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Fluid evolution from extension to compression in the Pyrenean Fold Belt and Basque-Cantabrian Basin: A review

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ABSTRACT

We propose a review to discuss the large number of studies dealing with the fluid history in extensional and compressional sedimentary basins that evolved along the Iberian-Eurasian plate boundary during the full Mesozoic-Cenozoic Wilson Cycle in the Pyrenean fold belt and the Basque-Cantabrian Basin. We integrate classic and modern geochemical and geochronological datasets used in fluid studies with the current tectonic knowledge of the studied area.

Late Hercynian fluid systems were dominated by Carboniferous-early Permian magmatic intrusions related to large-scale lithospheric delamination at the end of the collision, which caused the accumulation of skarns at depths of 8000–10,000 m during contact metamorphism. During the Mesozoic extension, early and widespread shallow burial dolomitization of Jurassic and Early-Cretaceous carbonates occurred at burial depths of 500–1000 m due to seawater influx. From Albian to Cenomanian, along the North Pyrenean extensional fault zone, contact metamorphism processes occurred in association with mantle-derived and deep-crustal fluids at temperatures higher than 300 °C, which interacted with Triassic evaporites and formation and marine waters and depths of 2000–3000 m. Away from this fault, fluid systems were dominated by hydrothermal dolomitization and the accumulation of Zn—Pb mineralization along diapir walls and faults, whereas in the less extended and proximal domains of the extensional system, fluids were formation waters at temperatures up to 150 °C. The Alpine compressional fluid history registers the increasing influence of meteoric fluids as the foreland basin became overfilled and fluid flow occurred at depths of 2.5–4 km in tectonic units detached in Triassic evaporites and of >4 km in units rooted at depth with the Paleozoic basement. Along and across strike differences in the fluid evolution of the Pyrenees are attributed to changes in the structure of the cover and basement tectonic units, the westward decrease of shortening and in the oblique directions of Upper Triassic successions, which acted as very efficient seals for deep-sourced fluids.

Subvertical walls of diapirs are baffles for fluid flow, whereas fracturing and deposition of porous halokinetic successions are effective conduits. Evaporite detachments compartmentalize paleohydrological systems during tectonic deformation, although they may be breached by fluids reaching lithostatic pressures. In large evaporite-bearing provinces, fluid systems may share common patterns during successive extensional and compressional tectonic events, as documented in the Western Mediterranean Mesozoic extensional rift system. In this area, metal-bearing and deep-sourced fluids interacted with Triassic sulphates and organic matter, triggering the accumulation sulphides in rock porosity. However, more research is needed in other large-scale evaporitic provinces of different ages to identify common fluid flow patterns.

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1. Introduction

The Iberia-Eurasia Plate Boundary, including the Pyrenees and the Basque-Cantabrian Basin, is a well-studied region due to its relatively simple evolution, preservation of foreland basins, its reduced size and its outstanding outcrop conditions preserving much of its pre-, *syn*- and post-tectonic history (e.g, Vergés et al., 1995, 2002; Carola et al., 2015; Muñoz et al., 2018; Teixell et al., 2018; Ford et al., 2022). Recent understandings of the tectonic evolution of the Pyrenean-Cantabrian belt respond to the reinterpretation of the Iberia and Eurasia margins as a diapiric province developed during the Late Jurassic-Early Cretaceous, as well as to the characterization of the controls of Mesozoic salt-related structures on the structural architecture of the Cenozoic fold and thrust belts (e.g., López-Mir et al., 2015; Saura et al., 2015; Cámara, 2017; Granado et al., 2018; Roca et al., 2011, 2021; Ducoux et al., 2022; Ramos et al., 2022; Cofrade et al., 2023; Casini et al., 2023). Other recent studies focused on deciphering the structure of the thinnest portions of the Iberia and Eurasia margins resulting in two contrasting models, which imply an asymmetrical faulting geometry of plate boundaries (e.g., Ford and Vergés, 2020; Pedrera et al., 2021) or symmetrical and smooth slopes separated by a continuous domain of exhumed mantle (e.g., Lagabrielle and Bodinier, 2008; Lagabrielle et al., 2010, 2016, 2020; Teixell et al., 2018;). All the tectonic knowledge of the Pyrenees and Basque-Cantabrian Basin was integrated with the analysis of *syn*-tectonic fluid flow along fault zones and fracture networks and the geochronology of fracture-filling carbonate cement (e.g., Banks et al., 1991; Travé et al., 1997; Cruset et al., 2020a, 2021; Motte et al., 2021; Hoareau et al., 2021; Muñoz-López et al., 2022).

The Iberia-Eurasia Boundary is a world-class case study for fluid behavior in extensional and compressional settings. Fluid flow studies are numerous and are distributed from the most internal Pyrenean Axial Zone to the most external Ebro and Aquitaine foreland basins across the Pyrenean Fold belt and Basque-Cantabrian Basin. Many of these fluid flow studies aimed to understand the formation of metalliferous deposits in the Paleozoic Axial Zone and the Mesozoic Basque-Cantabrian Basin and North Pyrenean Zone as well as the potential hydrocarbon resources in the adjacent fold and thrust belts (e.g., Soler et al., 1990; Cardellach et al., 1992; Beroiz and Permanyer, 2011; Perona et al., 2018). Fluid systems in the Pyrenees and Basque-Cantabrian Basin have been comprehensively analysed in >70 research articles since the earlier studies of Rouvier et al. (1985), Rye and Bradbury (1988), McCaig et al. (1990) and Travé et al. (1997) (Table S1). These authors applied fluid flow concepts and models generated for both extensional and compressional tectonic settings allowing to correlate the changes in both the composition and temperature of fluids with the propagation of normal and thrust faults (e.g., Baqués et al., 2010; Muñoz-López et al., 2020a; Fig. 1). Such large number of studies has settled an effective basis on how fluids evolve during the tectonic evolution since the end of the Variscan Orogeny and Mesozoic extension along the Pyrenees and Basque-Cantabrian Basin as well as the Cenozoic compression mostly studied in the South Pyrenean fold and thrust belt (e.g., Wickham and Taylor, 1985; Travé et al., 1997, 1998, 2000, 2007; Crognier et al., 2018; Cruset et al., 2021; Barré et al., 2021; Hoareau et al., 2021). However, some questions remain open about the fluid evolution of the Pyrenees and Basque-Cantabrian Basin: 1) what are the along- and across-strike fluid flow variations along the Pyrenean and Basque-Cantabrian extensional margins and subsequent fold and thrust belts?; 2) what are the controls of the pre-compressive structure and stratigraphy on the subsequent compressive fluid evolution?; and 3) Do these fluid flow variations respond to local particularities, large-scale tectonics and fluid events? or are common to other extensional and compressional systems worldwide?

The main objective of this review is to bring together the growing number of available results on fluid flow evolution across the Pyrenean system as a whole including both extension and subsequent compression histories. To do so, we will discuss the fluid history integrated within a large-scale tectonic frame during the complete Mesozoic-Cenozoic Wilson Cycle associated with the Iberian-Eurasian plate boundary since the end of the Variscan Orogeny and during the development of the Mesozoic extensional system and subsequent Alpine compression. The second aim of this study is to discuss the fluid flow patterns in the Iberia-Eurasia Plate Boundary considering their along- and across-strike fluid flow variations, controls of pre-compressive and evaporite units and structures and mechanical stratigraphy. We also discuss the origin of the particularities or similarities of the Pyrenean and Basque-Cantabrian fluid flow patterns with case studies from the Appalachians (e.g., Oliver, 1986; Evans and Hobbs, 2003), Rocky Mountains (e.g., Sheldon, 1967), Zagros Oman Mountains and Mexican fold and thrust belts (e.g.,

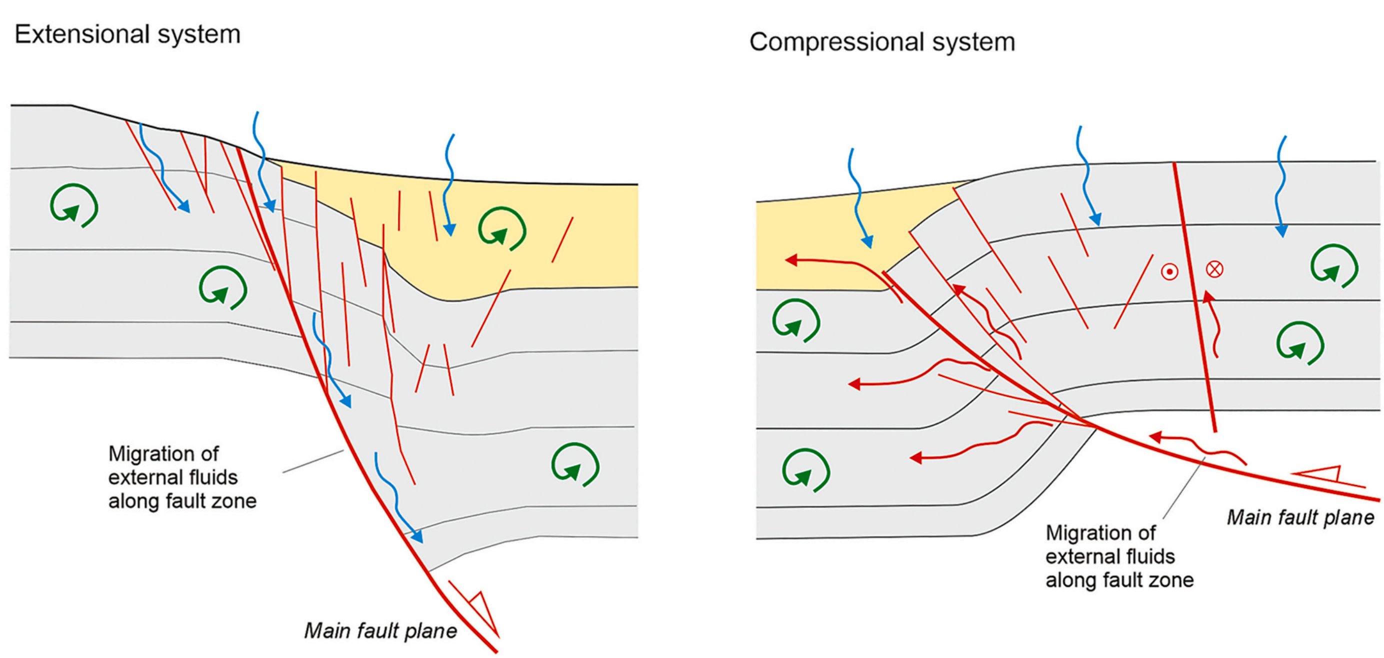

Fig. 1. Schemes showing the relationships between the evolution of fault zones in carbonate rocks and fluid flow. The extensional system is based on Baqués et al. (2010) and the compressional system on Muñoz-López et al. (2020a).

Lefticariu et al., 2005; Breesch et al., 2009; Sharp et al., 2010), and the folded orogenic systems shaping the Central and Western Mediterranean (Vilasi et al., 2006; Martín-Martín et al., 2015; Moragas et al., 2020).

2. Pyrenees and Basque-Cantabrian Basin Geological Setting

The Pyrenean and the Basque-Cantabrian fold and thrust belts are the consequence of the intra-continental collision between the Iberia and Eurasia plates during the Africa-Europe N-S convergence that initiated during the Late Cretaceous (Muñoz, 2002; Macchiavelli et al., 2017; Grool et al., 2018) (Fig. 2). This collision resulted in the underthrusting of the Iberian lithosphere beneath the Eurasian plate, according to deep seismic reflection and seismic data acquired along N-S cross-sections of the Pyrenees and Basque-Cantabrian Basin (Choukroune and Team, 1989; Muñoz, 1992; Díaz et al., 2012; Chevrot et al., 2015). Before the Pyrenean orogeny, the north Iberian and south Eurasian margins were separated by a Mesozoic rift system associated with the opening of the Bay of Biscay and the exhumation of

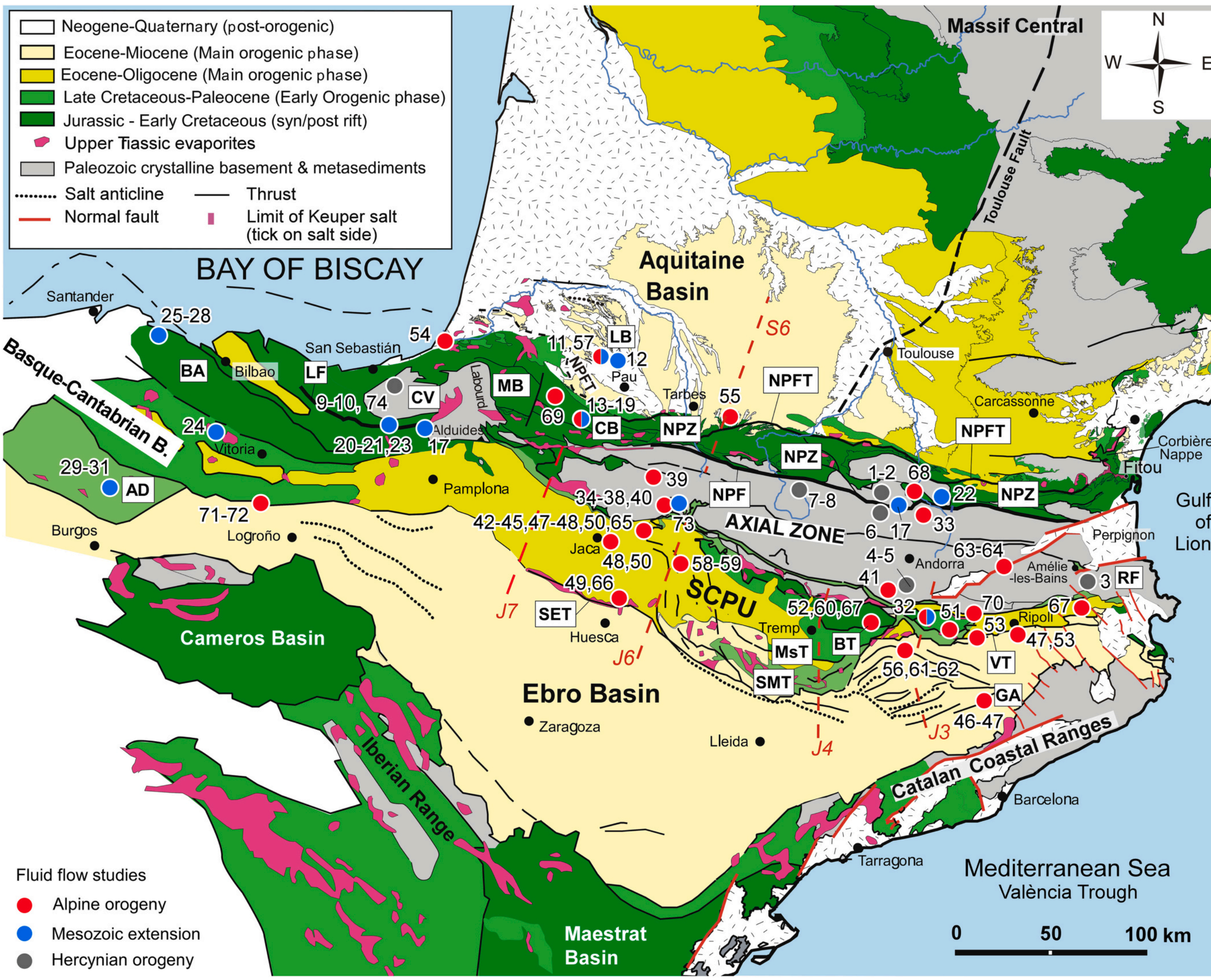

Fig. 2. Tectonostratigraphic map of the Pyrenees and Basque-Cantabrian Belt adapted from Ford and Vergés (2020). AD: Ayoluengo Salt Dome; BA: Bilbao anticline; BT: Bóixols thrust; CB: Chaînons Béarnais; CV: Cinco Villas Massif; GA: El Guix anticline; LA: Lacq Basin, LF; Leiza Fault; MB: Mauléon Basin; MT: Montsec thrust; NPZ: North Pyrenean Zone; NPFT: North Pyrenean Frontal Thrust; RF: Roc de Frausa; SCPU: South-Central Pyrenean Unit; SET: Sierras Exteriores thrust; SMT: Serres Marginals thrust; VF: Vallfogona thrust. The grey, blue and red symbols indicate the location of fluid flow studies treated in sections 3 and 4: 1 and 2) Wickham and Taylor (1985, 1987); 3) Aguilar et al. (2016); 4) Soler et al. (1990); 5) Cardellach et al. (1992); 6) Poitrenaud et al. (2021); 7 and 8) Cugerone et al. (2018, 2019); 9–10) Pesquera and Velasco (1989, 1997); 11) Elias-Bahnan et al. (2021); 12) Renard et al. (2019); 13) Motte et al. (2021); 14) Corre et al. (2018); 15) Lagabrielle et al. (2019b); 16) Nteme-Mukonzo et al. (2020); 17) De Felipe et al., 2017; 18) Salardon et al., 2017; 19) Incerpi et al., 2020; 20 and 21) Ducoux et al., 2019, 2021); 22) Quesnel et al., 2019; 23) Mendia and Ibarguchi, 1991; 24) Perona et al. (2018); 25) López-Horgue et al. (2010); 26) Nader et al. (2012); 27) Iriarte et al. (2012); 28) Dewit et al. (2012); 29) Alcalde et al., 2013; 30) Pérez-López et al., 2020; 31) Beroiz and Permanyer (2011); 32) Cruset et al. (2021); 33) McCaig (1988); 34) Grant et al. (1990); 35) Banks et al. (1991); 36, 37 and 38) McCaig et al. (1990, 1995, 2000); 39) Losh (1989); 40) Trincal et al. (2017); 41) Muñoz-López et al. (2020b); 42) Bradbury and Woodwell (1987); 43) Rye and Bradbury (1988); 44 and 45) Travé et al. (1997, 1998a); 46) Travé et al. (2000); 47) Travé et al. (2007); 48) Lacroix et al. (2014); 49) Beaudoin et al. (2015); 50) Crognier et al. (2018); 51) Cruset et al. (2020a); 52) Muñoz-López et al. (2022); 53) Cruset et al. (2018); 54) Tilhac et al. (2013); 55) Barré et al. (2021); 56) Cruset et al. (2016); 57) Bahnan et al. (2020); 58 and 59) Hoareau et al. (2009, 2015); 60) Nardini et al. (2019); 61 and 62) Sun et al. (2021, 2022); 63 and 64) Taillefer et al. (2017, 2018); 65) Lacroix et al. (2018); 66) Hoareau et al. (2021); 67) Muñoz-López et al. (2020a); 68 and 69) Laurent et al. (2021, 2023); 70) Caja et al. (2006); 71 and 72) Permanyer et al. (2015; 2018); 73) Subías et al. (2015); 74) Velasco et al. (1996). The dashed red lines indicate the location of cross-sections J3, J4, J6, S6 and J7 of Ford et al. (2022) in Fig. 5. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

asthenosphere mantle rocks from Aptian to Cenomanian. (Debroas, 1990; Jammes et al., 2009; Lagabrielle et al., 2010).

The Mesozoic extension in the Pyrenean and Basque-Cantabrian rift system represents the onset of the Alpine Cycle in the Iberia-Eurasia plate boundary and postdates the end of the Variscan orogeny that took place during the Late Carboniferous-Early Permian. This orogeny was characterized by the extensional collapse of the Variscan fold and thrust belt and the development of NW-SE and NNE-SSW extensional basins filled with detrital non-marine sediments (López-Gómez et al., 2021; Lloret et al., 2021). This extensional collapse was accompanied by a first stage of calc-alkaline magmatism (Maurel et al., 2004; Pereira et al., 2014). This collapse was followed by alkaline and mafic events related to the breakup of Pangea during the middle-late Permian beginning of the Alpine Cycle, which was generalized in the study area during the Early-Middle Triassic (Lloret et al., 2021 and references therein). The Jurassic-Cretaceous structure and evolution of the Pyrenean and Basque-Cantabrian extensional system was controlled by reactivated *E*-W, NW-SE and NE-SW Variscan faults, which in turn controlled the structural architecture of the Late-Cretaceous to Miocene fold and thrust belts (Bond and McClay, 1995; Vergés and Garcia-Senz, 2001; Cámara, 2017; Muñoz et al., 2018). Additionally, the accumulation of thick successions of Triassic evaporites also controlled the development of Late Jurassic-Early Cretaceous diapiric zones that were bounded by E-W, N-S and WNW-ESE listric faults (e.g., Saura et al., 2015; Cámara, 2017; Casini et al., 2023).The continuous extension and associated crustal thinning along the North Pyrenean Zone triggered thermal metamorphism processes along its contact with the Iberian plate(e.g., De Felipe et al., 2017; Teixell et al., 2018; Ducoux et al., 2019, 2021; Lagabrielle et al., 2020).

The studied area is divided into different domains. The first domain comprises the North Pyrenean Zone and Aquitaine Basin developed along the southern margin of Eurasia. The thick Mesozoic basins in the North Pyrenean Zone developed on or near to the plate boundary along the North Pyrenean Fault and therefore, display the most intense metamorphism and deformation during both Mesozoic extension and subsequent Cenozoic compression during the Alpine Cycle, respectively. This boundary continues to the west in the Basque-Cantabrian Basin in which the complex Bilbao anticline represents its westwards continuation (Ábalos, 2016; Cámara, 2017). The second domain is located to the south of the North Pyrenean Fault corresponding to the northern margin of Iberia, and comprise the large Mesozoic basins of the Southern Basque Cantabrian and Pyrenees.

During the Iberian-Eurasian convergence and continental collision, compression triggered the tectonic inversion of the Mesozoic and Carboniferous-Permian extensional sedimentary basins on both sides of the growing Pyrenean orogenic system (Choukroune and Team, 1989; Roure et al., 1989; Muñoz, 1992; Vergés et al., 2002; Muñoz, 2002). Contrarily, the Alpine compression was less intense in the Basque-Cantabrian belt and as a result, the Carboniferous-Permian basins were only partially inverted (Alonso et al., 1996; García-Espina, 1997; Pulgar et al., 1999; López-Gómez et al., 2019, 2021). In this context, the Pyrenees as a whole and the Basque-Cantabrian fold and thrust belt grew from Late Cretaceous to Miocene, accumulating a total amount of shortening of 125 km in the east (e.g., Macchiavelli et al., 2017). The Pyrenees displays a significant foreland basin on the Iberian lower plate in the south and a less developed retro-foreland on its northern side on the Eurasian upper plate (e.g., Muñoz, 1992; Grool et al., 2018). Towards the west, the Basque-Cantabrian fold belt is more subdued, and its southern foreland basin is poorly developed.

3. The late-Variscan and Mesozoic extensional fluid system

In this section, we summarize the numerous studies dealing with the fluid flow history of the North Iberian and Eurasian plate margins from the end of the Variscan orogeny to the Mesozoic extension during the early stages of the Alpine cycle. The fluid flow studies referred to sections 3 and 4 are indicated with a number along the text and in the map in Fig. 2.

3.1. Fluids in North Iberia related to the Variscan Orogeny

Fluid systems in the Iberia-Eurasia Plate Boundary during *syn*- to post-Variscan orogenic periods were mostly studied in the exposures of Paleozoic rocks in the Pyrenean Axial Zone, whereas few studies were carried out in the Paleozoic Cinco Villas Massif in Basque-Cantabrian Basin. These fluid systems were strongly influenced by extensive late Carboniferous-early Permian calc-alkaline magmatism from 310 to 276 Ma, emplaced during crustal thinning due to large-scale lithospheric delamination at the end of the collision between Avalonia and Laurasia (e.g., Maurel et al., 2004; Pereira et al., 2014; Laurent et al., 2017; Liesa et al., 2021; Lloret et al., 2021). The fluid flow research related to the Variscan Orogeny focused on regional and contact metamorphism processes, as well as the accumulation of metalliferous deposits.

In the Pyrenees, contact metamorphism of metasediments was described in the Trois Seigneurs Massif due to the heat produced by magmatism coupled with percolation of formation or marine fluids at depths between 6 and 12 km (Wickham and Taylor, 1985, 1987, 1–2 in Fig. 2). Contact metamorphism processes during Late Carboniferous magmatism were also described in the Roc de Frausa Massif and resulted in reactions of the expulsion of silica-rich fluids, as well as the partial melting of metasedimentary rocks at temperatures between 710 and 750 °C (Aguilar et al., 2016, 3 in Fig. 2). Contact metamorphism processes also triggered the accumulation of metalliferous ores during the later stages of the Variscan orogeny (Soler et al., 1990; Cardellach et al., 1992, 4 and 5 in Fig. 2). According to these authors, along the contact between the late Carboniferous Andorra-Mont Louis granite and the Devonian limestones, contact metamorphism and fluid-rock interaction controlled the accumulation of gold-bearing hedenbergite skarns at temperatures between 350 and 650 °C. Similarly, in the Salau skarn deposits, in the northern flank of the Pallaresa anticlinorium, skarns accumulated from magmatic fluids at a temperature of 850 °C during the Early Permian at 295 Ma as determined by U—Pb dating and fluid inclusion analyses in Au-W-bearing minerals (Poitrenaud et al., 2021, 6 in Fig. 2). These authors also reported that magmatic fluids released from deformed magmatic intrusions at temperatures between 400 and 450 °C migrated along strike-slip faults at depths between 8 and 10 km during the Permian at 289 Ma. In the Bossòst anticlinorium, Cugerone et al. (2018, 2021), 7 and 8 in Fig. 2) established that magmatic and metamorphic fluids at temperatures between 300 and 450 °C triggered the recrystallization and remobilization of earlier Pb—Zn mineralization during the Late Carboniferous at 309 Ma.

The scarce fluid flow studies of the Variscan history of the Basque-Cantabrian Basin were carried out in the Cinco Villas Massif. In this area, the Arditurri Pb-Zn-F-Ba deposits recrystallized and remobilized during the Late Variscan regional metamorphism (Pesquera and Velasco, 1989, 9 in Fig. 2). Contrarily, stratabound tourmaline accumulations in the same area recrystallized during metasomatism triggered by contact metamorphism processes between Late Variscan granite intrusions and metapelites (Pesquera and Velasco, 1997, 10 in Fig. 2).

3.2. Fluids during the Jurassic-Early Cretaceous rift-related extension of the Iberia-Eurasia Plate Boundary

During the first two decades of the 21st century, significant progress has been made in the study of the fluid flow evolution in the margins of the Iberia-Eurasia Plate Boundary during the Mesozoic extension. This progress was partly in line with its reinterpretation as an extensional system showing unequivocal halokinetic patterns denoting the diapiric evolution of the Pyrenean and Basque-Cantabrian extensional systems, possibly accompanied by hyperextension associated with mantle exhumation in the North Pyrenean Zone. (e.g., Jammes et al., 2009;

Lagabrielle et al., 2010, 2019a; López-Mir et al., 2016; Saura et al., 2015; Teixell et al., 2016, 2018; Cámara, 2017; Labaume and Teixell, 2020; Ford and Vergés, 2020; Hudec et al., 2021; Roca et al., 2021; Ducoux et al., 2022).

Fluid flow evolution linked to the Mesozoic extension of North Iberia and South Eurasia was analysed at different sectors of their margins and along the North Pyrenean Zone acting as a plate boundary. To the north, in the Eurasian Plate Margin, fluid flow studies focused on the Late Jurassic-Early Cretaceous carbonate reservoirs in the Lacq and Rousse hydrocarbon-prone basins deeply buried beneath the syn-orogenic sediments of the Aquitaine retro-foreland basin (Renard et al., 2019; Bahnan et al., 2020, 2021) (section 3.2.1.). Along the North Pyrenean Fault, those carbonates were also studied in the Chaînons Béarnais area (e.g., Salardon et al., 2017; Motte et al., 2021) (section 3.2.2.). Fluid flow in the North Iberian Plate Margin was studied in the southern side of the Pyrenees, where fluids migrating along Jurassic and Lower Cretaceous rocks have been studied in the Basque-Cantabrian Basin (section 3.2.3.), which preserves its Mesozoic fluid history given its least compressive deformation, as well as in the Upper Pedraforca thrust sheet (section 3.2.4.) (e.g., López-Horgue et al., 2010; Perona et al., 2018; Cruset et al., 2021). In this section, we review these studiesand place them within the context of the Mesozoic evolution of the Iberia-Eurasia extensional system.

The initial fluid events in Mesozoic extensional basins from the Iberia-Eurasia Plate Boundary were characterized by the widely distributed shallow burial dolomitization of Jurassic and Early Cretaceous carbonates due to seawater influx (Bahnan et al., 2021; Motte et al., 2021; Cruset et al., 2021). Subsequent fluid flow events were frequently linked to the tectonic and salt-related evolution of Jurassic and Early Cretaceous rift basins. These fluid flow events were characterized by significant and abrupt changes in the temperature and composition of fluids according to their structural position within the Iberia-Eurasia Plate Boundary. In this scenario, Mg-rich hydrothermal fluids, at temperatures of up to 250 °C interacting with Triassic evaporites, caused the recrystallization of earlier dolostones and limestones in the Lacq Basin, Chaînons Béarnais and northern sector of the Basque-Cantabrian Basin. Additionally, we will show the distribution of georesources such as hydrocarbons and ore accumulations.

3.2.1. Aquitaine Basin: The deep Lacq Reservoir

In the Lacq gas reservoir in the Aquitaine basin in the Eurasian Plate, the fluid flow study of Bahnan et al. (2021), 11 in Fig. 2) registered the progressive burial of Jurassic reservoirs (Fig. 3). In this section, we use the nomenclature proposed by these authors to describe the different fluid-related diagenetic phases (Dolomite 1 and 2 and Calcite 1 to 3). Shallow burial Dolomite 1 probably precipitated at shallow burial conditions from marine fluids, based on textural observations done by Bahnan et al. (2021,11) and the shallow marine conditions prevailing in the Lacq reservoir during the Late Jurassic-Early Cretaceous. This early dolomite was post-dated by Calcite 1 and 2, which possibly precipitated at deeper burial conditions from evolved meteoric fluids according to their crystal morphology and luminescence zoning reflecting changes in the redox conditions in a phreatic environment. Shallow Dolomite 1 was also recrystallized by deep-sourced formation fluids carrying oil, methane and hydrogen sulphide (Dolomite 2) at temperatures between 136 and 144 °C and with $\delta^{18}O_{fluid}$ between +8 and + 10 ‰ VSMOW (Bahnan et al., 2021, 11 in Fig. 2). According to these authors, these hot formation waters probably derived from the thermochemical reduction of Triassic evaporites during the Aptian rift-related deep burial of the Lacq gas reservoir at depths between 2500 and 3000 m. Oil, methane and hydrogen sulphide also migrated along fractures together with Ca-rich fluids, resulting in the precipitation of Calcite 3, at temperatures between 146 and 156 °C during the Albian and Cenomanian post-rift stage of the Lacq reservoir at depths between 3000 and 3200 m. Similarly, in the Rousse reservoir, Renard et al. (2019), 12 in Fig. 2) interpreted that extensive dolomitization of the Upper Jurassic limestones

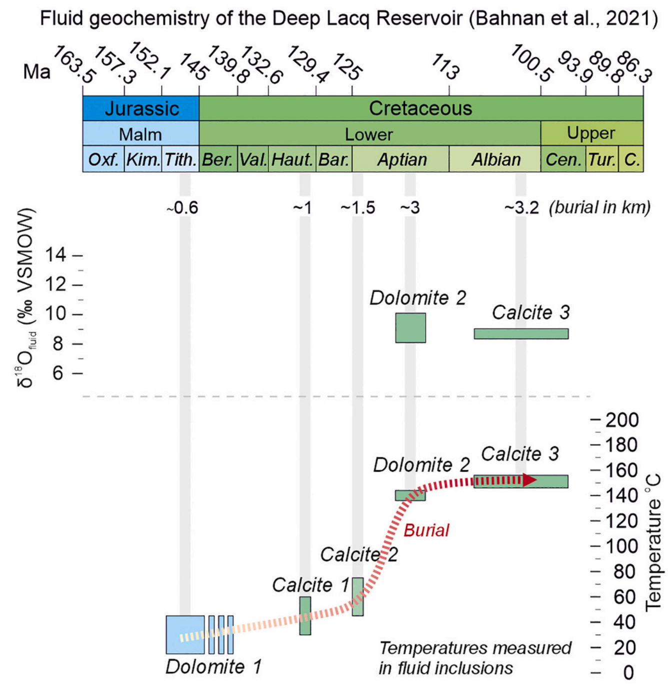

Fig. 3. Fluid temperature and $\delta^{18}O_{fluid}$ of carbonate cements and dolostones of the Lacq reservoir reported in Bahnan et al. (2021), 11 in Fig. 2).

occurred in a context of deep burial. These authors also described two additional fracture-related dolomitization events related to low-salinity hydrothermal fluids carrying methane and carbon dioxide taking place at around 105 Ma, at temperatures of around 150 °C, and at a depth of 2000 m with a thermal gradient of 75 °C/km.

3.2.2. North Pyrenean Zone

To the south of the Lacq reservoir, the Chaînons Béarnais area registers the fluid flow evolution of the Northern Pyrenees close to the North Pyrenean Fault (Motte et al., 2021, 13 in Fig. 2). These authors reported a Berriasian-Valanginian (Rd1) early and low-temperature dolomitization event of middle to Upper Jurassic limestones at burial depths between 500 and 1000 m caused by the influx of marine-derived waters with $^{87}Sr/^{86}Sr$ ratios between 0.707461 and 0.707798 (Fig. 4). A subsequent dolomitization event caused the recrystallization of the early dolostones and precipitation of dolomite cements (Rd2, Dc2, Dc3) from hydrothermal fluids with $\delta^{18}O_{fluid}$ values between +0.6 and + 11.6 ‰ VSMOW and $^{87}Sr/^{86}Sr$ ratios of up to 0.709001 during the Albian (Fig. 4). According to the same authors, the positive $\delta^{18}O_{fluid}$ and Sr isotopic ratios were consistent with heavy basinal fluids that interacted with the host rock. These fluids probably derived from Upper Triassic evaporites, which could have supplied Mg and clay dehydration in a context of sub-salt crustal thinning and supra-salt diapiric growth. According to Motte et al. (2021), 13 in Fig. 2), dolomite recrystallization of early dolostones and precipitation of dolomite cement occurred at depths between 1500 and 2000 m at a maximum temperature of 180 °C considering the Aptian geothermal gradient of around 80 °C/km estimated by Vacherat et al. (2014) and Hart et al. (2017) from thermochronological data in the extensional Mauléon Basin. A younger cement with $\delta^{18}O_{fluid}$ values between +6 and + 17 ‰ VSMOW, high chlorinity and $^{87}Sr/^{86}Sr$ ratios between 0.710043 and 0.710130 was interpreted by Motte et al. (2021) as the result of the mixing of formation waters influenced by Upper Triassic evaporites and crustal- and mantle-derived fluids at >300 °C that migrated along diapir walls from Albian to Cenomanian (Dc4 from Motte et al., 2021, 13 in Fig. 2) (Fig. 4). These mixed hot fluids also migrated throughout large extensional detachments along the mantle-crust contact involving Triassic evaporites (Corre et al.,

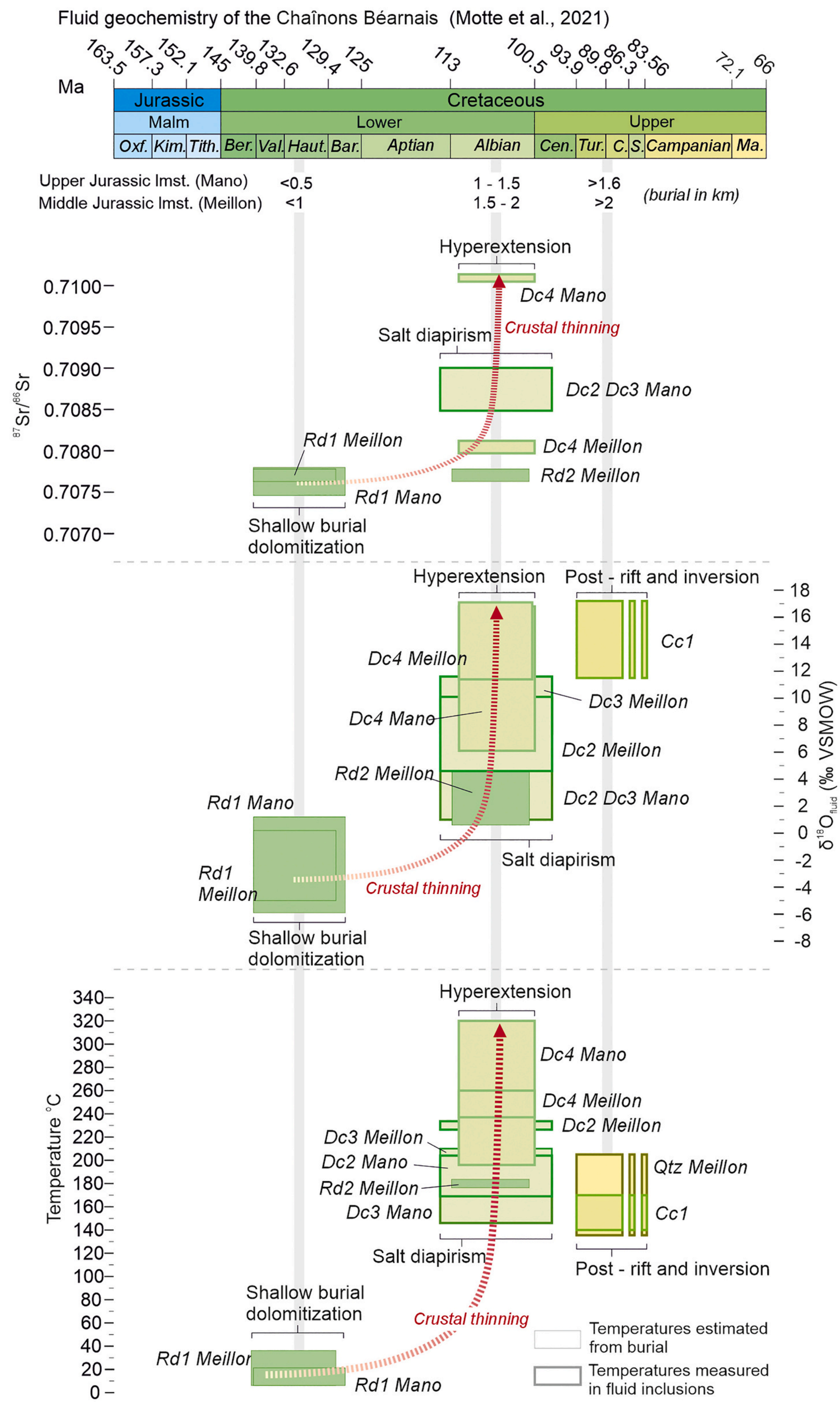

Fig. 4. Fluid temperature, $\delta^{18}O_{fluid}$ and $^{87}Sr/^{86}Sr$ ratios of carbonate cements and dolostones of the Chaînons Béarnais reported in Motte et al. (2021),13, in Fig. 2). Mano and Meillon refer to the Middle and Upper Jurassic formations described by Motte et al. (2021), 13 in Fig. 2), respectively.

2018; Lagabrielle et al., 2019b; Nteme-Mukonzo et al., 2020, 14, 15 and 16 in Fig. 2). Mantle-derived fluids, probably leached from serpentinized upper mantle peridotites, at temperatures between 200 and 300 °C, caused high-temperature and low-pressure metamorphism within the Triassic-Early Cretaceous sedimentary succession along the North Pyrenean Fault Zone (De Felipe et al., 2017; Salardon et al., 2017; Incerpi et al., 2020; Ducoux et al., 2019, 2021, 17, 18, 19, 20 and 21 in Fig. 2). Locally, in these regions, exhumed mantle rocks possibly acted as heat source for metasomatic reactions between high-salinity fluids derived from Triassic evaporites and metamorphic and igneous rocks emplaced during the Albian, which resulted in the accumulation of talc-chlorite mineralization (e.g., Trimouns deposit in the Saint Barthelemy Massif;

Quesnel et al., 2019, 22 in Fig. 2). Finally, fluid flow events in the Chaînons Béarnais since Turonian times were characterized by hot waters with temperatures between 135 and 200 °C and $\delta^{18}O_{fluid}$ values between +4.5 and + 17 ‰ VSMOW from which calcite and quartz cement probably precipitated in a post-rift scenario (Cal1 and Qtz1; Motte et al., 2021, 13 in Fig. 2) (Fig. 4).

As a summary, the fluid flow history of the North Pyrenean Zone reconstructed by Motte et al. (2021) in the Chaînons Béarnais (13 in Fig. 2), registers the progressive process of extension of the North European margin and the exhumation of the lithospheric mantle. These authors concluded that this process was recorded by the progressive increase of $^{87}Sr/^{86}Sr$ ratios from 0.707461 to 0.710043, $\delta^{18}O_{fluid}$ between +0.6 and + 17 ‰ VSMOW and fluid temperatures of up to >300 °C (Fig. 4), which register the progressive input of hot crustal-derived radiogenic fluids and hotter mantle-derived waters that caused high-temperature/low-pressure metamorphism. These temperatures, $\delta^{18}O_{fluid}$ and Sr isotopic ratios documented in the Chaînons Béarnais are higher than those of the Lacq reservoir in the Aquitaine Basin (up to 140 °C, +8 ‰ VSMOW and 0.709001). This suggests that during the Mesozoic extension, crustal thinning controlled the composition and temperature of fluids, in accordance to the location Lacq Reservoir in a more proximal and thicker crustal domain than the North Pyrenean Zone (Teixell et al., 2018; Ducoux et al., 2021).

3.2.3. Basque-Cantabrian Basin

In the northern sector of the Basque-Cantabrian Basin, high-temperature and low pressure metamorphic processes at temperatures of >300 °C, similar to that observed along the North Pyrenean Zone, were also described along the Leiza Fault, which represents the western equivalent of the North Pyrenean Fault (e.g., Mendia and Ibarguchi, 1991; De Felipe et al., 2017; Ducoux et al., 2019, 2021, 23, 17, 20 and 21 in Fig. 2). To the south of this fault, in the North Iberian Plate Margin, the Basque-Cantabrian sector is considered an important mining province in which Mississippi Valley Type Zn—Pb deposits accumulated in close association with diapiric structures involving Triassic evaporites and dolomitization processes controlled by faults, salt walls, and sedimentological variations of the carbonate rocks (e.g., Rouvier et al., 1985; Velasco et al., 1994; Dewit et al., 2012; Perona et al., 2018, 28 and 24 in Fig. 2). These authors interpreted that these ore accumulations and dolomitization occurred during the Mesozoic extension, although absolute dating studies have not been carried out.

The study of the hydrothermal dolomitization and associated Mississippi Valley Type Zn—Pb deposits of the Albian-Turonian Ramales carbonate platform and Murguía Diapir concluded that fluids migrating during salt-related extension had high-salinity, temperatures between 75 and 295 °C, $\delta^{18}O_{fluid}$ values between −5.7 and + 20. 73 ‰ VSMOW, and $^{87}Sr/^{86}Sr$ ratios from 0.708024 to 0.708262 (López-Horgue et al., 2010,; Nader et al., 2012; Iriarte et al., 2012; Perona et al., 2018, 24 25, 26 and 27 in Fig. 2). These isotopic values and fluid temperatures are similar to those reported in diapiric zones from the North Pyrenean Zone, pointing to a common fluid system controlled by salt-related structures and Triassic evaporites. The southernmost domains of the Basque-Cantabrian Basin were characterized by the generation and migration of hydrocarbons. The Polientes and Sedano minibasins represent a superb example of fluid flow compartmentalization by a polygonal network of Jurassic–Early Cretaceous growing diapiric salt-walls displaying a polygonal distribution of which the NNE-SSW trending Hontomín and Ayoluengo salt anticlines are the most significant (e.g., Beroiz and Permanyer, 2011; Cámara, 2017). The Ayoluengo salt dome, a productive oil field from 1967 to 2017, is now an active tectonic field for CO_2 storage management with Jurassic reservoirs at 1.5 km deep (e.g., Alcalde et al., 2013; Pérez-López et al., 2020, 29 and 30 in Fig. 2). In the Polientes and Sedano minibasins, the generation, expulsion and migration of hydrocarbons took place during Early-Late Cretaceous times, when the source rock was buried at 2300 m adjacent to Ayoluengo oil field and 4000 m adjacent to Hontomín gas field. The correlation of these findings with oil and asphalt seeps along the flanks of both minibasins indicated that oil migration was very effective vertically and horizontally with distances of up to 15 km in the case of the Hontomín salt dome (Beroiz and Permanyer, 2011, 31 in Fig. 2).

3.2.4. Southern Pyrenees

In the South Pyrenean fold and thrust belt, also in the North Iberian Margin, the evolution of fluid flow during the Mesozoic extensional events is scarcely studied, although it is preserved in the Jurassic to Early Cretaceous carbonate successions cropping out in the Upper Pedraforca thrust sheet, as documented in Cruset et al. (2021), 32 in Fig. 2). After shallow burial dolomitization, formation waters at 150 °C, probably seawater mixed with fluids derived from Upper Triassic evaporites, migrated along fractures linked to the Early Cretaceous diapiric growth (Fig. 5). As a consequence of these Early Cretaceous halokinetic movements recorded in the Upper Pedraforca and Organyà Basins, fluids at temperatures close to 125 °C interacted with highly radiogenic pre-salt rocks in primary welded regions. The absence of metamorphic processes, ore mineralization and hydrothermal fluid flow could indicate that the Mesozoic evolution of the Southern Pyrenees developed at the more external sectors of the Pyrenean extensional system that were affected by minor crustal thinning with respect to the North Pyrenean Zone and the internal sector of the North Basque-Cantabrian Basin.

4. Fluid flow during the Alpine Orogeny

Fluid flow in the North Iberian Plate Boundary during the Alpine Orogeny has been mostly studied in the South Pyrenean fold and thrust belt (e.g., Travé et al., 2007; Giménez-Montsant et al., 1999; Mansurbeg et al., 2009; Lacroix et al., 2018; Hoareau et al., 2021), although a few works have also been done in the North Pyrenean fold and thrust belt (Tilhac et al., 2013; Bahnan et al., 2020; Barré et al., 2021). The cross-sections in Fig. 6 show the location of most of the fluid flow studies, which cover the Axial antiformal stack, fold and thrust belt and Ebro foreland basin domains and allow to depict a general vision of the compressional Pyrenean fluid flow patterns that will be summarized in this section. We will review the fluid flow systems in each Pyrenean domain, their relationship with the mobility and/or accumulation of georesources such as hydrocarbons and the geochemical signature of fracture-filling cements.

4.1. Fluid flow in the Axial Zone antiformal stack

Pioneering research articles constraining the relationships between fluid flow and compressional deformation in the Pyrenean orogenic system were carried out in deep thrust fault zones of the Axial Zone. In these deep structures, fluids percolated through highly permeable microcrack networks in which quartz veins precipitated due to a pressure drop following an earthquake, whereas fluid pressure built-up during compressional tectonics promoted slow upward flow at low permeability conditions (McCaig, 1988, 33 in Fig. 2).

Studies done by several authors determined that fluids in deep basement faults could have derived from different sources. Grant et al. (1990), 34 in Fig. 2), Banks et al. (1991), 35 in Fig. 2) and McCaig et al., 1990, 1995, 2000, 36, 37 and 38 in Fig. 2) interpreted that those brines, probably derived from Triassic red-beds, migrated along thrust fault zones modifying the geochemical composition of host rocks, based on geochemical studies of carbonate and quartz veins and host rocks in the Aston Massif and Gavarnie thrust sheet. Contrarily, a different interpretation proposed that quartz veins probably precipitated from metamorphic fluids derived from devolatilization reactions of metasediments, which occasionally mixed with shallower fluids derived from overlying Mesozoic and Cenozoic carbonates, based on granitic outcrops from the central sector of the Axial Zone (Losh, 1989, 39 in Fig. 2). Circulation of fluids occurred under low-grade and reducing

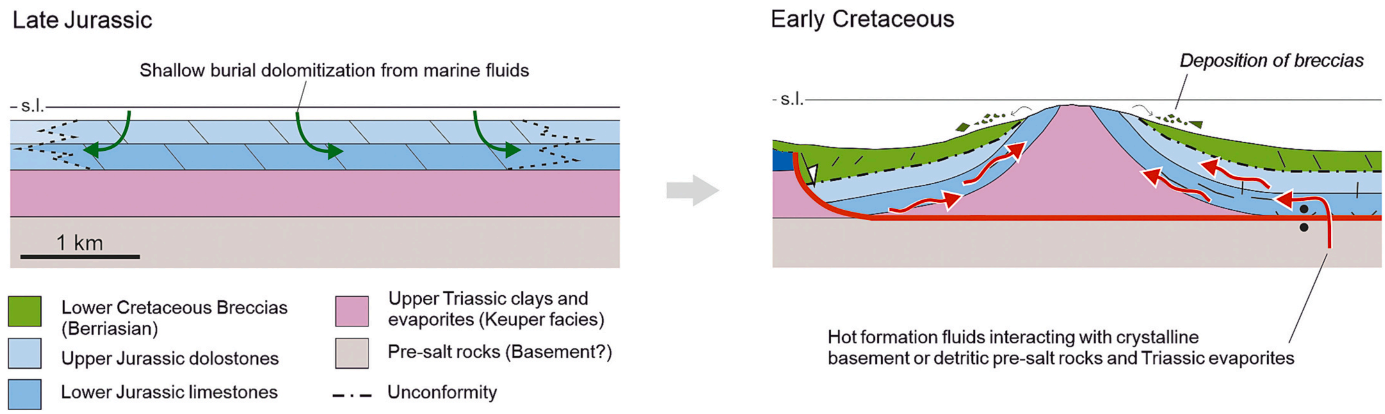

Fig. 5. Fluid evolution in the Upper Pedraforca basin during the Late Jurassic and Early cretaceous extension. Redrawn from Cruset et al. (2021), 32 in Fig. 2).

metamorphic conditions, as observed by Trincal et al., 2017, 40 in Fig. 2) in the Pic-de-Port-Vieux thrust zone. Metamorphic processes favoured the recrystallization of Mesozoic carbonates as well as hematite dissolution and chlorite formation within Paleozoic metasediments. In the reactivated Variscan Estamariu thrust in the eastern Axial Zone, the geochemical studies of calcite veins done by Muñoz-López et al., 2020b, 41 in Fig. 2) revealed that meteoric fluids, heated at depth and interacted with metamorphic rocks, migrated along this fault during the Pyrenean orogeny. As a result, the fluids reached temperatures between 50 and 100 °C and had $^{87}Sr/^{86}Sr$ ratios between 0.7400 and 0.7100. Muñoz-López et al., 2020b, 41 in Fig. 2) also compared the high $^{87}Sr/^{86}Sr$ ratios of vein cement in the Estamariu thrust with those measured in cement from multiple fault zones of the Pyrenean Axial Zone and cover thrust sheets. These authors observed that fracture-filling cements in fractures from the Axial Zone had $^{87}Sr/^{86}Sr$ ratios higher than 0.7100, evidencing the interaction of the migrating fluids with high radiogenic igneous and metamorphic rocks (e.g., Banks et al., 1991; McCaig et al., 1995; McCaig et al., 2000, 35, 36 and 37 in Fig. 2). Contrarily, cement precipitated from fluids that migrated through the sedimentary cover in the Pyrenees had $^{87}Sr/^{86}Sr$ ratios below 0.7100, reflecting the interaction of these fluids with the less radiogenic Mesozoic and Cenozoic sedimentary rocks. However, basement-derived fluids could have migrated a minimum distance of 5 km into the overlying South Pyrenean fold and thrust belt, creating vertical and lateral isotopic alteration fronts within the sedimentary pile of thrust sheets (Bradbury and Woodwell, 1987, 42 in Fig. 2).

The integration of the studies mentioned above allows us to build a fluid flow model of the Pyrenean Axial Zone (Fig. 7). In this model, Triassic red beds acted as a paleo reservoir for formation waters, which were expelled from rock pores during compressional tectonics coevally with the migration of metamorphic fluids derived from mineral reactions occurring at depths between 4 and 10 km (Grant et al., 1990; Banks et al., 1991; McCaig et al., 2000; 34, 35 and 36 in Fig. 2). These fluids, mobilized at shallow positions along fracture networks and thrust faults, triggering the recrystallization of Cretaceous carbonates overlying the Pyrenean basement. Locally, meteoric fluids could have percolated to the basement, interacting with igneous and metamorphic rocks and increasing their temperature and $^{87}Sr/^{86}Sr$ ratios to values higher than those of fluids migrating throughout the cover thrust sheets (Fig. 8).

4.2. Fluid flow in the Late Cretaceous to Miocene Pyrenean fold and thrust belt

Early studies on the fluid flow evolution of the Pyrenean fold and thrust belt were conducted on its southwestern side. Rye and Bradbury (1988), 43 in Fig. 2) determined a progressive decrease of the fluid-rock interaction as deformation in fracture networks progressed, using isotopic studies of calcite veins and host limestones in the Monte Perdido thrust system. Travé et al. (2007) integrated previous studies about the fluid flow evolution in the Axial Zone and South Pyrenean fold and thrust belt in a first attempt to review the evolution of the fluid systems during the Cenozoic compression performing a model in which palaeohydrological systems were compartmentalized by thrust faults in time and space. This model was characterized by: 1) evolved seawater mixed with deep-sourced fluids that migrated along thrust faults in the early Eocene marine foreland basin (Travé et al., 1997, 1998, 44–45 in Fig. 2); and 2) evolved meteoric fluids migrating in the late Eocene-early Oligocene non-marine Ebro foreland basin (Travé et al., 2000, 46 in Fig. 2). In a younger publication, Lacroix et al., 2014, 48 in Fig. 2) reached similar conclusions in their study of the evolution of fluid systems in Gavarnie, Cotiella, Jaca and Monte Perdido thrust faults, in the SW Pyrenees.

The development of recent methods of fluid analysis (e.g., clumped isotopes thermometry, Rare Earth Elements analysis, U—Pb dating of carbonate veins) allows the reconstruction of a more complex and accurate fluid flow history of the north and south Pyrenean fold and thrust belts from the Late Cretaceous to the Miocene that complements the previous model of Travé et al. (2007). One of the main characteristics of this fluid flow history is that it was sequential, with repetitive fluid flow patterns occurring during the progressive emplacement of Pyrenean thrust sheets and the development of related foreland basins. In each structure, fluid systems were mostly dominated by formation waters, which were stratigraphically segregated during the first stages of compression and mixed with marine, meteoric or hydrothermal fluids as deformation progressed (e.g., Lacroix et al., 2014; Beaudoin et al., 2015; Crognier et al., 2018; Cruset et al., 2020a, 51; Muñoz-López et al., 2022, 48, 49, 50, 51 and 52 in Fig. 2). The source of these fluids was mostly controlled by the presence or absence of salt detachments, the emersion and/or exhumation of thrust sheets and inverted rift-related basins as well as the stage of evolution of the Pyrenean foreland basin (marine underfilled vs. non-marine overfilled) (Cruset et al., 2018, 53 in Fig. 2).

Rift-related Upper Triassic evaporites and Eocene foreland evaporites played an important role, acting as detachment levels of thrust sheets as well as controlling the thrust front geometry and the tectonic style of deformation of the north and south Pyrenean fold and thrust belts (e.g., McCaig, 1986; Vergés et al., 2002; Sans, 2003; Muñoz et al., 2018; Ford and Vergés, 2020; Ducoux et al., 2021; Cofrade et al., 2023). These evaporites also had a strong impact on the composition of migrating fluids, as described in several Pyrenean compressional structures. One of these structures is the Larra/Eaux-Chaudes thrust system in the SW Pyrenees, where Triassic evaporites probably influenced the composition of hydrothermal fluids with temperatures of 215–270 °C and salinities of 2.2–5.7 wt% eq. NaCl, above the values of seawater (Crognier et al., 2018, 50 in Fig. 2). In their study of Upper and Lower Pedraforca thrust sheets in the SE Pyrenees, Cruset et al., 2020a, 2021,

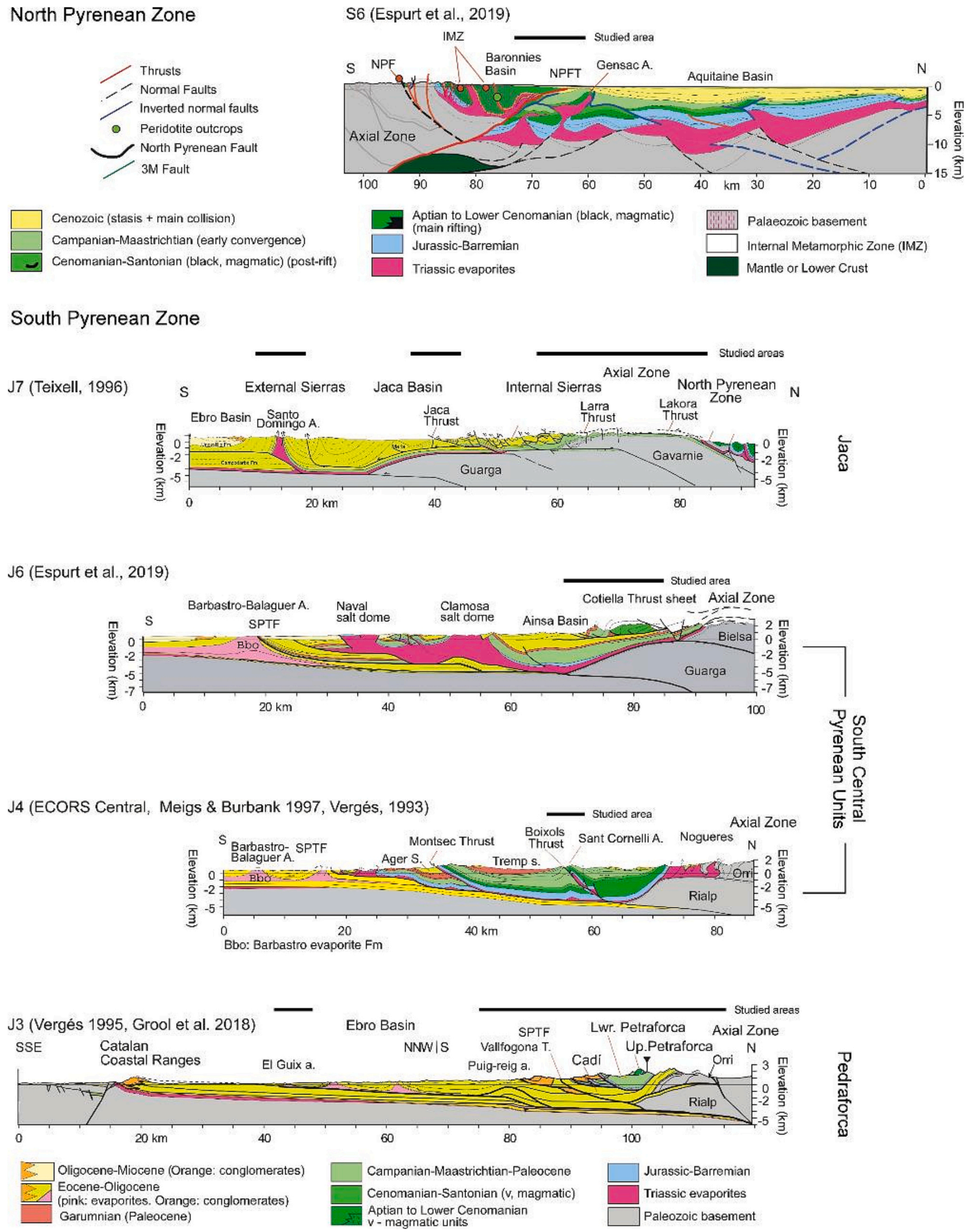

Fig. 6. Compilation of cross-sections across the Pyrenees located in Fig. 2. Sections are adapted from Vergés et al. (1995) and Grool et al. (2018) (J3); Vergés (1993); Meigs and Burbank (1997) (J4); Espurt et al. (2019a, 2019b) (J6 and S6); Teixell (1996) (J7). The black and thick horizontal lines englobe the equivalent position of the studied Pyrenean structures from a fluid flow approach.

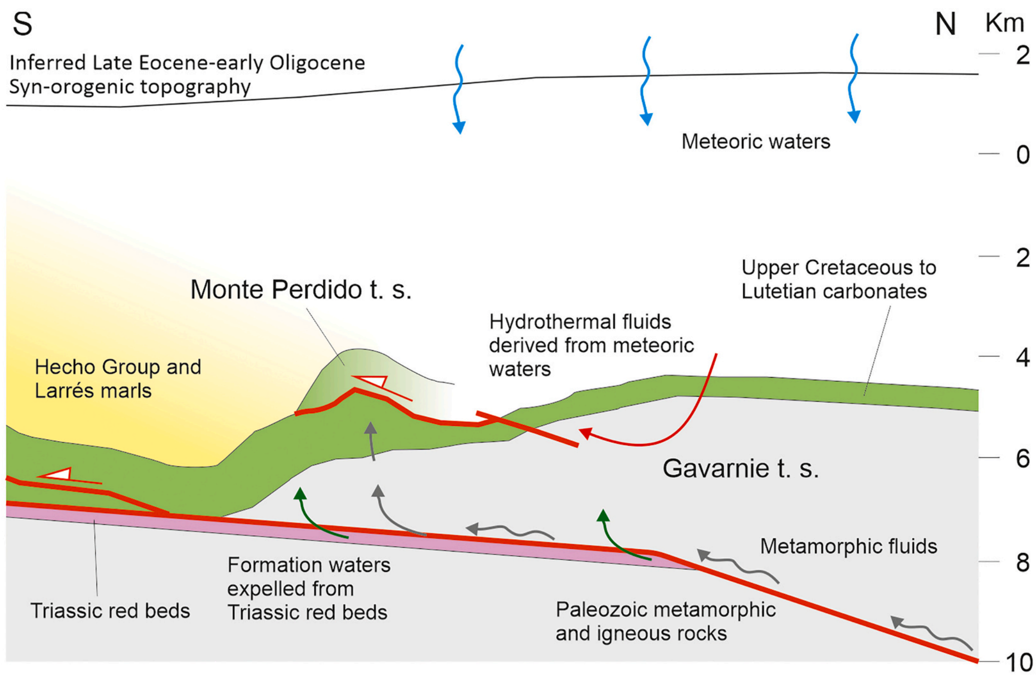

Fig. 7. Fluid flow model of the Pyrenean Axial Zone based on McCaig (1988), Grant et al. (1990), Banks et al. (1991), McCaig et al. (1990, 1995, 2000), Losh (1989), Trincal et al. (2017), Muñoz-López et al. (2020b) and Bradbury and Woodwell (1987) using the Gavarnie thrust sheet as an example. The cross-section is based on the restoration of Labaume et al. (2016) during the late Eocene-early Oligocene. In this model, metamorphic fluids derived from the basement and formation waters trapped in Triassic rock pores migrate into cover thrust sheets (in green). Also, meteoric waters percolate and are heated at depth, evolving into hydrothermal fluids. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

51 and 32 in Fig. 2) reported that formation waters with $\delta^{18}O_{fluid}$ values between +4 and + 9 ‰ VSMOW reached temperatures of 80–100 °C at burial depths of ~2.3 km. These authors concluded that these fluids possibly interacted with Upper Triassic evaporitic units acting as main thrust sheet detachment, migrated along major basal thrusts, background vein systems and faults as well as through secondary weld zones of squeezed rift-related diapirs. U—Pb dating of calcite veins precipitated from these fluids yielded ages between 70 and 25 Ma, reporting a homogeneous fluid system from Late Cretaceous to Oligocene. On the other hand, Travé et al., 2000, 46 in Fig. 2) and Cruset et al., 2018, 53 in Fig. 2) accounted for the influence of the foreland Eocene Cardona salt and Beuda gypsum on the composition of fluids migrating along fractures related to the growth of the El Guix anticline and the propagation of the Abocador thrust in the eastern sector of the Ebro foreland basin. These fluids had temperatures between 90 and 175 °C and $\delta^{18}O_{fluid}$ between +5 and + 14 ‰ VSMOW. In the North Pyrenean fold and thrust belt, fluids with salinity from 9 to 23 wt% eq. NaCl and a temperature between 79 and 102 °C, which was reached at a depth of <5 km, interacted or derived from Triassic evaporites and migrated along fold-related vein systems of the Saint-Jean-de-Luz basin (Tilhac et al., 2013, 54). Similarly, Barré et al., 2021, 55 in Fig. 2) reported the interaction of formation waters with Triassic evaporites during their migration along the North Pyrenean frontal thrust at depths of <2–3 km. The fluids resulting from this interaction had a salinity of 21 wt% eq. NaCl and temperatures between 63.1 and 145.1 °C.

In both the south and north Pyrenean fold and thrust belts, fluid systems were generally characterized by formation, meteoric and marine fluids confined above evaporitic units, which often acted as barriers for the input of deeper fluids (Fig. 9A). An example of these confined fluid systems was studied in the Upper Pedraforca thrust sheet by Cruset et al., 2021; 32 in Fig. 2). In this tectonic unit, formation waters, possibly influenced by Triassic evaporites, were heated at a depth of ~2.5 km at a temperature of 100 °C. These fluids had a $\delta^{18}O_{fluid}$ between +3.9 and 9.4 ‰ VSMOW, and progressively diluted to fluids at a temperature of 67 °C and a $\delta^{18}O_{fluid}$ of −0.3 ‰ VSMOW as the thrust front exhumed during the Eocene-Oligocene (Fig. 9A). The increasing influence of the detrital sediments derived from the unroofing of the Pyrenean Axial Zone also impacted the elemental composition of these fluids, resulting in a progressive decrease in the Y/Ho ratios (Fig. 9A). The absence or breaching of sub-horizontal fluid barriers could have facilitated the input of deep-sourced hydrothermal fluids into cover thrust sheets and foreland basins. This scenario was described in the Puig-reig anticline and Vallfogona thrust along the Eocene-Oligocene thick-skinned front of the SE Pyrenean fold and thrust belt by Cruset et al., 2016, 2018, 56 and 53 in Fig. 2). These authors concluded that tectonically-driven deep-sourced fluids at temperatures between 100 and 154 °C and $\delta^{18}O_{fluid}$ between +4.2 and + 12 ‰ VSMOW, possibly basement-derived (from depths of ~4 km), migrated from the internal and deeper positions of the Pyrenean orogen to the shallower thrust front (Fig. 9B). The progressive emersion of the Vallfogona thrust front resulted in the dilution of fluids, the decrease of their temperature and the increase of the $^{87}Sr/^{86}Sr$ ratios of fluids when they were in contact with non-marine *syn*-tectonic sediments (Fig. 9B). A similar scenario was reported in the Puig-reig-anticline in the Ebro foreland Basin, where fluids sourced from a depth of 4–5 km, at temperatures between 77 and 130 °C and $\delta^{18}O_{fluid}$ between +4.7 and 9.7 ‰ VSMOW migrated to the crestal normal faults buried at 1.7 km of burial (Cruset et al., 2016; 56 in Fig. 2). In the Larra and Monte Perdido thrusts, branched at depth with the Lakhora and Eaux-Chaudes basement thrusts, hydrothermal fluid flow at a depth of ~5 km was described by Crognier et al., 2018, 50 in Fig. 2). Likewise, thermal anomalies of up to 240 °C derived from hydrothermal fluid flow were identified in the lower depositional units of the Eocene Hecho Group turbidites, beneath the Monte Perdido thrust system by Labaume et al. (2016) based on vitrinite reflectance measurements. Thermal anomalies at depths between 1 and 2.2 km were also associated with hydrothermal fluids at temperatures between 60 and 90 °C in the Upper Lacq reservoir in the Aquitaine Basin during the Eocene thrusting (Bahnan et al., 2020, 57 in Fig. 2). These authors concluded that fluids were 30–40 °C hotter than their host rocks. In the Chaînons Béarnais, thermal disequilibrium between deep-sourced fluids and host rocks occurred during crustal thinning (Salardon et al., 2017; Motte et al., 2021 13 and 18 in Fig. 2).

The control of the stage of evolution of the Pyrenean foreland basins on fluid flow was especially well-documented in the south Pyrenean foreland basin, where its first stages occurred from Late Cretaceous to late Eocene, generally at underfilled marine foreland conditions (e.g., Vergés et al., 1995). In the Ainsa Basin, and under these conditions, Travé et al., 1997, 1998, 44–45 in Fig. 2) documented that connate

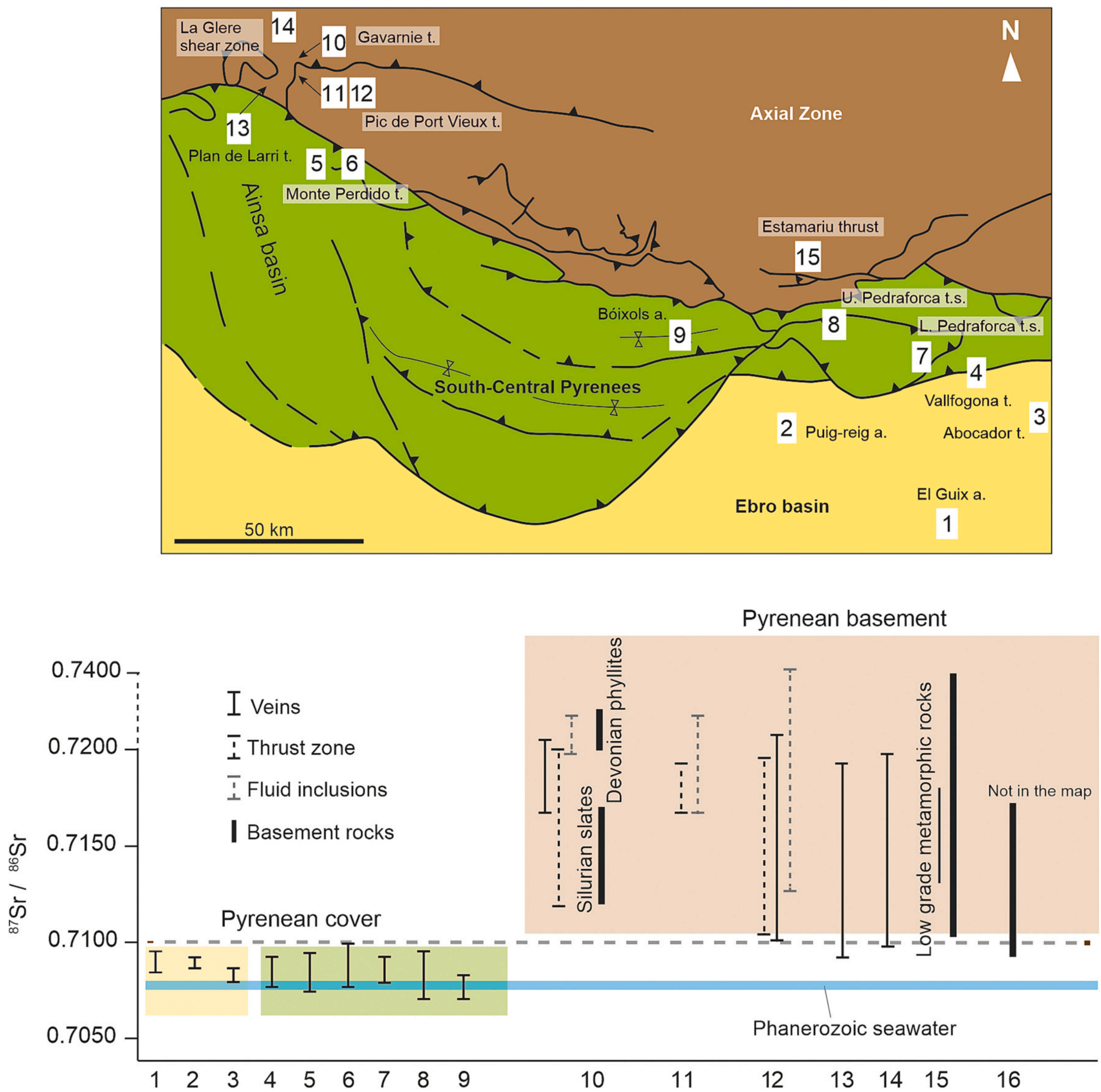

Fig. 8. Simplified geological map of the South Pyrenean fold and thrust belt showing the location of mineral veins and host rocks where 87Sr/86Sr ratios have been measured. The graph below is redrawn from Muñoz-López et al. (2020b) and shows the isotopic ratios of cover units (1–9) and the Axial Zone (10–16). Below, the thick blue line refers to the 87Sr/86Sr ratios of Phanerozoic seawater, whereas the dashed horizontal grey line marks the limit value between the basement and cover structures. 1) El Guix anticline (Travé et al., 2000), 2) Puig-Reig anticline (Cruset et al., 2016), 3) L'Escala thrust (Cruset et al., 2018), 4) Vallfogona thrust (Cruset et al., 2018), 5–6) Ainsa basin (McCaig et al., 1995; Travé et al., 1997), 7) Lower Pedraforca thrust sheet (Cruset et al., 2020a), 8) Upper Pedraforca thrust (Cruset et al., 2021), 9) Bóixols anticline (Muñoz-López et al., 2022), 10) Gavarnie thrust (McCaig et al., 1995), 11–12) Pic de Port Vieux thrust (Banks et al., 1991; McCaig et al., 2000), 13) La Glere shear zone (Wayne and McCaig, 1998), 14) Plan de Larri thrust (McCaig et al., 1995), 15) Estamariu thrust (Muñoz-López et al., 2020b), 16) Trois Seigneurs Massif (not in the map) (Bickle et al., 1988). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

seawater trapped within rock pores mixed with deep-sourced fluids migrating along the Cotiella thrust system at a depth between 500 and 2000 m during the early Eocene. The mixed fluid had temperatures between 87 and 183 °C and salinities from 6.8 to 8.5 wt% eq. NaCl. To the south of the Ainsa basin, Hoareau et al., 2009, 2015, 58–59 in Fig. 2) concluded that connate seawater was responsible for the growth of celestite and dolomite concretions within marls from the Sobrarbe delta system during shallow burial diagenesis (< 200 m depth), whereas meteoric flow from the coast to the delta front possibly occurred before celestite growth and resulted in the precipitation of calcite cement. Eastwards, in the Organyà Basin in the South-Central Pyrenees, Nardini et al., 2019, 60 in Fig. 2) reported that Late Cretaceous seawater at 87 °C and with a $\delta^{18}O_{fluid}$ between +0.6 and + 1.6 ‰ VSMOW trapped within and/or derived from marine carbonates migrated along strike-slip faults from deeper Late Cretaceous units to shallower non-marine sediments of the Paleocene Garumnian facies.

Travé et al. (2007) documented the increase of the $^{87}Sr/^{86}Sr$ ratios coupled with de depletion of the $\delta^{13}C$ of calcite veins and host rocks from the South Pyrenean foreland basin since the late Eocene. These authors interpreted this negative correlation with the increasing relief and denudation of the Pyrenean Axial Zone, the input of soil-derived CO_2, as well as the increasing influence of meteoric fluids during the closure of the Ebro Foreland basin and the beginning of its endorheic period. This continentalization and increasing meteoric influence were also documented in the same area by the progressive decrease of the Fe and Sr contents of calcite veins reported by Cruset et al., 2018, 53 in Fig. 2). In the Eocene-Oligocene fluvial successions folded in the Puig-reig anticline, along the footwall of the Vallfogona thrust, Sun et al., 2021, 2022, 61–62 in Fig. 2) determined that calcite cement precipitated in the interparticle porosity from meteoric fluids at temperatures

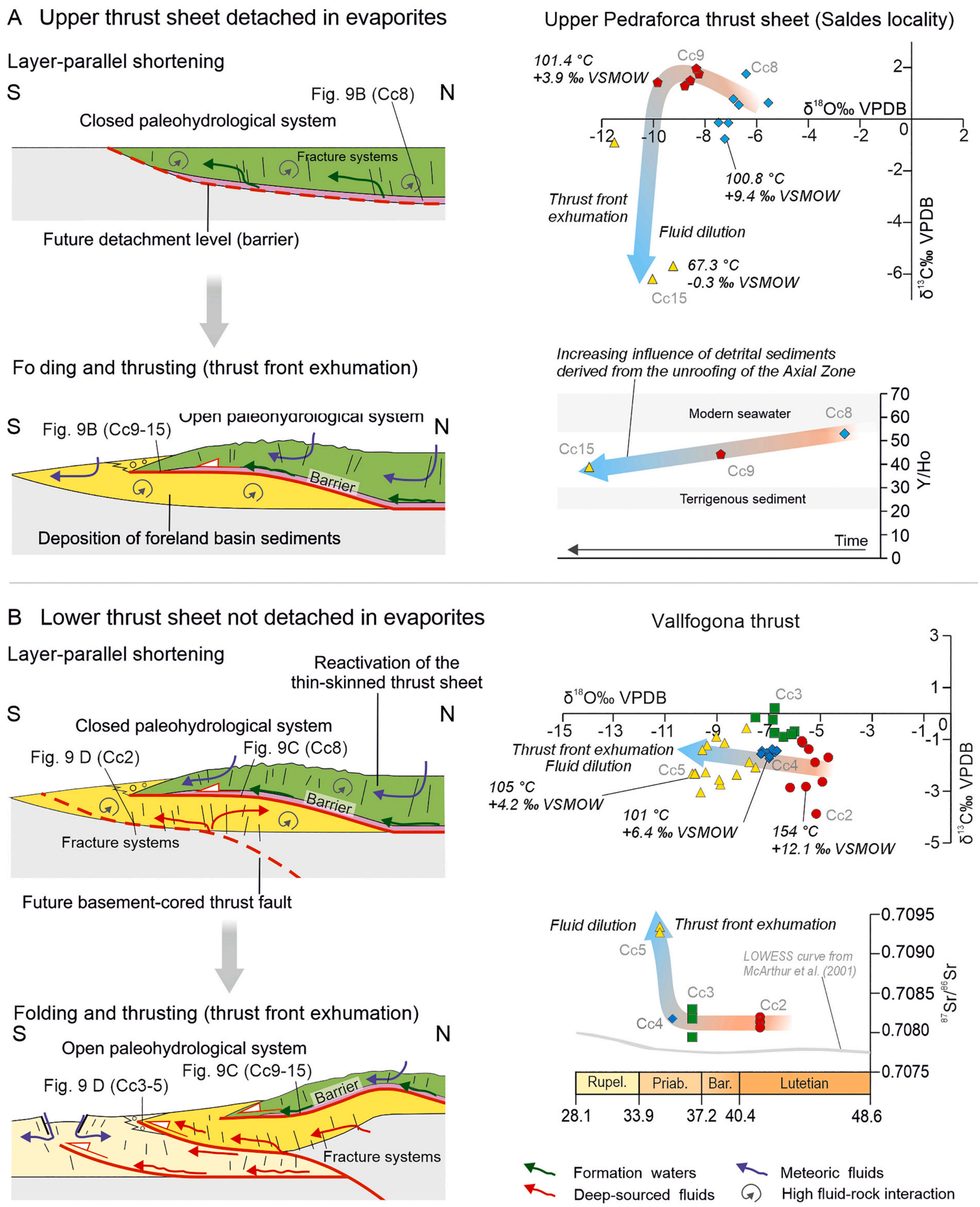

Fig. 9. Conceptual fluid flow models of the SE Pyrenees. A) thrust sheets detached in evaporite units. During the layer-parallel shortening, the fluid system is influenced by detachment levels and fluid-rock interaction is high. During folding and thrusting the fluid system was open to external fluids (e.g., meteoric waters) and the fluid-rock interaction decreased. The graphs on the right show the progressive $\delta^{18}O_{fluid}$ depletion and the decrease of the fluid temperature and Y/Ho ratios as the thrust front emerged using the Upper Pedraforca thrust sheet as an example (Cruset et al., 2021; 32 in Fig. 2). B) thrust sheets not detached in evaporites. The absence of impermeable detachments during both layer-parallel shortening and folding and thrusting facilitated the upward migration of hydrothermal fluids. The graphs on the right show the progressive $\delta^{18}O_{fluid}$ depletion and decrease of the temperature and $^{87}Sr/^{86}Sr$ ratios decrease as the thrust front emerged using the Vallfogona thrust as an example (Cruset et al., 2018; 53 in Fig. 2).

between 16 and 90 °C and with $\delta^{18}O_{fluid}$ between −4.8 and − 0.7 ‰ VSMOW. According to Cruset et al., 2016, 56 in Fig. 2), meteoric fluids in this anticline also migrated along crestal normal faults and mixed at depth with hydrothermal fluids at temperatures between 90 and 129 °C, whereas Travé et al., 2000, 46 in Fig. 2) indicated that only evolved meteoric waters migrated along inner arc thrust faults of the El Guix anticline along the tip-line of the Oligocene South Pyrenean front. Meteoric percolation also occurred in the South Pyrenean thrust sheets and North Pyrenean Zone. However, this percolation of meteoric fluids was not only restricted in time to the non-marine stage of the related Ebro and Aquitaine foreland basins, but also to the whole Late Cretaceous to Miocene fluid history of the Pyrenees due to the generation of high-topographic areas formed by the stacking and imbrication of emerging and exhuming thrust sheets (e.g., Beaudoin et al., 2015; Saldardon et al., 2017; Taillefer et al., 2017, 2018; Lacroix et al., 2018; Cruset et al., 2021; Hoareau et al., 2021; Muñoz-López et al., 2020a, 2022, 67 and 52;, 18, 32, 49, 63, 64, 65). In these emerged areas, karstic cement and sediments precipitated and accumulated, respectively in the whole Pyrenean thrust sheet system (Fig. 10). The U—Pb ages from 67 to 65 Ma (Muñoz-López et al., 2022, 52 in Fig. 2), from 61 to 14 Ma (Hoareau et al., 2021, 66 in Fig. 2) and from 47 to 34 Ma (Cruset et al., 2021, 32 in Fig. 2) measured in calcite cement precipitated from

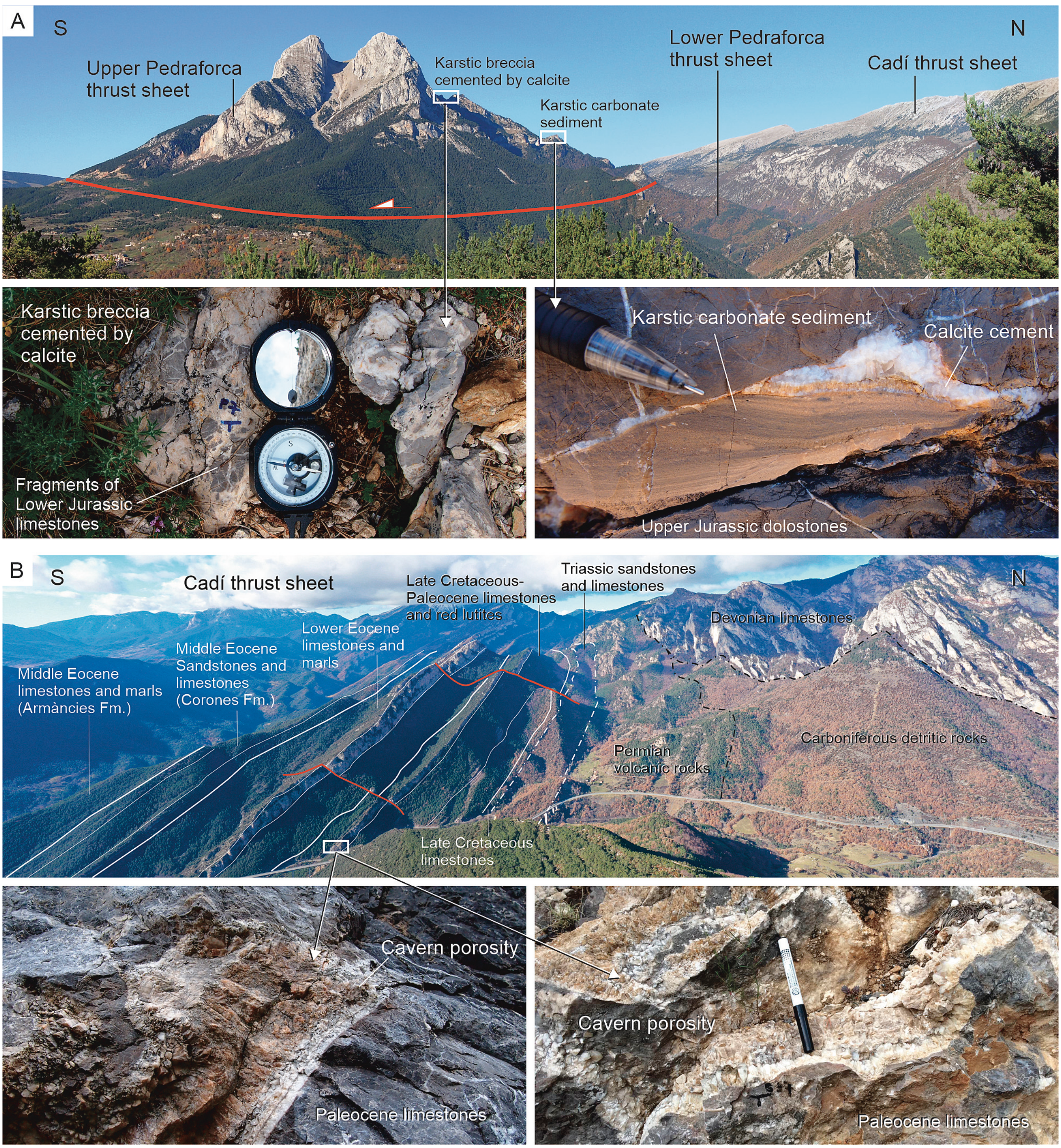

Fig. 10. Field images showing diagenetic patterns related to meteoric water migration in the southern Pyrenees. A) Meteoric calcite cement together with karstic sediments and between breccia clasts in Jurassic dolostones and limestones of the Upper Pedraforca thrust sheet. The fluids from which these cements precipitated were studied by Cruset et al., 2021, 32 in Fig. 2). B) Cavern porosity in Paleocene limestones filled by meteoric calcite at the northern sector of the Cadí thrust sheet.

meteoric fluids in the Upper Pedraforca and Bóixols thrust sheets and in the Sierras Exteriores revealed a continuous and long-lasting period of exhumation from Late Cretaceous to Miocene. In most of the aforementioned examples, karst systems are associated to meteoric fluids. Locally, in the North Pyrenean Zone, the upward migration of deep-sourced hydrothermal fluids derived from Triassic sulphates and interacted with surface waters also contributed to the dissolution of exhumed carbonates (Laurent et al., 2021, 2023, 68 and 69 in Fig. 2).

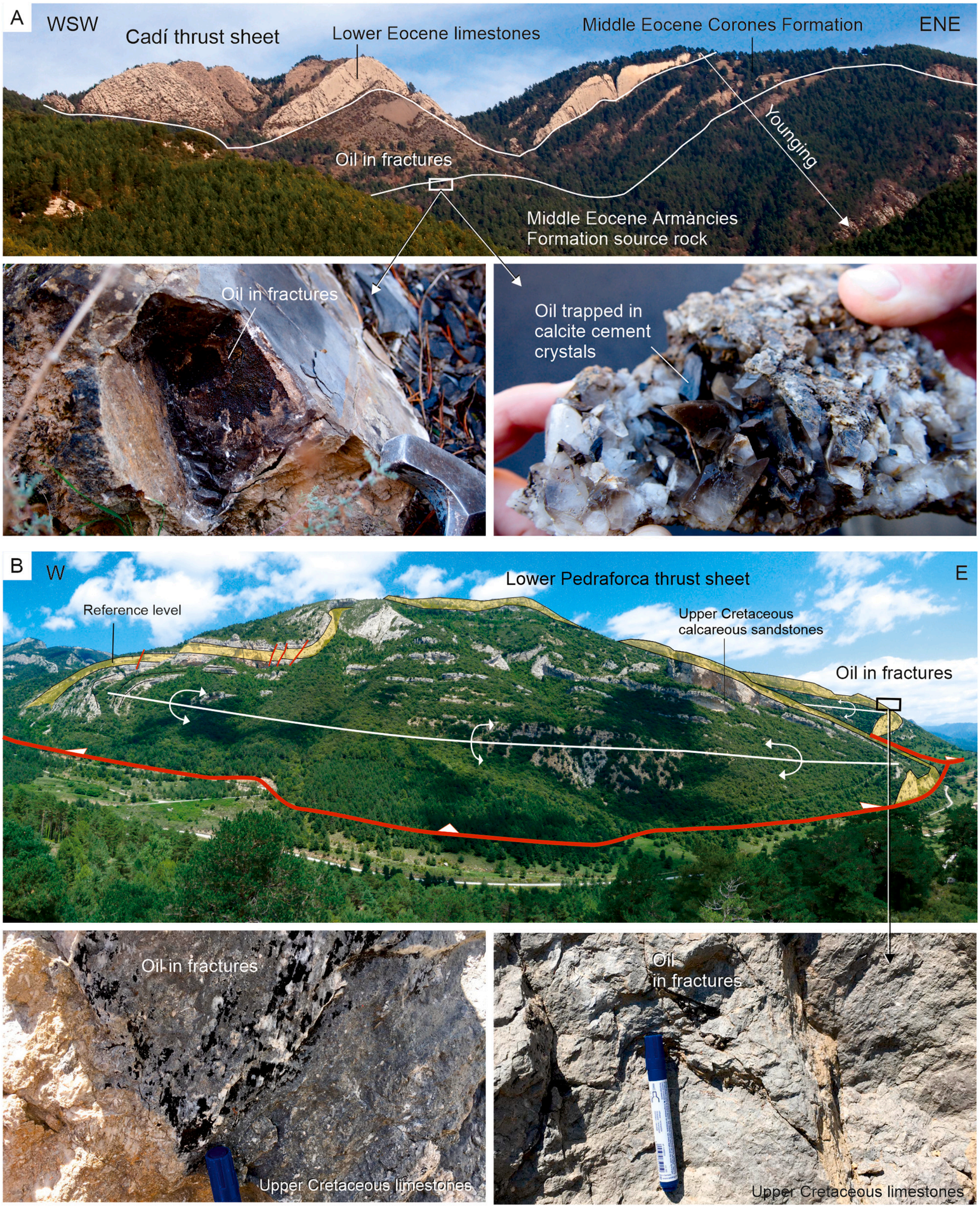

Fig. 11. Evidence of oil migration in the southern Pyrenees. A) Fractures containing oil and cutting the lower to middle Eocene carbonate platforms and slope deposits in the northern Cadí thrust sheet. Photo of the big black calcite crystals courtesy of Gemma Serrano. The oil is also trapped within calcite cement crystals. B) Oil accumulated within faults cutting Upper Cretaceous carbonates at the front of the Lower Pedraforca thrust sheet.

The relationships between Pyrenean tectonics and migration of fluid resources such as hydrocarbons have been observed in fractures cutting syntectonic lower to middle Eocene carbonate platforms and slope deposits in the Cadí thrust sheet (Caja et al., 2006, 70 in Fig. 2) (Fig. 11A). In this study, oil inclusions trapped within calcite cement and their intercrystalline porosity, allowed to determine that oil migration occurred at temperatures around 120 °C during and after calcite precipitation. Oil migration along fractures was coeval to the Lower Pedraforca thrust sheet emplacement during the Lutetian (Cruset et al., 2020a, 51 in Fig. 2) (Fig. 11B). Two pulses of oil migration were identified in the Basque-Cantabrian Basin (Permanyer et al., 2015, 2019, 71–72 in Fig. 2): a migration of immature condensates to light oils between Turonian and Campanian times at around 174 °C, partially synchronous to the opening of the Bay of Biscay; and a second entrapment of heavy to asphaltene-rich oils during the Cenozoic tectonic inversion of the Basque-Cantabrian Basin at 60–84 °C (Permanyer et al., 2019, 72 in Fig. 2). The composition and history of hydrocarbons in the large Basque-Cantabrian Basin indicate that their generation, migration and trapping vary in each of the oil plays depending on their structural position and their absolute distance with respect to heat sources during the opening of the Bay of Biscay but also on the singular characteristics of the source rocks of each salt-related minibasins encircled by diapiric walls (Beroiz and Permanyer, 2011, 31 in Fig. 2). Similar control of diapiric structures on hydrocarbon maturity and trapping has been reported to the north of the North Pyrenean frontal thrust in the Aquitaine basin, which is the most productive area of the Iberia-Eurasia collisional zone. In this basin, Upper Jurassic and Lower Cretaceous successions at depths of 4000–5000 m acted as preferential source rocks (Biteau et al., 2006), whereas *syn*-sedimentary unconformities within halokinetic sequences acted as effective seals for hydrocarbons, whose maturity began during the Mesozoic and was enhanced during the Alpine compression (Bourrouilh et al., 1995; Bourrouilh, 2012). According to the fluid inclusion analysis of Bahnan et al., 2021, 11 in Fig. 2) done in the Lacq Reservoir, the Late Jurassic-Early Cretaceous source rocks entered into the oil window during the Aptian-Albian at temperatures of 120–135 °C, and into the gas window during the Albian-Cenomanian at temperatures of 165–180 °C. The preservation of the petroleum systems developed in Mesozoic rocks from the Aquitaine basin was due to their location within the Pyrenean orogenic system, whose marginal position favoured the burial of the source rocks by foredeep deposits (Bourrouilh, 2012). Contrarily, in the Southern Pyrenees, hydrocarbon generation minimally survived due to long-lasting exhumation and the development of a high-taper orogenic wedge (Vergés and Garcia-Senz, 2001; Kendall et al., 2020). A lower exhumation and taper wedge due to lower tectonic shortening preserved the petroleum systems in the Basque-Cantabrian Basin.

The occurrence of metalliferous deposits in the Pyrenees and Basque-Cantabrian Basin is strongly controlled by fluid flow processes occurring in Paleozoic basement rocks. At the end of the Variscan Orogeny, regional and contact metamorphism-related processes may have triggered the interaction between igneous and metamorphic fluids and sedimentary rocks, resulting in the accumulation of Au-rich skarn deposits and sulphides (Cugerone et al., 2018, 7). On the other hand, during the rift-related extension, basinal brines percolated from Mesozoic strata (including Triassic evaporites) and scavenged metals in the Paleozoic basement rocks. As a result of this fluid-rock interaction, metal-rich fluids Zn—Pb and fluorite deposits accumulated. This process has been described in the Paleozoic rocks of the Pyrenean Axial Zone (e.g., Subías et al., 2015, 73) and the Cinco Villas Massif (Velasco et al., 1996, 74). Out of the Pyrenees, similar processes have been described in Paleozoic rocks from the Catalan Coastal Ranges (Piqué et al., 2008) and the Schwarzwald district of the Black Forest (Bons et al., 2014). The upward migration of metal-rich fluids to Mesozoic sedimentary rocks resulted in the accumulation of Zn—Pb sulphides in diapiric zones of the Basque-Cantabrian Basin according to the model of Rouvier et al. (1985).

5. Discussion

The fluid evolution of the Pyrenees shares common patterns with other compressional belts as has been reviewed by Beaudoin et al. (2023) using case studies from the Pyrenean, Apennines, Sevier, Rocky Mountains, Mexican, Appalachian, Oman, Albanides and Zagros fold and thrust belts. In these compressional belts, fluid systems were stratigraphically segregated during layer-parallel shortening and opened to external fluids during more advanced stages of folding according to the structural style of deformation (thin- vs. thick-skinned). However, the great number of publications dealing with the compressional fluid history of the Southern Pyrenees reveals variations in fluid evolution and distribution along and across the strike of this fold and thrust belt. In this section, we discuss the origin of these variations and if they respond to fluid patterns that are common in fold and thrust belts worldwide. We also discuss the controls of pre-compressive evaporite units and salt-related structures on fluid flow during extension and compression both at the local (single basin, diapir) and at the larger scale (extensional system, fold and thrust belt). Finally, it is discussed how the evolution of the pre- and *syn*-compression mechanical stratigraphy controls the connectivity of the paleohydrological systems.

5.1. Large-scale fluid evolution within the context of the Iberia and Eurasia Mesozoic extension

The review of the fluid evolution of the Mesozoic extensional systems in the Iberia-Eurasia Plate Boundary shows unequivocal and regional common trends related with early shallow burial dolomitization processes and the influence of Triassic Evaporite successions on fluid composition. However, differences in fluid composition and temperature arise when considering the structural position of each studied locality within the extensional system. The model in Fig. 12 summarizes the fluid flow knowledge of the Mesozoic Pyrenean and Basque-Cantabrian extensional system (Mendia and Ibarguchi, 1991; López-Horgue et al., 2010; Iriarte et al., 2012; Dewit et al., 2012; Clerc et al., 2015; De Felipe et al., 2017; Salardon et al., 2017; Perona et al., 2018; Ducoux et al., 2019; Quesnel et al., 2019; Renard et al., 2019; Incerpi et al., 2020; Cruset et al., 2021, Motte et al., 2021; Bahnan et al., 2021). In this model, we use for the North Pyrenean Zone the restored cross-sections of Teixell et al. (2018) and Ducoux et al. (2021). The first cross-section represents symmetrical and smooth slopes for the Iberian and Eurasian margins, whereas the second one is characterized by asymmetrical faulted margins. In both scenarios, plate margins are separated by the exhumed mantle. For the Basque-Cantabrian Basin and its equivalent, the South Pyrenean extensional basin, we use the restored cross-section of Cámara (2017), which represents the asymmetrical and faulted northern margin of Iberia.

Early dolomitization of carbonate platforms in the Mesozoic extensional system of the Pyrenees occurred at depths between 600 and 1000 m according to the available data in the Lacq reservoir in the Aquitaine Basin, Chaînons Béarnais along the North Pyrenean Zone and the Ramales Platform in the Basque-Cantabrian Basin (López-Horgue et al., 2010; Bahnan et al., 2021; Motte et al., 2021). According to the previous authors, dolomitization temperatures were controlled by sedimentary burial of the Jurassic successions during the Late Jurassic to Aptian.

In the North Pyrenean Zone and Basque-Cantabrian Basin along the North Pyrenean and Leiza faults, high-temperature and low-pressure metamorphic processes at temperatures of >300 °C occurred during hyperextension and mantle exhumation, which triggered the migration of deep crustal and mantle-derived fluids. Away from the North Pyrenean Fault, in the Iberian and Eurasian margins, fluids were colder and thermal metamorphism did not occur due to lesser crustal thinning. In these margins, Early Cretaceous hydrothermal dolomitization of Jurassic-Cretaceous limestones caused by basement-sourced fluids enriched in metals at temperatures between 150 and 250 °C interacting with Triassic evaporites, seawater and formation fluids occurred in the

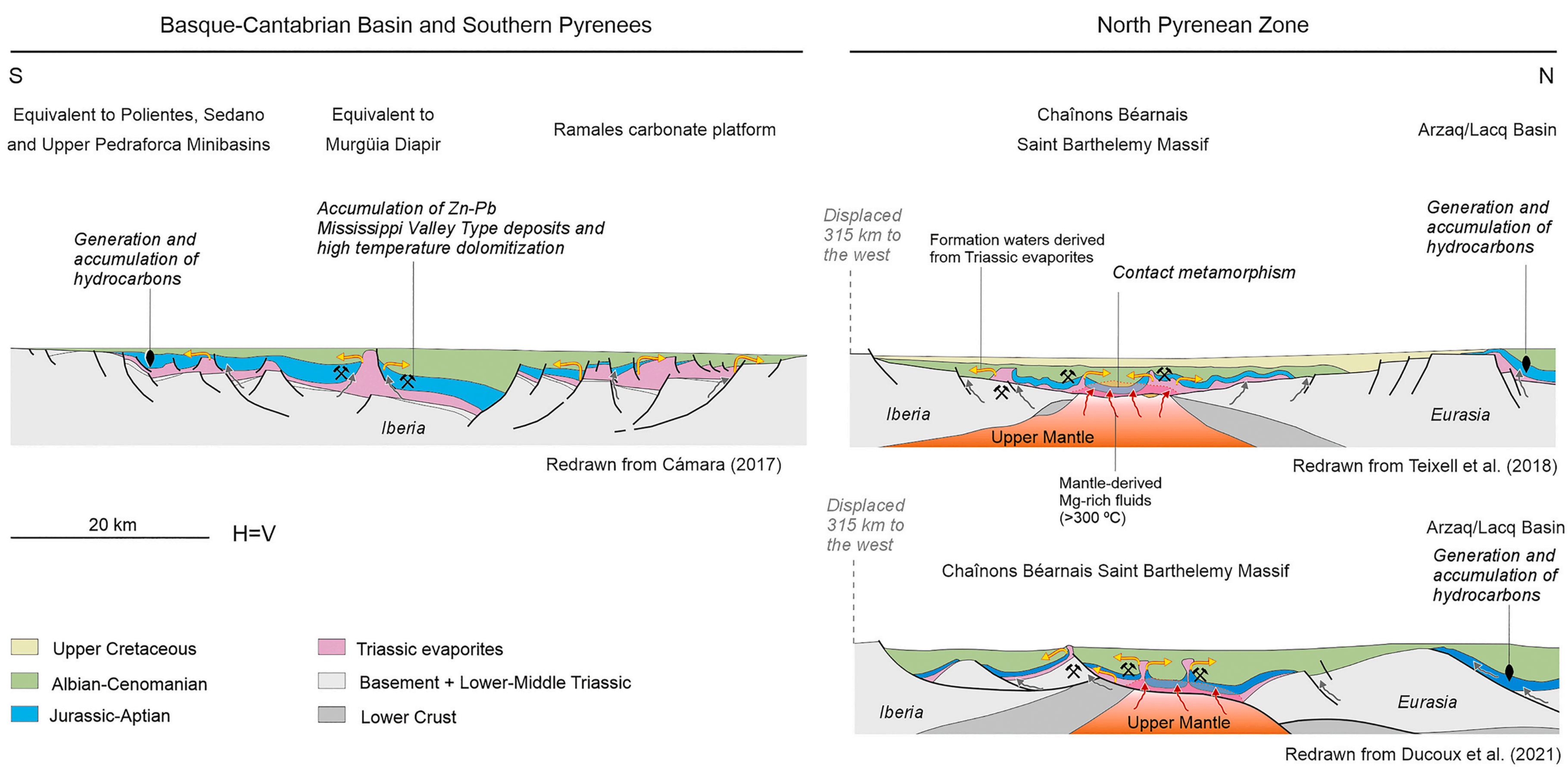

Fig. 12. Conceptual fluid flow model showing the types of fluids and related processes occurring during the development of the Pyrenean rift system from the Early to Late Cretaceous. The fluid flow data is based on Mendia and Ibarguchi (1991), López-Horgue et al. (2010), Iriarte et al. (2012), Dewit et al. (2012), Clerc et al. (2015), De Felipe et al. (2017), Salardon et al. (2017), Perona et al. (2018), Ducoux et al. (2019), Quesnel et al. (2019), Renard et al. (2019), Incerpi et al. (2020), Cruset et al. (2021), Motte et al. (2021) and Bahnan et al. (2021). The cross-sections are redrawn from Teixell et al. (2018) and Ducoux et al. (2021) for the North Pyrenean Zone and Cámara (2017) for the Basque-Cantabrian Basin.

Lacq reservoir, in the northern sector of the Basque Cantabrian Basin as well as during the earlier stages of extension in the Chaînons Béarnais along the North Pyrenean Zone. Associated to these fluid flow events, Zn—Pb mineralization accumulated. Finally, in the more proximal domains of the Basque-Cantabrian basin and Southern Pyrenees, neither hydrothermal dolomitization processes nor ore accumulations occurred, and formation waters at 150 °C derived from seawater interacting with Triassic evaporites were identified. The absence of hydrothermal fluid flow in this proximal domain agrees with a less extended and thicker crust than that of the North Pyrenean Zone, in which deep-sourced fluids enriched in metals migrated. Hydrothermal dolomitization triggered the recrystallization of early dolostones from Aptian to Turonian at burial depths from 1000 to 3000 m in the North Pyrenean Zone (Bahnan et al., 2021), from 1000 to 2000 m along the North Pyrenean Fault (Motte et al., 2021) and from 1400 to 1700 m in the Basque Cantabrian Basin (López-Horgue et al., 2010). In the crust-mantle detachment, fluid-rock interactions occurred at 5000–6000 m according to the calculations done by Nteme-Mukonzo et al. (2020) in the breccias cemented by quartz accumulated on top of the Urdach lherzolite. Fluid temperatures were controlled by crustal thinning, which triggered the migration of deep crustal and mantle-derived hot fluids to shallower positions, possibly overprinting normal geothermal gradients.

5.2. Along and across strike fluid flow changes in fold and thrust belts

Fluid flow variations along and across the strike of the Southern Pyrenees are well registered in the $^{87}Sr/^{86}Sr$ ratios, $\delta^{18}O_{fluid}$ and fluid temperatures measured in calcite veins along the eastern transect J3 and western transect J7 (Figs. 13 and 14). In this section, we relate the temporal and distribution patterns of these geochemical proxies along the Southern Pyrenees with its structure and tectonic evolution. These trends will be compared with those reported in other fold and thrust belts worldwide such as the Appalachians, Apennines, Bighorn Basin, Mexican fold and thrust belt and the Rocky Mountains.

The SE Pyrenees formed from Late Cretaceous to Oligocene by the stack of piggy-back thrust sheet sequence, whereas the structure of the SW Pyrenees is characterized by the imbrication of a piggy-back sequence of thrusts. In the SE Pyrenees, it is observed a progressive increase of the $^{87}Sr/^{86}Sr$ ratios of calcite veins in lower and younger thrust sheets since the middle Eocene (Fig. 13). This geochemical trend can be interpreted as the increasing interaction of fluids with radiogenic sediments forming the Pyrenean thrust sheets. These sediments derived from igneous and metamorphic rocks sourced from the progressive unroofing of the Axial Zone from lower Eocene to Miocene times as resolved by a large number of thermochronological data (e.g., Garwin, 1985; Fitzgerald et al., 1999; Maurel et al., 2002, 2008; Morris et al., 1998; Beamud et al., 2010; Rushlow et al., 2013). The increasing Sr isotopic ratios are also in agreement with the progressive closure of the SE Pyrenean foreland basin, culminating at around 36 Ma during deposition of the Cardona uppermost potash salt, as already inferred by Travé et al. (2007). In the SW Pyrenees $^{87}Sr/^{86}Sr$ data are scarce and do not allow to identify geochemical trends such as those of the eastern sector. The $\delta^{18}O_{fluid}$ of carbonate veins in both the SE and SW Pyrenees shows an increasing number of values within the range of meteoric waters (lower than 0 ‰ VSMOW) since the middle-late Eocene, which corresponds to the marine-continental transition in the South Pyrenean foreland basin. However, differences in the spatial distribution of the $\delta^{18}O_{fluid}$ values higher than +8 ‰ VSMOW and fluid temperatures measured in calcite veins have been observed from the comparison of the two sectors of the Pyrenees. In the SE Pyrenees, these higher values were measured in the lowermost Cadí thrust sheet and in the active margin of the Ebro foreland basin, whereas in the overlying Upper and Lower Pedraforca thrust sheets the $\delta^{18}O_{fluid}$ values and fluid temperatures are lower (Fig. 13). This distribution of isotopic and temperature values evidences a vertical increasing trend from upper to lower tectonic units. Contrarily, in the imbricated thrust sequence of the SW Pyrenees, higher $\delta^{18}O_{fluid}$ values and fluid temperatures were measured in the more internal Larra, Eaux-Chaudes and Cotiella tectonic units with respect to the southernmost Jaca Basin and Sierras Exteriores, revealing a horizontal and forelandward decreasing gradient (Fig. 14).

The $\delta^{18}O_{fluid}$ values and fluid temperatures measured in veins from the SE and SW Pyrenees also evidence along and across strike fluid flow differences that can be attributed to changes in the structure of basement and cover tectonic units and to the oblique directions of Upper Triassic evaporite successions that acted as barriers for deep-sourced fluids. In the SE Pyrenean pile of thrust sheets, the increasing vertical trend of these values could indicate the preservation of fluid systems of the upper tectonic units above pre-compressive Triassic evaporite detachments, which acted as a barrier for deeper hotter fluids migrating in the lower tectonic units not compartmentalized by impermeable detachments. These conditions are reflected in the high temperatures from 110 to 170 °C of hydrothermal fluids, possibly basement-derived, measured in veins from the lowermost Cadí thrust sheet and Ebro foreland basin contrasting with the lower temperatures between 30 and 90 °C of formation and meteoric waters measured in the overthrusting Upper and Lower Pedraforca thrust sheets (Fig. 13). Contrarily, in the SW Pyrenean imbricate, the $\delta^{18}O_{fluid}$ and temperature horizontal decreasing trend towards the foreland indicates the dilution of deep-sourced formation and metamorphic fluids derived from the internal Larra, Eaux-Chaudes and Cotiella tectonic units to the external Jaca Basin and Sierras Exteriores dominated by formation and meteoric fluids (Fig. 14). This horizontal trend is common in compressional settings, as described in other imbricated thrust systems from the Apennines, Rocky Mountains, Sevier and Mexican fold and thrust belts (Qing and Mountjoy, 1992; Fitz-Diaz et al., 2011; Beaudoin et al., 2014, 2015, 2023), whereas the vertical trend identified in the thrust sheet stack of the SE Pyrenees represents a particular case. In any case, the forelandward dilution of fluids requires the participation of compressional tectonic stresses (squeegee-type fluid flow) to mobilize the fluids to more external and cooler positions of the compressional system (Oliver, 1986; Ge and Garven, 1992).

In both the SE and SW Pyrenees, hot fluids tend to dilute towards the foreland as reported from several case studies that show similar patterns regardless fluid composition. In the Central Appalachians, hot fluids derived from the Valley and Ridge Province did not migrate significantly into the foreland Appalachian Plateau (Evans and Hobbs, 2003; Evans, 2010). In the Presqu'ile barrier in the Western Canada Sedimentary Basin and in the Bighorn Basin in the Sevier thrust belt, basement-derived hydrothermal fluids progressively diluted forelandward by their interaction with the sedimentary cover and cooler fluids (Qing and Mountjoy, 1992; Beaudoin et al., 2014). In the Ebro foreland basin, deep-sourced hydrothermal fluids were identified in the Puig-reig anticline at the footwall of the Vallfogona thrust in the Eastern Pyrenees (Cruset et al., 2016, 2018; Sun et al., 2022), whereas in the frontalmost El Guix anticline, only evolved meteoric fluids migrated through fractures (Travé et al., 2000). In the front of the Appalachians and Ouachita fold and thrust belts, tectonically-driven fluids controlled the lateral maturation of hydrocarbons and horizontal thermal gradients, characterized by hot fluids and gas accumulated in the internal domains and colder fluids and petroleum towards the foreland (Leach et al., 1984; Rowan et al., 1984; Oliver, 1986). The same authors also described a forelandward decrease of homogenization temperatures measured in ore deposits hosted in foreland sedimentary rocks. The occurrence of horizontal geothermal gradients reveals a temperature control on the maturation of hydrocarbons in foreland settings. Another control of the timing of maturation of hydrocarbons and preservation of petroleum systems is the competition between burial and erosion related to uplift, as discussed by Kendall et al. (2020) from the comparison of petroleum systems from the Pyrenean, Zagros, Sevier, Andean fold and thrust belts.

5.3. The impact of evaporites and salt-related structures on fluid flow

The pre- and *syn*-compressive evolution of fluid flow in the Pyrenees

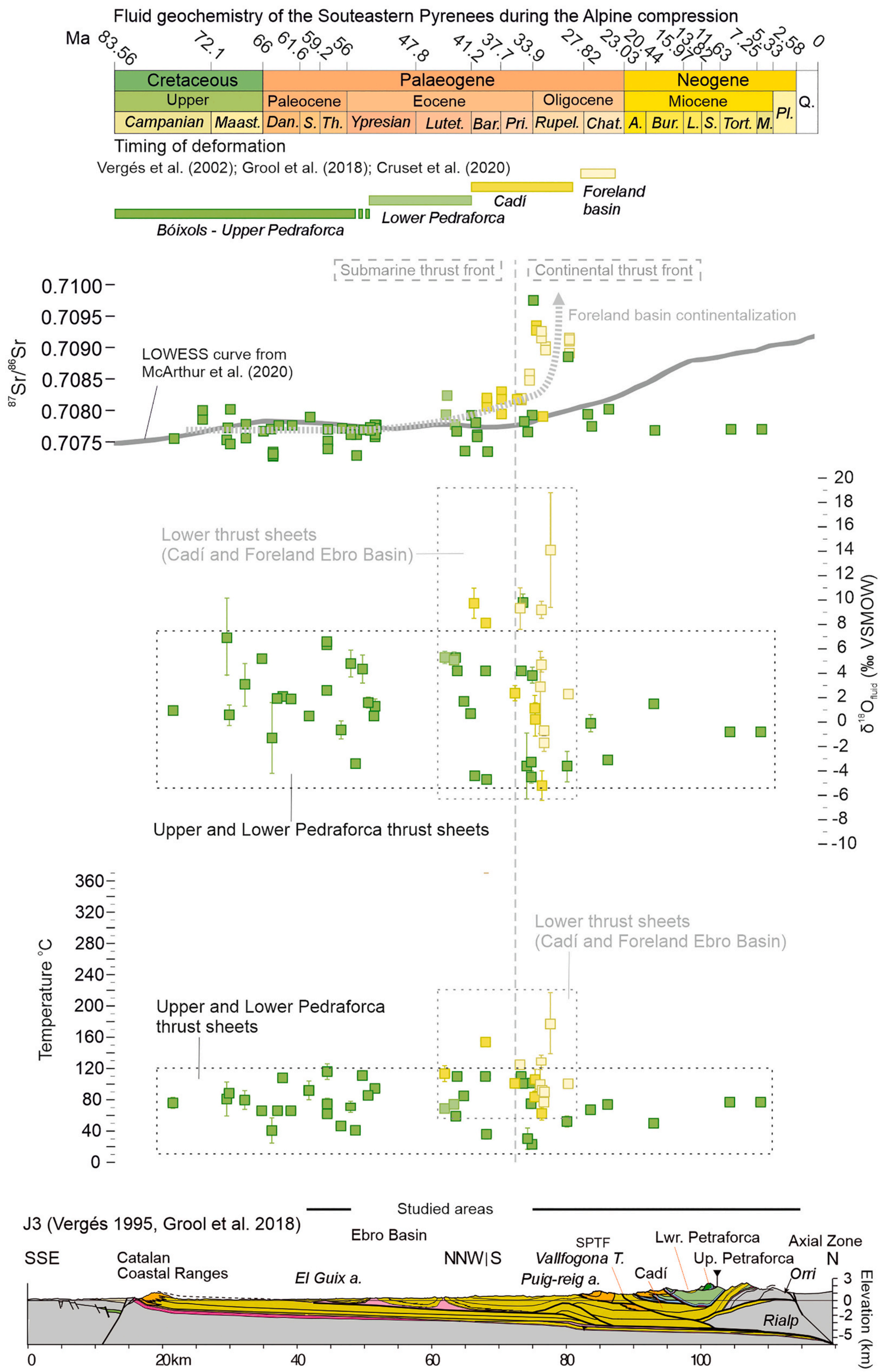

Fig. 13. Evolution of the fluid temperature, $\delta^{18}O_{fluid}$ and $^{87}Sr/^{86}Sr$ ratios of carbonate cement in the SE Pyrenees during the Alpine compression. Geochemical data and timing are based on Cruset et al. (2016, 2018, 2019; 2020a, b; 2021), Nardini et al. (2019) and Muñoz-López et al. (2020a, 2020b, 2022) The timing of thrusting is based on Vergés et al. (2002), Cruset et al. (2020b) and Muñoz-López et al. (2022). Cross-section J3 from Vergés et al. (1995) and Grool et al. (2018).

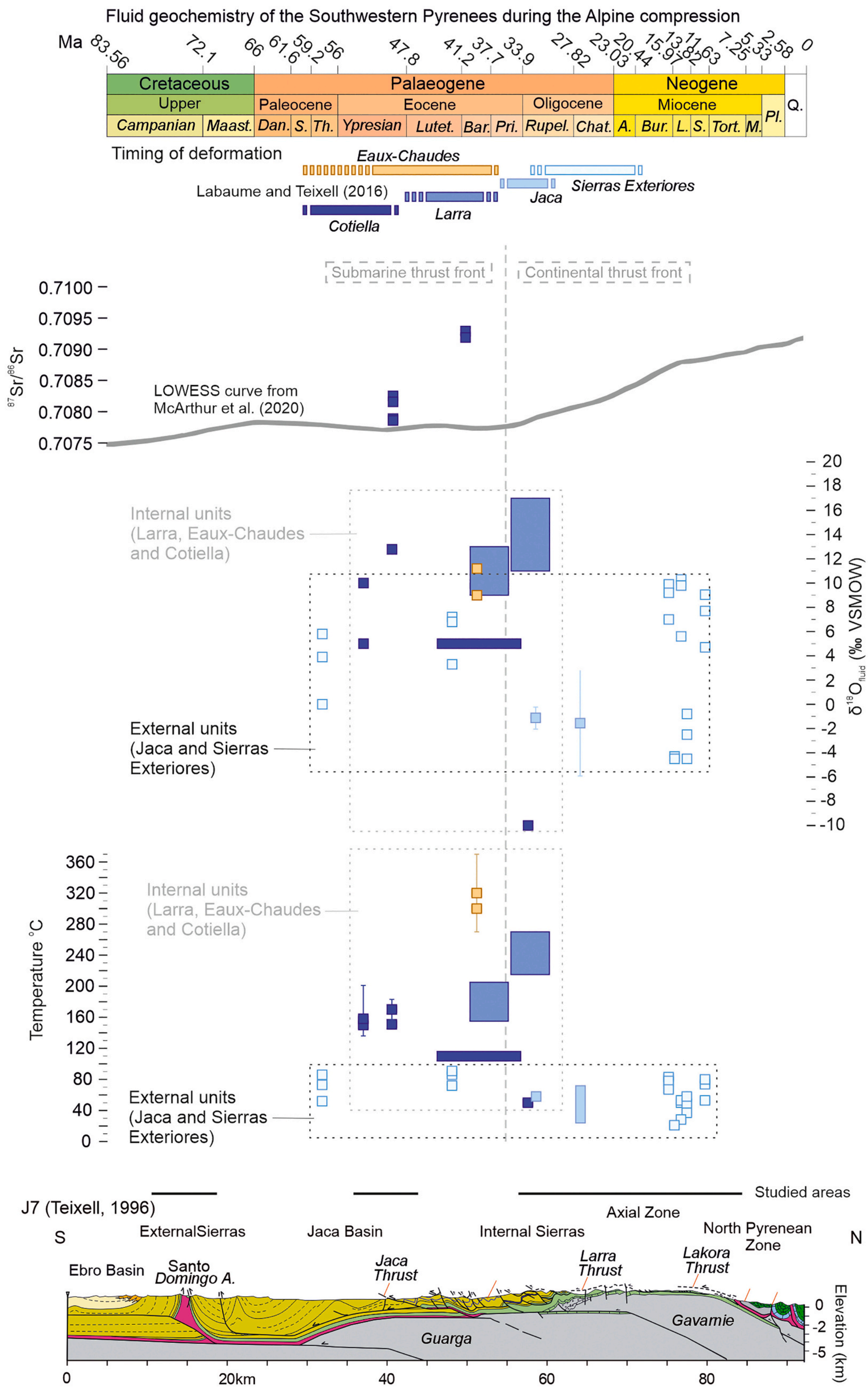

Fig. 14. Evolution of the fluid temperature, $\delta^{18}O_{fluid}$ and $^{87}Sr/^{86}Sr$ ratios of carbonate cement in the SW Pyrenees during the Alpine compression. Geochemical data and timing are based on Travé et al. (1997; 1998a; 2000; 2007), Lacroix et al. (2014, 2018), Trincal et al. (2017), Crognier et al. (2018) and Hoareau et al. (2021). The timing of thrusting is based on Vergés et al. (2002) and Labaume et al. (2016). Cross-section J7 from Teixell (1996). The colours of the data correspond to that of the different tectonic units in the upper part of the figure.

and Basque-Cantabrian Basin has evidenced a strong control of evaporite units and salt-related structures on fluid composition and mobility as well as on the accumulation of ore deposits and hydrothermal dolomitization processes. In this section, we summarize and discuss these controls using examples from the Central and Western Peri-Mediterranean domain, Paradox Basin, Oman Mountains thrust belt and the Gulf of Mexico.

Fracture systems and salt welds formed during pre- and syn-compressive diapirism are the main features channelling or blocking the mobility of fluids through sedimentary units flanking salt bodies. The La Popa salt weld in Mexico, acted as a vertical conduit and horizontal baffle for fluids of different sources possibly interacting with the post-rift Jurassic evaporites (Smith et al., 2012). Similarly, in the Central High Atlas of Morocco, North Pyrenean Zone and Basque-Cantabrian Basin, dolomitization processes of Lower Jurassic and Early Cretaceous carbonates occurred along diapir salt-walls, which were preferential conduits for Mg-rich fluids during the Early Jurassic and Early Cretaceous (e.g., Perona et al., 2018; Moragas et al., 2020; Motte et al.,

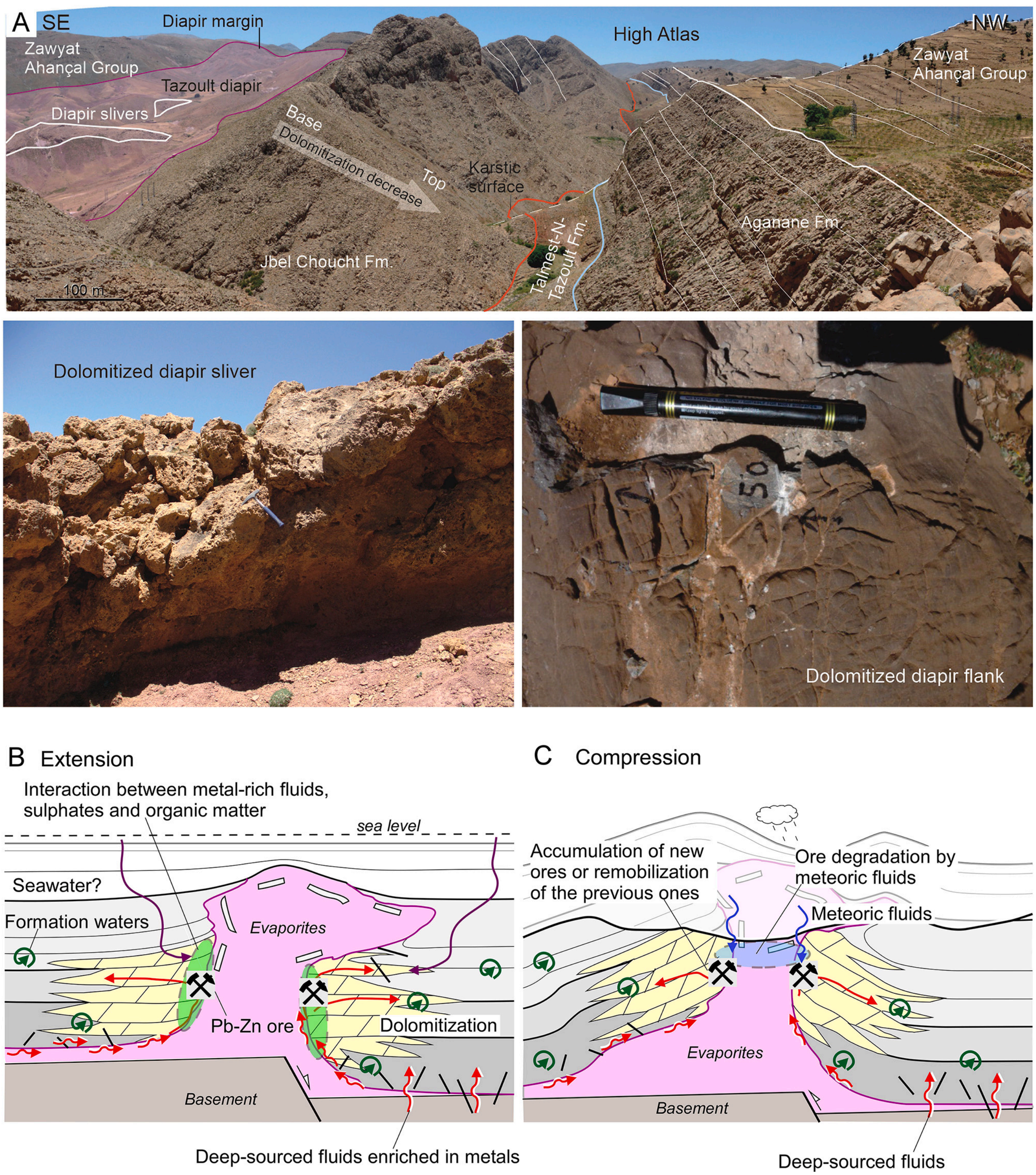

Fig. 15. A) Field images of replacive dolomite related to salt-related extension in the Tazoult Diapir, High Atlas (large-view image redrawn from Martín-Martín et al., 2017). Replacement by dolomite occurred both in carbonate slivers within the diapir core and their flanks. Photographs courtesy of Vinyet Baqués. B—C schematic cross-sections modified from Moragas et al. (2020) showing the fluid-rock interactions in diapirs in both extensional B) and compressional C) tectonic settings.

2021; Bahnan et al., 2021) (Fig. 15A). In the Paradox folded foreland Basin, major faults bounding the Gypsum Valley salt wall acted as drains for fluids of different sources, whereas in less deformed areas fluids were stratigraphically segregated (Lueck et al., 2022). This investigation also revealed that older fluid systems in the Gypsum Valley salt wall were characterized by hot brines interacting with Pennsylvanian evaporites, whereas younger systems were likely shallow meteoric fluids. Brines associated with the Cambrian evaporites in the Jabal Qusaybah anticline at the front of the Oman Mountains thrust wedge, migrated along faults during sediment compaction and/or tectonic deformation (Mozafari et al., 2017). In the northern Gulf of Mexico, crestal normal faults rooted at depth along the top of salt diapirs acted as preferential channels for the expulsion of deep hydrocarbons to the seafloor (Roelofse et al., 2020). Contrarily, in the sedimentary minibasins adjacent to salt diapirs, fluid flow was less effective through diffusion, although was also enhanced due to the formation of faults. The comparison of the aforementioned examples reveals that subvertical diapir walls act as vertical drains and horizontal barriers for fluid flow. However, the halokinetic fracturing of porous sedimentary rocks may enhance the migration of fluids across these barriers.

In the aforementioned examples, different types of evaporite minerals were involved in fluid flow (anhydrite, gypsum, halite, potash salt). However, these examples have been studied in subaerial exposures where the deformed evaporite successions are not always fully preserved due to dissolution and/or salt extrusion during tectonic compression. Despite this lack of preservation, it is possible to hypothesize the seal or conduit behavior of evaporite units from the study of salt mines and experimental studies. As an example, Davison (2009) concluded, from the comparison of evaporite basins from Northern Europe and North and South American Atlantic margins, that brittle deformation can affect even halite and carnallite, the most ductile evaporite minerals, at strain rates of $5 \times 10^{-9}\ s^{-1}$ at 25 MPa (equivalent to 1 km of burial). According to this author, these conditions require the impact of overpressured fluids during faulting. As a consequence, these overpressures may have facilitated the generation of open fracture networks that act as conduit for fluid flow. Similarly, overpressured fluids released from carnallite during its transformation to sylvite led to the growth of interconnected fracture networks (Connolly et al., 1997; Holness and Lewis, 1997; Schléder et al., 2008). A similar scenario is reported for gypsum. According to Davison (2009), the continuous burial and compaction of this sulphate results in the expulsion of fluids, which can induce the brittle deformation of the resultant anhydrite and the generation of fluid conduits. However, gypsum has a more complex behavior. In this line, Wang et al. (2019), based on triaxial tests on gypsum caprocks from the Tarim Basin, concluded that these rocks exhibit ductile deformation with increasing burial depth and confining pressures between 18 and 62 MPa. In this scenario, gypsum may act as a barrier for the migration of fluids.

Fluid-rock interactions occurring during the growth of structures involving evaporites can control the evolution of fluid systems beyond the fold and thrust belt and basin scale. One of the best examples where this scenario can be observed is the Mesozoic extensional rift system in southern Europe and North Africa related to the opening of the Central and Western Peri-Mediterranean domain, where thick successions of Upper Triassic evaporites were deposited (e.g., Turner and Sherif, 2007; Ortí et al., 2017; Soto et al., 2017). These evaporites played a significant role, in the rifting before and after the Late Jurassic, the Late Cretaceous post-rift and the Late Cretaceous-Cenozoic compression, forming thrust detachments and complex diapiric provinces characterized by the dolomitization of thick Jurassic and Cretaceous carbonate successions and the accumulation of Zn—Pb deposits (e.g., Rouvier et al., 1985). Dolomitization and metalliferous deposits resulted from the interaction of seawater or formation fluids in contact with Triassic sulphates with hydrothermal basement-derived fluids and organic matter during both extensional and compressional tectonic settings (Fig. 15B-C), as described in the Basque-Cantabrian Basin, the Aptian-Albian Benicàssim carbonate ramp cropping out along the Maestrat Basin in the Iberian Ranges, the Riópar deposits in the Prebetic domain, the Montnegre Massif in the central Catalan Coastal Ranges, in the Early Jurassic Causses Basin at the footwall of the Cévennes Fault, in the Ikkou Ou Ali ridge along the Moroccan High Atlas and in the Tell and Tunisian Atlas (Rouvier et al., 1985; Leach et al., 2006; Piqué et al., 2008; Mouttaqi et al., 2011; Gomez-Rivas et al., 2014; Martín-Martín et al., 2015; Bouhlel et al., 2016; Navarro-Ciurana et al., 2016a, 2016b, 2017; Perona et al., 2018; Rddad et al., 2018, 2019) (Fig. 15A-B). According to the previous authors, the migration of metal-bearing fluids, through porous rocks during tectonic deformation facilitated the accumulation of the Zn—Pb ores in fault zones, in fractures along diapir walls and unconformities, and in the interparticle porosity of detrital rocks and associated with replacive hydrothermal dolomite. The Zn—Pb deposits were occasionally altered by meteoric fluids and remobilized during Alpine compressional events (Navarro-Ciurana et al., 2016b; Garnit et al., 2018; Rddad et al., 2019) (Fig. 15C).

The link between Triassic sulphates (gypsum) and Mississippi Valley Type deposits identified in Iberian, south European Mesozoic extensional basins and the examples along the Atlas from Morocco to Tunisia evidences a common interpretation for the Central and Western Peri-Mediterranean orogens. This link also evidences a common paleohydrological system for each basin controlled by sulphates and characterized by similar fluids, fluid-related processes and mineralization accumulations occurring during both the Mesozoic extension and Alpine compression. Future research is needed to identify other possible large-scale fluid systems in other deformed areas that can be a potential source of critical raw materials.

5.4. Compartmentalization of fluid systems by stratigraphy and detachment levels

The stratigraphy of fold and thrust belts and the evolution of their mechanical behavior during tectonic deformation are elements that impact the distribution of fluids as well as their migration paths (e.g., Laubach et al., 2009; Cosgrove, 2015). The preservation or destruction of the mechanical stratigraphy allows the compartmentalization of fluid systems into different reservoir units or their mixing by the development of non-stratabound fractures, respectively, as described in the different scenarios presented in Fig. 16.

Fluid reservoirs can be homogeneous, with one single stratigraphic unit hosting one fluid system (Fig. 16A), or heterogeneous, with impermeable stratigraphic units compartmentalizing different fluid systems (Fig. 16B). Well-defined examples of fluid compartmentalization by impermeable units are well-described in the Pyrenean folds and thrusts detached on Upper Triassic evaporites, which favoured the segregation of fluid systems between upper and lower tectonic units (Beaudoin et al., 2015; Cruset et al., 2020a; Hoareau et al., 2021; Motte et al., 2021; Muñoz-López et al., 2022). Fluid compartmentalization by impermeable detachments has been also described in a significant number of folded areas of which we highlight those of the Jura Mountains in the Western Alps, detached on Triassic evaporites (e.g., Smeraglia et al., 2020), the front of the Sicilian fold and thrust belt, detached on late Oligocene-middle Miocene turbidites and Pliocene–Quaternary sands and shales (Dewever et al., 2013), the Patterson Creek anticline in the Central Appalachians (Evans et al., 2012), displaying a blind passive-roof duplex structure detached on Cambrian limestones and the Nuncios Fold Complex detached above Jurassic evaporites (Lefticariu et al., 2005; Fischer et al., 2009).

Fluid compartmentalization is also defined by interlayered impermeable units (i.e., clay-rich and evaporite units) (Fig. 16C), as described in fluid reservoirs from the Alberta Basin, Nuncios Fold Complex, Central Appalachians and Danish Central Graben (Bachu, 1995; Evans and Battles, 1999; Jakobsen et al., 2004; Fischer et al., 2009; Evans, 2010; Evans et al., 2012). In these reservoirs, abrupt changes in the rock porosity and permeability, as well as in fluid temperature and salinity are reported across stratigraphic boundaries that are separated by a few

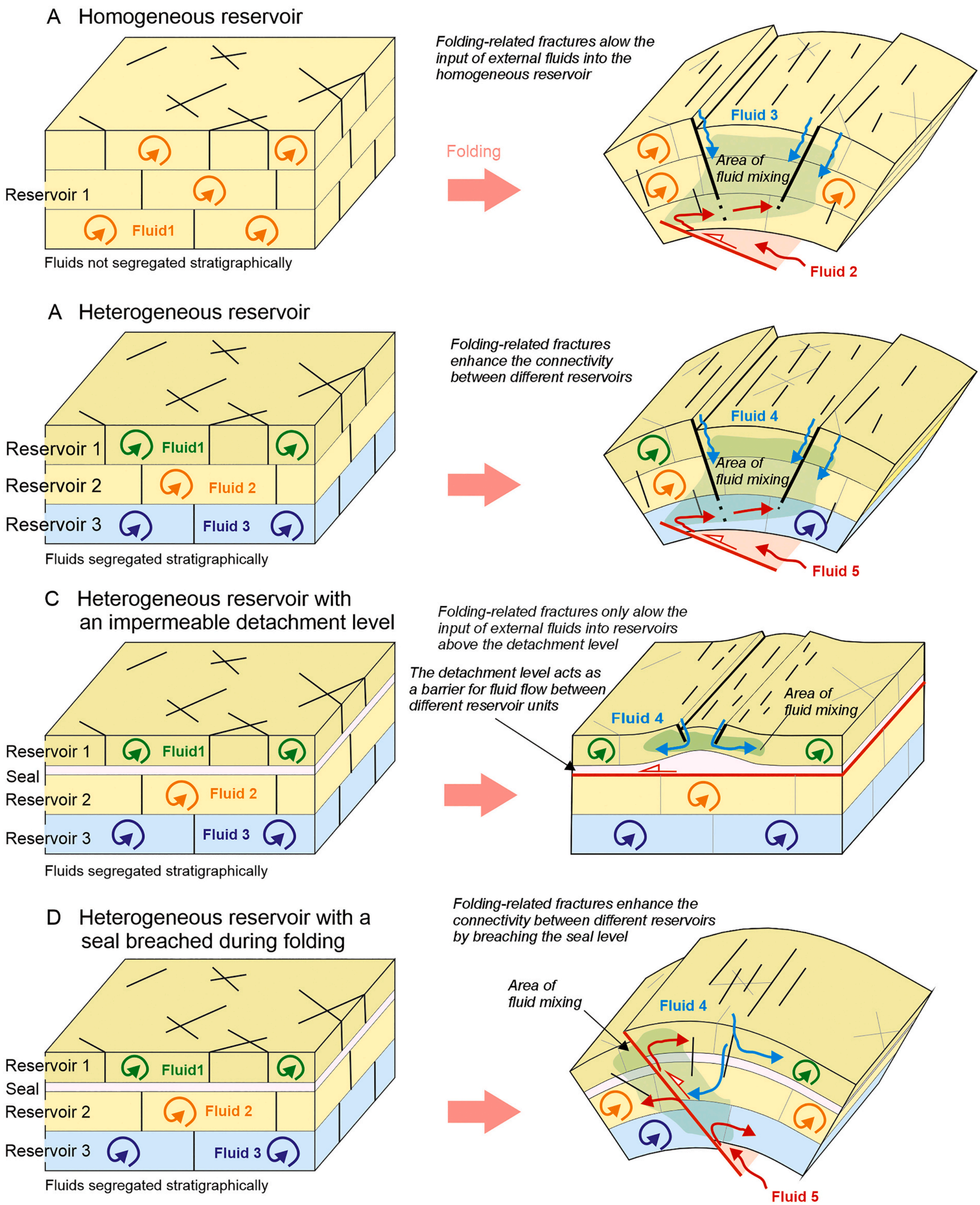

Fig. 16. Block diagrams showing four scenarios of fluid behavior according to the stratigraphy and fracturing history of different reservoirs. A) Slightly deformed homogeneous reservoir with fluids not segregated stratigraphically (Fluid 1). Folding-related fractures allow the input of external Fluids 2 and 3 into the reservoir and their mixing. B) Slightly deformed heterogeneous reservoirs with fluids segregated stratigraphically (Fluids 1 to 3). Reservoirs 1 to 3 are interconnected during folding.C) Slightly deformed heterogeneous reservoirs with an impermeable detachment level (Fluids 1 to 3). During folding, external Fluid 4 mixed with Fluid 1 in Reservoir 1 above the detachment level, whereas Reservoirs 2 and 3 were still undeformed and stratigraphically segregated. D) Slightly deformed heterogeneous reservoir with a seal and fluids segregated stratigraphically (Fluids 1 to 3). Folding-related fractures breach the seal and allow the interconnection between Reservoirs 1 to 3 and the mixing of Fluids 1 to 5.

meters. Burial diagenesis can also generate horizontal barriers for cross-formational fluid flow. From the comparison of examples of carbonate reservoirs from the Bighorn Basin, Zagros, Bahamas Bank, Marion Plateau and Finnmark Platform, Ehrenberg et al. (2006) concluded that calcite cementation produced by chemical compaction and early dolomitization are key processes controlling the development of stratigraphic barriers. In folded successions, the stratigraphic segregation of fluids remains stable during layer-parallel shortening, whereas fluid reservoirs can be interconnected by the development of cross-formational fracture systems during more advanced stages of folding as discussed in Evans and Fischer (2012) and Cruset et al. (2020a) (Fig. 16A, C and D). Fracture systems with good connectivity may cut seal units, favouring the migration of deep hydrothermal fluids and resources previously accumulated beneath non-permeable units, as observed in anticlines from the Bighorn Basin, the Paradox foreland basin and the Lurestan Zagros fold and thrust belt (Sharp et al., 2010; Casini et al., 2011; Beaudoin et al., 2014; Ogata et al., 2014). The fluid flow evolution and the relative extent of fluid-rock interaction during the growth of anticlines may be controlled by the scale and distribution of fracture networks, by the structural domain of the fold and by the folded stratigraphy, as concluded by Muñoz-López et al. (2022) in their study of the Bóixols-Sant Corneli anticline in the Southern Pyrenees. Thus, in the fold hinge, fluid systems are locally rock-buffered, whereas the paleohydrology in the fold limbs and along large thrusts, strike-slip and normal faults is open to the input of external fluids.

Changes in the mechanical stratigraphy also occur during the propagation of both extensional and compressional faults. In this line, through a review of papers reconstructing the evolution of fault zones and related fluid systems in carbonate rocks, Muñoz-López et al. (2020a) reported similar deformation and fluid-related processes associated with the evolution of both thrust and normal faults cutting carbonates (Stewart and Hancock, 1988, 1990; Vermilye and Scholz, 1998; Labaume et al., 2004; Baqués et al., 2010; Bussolotto et al., 2015; Muñoz-López et al., 2020b). In the models proposed by these authors, the initial stage of fault growth was characterized by the formation of extensional fractures and breccias in the fault tip, before compartmentalization of the fault slip plane. The fluids related to this early deformation stage had a similar geochemical composition to the host carbonate, indicating a local origin of the fluid and/or a relatively high extent of fluid-rock interaction. As deformation continued, the fault slip plane propagated along the process zone, allowing the circulation of external fluids that triggered the decrease of fluid rock interaction. However, the fluid-rock interaction is not homogeneous along a fault plain. Studies on the interactions between fluids and host-rocks during thrusting assessed in the Helvetic Alps, in the Canadian Cordillera and in the southern Pyrenees determined that fluid-rock interactions increased from fault cores to damage zones (Kirschner et al., 1999; Knoop et al., 2002; Muñoz-López et al., 2020a, 2020b). The propagation of faults may modify the tectonic fabric of deformed rocks and the compartmentalization of fluid systems. As an example, Curzi et al. (2023) measured a decrease in the permeability of marly rocks as they approach the core of the Sibilini Mts. thrust zone in the Central Apennines, and concluded that slip surfaces and S—C planes acted as barriers for fluid flow. In the same line, similar compartmentalization of fluid systems is reported by Muñoz-López et al. (2020a) in the Bóixols thrust in the Southern Pyrenees cutting limestones, although permeability measurements of this fault zone have not been done. The development of pressure-solution cleavage and the accumulation of non-soluble minerals in the fault core (i.e., clays) could have facilitated the compartmentalization of fluid systems in the footwall and hangingwall. In extensional settings, thick and clay-rich fault gouges developed in poorly-consolidated alluvial fan deposits of the Vallès-Penedès extensional basin also acted as seals (Cantarero et al., 2014a).

The compartmentalization of paleohydrological systems by impermeable layers may control the thermal equilibrium between fluids and host rocks. In their study of the Upper and Lower Pedraforca thrust sheets in the Southeastern Pyrenees, Cruset et al. (2020a, 2021) concluded that temperatures of *syn*-tectonic formation waters at temperatures between 70 and 100 °C were reached by burial considering: 1) the geological cross-sections of Vergés (1993); 2) the geothermal gradients of 25 and 35 °C/km estimated by Labaume et al. (2016) and Metcalf et al. (2009) for the Pyrenean compression; and 3) that Upper Triassic evaporite thrust detachments acted as barriers for the migration of deep-sourced fluids. To the west, in the Sierras Exteriores, Hoareau et al. (2021) also concluded from thermal modeling of fluid temperatures that formation waters and meteoric fluids at a depth of 4 km and with a temperature of <90 °C were in thermal equilibrium with tectonic units detached in Upper Triassic evaporites. Contrarily, the absence of these detachments in the Jaca Basin in the Southwestern Pyrenees and in the Cadí thrust sheet and the northern margin of the Ebro foreland basin to the east facilitated the migration of deep-sourced fluids at temperatures between 100 and 240 °C, possibly derived from the Paleozoic basement, to the shallow thrust front and triggered the development of thermal anomalies (Labaume et al., 2016; Cruset et al., 2016, 2018). Out of the Pyrenees, Smeraglia et al. (2018) concluded that mantle-derived fluids migrated to shallow depths of up to 300 m during the co-seismic to the post-seismic phase of the deep-seated extensional Val Roveto Fault in the Central Apennines. Likewise, in the Catalan Coastal Ranges, the Neogene Vallès extensional fault acted as a conduit for ascending hot fluids that produced anomalous geothermal gradients along fractures (Cantarero et al., 2014b). Cross-formational faulting during the more advanced stages of folding in the Appalachians and Bighorn Basin resulted in the input of squeezed external fluids in thermal disequilibrium with their host rocks (Evans et al., 2012; Beaudoin et al., 2014).

6. Concluding remarks

Fluid analysis in multiple sedimentary basins reports on their relationships with the tectonic evolution associated with the Mesozoic-Cenozoic Wilson Cycle along the Iberian-Eurasian plate boundary zone, based on the combination of existing geochemical and geochronological datasets. The oldest post-Variscan fluids were related to post-orogenic magmatic intrusions interacting with Devonian carbonates that triggered the accumulation of skarn deposits during contact metamorphism and recrystallization of previous ores within the present Pyrenean Axial Zone.

Jurassic-Early Cretaceous carbonates show an early, low-temperature and shallow-burial dolomitization event controlled by seawater influx. This event was widely distributed along the Aquitaine Basin at the southern margin of Eurasia, the Pyrenean extensional system along the North Pyrenean Fault Zone and the Southern Pyrenees and Basque-Cantabrian Basin at the northern margin of Iberia. In the hyperextended North Pyrenean domain, Aptian to Cenomanian fluid events were characterized by hydrothermal dolomitization and the accumulation of Zn—Pb and talc-chloride mineralization. These events were associated with high-temperature and low-pressure metamorphism linked to mantle-derived fluids at temperatures of >300 °C, which interacted with Upper Triassic evaporites, formation waters, seawater and deep crustal fluids. The proximal domains of the Pyrenean extensional system were characterized by colder formation waters at temperatures of up to 150 °C, which possibly interacted with Triassic evaporites along diapir walls and pre-salt rocks in primary welded zones, and the generation of hydrocarbons.

The Pyrenean foreland basin underfilled marine stage was characterized by seawater and formation fluids migrating along fractures and trapped within syn-tectonic rock pores. The progressive transition to the overfilled stage was differentiated by the increasing influence of meteoric fluids, which percolated in the emerging tectonic domains. The Axial Zone antiformal stack, characterized by a significant crustal thickening, represents the source of hot metamorphic and formation fluids during Alpine foreland fold and thrust belt stage on both sides of the Pyrenees.

Variations in the structural complexity along the strike of a mountain belt can produce significant variations in the fluid flow evolution as observed in the Southern Pyrenees. In the east, the tectonic structure is characterized by a particular system of superposed thrust sheets, in which fluid systems progressively increase in temperature and $\delta^{18}O_{fluid}$ from the upper units to the lower units. In the SW Pyrenees, the imbricate thrust system controlled the horizontal gradients with higher fluid temperatures and $\delta^{18}O_{fluid}$ from deeper and internal to shallower and external units. Along-strike fluid changes might be affected by cover and basement changes in structure, a westward decrease of shortening and oblique directions of Upper Triassic evaporites that acted as very efficient seals for the upward migration of deep-sourced fluids.

Subvertical walls of diapirs can act as baffles for fluid flow, whereas halokinetic fracturing and sedimentary units displaying good porosity are effective conduits. Evaporite detachments, fault zones and burial diagenesis generate horizontal fluid barriers that compartmentalize paleohydrological systems that are breached if fluids reach lithostatic pressures. Evaporite successions can control fluid flow events and accumulation of economic resources in large provinces during long periods of time and under both extension and subsequent compression (e.g., Western Mediterranean Mesozoic extensional rift system).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data is derived from other publications cited in the manuscript

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.earscirev.2023.104494.

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