formed a basement high during the rifting event, as suggested by the lack of pre- and syn-rift sediments. As for the IMZ, a second small basin filled by Mesozoic sediments is pinched between the Trois-Seigneurs and Arize massifs further north. The recorded  $T_{\text{max}}$  of  $348 \pm 7 \,^{\circ}\text{C}$  in this area indicates a lower intensity of the HT/LP metamorphism within this basin compared to the IMZ (Fig. 10b).

The southern margin of the Arize Massif, composed of metasediments, shows  $T_{\text{max}}$  exceeding 600 °C. A thermal break can be observed in the central part of the massif where  $T_{\text{max}}$  rapidly decrease to ~400 °C. From this break,  $T_{\text{max}}$  gently decreases under 300 °C, in agreement with the  $T_{\text{max}}$  close to 300 °C recorded in the syn-rift sediments of the Camarade Basin in the syn-rift sediments (Fig. 10). Interestingly, measured  $T_{\text{max}}$  in the northern margins of the Arize Massif composed of Devonian sediments are significantly lower than the  $T_{\text{max}}$  observed in the same stratigraphic levels of the Axial Zone. The Aquitaine Foreland Basin has not recorded the HT/LP metamorphism, with  $T_{\text{max}} < 200$  °C measured in the early syn-convergence sediments (Santonian to Maastrichtian in age).

### 6 Discussion

### 6.1 Time constraints on the HT/LP metamorphic event

The most constraining observations regarding the timing of the HT/LP metamorphic event come from the Vitrinite

Reflectance values obtained from boreholes data. They provide crucial information on the vertical maturation trends within both the Mauléon and Arzacq basins located in the Western Pyrénées (Figs. 2, 3 and 4). The depth evolution of R<sub>o</sub> values generally shows a break between two linear trends corresponding to the late syn-rift and the early post-rift sequences (Cenomanian). The lower trend affects the pre- and syn-rift depositional sequences and represents a higher thermal maturity gradient in comparison with the upper trend. Such a vertical evolution in the absence of a major erosional unconformity can be interpreted as a decrease of the thermal gradient in between the two sequences (i.e. syn-rift/post-rift and syn-orogenic time, e.g. Allen and Allen, 2013; Dow, 1977). It can also be noticed that the vertical trends of the R<sub>o</sub> values are affected by the Pyrenean orogenic deformation. This is first demonstrated by the Bellevue-1 well where the vertical trend of the R<sub>o</sub> is not depth-dependent along the drilled vertical fold limb (base of the well; Fig. 2b). Isovalues of R<sub>o</sub> seem to be bed-parallel and subsequently folded during the tectonic inversion. In the Orthez well, it can also be shown from the sudden vertical decrease of Ro values across the NPFT that the maximum maturity was locally reached before thrusting (Saspiturry et al., 2020). From the map in Figure 4a displaying the spatial distribution of R<sub>o</sub> data as a function of the stratigraphic interval (*i.e.* not burial-dependent), a strip of high values anomalies (ranging between 0.9 to 2.29%) can also be reported within early syn-orogenic deposits in the vicinity of the NPFT (*i.e.* along the Grand-Rieu ridge, Figs. 4a and 9). This narrow strip of high temperatures will be discussed in more detail in the following section as a likely signature for early orogenic hydrothermal circulations (Bahnan et al., 2020; Renard et al., 2019; Salardon et al., 2017).

These new data are in general agreement with the literature that directly or indirectly suggests that the HT/LP metamorphic event is Upper Cretaceous in age throughout the Pyrénées (Albarède and Michard-Vitrac, 1978a, 1978b; Casquet *et al.*, 1992; Chelalou *et al.*, 2016; Clerc *et al.*, 2015; Golberg and Maluski, 1988; Golberg *et al.*, 1986; Montigny *et al.*, 1986). The timing of the HT/LP metamorphic event is also supported by the subsidence analyses from the Aquitaine Basin showing that a part of the syn-orogenic subsidence is related to thermal relaxation and require a late rift residual thermal anomaly reaching the Aquitaine Basin (Angrand *et al.*, 2018; Brunet, 1984).

# 6.2 Intensity and spatial distribution of the HT/LP event

Generally, it has been reported that the H*T*/L*P* metamorphic oveprint is restricted to the NPZ and that the maximum intensities (>500 °C on pre-rift strata by RSCM) was reached within the IMZ (*e.g.* Clerc *et al.*, 2015). It has also been proposed, based on these maximum values collected at the present-day surface, that there is a progressive decrease of the intensity of the thermal anomaly from east to west along the Pyrénées reaching a minimum in the Mauléon Basin (Clerc and Lagabrielle, 2014; Clerc *et al.*, 2015). Our dataset is nuancing these conclusions. Similar high  $T_{max}$  measurements were undifferently obtained from the Eastern and Central Pyrénées as well as from the Basque-Cantabrian Basin (Nappe des

Marbres unit) located at the SW edge of the study area (>500 °C on RSCM data), *i.e.* further west from the Mauléon Basin. Moreover, once considering wells data from the Mauléon Basin, a downward increase of T<sub>max</sub> values is revealed by  $R_o$  measurements traduce a much higher  $T_{max}$ values at the base of the basin (~400-500 °C) from which the deepest (and hotter) levels have never been reached by drillholes. These high temperatures are consistently reached by pre- to syn-rift strata deeply buried underneath the northern (and deepest) part of the Mauléon Basin, partly outcropping in the Chaînons Béarnais fold and thrust belt, at the easternmost termination of the Western Pyrénées (Fig. 9). Such palaeo-temperatures also explain why oil and gas exploration has been generally discarted from the NPZ. These results are in agreement with Saspiturry et al. (2020) proposing similar maximum temperatures distribution within the Mauléon Basin. In order to enlarge this spatial distribution at the scale of the entire rift system and for the whole Pyrenean-Cantabrian belt, we propose in this study an interpolated map combining surface and subsurface data to better locate  $T_{\text{max}}$ values reached at the base of the Pyrenean rift basin (Fig. 11). From this dataset, it can be proposed that the E-W variations of surface  $T_{\text{max}}$  values may not be due to the intensity of the thermal anomaly itself (i.e. "hot" versus "cold" rifting as addressed by Clerc and Lagabrielle, 2014) but are rather due to the burial conditions of outcropping pre- and syn-rift sediments along the NPZ. In other words, as observed in the geological cross-sections (Figs. 8-10), Eastern and Central Pyrénées outcrops are mostly made of discontinuous remnants of the former basal part of deeply buried rift basins and its exposed underlying basement (i.e. younger deposits were eroded) whereas the Western Pyrénées do preserve a more complete, less deformed and shallower rift sedimentary succession that were located closer to the seabed at the thermal climax. In the Western Pyrénées former rift basins were sampled as a pop-up above the colliding European and Iberian continents. This situation is partly due to the low frictional Upper Triassic evaporites that acted as a basal décollement that efficiently decoupled thick-skinned from thin-skinned deformations during basin inversion. It partly explains the preservation of rift architecture like in the Chaînons Béarnais characterized by tightened salt diapirs/ walls bracketting inverted minibasins on top of the forming thick-skinned orogen (e.g. Izquierdo-Llavall et al., 2020; Labaume and Teixell, 2020). According to our field observations, the outcropping rocks in the Chaînons Béarnais mostly represent the shallow part of the basin made of a succession of tightened minibasins and salt walls above a thick pile of Mesozoic sediments (Fig. 9) (Labaume and Teixell, 2020). Further west, the Mauléon Basin mainly exposed at the surface even more preserved late syn-rift to post-rift sediments above up to 5 km of syn-rift sediments at the depocenter as suggested by wells (e.g. Lescoutre et al., 2019; Saspiturry et al., 2020). From this point of view, the Western Pyrénées contrasts with the Central and Eastern Pyrénées from the exposed structural level at the present-day surface (deep versus shallow parts of rift basin infill) (e.g. Ford et al., 2016; Mouthereau et al., 2014). Such a difference may be due to different orogenic processes (orogenic non-cylindricity, e.g. Muñoz, 2002; Roca et al., 2011), to a lateral change in the preorogenic rift geometry (rift non-cylindricity, e.g. lateral change in the rift polarity of Tugend et al., 2015), or a combination of these two with different modes of rift inversion along strike (e.g. Chevrot et al., 2018). Because of this preservation issue, reaching the N-S thermal architecture within the former rift basins can be done in detail within the Western Pyrénées, where the thermal record is preserved at depth. From our dataset, it can be shown that the Vitrinite Reflectance maturity profiles with depth are much steeper across the Arzacq Basin (e.g. Thèse well in Fig. 9) than along the Grand Rieu ridge and the northern slope of the Mauléon Basin (Fig. 11) with similar burial depth. The Grand Rieu ridge record is even more discriminant. As shown by onlapping post-rift sediments above the erosional surface truncating the pre- and syn-rift sequences along this rift relief (Biteau et al., 2006; Canérot et al., 2005; Labaume and Teixell, 2020; Masini et al., 2014; Teixell et al., 2016), the syn- and post-rift burial was extremely limited. From the Grand Rieu ridge southwards, high  $T_{\text{max}}$ values are rapidly increasing toward the juxtaposed deepest part of the Mauléon Basin. In agreement with Lescoutre et al. (2019) conclusions, this dataset further indicates that syn-rift heat flow may have been significantly higher toward the more distal and more extended rifted domains. This latter sampled by the NPZ was characterized by exhumed mantle (Debroas et al., 2010; Fortané et al., 1986; Jammes et al., 2009; Lagabrielle et al., 2010; Masini et al., 2014) and late Albian/ Cenomanian alkaline magma intrusions (e.g. Azambre and Rossy, 1976; Azambre et al., 1992; Montigny et al., 1986; Rossy et al., 1992). Such a thermal climax may reflect the increased lithospheric thinning in this direction as well as the spatial migration of deformation from rift borders inward (Lescoutre et al., 2019; Tugend et al., 2015). This interpretation is in agreement with the former width of the rifted domain affected by the Pyrenean HT/LP event revealed by our dataset (Fig. 12) including both the Southern Arzacq and the Mauléon basins as well as the northern part of the Axial zone It should be noticed that evidence for a hot thermal gradient were also discovered in the northern border of the Axial zone (less intense than the former hyperextended rift domain, e.g. Airaghi et al., 2020; Bellahsen et al., 2019). At present, this area is  $\sim 60 \text{ km}$  wide in the N-S section in the Western Pyrénées (Figs. 9 and 12a, b) not considering the orogenic shortening accommodated within the Mauléon Basin. Accounting for the Pyrenean shortening accommodated within this domain, the initial N-S width of the metamorphic domain should have been in the order of 80-100 km (Figs. 12a, b) (Jammes et al., 2009; Labaume and Teixell, 2020; Teixell et al., 2016).

Replacing the IMZ in respect of the entire rift system may also be meaningful to address rift to orogenic processes. For this purpose, it should be first acknowledged that the zone affected by the HT/LP metamorphism within the Western Pyrénées does not corresponding to an entire rift section between the stable European and Iberian crusts. The entire N-S rift section within the Western Pyrénées (Fig. 12e, f) is made of two juxtaposed rift depocenters overlying thinned crustal (and locally mantle) domains: the Mauléon and Arzacq basins. The Mauléon Basin, in the southern half of the rifted section, is entirely recording the HT/LP metamorphism that increasing northwards and with an increased sedimentary burial along the tapering Iberian crust from South to North. The basement of the Mauléon Basin was made of the crustal necking zone





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**Fig. 12.** Comparison of rift-related domains with the spatial distribution of the HT/LP metamorphism for the present-day structure and for the end of rifting restoration. (a) Rift-related map of the Pyrenean-Cantabrian belt modified from Tugend *et al.* (2014) and Lescoutre and Manatschal (2020). (b) The same rift-related map associated with the distribution of the HT/LP metamorphism shown in the Figure 11. (c) map of the restored Pyrenean-Cantabrian rift system modified from Tugend *et al.* (2014) and Lescoutre and Manatschal (2020). (d) The same map of the restored rift associated with the distribution of the HT/LP metamorphism. (e) Crustal-scale cross-section restored at the end of the rifting modified from Masini *et al.* (2014), Gomez-Romeu *et al.* (2019) and Ducoux *et al.* (2021b), (f) associated with isotherms of the HT/LP metamorphism.

(represented by the northern tip of the Axial zone, the Lakhora thrust sheet and the Aldudes massif), the hyper-thinned crust and the exhumed mantle domain (Chaînons Béarnais and Northern Mauléon Basin) from south to north. Thermochronological results performed along the Iberian necking zone of the Mauléon Basin document geothermal gradients up to 80 °C/km (Hart *et al.*, 2017) in agreement with the recent  $T_{\text{max}}$ dataset of Saspiturry et al. (2020). This thermal record contrasts with the European side of the rift system. From north to south, the European necking zone (zone of crustal thinning sensu Manatschal, 2004) corresponds to the southward tapering continental crust underneath the Arzacq Basin that form coevally to its Iberian conjugate during the Aptian - Early Albian phase of rifting (Masini et al., 2014; Tugend et al., 2014). Both subsidence analysis (e.g. Brunet, 1984) and more recent geophysical images (Chevrot et al., 2018; Wang et al., 2016) indicate that the European crust thins by a factor of 2 from the Aquitaine plateform in the north to the Grand Rieu ridge in the south (made itself of a thinned piece of European

crust). Despite the intensity of crustal thinning, our dataset shows that the European necking zone was not significantly affected by the HT/LP metamorphic event showing that the northern half of the rift section was located away from the rift thermal anomaly. Whatever the underlying rift-related process, it indicates a first order thermal asymmetry at the scale of the complete rift system. From the available pure sheardominated rift models (e.g. Labaume and Teixell, 2020; Lagabrielle et al., 2020; Saspiturry et al., 2020; Teixell et al., 2016), similar thermal records would be expected for equivalent stretching/thinning factors (i.e. equivalent structural domains for conjugate pairs of rifted margins). They cannot easily explain the diverging thermal evolutions recorded by the European and Iberian necking zones and thinned crustal domains arising from our dataset. Former authors were reporting that there is a misfit between the upper crustal stretching (limited number of normal faults) and the observed crustal thinning along the Arzacq Basin (e.g. Masini et al., 2014; Saspiturry et al., 2019). If they disagree on the regional rift geometry, they all converge on proposing depthdependent processes to thin the European crust from north to south, that is a diagnostic criterion for a simple shear component of rifting (heterogenous vertical thinning) and fits with an upper plate relative position at the considered location (e.g. Lister et al., 1986). As for the thermal record, these structural observations within the Arzacq Basin therefore diverge with those from the Mauléon Basin too, where evidence for high-strain top-basement extensional structures (detachment faulting) and exhumation of deep crustal rocks make a large consensus at present (e.g. Lagabrielle et al., 2020; Masini et al., 2014; Saspiturry et al., 2020). Both the structural and thermal observations were recently simulated by thermo-mechanical modeling experiments (Lescoutre et al., 2019) and is in good agreement with our palaeothermal dataset. These models generally predict a persisting and high lateral thermal gradient at the tip of the upper plate margin what is best exemplified by the Grand Rieu ridge tectonothermal record (Figs. 9, 11 and 12). Therefore, we think that the varying spatial tectono-thermal record across the Western Pyrénées are indicating a rift asymmetry where the Iberian side would represent a lower plate evolution whereas Europe would represent its upper plate counterside (Gómez-Romeu et al., 2019; Masini et al., 2014; Tugend et al., 2014). The total initial width of the entire rift system, including the lower and upper plate crustal tapers may therefore be >140 km in the Western Pyrénées. It falls into similar numbers regarding the Pyrenean-like Porcupine Basin (NW Ireland) where hyperextension rifting was presumably stopping at a mantle exhumation stage (Reston et al., 2001; Watremez et al., 2018). A similar asymmetric thermal architecture is also revealed from the Basque-Cantabrian Basin data (Ducoux et al., 2019) located further to the SE. Even more heavily affected by Pyrenean deformations, the same first order thermal asymmetry can also be unraveled from the Central and Eastern Pyrénées dataset where the sharp lateral thermal gradient is however switching on the opposite border of the rift as the highest  $T_{\text{max}}$  values are located along the southern side of the NPZ juxtaposed to the North Pyrenean Fault (e.g. Tarascon Basin, Figs. 11 and 12). Already known for its present-day non-cylindricity of the orogenic structure (e.g. Chevrot et al., 2018), such a lateral shift of the rift thermal structure may further support that the non cylindricity may also be inherited from the Pyrenean rift history (e.g. reversal of rift polarity of Tugend et al., 2015 and Chevrot et al., 2018; Fig. 12).

# 6.3 Evidence for a significant role of hydrothermal fluids for the Pyrenean thermal record

The regional scale of the HT anomaly discussed above is further complicated by smaller-scale spatial variations recorded in the  $T_{\rm max}$  values. First, a small strip of high R<sub>o</sub> values (1.0 to 2.58%; 150 to 250 °C eq) is observed can be reported from the post-rift and early syn-orogenic deposits along the NPFT above the Grand Rieu ridge of the Western Pyrénées (Figs. 4a and 9). A likely possibility is that this area may have been anomalously buried underneath a thick pile of sediments emplaced by either sedimentation or thrusting (or a combination of both) which was subsequently eroded. It should be noticed that much lower  $T_{\rm max}$  values are recorded both northward (La Commande-101 and Pont-d'As-5 wells) and southward (Cardesse-2 well) in spite of a deeper burial depth indicated by the local Pyrenean tectono-stratigraphic architecture (Fig. 9). As it is geometrically related to the NPFT, an alternative scenario is that this HT imprint is rather due to hydrothermal circulations along the NPFT that was channeling rising hot fluids to this precise location (Fig. 9). Contrasting with the rest of the area, this thermal record is early orogenic as it affects latest Cretaceous deposits but no younger units. Therefore, this HT is coeval with the northward underthrusting of a part of the former hyperextended basement of the Mauléon Basin underneath Europe below the Grand Rieu Ridge (e.g. Gómez-Romeu et al., 2019; Tugend et al., 2014). From the record of the Mauléon Basin as well as the rest of the NPZ, it was demonstrated that this basement was made of highly hydrated minerals (serpentinized mantle, altered continental crust, Clerc et al., 2013; Corre et al., 2018; DeFelipe et al., 2017; Incerpi et al., 2020b; Monchoux, 1970; Quesnel et al., 2019). Thus, we further speculate that those fluids may have been sourced by the syn-orogenic dehydration of these rocks during their prograde metamorphic evolution at depth. Such a complex fluid-controlled thermal regime was already well identified within syn-rift times (Bernus-Maury, 1984; Clerc et al., 2015; Corre et al., 2016; Golberg, 1987; Incerpi et al., 2020a; Lagabrielle et al., 2019a; Motte et al., 2021; Mukonzo et al., 2021; Renard et al., 2019; Salardon et al., 2017). Scapolites observed in the syn-rift sequence are evidence for salt-rich fluids having circulated through the sedimentary succession, which originated from Upper Triassic evaporites (Clerc et al., 2015; Golberg and Leyreloup, 1990).

In the hotter and more distal part of the rift located north of the Mauléon Basin and south of the Grand Rieu ridge, it has already been noticed that the HT record is spatially associated with occurences of late rift alkaline magma (the so-called "Episyenites"), hyperthinned continental crust and exhumed mantle rocks (serpentinites and ophicalcites, Clerc et al., 2013; DeFelipe *et al.*, 2017). All of those rocks imply a significant magmatic heat advection and hydrothermal activity that should strongly impact the thermal regime of the rift system. We cannot exclude that the syn-convergence HT record may relate to the remaining rift-related heat-flow and the delay of thermal relaxation (e.g. Bellahsen et al., 2019; Caldera et al., 2021; Vacherat et al., 2014). All of these observations in pre- and syn-rift sequences are commonly indicating a major role of fluids influencing the thermal record. These syn-tectonic thermal regimes (either under rifting or early orogenesis) were not entirely conductive but also imply fluid flow. In such a context, the resulting geotherm may not have been a linear depth-dependent function but should strongly vary both vertically and laterally as a function of the geometries of convection cells (e.g. Incerpi et al., 2020a, 2020b; Pinto et al., 2015), what could explain the apparent rapid lateral variations of measured  $T_{\text{max}}$  values in our dataset at a more local/subbasin scale.

Different thermal records of the Mauléon and Arzacq basins may also be related to different fluid-related thermal regimes and may be due to different structural settings with respect to hyperextension rifting. As proposed by Pinto *et al.* (2015) and Incerpi *et al.* (2020b) in the Alps and by Incerpi *et al.* (2020a) in the Pyrénées, hydrothermal fluids along hyperextended domains of rifts transport the heat from the

active part of detachment faults along the fault plane and therefore reach the exhumed domain and the supra-detachment rift basin. This asymmetric situation makes that the thermal effect of incoming "hot" fluids is more favorably recorded within supra-detachment basins (along the footwall of the detachment faults) than within basins located in the hangingwall of detachment systems, as for instance the Arzacq Basin (Fig. 12e, f). If valid, this model strongly supports a supradetachment thermal record for the Mauléon Basin along which fluids may have intensively interacted with the sediments south of the Grand Rieu ridge (Fig. 12e, f). As discussed by Lavier et al. (2019) and proposed in the Pyrénées by Clerc et al. (2015), it may indicate that the thermal maximum could be delayed because of the decrease of hydrothermal circulations through time. While decreasing, the thermal regime becomes more conductive, implying a heating phase because of the low thermal conductivity of sediments (i.e. blanketing effect) that could exceed and erase the older thermal record.

The Arzacq Basin thermal record would rather fits, by contrast, with an upper plate situation record as only its southern tip records a late thermal H*T* event. The more limited effect of fluid circluations in this area may have promoted a continuous and progressive conduction-dominated syn to post-rift thermal regime. As being mostly conductive, an upper plate situation with respect to the detachment systems of late rifting (above the "exhumation channel" of Brune *et al.*, 2014) would imply a maximum delay in the recorded  $T_{\text{max}}$ . (*i.e.* post-rift) in respect of active rifting. Such a post-rift thermal maximum is documented in this area along the southern end of the Arzacq Basin as well as along the Grand Rieu ridge and further supported by an "anomalous" late to post-rift uplift (in the order of ~2 km, see Renard *et al.*, 2019 for more details).

# 6.4 Role of sediments for hyperextended rift thermal record

Of course, sediment burial is needed to record a  $T_{\text{max}}$ , whatever the gradient and thermal regime in space and time. More than a thermal tape-recorder, sedimentation also actively interacts with the thermal regime of rift systems. As globally discussed by Lavier et al. (2019) for hyperextended rifting, sediments induce a so-called blanketing effect (e.g. Blackwell and Steele, 1989; Callies et al., 2018; Lucazeau and Le Douaran, 1985; Nunn and Lin, 2002; Pollack and Cercone, 1994; Wangen, 1995) resulting from their low initial heat-flow and their low thermal conductivity above a thinned lithosphere and a high basement heat-flow. This effect widely described for the Pyrenean rift (e.g. Asti et al., 2019; Clerc and Lagabrielle, 2014; Clerc et al., 2015; Lagabrielle et al., 2019b), is characterized by progressive heating and a delayed thermal climax during heat conduction across a rift basin. While interacting with hyper-extension processes, the blanketing effect could lead to an extremely complex spatial thermal structure (Lavier et al., 2019). During hyperextension, this is mostly due to the spatial tectonic migration toward the rift axis forced by detachment faulting (i.e. toward the future ocean if a breakup is recorded, e.g. Brune et al., 2014; Péron-Pinvidic and Manatschal, 2019 and references therein). Accounting for thermal blanketing with tectonic migration and increased

lithospheric thinning, the recorded  $T_{\text{max}}$  are expected to get younger and more intense toward the rift axis, and therefore might even post-date rifting in the younger and more distal part of a hyperextended rift because of the conduction delay. This tendency should even be reinforced by higher sedimentation rates and/or thicker sedimentary cover. In the Pyrénées, despite rift basins were underfilled as shown by the development of deepwater conditions, the distal part of the hyperextended rift was likely covered by a pre-, syn- and post-rift sedimentary cover locally exceeding 5 km in thickness before the onset of shortening (Debroas, 1990; Ducoux et al., 2021b; Labaume and Teixell, 2020; Rougier et al., 2016). Effectively corresponding to a sedimented hyperextended rift system and in agreement with former studies (e.g. Angrand et al., 2018; Clerc et al., 2015; Hart et al., 2017; Jourdon et al., 2019; Saspiturry et al., 2020; Ternois et al., 2019; Vacherat et al., 2014), our dataset strongly confirms that a late to post-rift (and a likely early orogenic) HT record is consistently recorded along the entire NPZ of the Pyrénées and the northern side of the Cantabrian belt (Ducoux et al., 2019). It is noteworthy that such a delayed Mid-Cretaceous thermal climax is not only restrictred to the Pyrenean record sensu stricto but was also reported from the nearby but slightly older Cameros Basin located further south (Rat et al., 2019) (Fig. 11). In this case, an even thicker sedimentary cover accumulated within the rift basin during and after the rifting (Omodeo-Salé et al., 2017) and could have contributed to further delay the recorded  $T_{\text{max}}$ with respect to its initial rifting evolution.

The effect of sediments for the rift-related thermal record is also evidenced while comparing the Pyrenean record to its Alpine analogue. Of Jurassic age, the Alpine Tethys rifting came to an even more mature stage of divergence as it experienced, at least, an incipient seafloor spreading stage as recorded by the presence of ophiolites (e.g. Bernoulli et al., 2003; Decandia and Elter, 1972; Froitzheim and Manatschal, 1996; Lagabrielle and Cannat, 1990; Lemoine et al., 1986; McCarthy et al., 2018, 2020; Picazo et al., 2016). However, with the exception of direct indicators within basement rocks or indirect diagenetic/fluid inclusion contents of syn- and postrift sediments (e.g. European necking zone: Barale et al., 2016; Beltrando et al., 2012, Beltrando et al., 2014; Decarlis et al., 2017; Ewing et al., 2013; Rossetti et al., 2015; Seymour et al., 2016; Adriatic supra-detachment distal margin units: Coltat et al., 2020; Incerpi et al., 2020b; Pinto et al., 2015), there is little evidence for a Pyrenean-like HT-LP rift-related metamorphism in the Alpine Tethys rift systems. Even though it could have been overprinted by subduction and collisional overprints (Gabalda et al., 2009), it should be noticed that hyperextension rifting ahead of the Alpine necking zones was under a severe sediment starvation during and after rifting across the different segments of the Alpine Tethys (e.g. Lemoine et al., 1986; Manatschal, 2004; Manatschal and Bernoulli, 1999; Masini et al., 2012, 2013; Mohn et al., 2010; Ribes et al., 2020). This characteristic is primarily shown by long-standing hiatuses and condensed levels on rift-related palaeo-highs (e.g. Dumont et al., 1984; Lemoine and Trümpy, 1987; Trümpy, 1949), directly overlained by radiolarian cherts or marbles above the Alpine ocean-continent transition (e.g. Alpine European margin: Florineth and Froitzheim, 1994; Lemoine et al., 1987; Alpine Adriatic margin: Hermann and Müntener, 1996; Manatschal and Nievergelt, 1997; Ligurian



**Fig. 13.** Comparison between two conceptual models displaying respectively a high sedimentary budget rifted margin (*e.g.* the Pyrenean belt) and a low sedimentary budget rifted margin (*e.g.* the Alps). Associated logs display recorded  $T_{\text{max}}$  at equivalent heat flow and lithospheric thinning. Observed  $T_{\text{max}}$  influence the rheology of the basement which becomes ductile beyond of 350 °C for a typical felsic continental crust. In the case of high sedimentary budget, the recorded  $T_{\text{max}}$  at the top basement exceeds 400 °C, while for the case of low sedimentary budget, the recorded  $T_{\text{max}}$  are close to 100 °C.

Tethyan margins: Brovarone et al., 2011; Marroni et al., 1998; Molli, 1996) and discontinuous layers of pre- and syn-rift sedimentary slivers along hyperthinned crustal domains (Beltrando et al., 2014). This low-sedimentation rate is partly due to the persisting syn- and post-rift marine environments along sourcing areas and the occurrence of more proximal troughs trapping detrital sedimentation. It should also be replaced in the context of a wider Alpine oceanic basin (hundreds of kilometers wide, Vissers et al., 2013) with respect to the ~100 km wide aborted Pyrenean rift system (McCarthy et al., 2020). Although the Alpine margins may have recorded similar heat flows and hydrothermal circulations at the top of the basement, it cannot be efficiently recorded within a thin if not simply absent sedimentary cover, most of the syn-rift heat being directly brought to the seawater (Pinto et al., 2015). Thus, the comparison between the Pyrenean and Alpine examples strongly highlights that the top-basement  $T_{\text{max}}$ during rifting is obviously influenced by the syn-rift sedimentary budget within the hyperextended domain (Fig. 13). For similar thinning factors, it also implies that a felsic crust under hyperextension can easily reach and stay longer in the ductile field (*i.e.* >350 °C) for a highlysedimented rift system than for a poorly-sedimented scenario, as suggested by recent studies (Asti et al., 2019; Clerc and

Lagabrielle, 2014; Clerc et al., 2015; Lagabrielle et al., 2019b). In other words, high sedimentary budgets for similar topbasement heat flows should delay the crustal embrittlement during hyperextension. The resulting  $\sim 10$  km crustal thickness considered as a "thinning threshold" allowing the crustal embrittlement required for mantle exhumation proposed from both the Alpine and Iberia-Newfoundland sediment-poor cases (Manatschal et al., 2001; Müntener et al., 2000; Pérez-Gussinyé et al., 2003) may not be applicable for higher syn-rift sedimentary budgets like in the Pyrénées. Out of Western Europe and from this point of view, the observed extremely thinned (<5 km) but still mantle-decoupled crust of the distal part of the Lower Congo rift basin of the South Atlantic (Congo and Angola margins) described by Clerc et al. (2018) may not only be related to an abnormal late rift thermal gradient (i.e. hot versus cold rifting) but rather to an extremely high sedimentary budget as shown by its 7-8 km thick syn-rift burial.

### 7 Conclusions

This study provides an unprecedented database of thermal constraints of the entire Pyrenean hyperextended rift system

inverted and passively integrated in the Pyrenean orogenesis. This database is made of a compilation of surface and subsurface measurements of peak metamorphism temperatures  $(T_{\text{max}})$  archived by the organic matter originating from pre-rift to syn-orogenic sedimentary deposits of different rift-related and alpine structural units. Temperature constraints all derived from RSCM and Vitrinite Reflectance  $(R_0)$  methods harvested from all current available academic and industrial studies. The database described and analysed in this study enables investigating the spatial and temporal thermal record of a hyperextended rift system reaching a mantle exhumation stage, but which failed to reach a seafloor spreading stage. If its socalled HT/LP Pyrenean metamorphism is its most famous evidence, the spatial and temporal resolution of our study considerably refines its spatial and stratigraphic distribution and provides new data on its intensity, which can be then replaced in a rift and orogenic tectonic context after decades of research in the area. From our analysis we show that:

- at the rift scale, the depth  $T_{\text{max}}$  profiles recorded by  $R_o$  values along drillholes generally show a thermal break between the end of the syn-rift and the early post-rift sequences (*i.e.* at post-rift time) that is affected by Pyrenean deformations. It indicates that the effect of the H*T*/L*P* metamorphic event is mostly recorded within preand syn-rift sediments;
- this regional trend is however not respected along a small HT-strip within early orogenic sediments along the North Pyrenean Frontal Thrust and above the Grand Rieu ridge. Linked to previous studies, we attribute this HT-stripe to the percolation of hot hydrothermal fluids sourced from the dehydration of underthrust basement and/or sediment rocks at depth during the early orogenic stage ("continental subduction");
- the same intensity of the HT/LP metamorphism (reaching  $\sim$ 500 °C) is recorded all along the Pyrénées from the Boucheville and Bas-Agly areas in the east to the Cantabrian belt in the west for similar burial and riftrelated structural settings. They are consistently recorded at the basement-sediment interface underneath the most distal part of the hyperextended domain (in the vicinity of exhumed crustal or mantle rocks). This thermal peak is recorded underneath the northern border of the Mauléon Basin (calibrated by wells) and is shifted across transfer zone on the opposite side of the NPZ along the Central and Eastern Pyrénées where it outcrops at the surface. It suggests that all of the Pyrenean rift system can reach equally "hot"  $T_{\text{max}}$  from east to west and that the rift system may have recorded a major change in the rift paleogeography (e.g. Chevrot et al., 2018; Tugend et al., 2015), that in turn played a role during their orogenic shortening;
- along N-S-striking sections and at the scale of the entire rift system (*i.e.* necking zone to necking zone), the thermal structure appears asymmetric and reveals different horizontal thermal gradients between the northern and southern borders of rift basins as observed in the Basque-Cantabrian, Mauléon-Arzacq and Tarascon rift segments. This may be linked with an asymmetry of the former hyperextended rift system as proposed by former studies (Lescoutre *et al.*, 2019);

- by comparing the Pyrenean and Alpine hyperextended rift systems, we can conclude that this high thermal imprint is strongly influenced by fluid advection during hyperextension and may be a characteristic feature of hyperextended rift settings with a high sedimentation rate promoting thermal blanketting. Because of tectonic migration or cessation, the persisting late to post-rift Pyrenean HT record suggests that the decrease of heat convection led to a more conductive post-rift thermal regime and a delay of peak metamorphism.

### **Supplementary material**

**Fig. S1.** N-S seismic composite line (82-BUZ-11, 2005-Rontignon-Lacq-Merge, 82-SVG-08, 82-SVG-008, 82 SVG-03, Pecorade and AG1-V9 seismic lines provided by Total, R&D) across the Western Pyrénées as well as the location of the different used wells (at the top). Line drawing and geological interpretation of the seismic composite line with all tectono-stratigraphic units presented in the paper (at the bottom).

**Table S1.**  $R_o$  values from the Les Cassières-2 borehole and estimated temperatures calculated with the formulas provide by Barker and Pawlewicz (1994).

**Table S2.**  $R_o$  values from the Bellevue-1 borehole and estimated temperatures calculated with the formulas provide by Barker and Pawlewicz (1994).

**Table S3.** R<sub>o</sub> values from the Orthez-102 borehole and estimated temperatures calculated with the formulas provide by Barker and Pawlewicz (1994).

**Table S4.** All R<sub>o</sub> data used to build maps of spatial distribution of thermal maturity of organic matter by each tectonostratigraphic unit.

**Table S5.** RSCM peak temperatures from this study measured in the Pyrenean-Cantabrian belt. The parameters RA1<sub>Lahfid</sub> (Lahfid *et al.*, 2010) and R2<sub>Beyssac</sub> (Beyssac *et al.*, 2002) are used to estimate temperatures <320 and >330 °C, respectively. RA1<sub>Lahfid</sub>, R2<sub>Beyssac</sub> and T are expressed in terms of mean values and SD of all the data obtained for each of the 208 samples from the whole Pyrénées. The total number of spectra and the number of used spectra is detailed. Standard errors (SE) are given for all the temperatures (SD divided by the square root of the number of measurements).

The Supplementary Material is available at https://www.bsgf. fr/10.1051/bsgf/2021029/olm.

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