Paleomagnetism from multi-orogenic terranes is "not a simple game": Pyrenees' Paleozoic warning

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Summary

Paleomagnetism is a versatile tool in the Earth sciences: it provides critical input to geological time scales and plate tectonic reconstructions. Despite its undeniable perks, paleomagnetism is not without complications. Remagnetizations overprinting the original magnetic signature of rocks are frequent, especially in orogens which tend to be the areas with better rock exposure.

Unraveling the magnetic history of the rocks is a complicated task, especially in areas that underwent several orogenic pulses. In turn, constraining the timing of remagnetization represents an opportunity to solve post-magnetization structural and tectonic kinematics. Here, we evaluate the magnetization history of Silurian-Devonian carbonates from the Axial Zone of the Pyrenees. The Pyrenees are a multi-orogenic mountain belt where Silurian-Devonian rocks have seen the Variscan collision (late Paleozoic), the opening of the Atlantic / Bay of Biscay (early Cretaceous) and the Alpine orogeny (late Cretaceous to Miocene). Our results show widespread remagnetization(s) carried by magnetite and pyrrhotite in the Silurian-Devonian series of the Pyrenees. The majority of the samples show a post-folding but pre-alpine tilting magnetization. Considering the equatorial inclinations found in such samples, we suggest that they likely acquired their magnetization during the late Carboniferous and early Permian times. Two of the studied sites (located at the western Axial Zone) were subsequently remagnetized at the end of the Alpine orogeny. The paleomagnetic results constrained that the Variscan orogeny was responsible for the main folding event affecting Paleozoic rocks in the Axial Zone, whereas the Alpine orogeny produced the large-scale thrusting and antiformal stacking of these units. In addition, we observed a general clockwise rotational pattern which could be related with the formation of the Cantabrian Orocline and/or rotations associated with the Alpine orogeny. The Silurian-Devonian carbonates are thus useful to understand the tectonic evolution of the Pyrenean mountain range after a systematic combination of paleomagnetism with structural and petrological observations. In contrast, the secondary character of magnetization and complications associated with the Variscan tectonics indicate that a reassessment of Siluro-Devonian poles from the Variscan elsewhere in Europe might be appropriate.

Keywords

Remagnetization, Pyrenees, Variscan, Alpine, Silurian-Devonian, orocline

1. Introduction

The Earth's magnetic field has left a remnant signature in the geological record through eons. These magnetic signals in the rock archive have been crucial to almost any field of Earth Sciences, from the development of plate tectonics (e.g., Vine and Matthews, 1963), to the development of global time scales (e.g., Kuiper et al., 2008) or the origin and evolution of the core (e.g., Biggin et al., 2015). Paleomagnetism is still the only available technique that can quantify pre-Jurassic paleolatitudes (Domeier and Torsvik, 2019), intensities of the past magnetic field, or global reference times through reversals. The paleomagnetic imprint in rocks can last billions of years but may be also fragile. For example, remagnetizations that overprint or even delete the original magnetic signature are ubiquitous, especially in orogenic belts (e.g., Pueyo et al. 2007, 2016a; Van der Voo and Torsvik, 2012; Huang et al. 2017). The majority of the studies associated with the preservation and reacquisition of a magnetic remanence in rocks are relatively recent. Although remagnetizations were initially recognized already during the 1960's and remarked its importance in the 1980’s (McCabe et al., 1983; McCabe and Elmore, 1989), they have been studied in particular detail only from the first two decades of the 21st century onward (c.f. van der Voo and Torsvik, 2012). Since then paleomagnetists and rockmagnetists have realized to the full that a plethora of chemical and physical processes are capable of resetting the magnetic signature in a rock (e.g., Jackson et al., 1993; Weil and van der Voo., 2002; Dekkers, 2012; Pastor-Galán et al., 2017; Aubourg et al., 2019; Huang et al., 2020).

Remagnetization is often deemed a problem because it interferes with paleogeographic reconstructions that rely on the analysis of primary natural remanent magnetization (NRM), i.e. the age of the NRM is the same as the age of the sampled rock unit. Despite the perceived loss of information, remagnetized rocks do represent valuable sources of geological information when it is possible to retrieve precisely the timing of the resetting of the original NRM acquisition, i.e. the remagnetization. Remagnetized rocks have been successfully used to unravel paleolatitudes of orogenic processes, orogenic kinematics, as geothermometers, to reconstruct inverted basins, … (e.g., Dinarès-Turell and García-Senz, 2000, Huang et al., 2015; Villalaín et al., 2016; Aubourg et al., 2019; Izquierdo-Llavall et al., 2020). Rocks with complex orogenic histories – the rule in many orogens – present a myriad of complications including the timing of their NRM acquisition. In such settings, paleomagnetism can be an excellent tool to understand multiphase orogenic systems (and remagnetizations helpful to observe snapshots of deformation histories) if wisely used in concert with other geologic tools.

Many paleomagnetic studies were performed before our current understanding of remagnetization processes and for a given region, time frame, or lithology, those older paleomagnetic data may be the only data available. There is an evident need to review and critically reassess such paleomagnetic information: regional and even global geologic interpretations are still grounded in them. In this paper, we reappraise the paleomagnetism from Silurian-Devonian limestones in the Pyrenean mountain belt, from which only three rather limited studies have been previously published (Tait et al., 2000; Gil-Peña et al., 2006; Izquierdo-Llavall et al., 2020). From these datasets, Tait el al. (2000) interpreted that the magnetic signal carried by Silurian-Devonian units was primary and subsequent plate reconstructions (e.g. Domeier and Torsvik, 2014) have relied on this suggested magnetization age to create the Devonian paleogeography of Europe, but that date might have been remagnetized or not corrected properly as witnessed by several younger studies (e.g. Izquierdo-Llavall et al., 2018 and references therein). Our results, together with an enhanced geologic and paleomagnetic knowledge of the orogen, show that the sampled Silurian-Devonian carbonates were completely remagnetized during the late Carboniferous and Early Permian, when they could experience significant clockwise vertical axis rotations. These rocks were partially overprinted yet another time during Cretaceous and Cenozoic times, and experienced extension, compression, tilting and vertical axis rotations related to both the opening of the Bay of Biscay in the Late Mesozoic and the Cenozoic building of the Alpine chain in the Pyrenees. The magnetization history of the Pyrenees is a warning for paleomagnetists beautifully illustrated in Chris Scotese's anagram: PALEOMAGNETISM = NOT A SIMPLE GAME (Van der Voo, 1993).

2. The Pyrenean mountain belt

The Pyrenees are a mountainous barrier that separates the Iberian Peninsula from the rest of Eurasia. They are a prime example of the superposition of different tectonic events: (1) the multiphase late Paleozoic Variscan orogeny; (2) the Jurassic and early Cretaceous major extension resulting into the formation of oceanic crust in the Atlantic Ocean and the Bay of Biscay and the exhumation of the mantle between the Neotethys and Atlantic Oceans along the current Pyrenees; (3) the closure of this previously opened seaway in the late Cretaceous; and (4) the final collision between the Iberian and Eurasian plates during the Cenozoic (Muñoz, 1992; 2019 and references therein).

2.1 Paleozoic History: Variscan cycle

The tectonic evolution of the Paleozoic era was dominated by the progressive amalgamation of most continents into Pangea (e.g., Domeier and Torsvik, 2014; Domeier, 2016), the latest supercontinent (Pastor-Galán et al., 2019a). In western Europe the Pangean amalgamation history is recorded in the Variscan orogen, which sutured the continents of Gondwana and Laurussia along with a variable number of smaller plates that likely drifted away from Gondwana (e.g., Nance et al., 2010). On the basis of paleomagnetic data, Iberia has been considered part of a ribbon continent (usually named Armorica, Galatia, or Hun) that detached from Gondwana and drifted to the north or northwest in the Late Silurian or Early Devonian (e.g., van der Voo, 1993; Tait et al., 2000; Tait, 1999; Stampfli et al., 2013; Domeier and Torsvik, 2014). Other authors, however, place Iberia along the passive margin of Gondwana throughout the Paleozoic based on the fossil record or the provenance of detrital zircons (e.g., Robardet, 2003; Pastor-Galán et al., 2013a). Convergence leading up to the Variscan orogen started ca. 420 Ma (e.g., Franke et al., 2017) and continued until the complete consumption of the Rheic ocean and other minor oceanic basins that existed between Gondwana and Laurussia at ca. 280 Ma; (e.g., Pastor-Galán, 2020). The final continent–continent collision was diachronic and became progressively younger westwards (in present-day coordinates) with Devonian continent–continent collision along the eastern boundary of the Variscan orogen, progressing to earliest Permian ages in the westernmost sector (e.g., Pastor-Galán et al., 2020 and references therein).

Iberia has the largest exposure of the Variscan orogen in Europe, and an almost continuous cross section of the orogen (e.g., Azor et al., 2019). The majority of the Paleozoic outcrops in Iberia contain Gondwanan affinity rocks (e.g., Pastor-Galán et al., 2013a; Casas et al., 2019) and only a little sector of Southwest Iberia shows Laurussian affinity (e.g., Pérez-Cáceres et al., 2017). Geographically, the external zones of the Gondwana margin are nested to the north into the core of the Cantabrian Orocline (Fig. 1A), whereas the hinterland zones are to the west, center and northeast of Iberia (Fig. 1A; e.g., Azor et al., 2019). The stratigraphy of the Gondwanan authochton consists of Neoproterozoic arc rocks (e.g., Fernández-Suárez et al., 2014), which evolved to a rift-to-drift sequence during the Cambrian to early Ordovician and then to an Ordovician to late Devonian passive margin basin sequence (e.g., Gutiérrez-Alonso et al., 2020). During the Carboniferous and early Permian, the rocks recorded up to 6 phases of deformation (e.g., Dias da Silva et al., 2020; Pastor-Galán et al., 2020 and references therein), metamorphism (e.g., Ribeiro et al., 2019) and synorogenic sedimentation processes that evolved to post-orogenic and intracontinental style basins during the Permian (e.g., Oliveira et al., 2019).

The trend of the Variscan belt in north Iberia follows a "C" shape known as the Cantabrian Orocline (e.g., Pastor-Galán et al., 2020). The Cantabrian Orocline seems isoclinal, formed by a northern and a southern E-W-trending limbs, but this is likely the product of a retightening during the Alpine orogeny (e.g., Pastor-Galán et al., 2011; Leite Mendes et al., 2021; Fig. 1A). All kinematic data studied so far support a model in which the Cantabrian Orocline formed due to secondary vertical-axis rotation in a period of time later than 315 Ma and earlier than 290 Ma. Overall, the southern limb of the orocline rotated counterclockwise (CCW) and the northern limb clockwise (CW; e.g., Weil et al., 2013). Orocline formation postdates the main Variscan orogenic phases (e.g., Pastor-Galán et al., 2015a). The development of the Cantabrian Orocline implies the existence of a roughly linear orogenic belt during the early Variscan closure of the Rheic Ocean (with an approximately N–S orientation in present-day coordinates), which was subsequently bent in map-view into an orocline during the late stages of Pangea's amalgamation. This interpretation is grounded in extensive paleomagnetic, structural and geochronological studies (e.g., Weil et al., 2001; Pastor-Galán et al., 2014; Shaw et al., 2015; Gutiérrez-Alonso et al., 2015; Pastor-Galán et al., 2018).

The Palaeozoic rocks of the Pyrenees form the backbone of the Pyrenean chain and crop out in two areas (the Axial Zone and the Basque Massifs, to the east and west, respectively) that define an E-W elongated strip unconformably overlain by Mesozoic and Cenozoic rocks (Fig. 1A and B). These outcrops are geographically disconnected from neighboring Paleozoic outcrops of the Catalan Coastal Range and Balearic zone to the southeast, the Mouthoumet and Montagne Noire (southern

French Central massifs) to the north, Corsica-Sardinia to the east and the Iberian Massif to the west and southwest. The pre-Permian rocks of the Pyrenees recorded a polyphase deformation during the Variscan orogeny with metamorphism that ranges from absent to high grade (e.g. Casas et al., 2019). So far, no relics of early Variscan deformation and/or subduction related high pressure metamorphism have been found. Most palaeogeographic reconstructions suggest that the Pyrenean Paleozoic outcrops (Fig. 1) may be equivalent to the northern branch of the Cantabrian Orocline (e.g. García-Sansegundo et al., 2011; Pastor-Galán et al., 2020). Deformation, structural style and metamorphic grade show important differences along strike in the Pyrenees (Autran and García-Sansegundo, 1996; Debon and Guitard, 1996) and a N-S zonation, with fold-related cleavage being pervasive in the central and northern part of the Axial Zone but poorly developed in the south (García-Sansegundo et al., 2011). The superposition of the later Mesozoic extension and subsequent Alpine orogeny markedly complicate an integral interpretation of the Variscan portions of the Pyrenees (see Casas et al., 2019). As a consequence, a comprehensive scheme integrating all the available data is lacking despite decades of geological research (e.g., de Sitter and Zwart, 1959; Kleinsmiede, 1960; Zwart, 1979; 1986). In general terms, the Silurian, Devonian and Carboniferous successions show no to low-grade metamorphism and are composed of carbonates and shales (e.g., Casas et al., 2019). During the Carboniferous and Early Permian, the Pyrenees recorded an intense igneous activity including syn- to post-kinematic plutonism and volcanism (Fig. 1B; e.g., Gleizes et al., 1997, 2003; Denèle et al., 2011, 2014; Porquet et al., 2017), sometimes interpreted as subduction related (e.g., Pereira et al., 2014).

2.2 Mesozoic to present day evolution: Alpine cycle

The final break-up of Pangea is marked with the opening of the Central and South Atlantic from late Triassic times onward (e.g., Müller et al., 2019). During the Jurassic, Iberia was attached to Europe and North America as another piece of Laurasia. In the Cretaceous, the breakup and spreading in the North Atlantic led to the separation of the Iberian microplate from Eurasia, North America and Africa (e.g., Vissers and Meijer, 2012). During the North Atlantic breakup, the Bay of Biscay opened, leading to approximately 35° of counterclockwise (CCW) rotation of Iberia (Van der Voo, 1969; Neres et al., 2013; Nierrengarten et al., 2018) probably during the Aptian (Juárez et al., 1998; Gong et al., 2008). The opening of the Bay of Biscay to the west got recorded in the Pyrenees with the formation of a hyperextended margin with mantle exhumation during the Albian–Cenomanian (e.g. Lagabrielle et al., 2010). The rotation of Iberia, together with the increased convergence between Africa and Eurasia culminated in the collision between Iberia and Eurasia to form the Pyrenean range during Late Cretaceous-Miocene in the frame of the Alpine orogeny (e.g., Muñoz, 2019).

Paleozoic rocks in the Pyrenees are involved in a doubly-verging, asymmetric fold-and-thrust system with the main Alpine thrust sheets in the Axial Zone being South-verging (Muñoz 1992, 2019; Barnolas et al., 2019). They provoked over 100 km of N-S shortening and the southward displacement of the Palaeozoic rocks in the Axial Zone over the autochtonous basement in the southern Pyrenean foreland. The outcrops of Paleozoic rocks in the Axial Zone have witnessed more than 7000 meters of basement stacking (measured between the Balaïtous peak where the top basement crops out and the San Vicente drill core; Fig. 1B; Lanaja, 1987). The geometry and kinematics of the thrust units affecting the Paleozoic rocks are not fully understood because of the complex superposition of deformation events and the unclear relationships with cover units where syntectonic sedimentation plays a key role to assign kinematic ages (Oliva-Urcia, 2018). Numerous structural studies (e.g., Muñoz et al., 1986, 2019; Muñoz, 1992; Puigdefábregas et al., 1992; Teixell, 1996, Millán et al., 2000; Martínez-Peña and Casas, 2003; Casas et al., 2003; Millán et al., 2006; Labaume et al., 2016; Labaume and Teixell, 2018) have identified a general Alpine piggy-back thrust sequence affecting the Paleozoic rocks (Fig. 2). Besides, numerous fission-track data on granites (Fitzgerald et al., 1999; Jolivet et al., 2007), on detrital rocks (Beamud et al., 2011; Bosch et al., 2016; Labaume et al., 2016) as well as $^{40}Ar/^{39}Ar$ and U/Pb dating of samples directly taken from fault planes (Abd Elmola et al., 2018) and calcite veins (Hoareau et al., 2021, see also recent reviews by Oliva-Urcia, 2018 and Calvet et al., 2020, and references therein), have improved the knowledge on the chronology of emplacement and exhumation of the basement units and their relationship with the cover ones. Basement thrusts partly reactivated previous Variscan, late-Variscan and/or Mesozoic structures. The Alpine structure defines an imbricate thrust system (Fig 2) with a progressively increasing vertical overlap between the basement units from west (Fig. 2A; Teixell, 1996) to east where thrusts define an antiformal stack (Fig. 2C; Muñoz, 1992). Basement units in the west include four thrusts in the central sector (Fig. 2b): Millares, Bielsa , Gavarnie and Guarga; and an variable number in the western sector (Fig. 2a), including the Lakora-Eaux Chaudes, Gavarnie, Guara-Gedré, Fiscal-Broto and Guarga thrusts. In the eastern sector (Fig. 2c) three main basement units are recognized, from the North to the South: Nogueras-Gavarnie, Orri and Rialp. In the South Pyrenean Zone, all these basement units connect with an imbricate fold-and-thrust system with different Mesozoic and Cenozoic décollements (Upper/Middle Triassic evaporites, Cretaceous shales, and Eocene marls and evaporites); their geometry is controlled by salt-tectonics and Mesozoic inheritance (Millán et al., 2000; Huyghe et al., 2009; Labaume et al., 2016; Oliva-Urcia, 2018; Labaume and Teixell, 2018; Calvín et al., 2018; Santolaria et al., 2020; Muñoz et al., 2021).

Thrusting was associated with important foreland flexure and foreland succession deposition both in the Northern Pyrenees (France; Biteau et al., 2006) and the Southern Pyrenees (Spain; Puigdefàbregas, 1975). Foreland deposits partly covered the Paleozoic units of the Axial Zone in the early orogenic stages and were subsequently exhumed and eroded (Beamud et al., 2011; Fillon and Van der Beek, 2012). The early, maximum burial conditions in the sampled portion of the Axial

Zone are partly constrained by paleothermal studies in the overlying Meso-Cenozoic cover units (Izquierdo-Llavall et al., 2013; Labaume et al., 2016). They indicate Cenozoic-age, maximum temperatures of 160-190 ºC in the Upper Cretaceous units of the western Axial Zone (Izquierdo-Llavall et al., 2013) that increase up to ~250 ºC in the Eocene turbidites to the center of the Southern Pyrenean Zone. These values indicate that temperatures in the underlying Paleozoic rocks could range between 200 and 250 ºC during the Cenozoic. Locally, around the Panticosa intrusion, peak burial temperatures may have reached about ~300 ºC during the Oligocene (Bosch et al., 2016). In the central Axial Zone, thermal models for the Paleozoic units of the Gavarnie and Orri units reveal peak temperatures below 300ºC that were attained during the Early Paleogene (Waldner, 2019). Cenozoic burial favored the development of Alpine cleavage in the western Axial Zone (Choukroune and Séguret, 1972; Matte et al., 2000). Conversely, in the central Axial Zone Alpine cleavage developed only locally, the main cleavage being Variscan in age (Muñoz, 1992).

2.3 Paleomagnetism in the Pyrenees

Paleomagnetic investigations in the Pyrenees commenced with the pioneering studies of Van der Lingen (1960) and Schwarz (1963) in some Paleozoic rocks from the center of the Pyrenees. The available database has grown substantially during the following decades due to the excellent outcrop conditions (including world class stratigraphic sequences), the general exposure of synorogenic material throughout the chain allowing an accurate dating of deformation, the existence of well-exposed zones of lateral transference of deformation, etc. At present, the Pyrenean chain represents one of the most densely and homogeneously sampled paleomagnetic databases worldwide (Pueyo et al., 2017). Despite the quality and amount of paleomagnetic data in the Pyrenees, the Pyrenean Paleozoic rocks have remained largely unexplored. In the Axial Zone, very few data are known from sites older than Permian–Triassic red beds (Van Dongen, 1967; McClelland and McCaig, 1988, 1989;

Keller et al., 1994; Tait et al., 2000; Gil-Peña et al., 2006; Izquierdo-Llavall et al., 2014, 2020; Ramón et al., 2016). And, to our knowledge, in the Pyrenees only three paleomagnetic studies collected and analyzed a limited number of sites dating from older than late Carboniferous (Stephanian) age: Tait et al. (2000), Gil-Peña et al. (2006), and Izquierdo-Llavall et al. (2020). The two latter studies found the rocks remagnetized in the Late Carboniferous and Paleogene, respectively. Previous studies in late Carboniferous and early Permian rocks (Izquierdo-Llavall et al., 2014) revealed shallow inclinations and clockwise rotations of ~40°. Gil-Peña et al. (2006) showed analogous rotations (~50° CW) for the Ordovician rocks, that probably were remagnetized during late Carboniferous times.

The relatively good paleomagnetic control on undeformed areas in the vicinity of the Pyrenees (e.g., Garcés et al., 2020; Oliva-Urcia and Pueyo, 2019) allows to define a reliable reference paleomagnetic direction and to understand the post-Variscan (late Permian to Eocene) magnetization and tectonic history during the Alpine orogeny in the Pyrenees. In the South Pyrenean Zone, significant rotations both CW and CCW (40-60°) derived from proven primary paleomagnetic records from rocks of ages ranging from Permo-Triassic (e.g., Larrasoaña et al., 2003) to Oligocene (e.g., Sussman et al., 2004) are found in the most external cover units in relation to lateral ramps in thrust sheets. These rotations are especially evident nearby the boundaries of the so-called South Pyrenan Central Unit (e.g., Sussman et al., 2004; Mochales et al., 2012, 2016; Muñoz et al., 2013; Rodriguez-Pintó et al., 2016) but also in the most external thrust units (Pueyo et al., 2021a, 2021b). Other moderate vertical axis rotations (15-25° CW and CCW, locally attaining ~40°) occur in the Permo-Mesozoic structural units immediately to the south of the Axial Zone: The Internal Sierras (Larra-Monte Perdido units; Oliva-Urcia and Pueyo, 2007a, 2007b; Oliva-Urcia et al., 2008; Izquierdo-Llavall et al., 2015), the Nogueras thrust and underlying units (McClelland and McCaig, 1988, 1989; Dinarès et al., 1992; Oliva-Urcia et al., 2012; Izquierdo-Llavall et al., 2018) and in the eastern Cadi unit (Dinarès et al., 1992; Keller et al., 1994; Pueyo et al., 2016b). In the North Pyrenean Zone (France, Fig. 1B), paleomagnetic data are scarcer and evidence strong (over 70° CW in Aptian-Albian rocks, Oliva-Urcia et al., 2010; Rouvier et al., 2012) to null (Izquierdo-Llavall et al., 2020) vertical axis rotations in different areas.

Early Cretaceous remagnetizations are relatively common in the Cretaceous basins deformed to the south of the Axial Zone (e.g. Larrasoaña et al., 2003; Garcés et al., 2016). However, this event have been described, so far, only out of the Axial Zone. These remagnetizations are local and affect compartimentalized and highly subsident basins developed under high thermal gradient conditions (e.g., Lagabrielle et al., 2010) which played a key role to chemically remagnetize these rocks (e.g., Dinarès-Turell and García-Senz, 2000; Gong et al., 2008; 2009). On top of that, Cenozoic remagnetizations have been described in the Meso-Cenozoic units just above the Axial Zone (Oliva et al., 2008; 2012; Izquierdo-llavall et al., 2015). These Cenozoic remagnetizations likely occurred due to the burial associated with the development of the Pyrenean orogenic wedge and southern foreland basin. The wedge generated important lithostatic and tectonic load in the internal units until the final collision, continentalization and exhumation of the Paleozoic rocks during Oligocene-Miocene times.

Most of the paleomagnetic data to the south of the Axial Zone recorded, at least partially, Eocene secondary magnetizations (pre, syn and postfolding). Remanent magnetization in the Mesozoic units immediately to the North of the Axial Zone is in general terms post-folding and has been interpreted as a Cenozoic chemical (Oliva-Urcia et al., 2010) or thermal (Izquierdo-Llavall et al., 2020) remagnetization.

3. Sampling, methods and results

We drilled in 19 limestone sites from the Silurian or Devonian, one site of a late Carboniferous-early Permian granite (OG01, Panticosa intrusion) and one Permian dyke that intruded the surroundings of site OG12 (OG12dyke; Fig. 1B; Table 1) with a petrol-powered drill, in total 240 cores. We also collected 6 oriented hand samples (from the OG07 and OG08 sites, three samples each). Sites are distributed along–strike the southern and central Axial Zone from the Gallego valley in the west to the Valira valley in the east in eight different valleys (Fig. 1B; the kml file sample_locations.kml with exact locations is in the Supplementary material). Sites BN1 and OR15 were collected in the same area of sites published in Tait et al. (2000). Several sites allowed field tests: five site-scale fold-tests could be obtained (OG2; OG11; OG13; OG14; OG19); two tilt tests between sites within the same thrust unit (OG3-4; BN1-OR15), and two sites with a baked contact test (OG12 and OG17). We performed all analyses at Paleomagnetic Laboratory Fort Hoofddijk, Universiteit Utrecht, The Netherlands.

Our sample collection comes from fresh, non metamorphic and weakly or non-internally deformed sites. Limestones were sedimented in shallow waters usually associated with clastic and pelitic sediments (Casas et al., 2019). Most limestone sites contained variable amounts of organic matter visible while drilling. A few of these sites show a spaced solution cleavage and evidence of recrystallization. We collected the bedding orientation and, when observable, that of the pressure-solution cleavage (Table 1).

3.1 Alpine tilt estimation

The sampled rocks were affected by both Variscan and Pyrenean orogenies (Alpine). In the Axial Zone, the Pyrenean orogeny produced the development of a south vergent fold-and-thrust system.

Its geometry has been extensively investigated in previous studies that reconstruct the Axial Zone structure using a combination of surface data and seismic profiles (Labaume et al., 1985; Cámara and Klimowitz, 1985; Muñoz, 1992; Teixell, 1996; Teixell and Muñoz, 2000; Matínez-Peña and Casas-Sainz, 2003; Izquierdo-Llavall et al., 2013; Labaume et al., 2016; Muñoz et al., 2018; Labaume and Teixell, 2018). These studies reveal that the Axial Zone evolves laterally from an imbricate thrust system in the West (Fig. 2a, Teixell, 1996) to an antiformal-stack in the East (Fig. 2c, Muñoz, 1992). Alpine thrusts are related to kilometric-scale basement folds in their hangingwalls. Basement folding is well recorded by the Mesozoic units unconformably overlying the Paleozoic rocks and resulted in dominantly southward and northward tilts in the southern and northern part of the Axial Zone, respectively.

We used the available geological maps (GEODE, Robador et al., 2019) , including numerous bedding data, and published cross-sections to discriminate the effects of Variscan and Pyrenean orogenies and estimate the alpine tilt related to basement thrusting in our sampling sites. In the sites located in the southern part of the Axial Zone, alpine dip directions and dips (Table 1) were estimated from the average orientation of bedding in the Mesozoic units unconformably overlying the Paleozoic and tested against the dip estimated in cross-sections. Sites in the central part of the Axial Zone (OG11 to OG19) are to far away from Mesozoic cover units. In these sites, we defined dip directions as perpendicular to alpine thrusts in the Paleozoic rocks. Following the dip direction we projected our sampling points into the traces of previously published cross-sections to obtain the dip (Fig. 2). Alpine dips were also estimated considering the Mesozoic geometries reconstructed above the topography in such cross-sections (Fig. 2). Figure 2 provides a general picture of the structural position of the sampled sites (see corresponding thrust units in Table 1) and the regional alpine tilt recorded by the Mesozoic cover of the Axial Zone. Cross-sections run perpendicular to the main alpine structures and therefore show true alpine dips. Apart from the regional cross-sections shown in Figure 2 (Teixell, 1996; Muñoz, 1992; Martínez-Peña and Casas-Sainz, 2003), we also took into account additional cross-sections located closer to the sampling sites (Izquierdo-Llavall et al., 2013, 2018; Labaume and Teixell, 2018).

3.2 Paleomagnetic and Rock magnetic Methods

Knowing when and how rocks magnetized is crucial to obtain an appropriate interpretation of paleomagnetic results, especially in terms of plate and structural kinematics. In this paper we combined rock magnetic, paleomagnetic and structural geology analyses to unravel the magnetization history of the rocks.

Rock magnetism studies the magnetic properties of rocks and their magnetic minerals. The different magnetic properties of rocks, such as magnetic hysteresis, susceptibility and its anisotropy, magnetization vs. temperature (thermomagnetic analysis), can inform about the mineral(s) carrying the magnetic remanence and their crystal structure and grain size. This information is the most important to understand the geological proceses involved in the magnetization of the rocks, and eventually also a key to unravel magnetization timings. In this research we have performed a series of rock magnetic analyses to fully characterize the magnetic mineralogy of the studied samples as a step towards understanding the magnetization process and timing. We measured 18 high-field thermomagnetic runs in an in-house-built horizontal translation-type Curie balance with a sensitivity of approximately $5*10^{-9}Am^2$ (Mullender et al., 1993) and one in an AGICO KLY-3 susceptibility bridge with a CS2 furnace attachment with nominal sensitivity ($5*10^{-7}$ SI) and air forced into the tube. This latter analysis included two heating and cooling cycles. We also analyzed 20 magnetic hysteresis loops and one first order reversal curve (FORC) diagram. They were measured at room temperature with an alternating gradient force magnetometer (MicroMag Model 2900 with 2 Tesla magnet, Princeton Measurements Corporation, noise level 2×10^{-9} Am^2). Finally, we obtained 88 isothermal remanent magnetization (IRM) acquisition curves from our Pyrenean limestone samples. Curves were obtained with the robotized magnetometer system of Utrecht University (Mullender et al., 2016).

Our paleomagnetic analyses were focused towards determining the Natural Remanent Magnetization (NRM) of the rocks. NRM provides information about ancient latitudes (inclination of the magnetic remanence) and rotations (declinations with respect to the past north) so that we can constrain the magnetization timing. The NRM of the sample collection was investigated through alternating field (AF) demagnetization and thermal demagnetization. AF demagnetization was carried out using the aforementioned robotic 2G-SQUID magnetometer, through variable field increments (4–10 mT) up to 70–100 mT. In all limestone samples, where high-coercitivity, low-blocking temperature minerals (i.e. goethite, titano-hematite) were expected, a heating step to 150 °C was performed previous to the AF demagnetization. At the same time this enhances the distinction between secondary and characteristic NRM components determined with AF demagnetization (van Velzen & Zijderveld, 1995). In samples demagnetized thermally, a stepwise thermal demagnetization was carried through 10–100 °C increments up to complete demagnetization. Principal component analysis (Kirschvink, 1980) was used to calculate magnetic component directions from orthogonal vector end-point demagnetization diagrams (Zijderveld, 1967) with the online open-source software Paleomagnetism.org (Koymans et al., 2016; 2020).

Anisotropy of magnetic susceptibility (AMS) measures the induced magnetization in a rock when applying a magnetic field in different directions, defining an ellispsoid (e.g. Parés, 2015). The shape of the AMS ellipsoid depends on the crystallographic preferred orientation of the minerals; the shape, size, and preferred orientation of mineral grains; the occurrence of microfractures, its distribution and size… Frequently it is a good proxy for sedimentary and tectonic fabrics that are not visually obvious, but it is also a powerful method to investigate the effect of deformation on the NRM. We determined the composite fabric of the paramagnetic, diamagnetic and ferromagnetic grains by measuring the AMS of 148 samples from our collection with an AGICO MFK1-FA susceptometer (nominal sensitivity $2*10^{-8}$ SI).

3.3 Rock magnetism results

3.3.1 Thermomagnetic analyses

We placed between 50–100 mg of powdered sample material from representative samples into quartz glass cup holders that hold the sample with quartz wool. We programmed stepwise thermomagnetic runs with intermittent cooling between successive heating steps. The heating and cooling segments were 150, 100, 250, 200, 300, 250, 400, 350, 520, 450, 620, 550, 700º C and finally back to 25° C, respectively. Heating and cooling rates were 10° C min^{-1}. Many samples show a paramagnetic contribution, sometimes uniquely (OG19; Supplementary File SF1), sometimes with a more or less noteworthy alteration reaction at about 400–450° C. This indicates the presence of non-magnetic sulfides (likely pyrite) that oxidize to magnetite during the thermomagnetic run (Fig. 3; OG14; Supplementary File SF1). Some samples show a small but sharp decay between 500 and 600° C, indicating the presence of magnetite (Fig. 3, OG13), others show the presence of pyrrhotite with a sudden increase at ~300°-320° C followed by a sharp decrease afterwards (OG8 in Fig. 3; OG06 and OG07 in SF1). All samples showing pyrrhotite contained a less important, but observable, content of pyrite. In the susceptibility vs. temperature curve (Fig. 3, BN1), pyrrhotite is observable during cooling but it likely formed as a secondary mineral during one of the heating cycles.

3.3.2 Magnetic hysteresis

Representative samples with masses ranging from 20 to 50 mg were measured using a P1 phenolic probe. Hysteresis loops were measured to determine the saturation magnetization (Ms), the saturation remanent magnetization (Mrs), and the coercive force (Bc). These parameters were determined after correcting for the paramagnetic contribution. The maximum applied field was 0.5 T. The field increment was 10 mT and the averaging time for each measurement was 0.15 s. We found different loop shapes (Fig. 4, SF-2): (i) Loops that do not saturate at 0.5 T with a pseudo-single-domain like shape which points to the presence of a relatively hard magnetic carrier likely pyrrhotite (Fig. 4, OG08) and (ii) typical magnetite-like pseudo-single domain loops (Fig. 4, OG19). We performed a first order reversal curve (FORC) diagram (Fig. 4, BN1-3) that shows a mixture between superparamagnetic and single domain behaviour.

3.3.3 Isothermal Remanent Magnetization (IRM)

Before the actual IRM acquisition, samples were AF demagnetized with the static 3-axis AF protocol with the final demagnetization axis parallel to the subsequent IRM acquisition field, a procedure that generates IRM acquisition curves with a shape as close to a cumulative-lognormal distribution as possible (Egli, 2004; Heslop et al., 2004). IRM acquisition curves consist of 61 IRM levels up to 700 mT. The shape of IRM curves is approximately a variably skewed cumulative log-Gaussian function which may contain more than one coercivity phase. IRM component analysis enables a semi-quantitative evaluation of different coercivity components (magnetic minerals or particle sizes) to a measured IRM acquisition curve. Every skewed log-normal curve is characterized by four parameters: (1) The field ($B_{1/2}$) corresponding to the field at which half of the saturation isothermal remanent magnetization (SIRM) is reached; (2) the magnitude of the phase (Mri), which indicates the contribution of the component to the bulk IRM acquisition curve; (3) the dispersion parameter

(DP), expressing the width of the coercivity distribution of that mineral phase and corresponding to one standard deviation of the log-normal function (Kruiver et al. 2001; Heslop et al. 2002); and (4) the skewness of the gaussian curve (Maxbauer et al., 2016). IRM curve unmixing was performed with IRM MaxUnmix package (Maxbauer et al., 2016). The interpretation of the coercivity components in terms of mineralogy and grain size is usually done in concert with thermomagnetic curves.

Results from individual samples are characterized by two main IRM components: (a) a relatively soft component (C1 in Fig 5A) with $B_{1/2}$ between a minimum value of 23 and a maximum of 74 mT, but generally ~40 mT and dispersion parameter (DP) of ~ 0.33 and 0.38 (log units); and (b) a high coercivity component (C2) with a high $B_{1/2} > 200$ mT and DP ~0.5 (log units). Both components are present in all samples but in varying proportions (Fig. 5A) of the SIRM. We performed end-member modeling in all of the same lithology Silurian-Devonian samples (following the steps of Gong et al. (2009a) but without a 150 °C preheating of the samples) to fully characterize the IRM set of samples. The program (Heslop and Dillon, 2007) to interpret the IRM acquisition curves uses the algorithm developed by Weltje (1997). End-member modeling assumes that the measured data can be represented by a linear mixture of a number of invariant constituent components, which are referred to as endmembers. The algorithm dictates that input IRM acquisition curves are monotonic; the curves were smoothed when appropriate to enforce them being monotonic. By least-squares minimization calculated normative compositions are optimized to the measured IRM acquisition curves, eliminating the need for prior knowledge of end-member properties (cf. Weltje, 1997). For further information about this technique in the framework of remagnetization see the review by Dekkers (2012). We found that a two end-member model shows an acceptable r^2 value of 0.6. Models with 3 to 9 end members show slightly better fits ($r^2 = 0.73$ to 0.88 respectively) although improvement is not deemed that significant (see discussion). The two end members are a soft (42 mT component with a DP = 0.36) and a hard (a 200mT component and DP = 0.35) component, analogous to C1 and C2 in the individual IRM curves analyzed (Fig. 5C).

3.4 Paleomagnetism results

A minimum of five demagnetization steps was considered to characterize a remanent component. In specimens where directions were difficult to isolate, we used the approach of McFadden and McElhinny (1988) in combining great circles and linear best fits (set points). The Virtual Paleomagnetic Directions (VPD) software was also used (Ramón et al., 2017) at the site level (stacking routine, linearity spectrum analysis, and the virtual direction methods by Scheepers and Zijderveld (1992); Schmidt (1982) and Pueyo (2000), respectively) to confirm the means derived from PCA analyses of individual specimens. Representative Zijderveld diagrams are shown in Fig. 6. For the complete analyses, the reader can check the paleomagnetism.org files associated with this paper (check persistent identifier -PID- in the acknowledgements) and Supplementary File SF3.

Mean directions and uncertainties of each component were evaluated using Fisher's statistics (1953) of virtual geomagnetic poles (VGPs). We applied a fixed 45° cut-off to the VGP distributions of each site. In addition, we used the Deenen et al. (2011) criteria to evaluate the scatter of VGPs. As a general rule, if scatter is—mostly—due to paleosecular variation (PSV) of the geomagnetic field, the associated VGP distribution is roughly circular in shape. However, internal deformation, vertical axis rotation or inclination shallowing may add anisotropy to the scatter. In such cases, VGP distributions will show a certain degree of elongation or are otherwise not spherically uniform. Many samples show a NRM component with very low unblocking temperatures and low coercivities (100–180 °C or 10–12 mT). We consider this component as a viscous remanent magnetization (VRM), because of its similarity to the recent field (Fig. 7 and Supplementary File SF3-A and B).

After VRM removal, the samples show a single NRM component (Fig. 6), generally trending to the origin, regardless of the mineral, magnetite (usually fully demagnetized at 40-60 mT and 500-580° C) and/or pyrrhotite (fully demagnetized at 330° C and little to barely demagnetized in AF). This characteristic remanent magnetization (ChRM) clusters well in all the sites (concentration parameter $k > 8$, but generally over 15; Table 2; Supplementary file SF3-B) with the exceptions of sites OG02 and OG08 (Figs. 8 and 9; Table 2). In addition, there are three sites with less than seven samples passing the 45° cut-off (OG05, OG06, OG14) and therefore their statistical parameters are not reliable. These five sites were excluded from further interpretation. The remaining 15 sites show quite variable ChRM declinations and inclinations which appear to be only comparable between sites within the same thrust unit (Figs. 2, 8 and 9, Tables 1 and 2). To account for the different events of deformation we have used bedding corrections (Table 2) and fold tests (Fig. 10). In addition, we also performed inclination only statistics (Enkin and Watson, 1996; Arason and Levi, 2010) to eliminate clustering problems related to vertical axis rotations using both the bedding parameters and our inferred Alpine corrections (see top of section 3) (Tables 1 and 3).

In geographic coordinates sites that passed the quality filter show clusterings that range from k (concentration parameter) ~ 8 (OG10 and 11) to k ~188 (OG19). Site average declinations range from 125° to 297°, the majority of them in the south quadrants with sites OG15 and BN1-OR15 (combined sites separated by 100 m) being the only exceptions (Fig. 8 and 9; Table 2). Inclinations range from -50° to 50°. Bedding correction significantly changes the distribution of the site averages, but the scattering in declinations (from 111° to 289°) and inclinations (-65° to 56°) remains (Table 2), which means that magnetization timing is not the same for all samples and/or structural complications are larger than folding.

All fold-tests whose samples passed the aforementioned quality criteria (Fig. 10; OG03-04; OG11; OG13 and OG19) were performed in folds with weakly plunging axes (Table 1; Fig. 11) with the exception of OG11, which in turn is the only one that is not negative (Fig. 10). OG11 shows a better clustering (τ1) after tilt correction, however, the fold-axis in site OG11 is steeply plunging (the only case; Fig. 10). The performed fold-tests restore deformation as if axes were horizontal. Steeply plunging axis' folds, therefore usually yield false positives and negative foldtests (e.g. Pueyo et al., 2016a). A possibility to decipher the magnetization timing is pre-correcting the plunge of the fold axis before the fold-test. The declination results, in this case, will bring spurious rotations and wouldn't be trustworthy. After back-tilting the plunging-axis in OG11 (azimuth/plunge = 005/42), the fold test remains indeterminate, in this case with a greater clustering (τ1) before tilt correction (Figs. 10 (structural correction panel) and 11). The statistics of all sites that pass our quality criteria ($n \geq 7$ and $k > 8$) yield close to random distributions both considering all specimens ($k = 1.63$ and $K = 1.65$; Supplementary File SF3-A) and the mean of site averages ($k = 1.95$ and $K = 2.5$; Table 4). As expected from the negative within-site fold-tests, the concentration parameter does not change after bedding correction neither in all specimens together ($k = 1.51$ and $K = 1.59$; Supplementary File SF3) nor the mean of site averages ($k = 1.57$ and $K = 1.64$; Table 4).

Inclination only statistics are independent to differential vertical axis rotations since declinations are not taken into account (e.g. Enkin and Watson, 1996). Inclination only statistics were performed on site averages to avoid weighting based on number of specimens (Table 4). The concentration parameter (k) equals to 0 in geographic coordinates and 2.17 in tilt corrected coordinates, both figures representing very poor clusterings. k becomes close to 4 if we do not account for OG3 and OG4, which follow a different trajectory and may represent a different magnetization event (see explanation below). In contrast, when correcting the studied samples exclusively for our inferred

Alpine tilt, the inclination only concentration parameter is ~4, but becomes ~14 when excluding OG3 and OG4 (Table 4). An inclination only tilt test without OG3 and OG4 shows a best fit for a 110% correction, both using the Enkin and Watson, 1996 approach (with a maximum clustering around k ~12) and a stepwise untilting following Arason and Levi (2010) inclination only statistics with a maximum at k ~15 (Fig. 12). OG3 and OG4 share a common true direction in geographic coordinates and their Alpine tilt correction does not change them too much (Fig. 13)

3.5 Anisotropy of the Magnetic Susceptibility (AMS) results

We measured the anisotropy of the magnetic susceptibility in 148 samples from most sites to explore possible causes for the variety of ChRM directions found (Table 2). The degree of anisotropy (P) appears to be generally low (< 1.05; Fig. 14, Supplementary file SF5) although some individual samples showed up to 1.5. The samples' three principal ellipsoid axes (Kmax, Kint, and Kmin) mimic the bedding (S0, Kmax-Kint fall within the bedding plane and Kmin is perpendicular) in six sites (OG09, 15, 16, 17, 18, 19); in three samples the AMS ellipsoid corresponded to the foliation S1 (OG06, 10 and 13); and others showed a quasi-random pattern (OG03, 05, 11, 14) both in geographic and tectonic coordinates (Fig. 14). No observed AMS fabric (not Kmax, Kint, or Kmin axes) from the datasets studied coincides with the ChRM directions, suggesting that NRM and ChRM are not significantly biased by rock fabric.

4. Discussion

The paleomagnetic and rock magnetic results obtained from the Silurian-Devonian limestones in the Pyrenees certify that the Paleozoic rocks from this mountain belt have been subject to at least one widespread remagnetization event. Many samples contain a VRM that is similar to the recent geoaxial dipole for recent times in the Pyrenees (Fig. 7). Apart from this VRM, all rocks, regardless of their magnetic mineralogy, show a single stable component heading to the origin, with the exception of samples not delivering results (Fig. 6 and 8). This component is not deviated toward bedding/cleavage planes (as inferred from AMS patterns, Fig. 14) and displays negative fold tests (Fig. 10; perhaps a syn-folding remagnetization in the case of site OG11 where fold test results are indeterminate).

4.1 Rock magnetism

Rock magnetic analyses show that both pyrrhotite and magnetite are the magnetic carriers in the Silurian-Devonian limestones whereas magnetite is the carrier in the Panticosa late Carboniferous-Permian granite (OG1) and sampled dyke (OG12dyke). All limestone sites contain variable amounts of pyrrhotite and magnetite as shown both in thermomagnetic curves (Fig. 3), in IRM acquisition curves and during NRM demagnetization (Figs. 5 and 6). We applied the IRM end-member modeling technique in an attempt to discriminate between different remagnetization events in the Pyrenees. The two end-member model with a reasonably high r^2 value of 0.65 is our preferred model. Models with 3 to 9 end members evidently show slightly better fits ($r^2 = 0.73$ to 0.88 respectively). However, neither the fit improves significantly, nor the shape of the end members shows more or less anticipated IRM acquisition curves for any particular mineralogy (Fig. 5B). In addition, most of the additional end members seem to represent the variable coercivity windows of magnetite (e.g. the 4 endmember solution in Fig. 5B: three of the endmembers (EM1-3) represent magnetite and do not deliver any particular meaningful result). All samples contain a significant amount of those additional endmembers (varying from 10% to 60%) indicating that a variable grain-size or compositional (Ti-)magnetite is present in virtually every sample. The two end-member model further distinguishes a 42 mT component with a DP = 0.36 (C1), which is typical for magnetite and a 200mT component and DP = 0.35 (C2), which we interpret as pyrrhotite (Fig. 5C). The two end members are in agreement with individual sample fits, but end-member IRM acquisition curves describe much better the IRM properties of each magnetic phase (Fig. 5). We interpret the soft component (C1) as magnetite varying from coarse to very fine grained (i.e. lower and higher coercivity respectively) as supported by hysteresis loops (Fig. 4 and SF2). It is reasonable that the high coercivity component (C2) reflects the observed pyrrhotite in the thermomagnetic curves as SD pyrrhotite has a rather high coercivity (Dekkers, 1989).

The presence of variable amounts of pyrrhotite and magnetite in all Silurian-Devonian samples studied suggests that this is a common feature for the Paleozoic sedimentary and metasedimentary units of the mountain belt. Similar to the Pyrenees, pyrrhotite is the most common magnetic carrier in other limestone formations of the Iberian Variscides heavily affected by late Carboniferous magmatism (Pastor-Galán et al., 2015a; 2016; 2017; Fernández-Lozano et al., 2016).

In general terms, the occurrence of pyrrhotite in limestones is a sign of their remagnetization. Pyrrhotite is a frequent secondary mineral which is formed in low-grade metamorphic rocks under reducing conditions (Crouzet et al., 2001; Aubourg et al., 2012, 2019; Izquierdo-Llavall et al., 2020) or in the presence of non oxidizing magmatic fluids (Pastor-Galán et al., 2016). The ocurrence of pyrrhotite has been used as a geothermometer; increasing burial enhances the transformation of magnetite to pyrrhotite and the progressive replacement of magnetite-carried magnetizations by pyrrhotite-carried remagnetizations (e.g. Aubourg et al., 2019). In clay-rich rocks, magnetite and pyrrothite coexist at burial temperatures <340 ºC whereas at ~350 ºC the concentration of magnetite decreases drastically and pyrrothite becomes the dominant magnetic mineral (Aubourg et al., 2019). Izquierdo-Llavall et al. (2020) estimated the peak temperatures (~350 – 450 °C) in the

North Pyrenean Zone (between the northermost sides of cross sections 2a and 2b in Fig. 1) following the magnetite –pyrrhotite transformations. In contrast, the temperature estimates for the sampled area, in the southern Pyrenees (Izquierdo-Llavall et al., 2013; Labaume et al., 2016) are much lower (below ~300 ºC) and the burial/thermal effect in the formation of pyrrothite is expected to be less important. Taking that into account, we suggest that in our samples pyrrhotite could be mainly formed by a fluid induced chemical remagnetization during the latest stages of the Variscan orogeny. However, our westernmost sampled units (OG1-OG4) may have surpassed the Curie temperature of pyrrhotite (~320° C; e.g. Dekkers, 1989) during the Cenozoic burial (Bosch et al., 2016), and could, therefore, carry a Cenozoic TRM.

In pyrrhotite, the magnetic easy direction is confined to the basal crystallographic plane which implies an intrinsically strong anisotropy because of the 'hard' crystallographic c-axis (Schwarz and Vaughan, 1972; Schwarz, 1974). When pyrrhotite grows oriented in a preferred fabric (e.g. S1 or S0), the direction of the magnetic remanence can be biased towards the fabric plane (Fuller, 1963). The studied samples occasionally show pressure solution cleavage and a widespread presence of pyrrhotite as a partial or main carrier of the NRM. AMS fabrics revealed that pyrrhotite is not oriented according to the S1 fabric, and therefore preclude major biases in the magnetic remanence of such secondary sulfides. Besides, although our AMS results are frequently consistent with bedding (S0), the magnetic remanence is not contained within bedding planes (Fig. 14; Table 2) which also impedes a deviation of the ChRM towards S0.

4.2 Paleomagnetism and timing of remagnetization

We only considered sites for interpretation with at least 7 specimens passing the VGP's 45° cut-off criterion and with the concentration parameter k > 8. Sites OG02, 05, 06, 08, 14 and the dyke in OG12 do not pass these criteria and are not considered for further interpretation (Table 2). The rest of the sites show k values that range from barely above 8 in geographic coordinates (OG10 and 11), which could be expected also from detrital magnetizations, to over 40 (OG12, OG15, OG17-OG19) in geographic coordinates, which is unlikely in primary magnetizations of sediments (e.g., Deenen et al., 2011). Site averages point generally to ESE-WSW with both positive and negative inclinations in geographic coordinates with the only exceptions of OG15 and BN1-OR15 which point WNW (Fig. 8 and 9; Table 2), something that does not change after bedding correction. With the exception of OG11, we have documented post-folding magnetizations. Importantly, the fabrics do not show consistency with NRM directions (AMS fabric coincident with S1 does not occur and ChRMs are not within bedding planes) suggesting that any potential bias caused by a preferred orientation of pyrrhotite particles is not significant. Therefore, a post Variscan folding (i.e. late Carboniferous) is the oldest possible age for the magnetization since no earlier folding event has been described in the Pyrenees. The OG11 fold, which has a steep plunging axis (Fig. 11), yielded an inconclusive fold-test, but shows better clustering after tilt correction (Fig. 10). Classical fold-tests assume horizontal axes and performing them in steeply plunging axis' folds is unreliable (e.g. Pueyo et al., 2016a) In the case of OG11, pre-correcting the fold axis plunge produces another inconclusive result (Fig. 10). However, declinations in geographic, standard bedding correction, and Alpine tilt correction remain around 200° (Table 2 and 3) and inclinations are in all cases relatively shallow (between -9 and 20), suggesting that OG11 acquired its NRM during a reversed chron during a time when Iberia was at equatorial latitude: during late Carboniferous or Permian times regardless of the result of the foldtest (e.g. Pastor-Galán et al., 2018).

When considering all site paleomagnetic directions, we found that declinations and inclinations are only compatible in geographic coordinates for those sites within the same tectonic unit (Figs. 1, 2, 8, 9; Table 2). Such a directional pattern may be indicative of (a) different timing of NRM acquisition for each tectonic unit, (b) differential vertical axis rotations between units, (c) post-magnetization differential tilting between units or (d) a combination of the previous processes. To distinguish between these options inclination only statistics are appropriate (e.g. Enkin and Watson, 1996). Inclination only statistics do not consider declinations and therefore are independent of variations due to differential vertical axis rotations. In order to evaluate potential timing of magnetization we performed statistical analyses in geographic coordinates, after bedding correction, and also after correction of the tilt related to the emplacement of Alpine basement thrusts (Figs. 2, 12, 13; Tables 3 and 4). The concentration parameter of inclination data (k) is 0 in geographic coordinates (Table 4), which could mean that (i) sites magnetized at significantly different geological times when Iberia was at very different latitudes, and/or (ii) Alpine tilting postdates the magnetization and therefore it has a strong influence on the inclinations. Inclination only k is still too low (minimum k ~ 8 to consider an acceptable clustering) after bedding correction (~2) but also after Alpine tilt correction (~4).

Two sites from the Gavarnie thrust unit (OG03 and OG04; Fig. 8 and 13, geographic coordinates) move in a very different direction both during bedding and Alpine tilt corrections. In contrast, OG01 (Panticosa granite, late Carboniferous-Permian, Denèle et al., 2012), which is in the same Gavarnie thrust unit, correlates well with the rest of the sites in all thrust units after Alpine tilt correction but not with the neighboring OG03 and OG04. Thus, those sites might have acquired their magnetization at a significantly different time than the rest. After removing OG03 and OG04, the inclination only concentration parameter still indicates poor clustering after bedding correction (k ~ 4). However, when correcting only for the Alpine tilt k becomes 14.42 and a positive inclination only tilt test with a maximum in a 110% untilting is obtained (95% between 58 and 150; Fig. 12; Table 4). After the Alpine tilt correction, the mean inclination is 5° ± 11 (Table 4) and all included sites show SW to SE declinations (Fig. 12). Despite the positive result, our Alpine tilt correction should be taken cautiously. We considered only regional tilt values, which were inferred from the average orientation of the overlying Mesozoic units and thrust slopes in geological maps and cross-sections (Fig. 2). Our estimated values took into account the kilometric-scale, thrust-related folding of the basement but can not consider the potential contribution of Alpine, outcrop-scale folding of the Paleozoic strata.

We believe, however, that the inclination only k value of 14.42 together with the obtained shallow inclinations and southerly declinations are sufficiently convincing to argue for a common timing of NRM acquisition for the samples included in the tilt test (Fig. 12). The results imply a postfolding but a pre-Alpine tilt NRM. The shallow inclinations suggest that Iberia was located at equatorial latitudes and the southerly declinations suggest that this occurred during a reverse chron. We therefore suggest that all samples that passed the quality criteria, with the exception of sites OG03 and OG04, magnetized during the latest Carboniferous to middle Permian times during the Kiaman reverse superchron, when Iberia was indeed located at equatorial latitudes (Weil et al., 2010). The late Carboniferous and early Permian times in the Pyrenees are characterized by widespread intrusions and volcanism (Panticosa granite, for example, OG01; Gleizes et al., 1998). We hypothesize that the remagnetization mechanism in the Pyrenees was triggered by fluids associated to the magmatic activity analogously to the remagnetizations observed in the Central Iberian Zone of west Iberia (e.g. Fernández-Lozano et al., 2016; Pastor-Galán et al., 2016; 2017).

To further test the hypothesis of a post-Variscan but pre-Alpine orogeny remagnetization with the exception of OG03 and OG04, we have plotted the paleolatitudes and declinations of OG sites compared to the Global Apparent Polar Wander Path (GAPWaP) of Torsvik et al. (2012) rotated to Iberian coordinates (Pastor-Galán et al., 2018). Paleolatitudes of OG sites show a good fit and variable clockwise rotations with respect to the declinations expected in Iberia for Carboniferous and early Permian times after Alpine tilt correction, with the exception of OG03 and 04 (Fig. 15 A). Whereas, both paleolatitudes and declinations are scattered in geographic coordinates, in contrast to the expected general good fit if remagnetization had happened after the Alpine orogeny, when Iberia was tectonically stable (Fig. 15 B). In geographic coordinates only three sites fit in both paleolatitude and declinations with the post-Alpine GAPWaP segment: OG3, OG4, and OG16. However, we interpret OG16 as a Permian remagnetization since it fits much better with its neighboring sites OG17, 18 and 19 after the correction of the alpine tilt.

OG03 and OG04 show a negative fold test (Fig. 10) and a common true mean direction in geographic coordinates (Fig. 13). Their paleomagnetic direction is, however, significantly different from OG01 that is a late Carboniferous-early Permian site located in the same thrust sheet. OG03 and OG04 show declinations to the south (both in geographic coordinates and after Alpine tilt correction) and upward inclinations of -43° and -48° (geographic coordinates) or -60° and -68º (after the restoration of the inferred Alpine tilt respectively). Both geographic and Alpine tilt corrected data indicate a remagnetization when Iberia was located at latitudes between 25° and 50° during a reverse chron (Fig. 15A, B). OG03 and 04 results fit best with a remagnetization that postdates the Cretaceous normal superchron (e.g. Izquierdo-Llavall et al., 2015 and references therein) (Fig. 15). Inclinations >60° (i.e. after Alpine tilt corrections) would only be possible when Iberia was located at similar latitudes as at present, fairly long after the Alpine orogeny which ended at Miocene times. In such case, no correction at all should be applied. Therefore, the magnetization has to be syn- to post Alpine tilting to achieve a good paleolatitudinal fit. Most structural units just above the basement units are remagnetized by tardi-orogenic burial remagnetizations (post-, syn- and pre-Alpine folding) that affected all kinds of rocks (limestones, calcarenites, redbeds...) (Dinarès et al., 1992; Dinarès, 1994; Keller et al., 1994; Oliva-Urcia and Pueyo, 2007b; Izquierdo-Llavall et al., 2015, Mujal et al., 2017 etc.) In addition, the burial temperature of OG03 and OG04 could have been sufficiently elevated to trigger a thermal remagnetization after tilting during the unroofing and exhumation of the Axial Zone. With all this elements, we tentatively favor a late orogenic (Eocene) age which would allow inclinations around 50° (close to those obtained in geographic coordinates; Fig. 15B).

4.3 Tectonic significance

The hypothesis of a detachment and northward drift of a peri-Gondwana microntinent (Armorica s.l.) during the late Silurian or early Devonian (e.g. Torsvik et al., 2012; Stampfli et al., 2013; Domeier and Torsvik., 2014; Franke et al., 2017) is grounded largely on the basis of paleomagnetic data from the Silurian and Devonian rocks of the Pyrenees (Tait et al., 2000), Brittany (Tait et al., 1999) and Bohemian Massif (Tait et al., 1994). Our results show that pervasive remagnetizations have affected the Silurian-Devonian limestones of the Pyrenees during, at least, two episodes: late Carboniferous-Early Permian and at the end of the Alpine orogeny. Our site BN1-OR15 was collected in the vicinity of those from Tait et al. (2000) and shows the same direction in geographic coordinates (Fig. 9), but within-site clustering worsens after bedding correction (Table 2). After Alpine tilt correction, however, a very good fit results with the majority of our Silurian-Devonian collection. In addition, site BN1-OR15 contains pyrrhotite (Fig. 3), a feature common to all the other samples studied (Fig. 9) and a secondary mineral in (meta)sediments indicative of remagnetization (e.g. Pastor-Galán et al., 2017; Izquierdo-Llavall et al., 2020). We therefore conclude that the originally published data by Tait et al. (2000) also reflect a remagnetization. Since the other paleomagnetic results from the putative Armorican s.l. continent are similar to those from the Pyrenees and come from areas with intense Carboniferous deformation and enhanced thermal activity, we suspect that they could be remagnetized as well. We request using those paleolatitudes (Tait et al., 1994; 1999) with caution; their paleomagnetic veracity warrants to be reassessed.

Despite the inherent loss of information due to the remagnetization, especially regarding the potential paleolatitudinal constraints, the Silurian-Devonian rocks of the Pyrenees do provide interesting insights. Based on the inclination data we interpret that the sampled rocks mostly remagnetized during late Carboniferous and Permian times when Iberia was located around the equator, thus previous to the Alpine orogeny. Our paleomagnetic results show a positive inclination only fold test when correcting the Alpine tilt inferred from cross-sections and geological maps but negative outcrop scale fold tests when using the bedding parameters. Thus, paleomagnetism in combination with detailed structural analysis, is a reliable tool to unravel the deformation style of multi-phase orogens like the Pyrenees. With our paleomagnetic data we now can separate the effects of Alpine and Variscan orogeny in the Silurian-Devonian carbonate series, something that classically is deemed challenging (e.g. Casas et al., 2019). With our data we can say that the Variscan orogeny was responsible for the main folding event observed in the Silurian and Devonian rocks since all remagnetizations are post-folding (Figs. 10 and 12).

After the Alpine tilt correction, we obtained a relatively good agreement in inclinations, but declinations are still really scattered from SE to SW. Alpine vertical axis rotations in the Pyrenees are frequent and very variable with magnitudes ranging from a few degrees to up to 80° both clockwise and counterclockwise (e.g. Sussman et al., 2004; Rodríguez et al., 2016). Although it is plausible that the basement also underwent significant vertical axis rotations, very little is known about the rotational activity of Pyrenean basement thrusts during the Alpine orogeny. The declinations observed in the Silurian-Devonian rocks of the Pyrenees are generally clockwise rotated with respect to the Permian reference pole for stable Iberia (Weil et al., 2010; Oliva et al., 2012): These rotations range from a few degrees to ca. 90° (Fig. 1). We note that such results are also in line with the paleomagnetic results from Carboniferous and Permian igneous rocks in the Pyrenees and Catalan coastal ranges (Edel et al., 2018). The Pyrenees lay in the northern branch of the Cantabrian Orocline, which rotated clockwise during the Late Carboniferous and Early Permian (e.g. Pastor-Galán et al., 2015b; Pastor-Galán, 2020). Edel et al. (2018) interpreted their results as consistent with the rotations expected in the northern branch of the Cantabrian Orocline. Izquierdo-Llavall et al. (2014) also found similar data in late Carboniferous and Early Permian rocks of the Pyrenees and interpreted them as an Alpine rotation. We would like to remain wary about their meaning since the variety of rotations found might be reflecting: (i) Differential timing of the remagnetization which occurred widely during the Cantabrian Orocline formation as observed in other areas of Iberia (e.g. Pastor-Galán et al., 2017; 2020); (ii) Vertical axis rotations associated with the Alpine orogeny (Izquierdo-Llavall et al., 2014); or (iii) a combination of both processes where the Alpine rotations may be opposite to and/or in the same sense as the late Carboniferous clockwise rotations (Fig. 15).

4.4 A blessing in disguise

The Pyrenees are a multi-orogenic mountain range whose kinematics is often complicated due to the superposition of different deformation events. Accumulation of geological processes, many of them involving relatively high temperatures and fluid percolation increases the chances of remagnetization for the rocks involved in the orogenies. In fact, we suspect that the majority of the Silurian-Devonian carbonate series of the Pyrenees won't preserve any primary and syn-sedimentary magnetization. This makes the Pyrenees, despite the great outcrop quality and quantity, a bad candidate to study pre-Variscan plate motions and kinematics. However, we found generally strong magnetizations containing univocal ChRMs, which makes paleomagnetism of Paleozoic series in the Pyrenees useful for post-Variscan tectonic studies. Our paleomagnetic data, in combination with detailed structural observations, has proven an efficient way to unravel the complex tectonic evolution of the Axial Zone of the Pyrenees. We think that at least the Silurian-Devonian carbonate rocks, but likely other Paleozoic series and igneous rocks are excellent targets to study: (1) the Variscan - Alpine structural relationships, (2) the Alpine rotational history of the basement thrusts; and, perhaps, (3) the late Variscan deformation events leading to the final amalgamation of Pangea.

5. Conclusions and caution for paleomagnetists

- The Silurian-Devonian carbonate series of the Pyrenees show varying amounts of pyrrhotite, a secondary magnetic mineral, and negative fold tests using bedding parameters, which indicate widespread remagnetization(s).
- The majority of sites that passed the quality criteria (n >= 7 and k > 8) show a positive inclination only fold test when correcting the alpine tilt (with the exceptions of OG03 and OG04). The obtained inclinations are southerly and very shallow, constraining the remagnetization to a reverse chron when Iberia was around the equator, only possible during late Carboniferous or early Permian times.
- Sites OG03 and OG04 (Western Axial Zone, Gavarnie thrust sheet) were likely remagnetized after the main Alpine thrusting, during a pervasive burial remagnetization widely observed in the Internal Sierras and other along-strike equivalent units (Bóixols, Cadí).
- Paleomagnetism from the Silurian and Devonian rocks suggests that the Variscan orogeny was responsible for their main folding event, whereas the Alpine orogeny produced their thrusting and antiformal stacking.
- Our results also show general clockwise rotations which may be consistent with the northern branch of the Cantabrian Orocline. These rotations may as well represent Alpine vertical axis rotations or a combination of both.
- Given the generally good paleomagnetic quality of the Devonian carbonates, they could be targeted to study the Alpine imprint on Paleozoic rocks and thus, to unravel the rotational history of basement thrusts.

- The widespread remagnetizations found in the Paleozoic of the Pyrenees indicate that paleolatitudes inferred for Silurian and Devonian times from the studied rocks are very unlikely original and should be taken very cautiously. We urge a reassessment of Siluro-Devonian poles from the Variscan in Europe.
- Paleomagnetism from multi-orogenic areas is NOT A SIMPLE GAME. However, the systematic combination of paleomagnetism with detailed structural observations, seems to be a foremost way to unravel complex tectonic evolutions.

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Data availability statement

All data is included in the paper and supplementary materials. In Addition, paleomagnetic data is stored in paleomagnetism.org under the persisitent identifier (PID)

1871091757a6ef9d46ac59bb9f35c7a6387bc8fcece7bf0715eaba29164fbc7e and can be accessed in the link https://www.paleomagnetism.org/library/.

Figure and Table captions

Figure 1: A) Simplified map of the Iberian Peninsula showing the main Paleozoic outcrops and the areas affected by the Alpine orogeny (after Pastor-Galán et al., 2020). B) Geological map of the Pyrenees (modified from Barnolas et al., 2008 according to Choukroune and Seguret, 1973) showing our sampling locations (red dots) and the extensive paleomagnetic studies in the Pyrenees focused mainly on Permo-Triassic rocks (blue dots). Lines show the trend of the cross sections in figure 2.

Figure 2: Cross sections through the Pyrenees with projected positions of our sampling sites. The cross-section in a) is from Teixell (1996) whereas cross-sections in b) and c) are from Martínez-Peña and Casas-Sainz (2003) and Muñoz (1992), respectively.

Figure 3: Selected magnetization vs. temperature curves (OG8, OG13, OG14) and susceptibility vs. temperature (BN1). Note that magnetic and non magnetic sulfides are common. All measurements performed are available in supplementary file SF1.

Figure 4: Selected slope corrected hysteresis loops and FORC diagram (plotted with FORCINEL (Harrison and Feinberg, 2008, smoothing factor 13). All measurements performed are available in supplementary file SF2.

Figure 5: A) Unmixing of IRM acquisition curve for three samples showing different proportions of a ‘soft’ mineral that saturates below 75 mT (magnetite) and a ‘harder’ one that saturates over 200 mT (pyrrhotite). B) Results from the endmember modeling showing our preferred two end-

member solution (left) and the four end-member solution (right). C) Unmixing of the IRM synthetic acquisition curve from the two end members showing the same two magnetic mineralogies as in the forward modeling of different samples: magnetite to the left and pyrrhotite to the right.

Figure 6: Examples of ‘Zijderveld’ vector-end point plots (Zijderveld, 1967) for selected samples. All samples plotted in geographic coordinates. Close-open circles represent declination and inclination projections respectively. Complete analyses are available in paleomagnetism.org through the Persistent identifyer PID given in the acknowledgements.

Figure 7: Viscous remanent magnetization (VRM) from all samples is compatible with the present-day field (geographic coordinates). Red dots are those that fall outside of the 45° cut-off. Uncertainty envelope is in both cases VGP A95. The rather large scattering is likely due to the small number of demagnetization levels containing the VRM (3-4) and the possible migration of the VRM during transport, storage and analysis.

Figure 8: Directional and VGP results in geographic coordinates of sites OG01 to OG11. Uncertainty envelope is in both cases VGP A95. Red dots are those that fall outside of the 45° cut-off. Sites OG02, 05 and 08 did not provide statistically meaningful results and were not interpreted.

Figure 9: Directional and VGP results in geographic coordinates of sites OG12 to OG19 and BN1-OR15. Uncertainty envelope is in both cases VGP A95. Red dots are those that fall outside of the 45° cut-off. Sites OG02, 05 and 08 did not provide statistically meaningful results and were not interpreted.

Figure 10: Within-site fold tests. All are negative but OG11, which is inconclusive.

Figure 11: Pi diagrams for the studied folds. Only OG11 shows a steeply plunging axis.

Figure 12: Results after the inferred Alpine tilt correction. Permian reference declination for Iberia is after Weil et al. (2010). The results show a positive inclination only tilt test following the methodology of Enkin and Watson (1996) (selected bootstraps in thin gray lines) and Arson and Levi (2010) approach (dashed line).

Figure 13: OG03 and OG04 show a common true mean bootstrapped direction (after Tauxe, 2010) in geographic coordinates (and Alpine tilt corrected, since it is the same for both).

Figure 14: Results from the anisotropy of the magnetic susceptibility analyses. Magnetic fabrics represent bedding or S1 cleavage. Magnetic fabric directions do not coincide with the paleomagnetic directions, which allows us to discard an internal deformation control of the paleomagnetic remanence.

Figure 15: Figure showing paleolatitudes (up) and declinations (down) of OG sites compared to the GAPWaP of Torsvik et al. (2012) rotated to Iberia (following Pastor-Galán et al., 2018). A) After Alpine Tilt B) Geographic coordinates.

Table 1: Site Location and key structural data from each site.

Table 2: Paleomagnetic results for ChRM component for all sites in geographic and tilt (bedding) corrected coordinates.

Table 3: Paleomagnetic results (ChRM) after the Alpine tilt correction (tilt associated to the emplacement of the thrusts).

Table 4: Concentration parameters of inclination only statistics from the mean values of the sites.

Supplementary Files:

SF1: All thermomagnetic curves analyzed.

SF2: All Hysteresis loops analyzed.

SF3: A) Synthesis of the paleomagnetic results (can be opened with paleomagnetism.org). B) Extra plots and table showing the mean VRM per site and the mean ChRM per site in tectonic coordinates.

SF4: KMZ file (Google Earth) with the sampled locations.

SF5: Graphics showing all results of the AMS analyses.

SF6: Zip file containing all obtained raw results.

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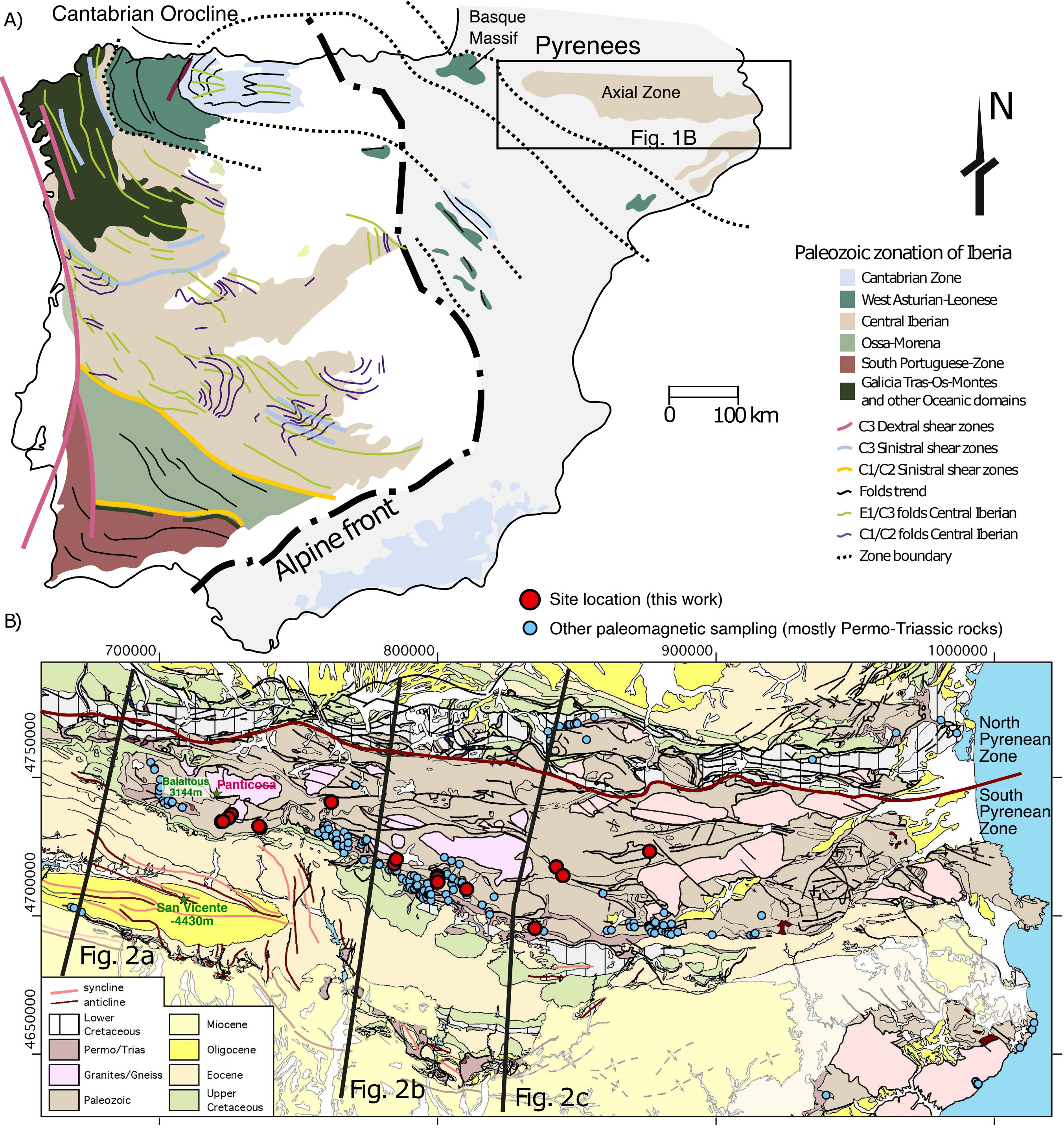

A)
Cantabrian Orocline
Basque Massif
Pyrenees
Axial Zone
Fig. 1B
N
Paleozoic zonation of Iberia
Cantabrian Zone
West Asturian-Leonese
Central Iberian
Ossa-Morena
South Portuguese-Zone
Galicia Tras-Os-Montes and other Oceanic domains
C3 Dextral shear zones
C3 Sinistral shear zones
C1/C2 Sinistral shear zones
Folds trend
E1/C3 folds Central Iberian
C1/C2 folds Central Iberian
Zone boundary
0
100 km
Alpine front
B)
Site location (this work)
Other paleomagnetic sampling (mostly Permo-Triassic rocks)
700000
800000
900000
1000000
4750000
4700000
4650000
North Pyrenean Zone
South Pyrenean Zone
Balaitous 3144m
Panticosa
San Vicente -4430m
Fig. 2a
Fig. 2b
Fig. 2c
syncline
anticline
Lower Cretaceous
Permo/Trias
Granites/Gneiss
Paleozoic
Miocene
Oligocene
Eocene
Upper Cretaceous

Figure 1: A) Simplified map of the Iberian Peninsula showing the main Paleozoic outcrops and the areas affected by the Alpine orogeny (after Pastor-Galán et al., 2020). B) Geological map of the Pyrenees (modified from Barnolas et al., 2008 according to Choukroune and Seguret, 1973) showing our sampling locations (red dots) and the extensive paleomagnetic studies in the Pyrenees focused mainly

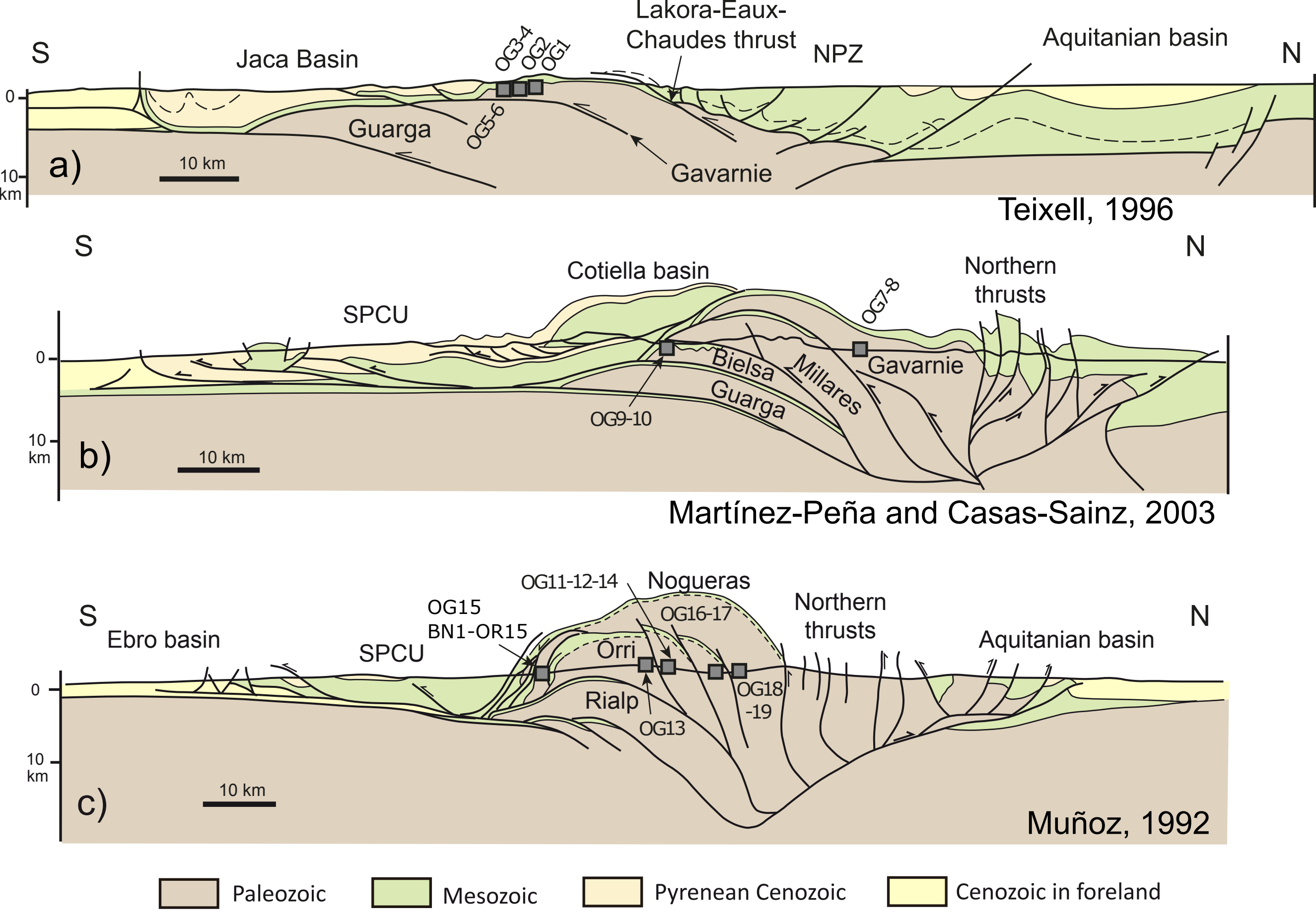

S
Jaca Basin
OG3-4
OG2
OG1
Lakora-Eaux-Chaudes thrust
NPZ
Aquitanian basin
N
0
10
km
a)
10 km
Guarga
OG5-6
Gavarnie
Teixell, 1996
S
N
Cotiella basin
SPCU
OG7-8
Northern thrusts
0
Bielsa
Millares
Gavarnie
Guarga
OG9-10
10
km
b)
10 km
Martínez-Peña and Casas-Sainz, 2003
OG11-12-14
Nogueras
OG15
BN1-OR15
OG16-17
Northern thrusts
S
N
Ebro basin
SPCU
Orri
Aquitanian basin
0
Rialp
OG13
OG18
-19
10
km
c)
10 km
Muñoz, 1992
Paleozoic
Mesozoic
Pyrenean Cenozoic
Cenozoic in foreland

Figure 2: Cross sections through the Pyrenees with projected positions of our sampling sites. The cross-section in a) is from Teixell (1996) whereas cross-sections in b) and c) are from Martínez-Peña and Casas-Sainz (2003) and Muñoz (1992), respectively.

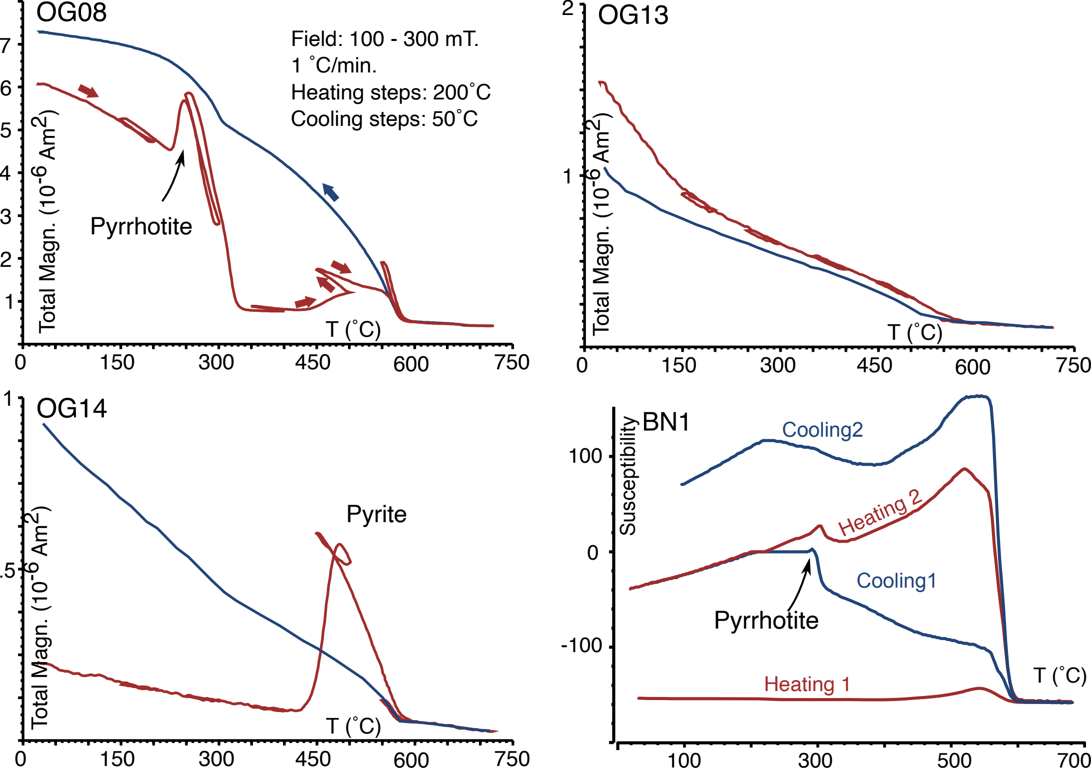
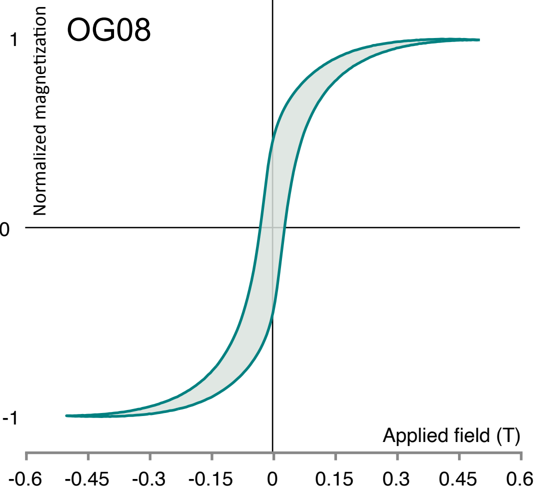
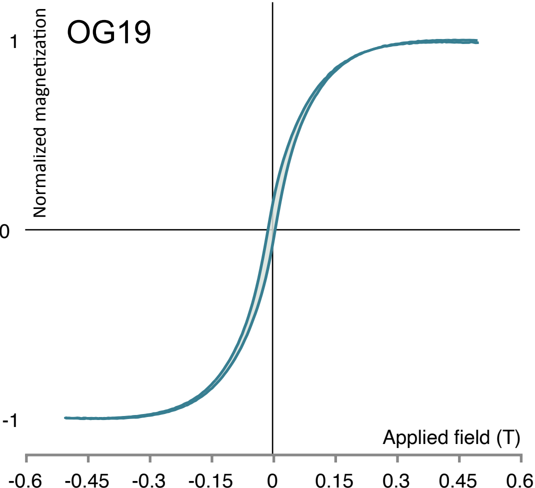

OG08
Field: 100 - 300 mT.
1 ˚C/min.
Heating steps: 200˚C
Cooling steps: 50˚C
Total Magn. (10^{-6} Am^2)
Pyrrhotite
T (˚C)
7
6
5
4
3
2
1
0
150
300
450
600
750
OG13
Total Magn. (10^{-6} Am^2)
T (˚C)
2
1
0
150
300
450
600
750
OG14
Pyrite
Total Magn. (10^{-6} Am^2)
T (˚C)
1
.5
0
150
300
450
600
750
BN1
Susceptibility
Cooling2
Heating 2
Cooling1
Pyrrhotite
Heating 1
T (˚C)
100
0
-100
100
300
500
700

Figure 3: Selected magnetization vs. temperature curves (OG8, OG13, OG14) and susceptibility vs. temperature (BN1). Note that magnetic and non magnetic sulfides are common. All measurements performed are available in supplementary file SF1.


OG08
Normalized magnetization
1
0
-1
Applied field (T)
-0.6
-0.45
-0.3
-0.15
0
0.15
0.3
0.45
0.6


OG19
Normalized magnetization
1
0
-1
Applied field (T)
-0.6
-0.45
-0.3
-0.15
0
0.15
0.3
0.45
0.6

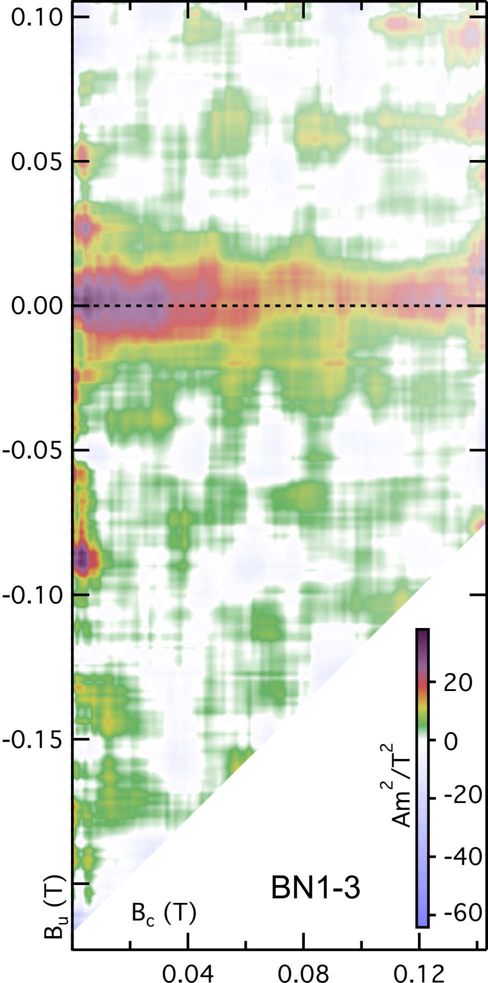

0.10
0.05
0.00
-0.05
-0.10
-0.15
B_u (T)
B_c (T)
0.04
0.08
0.12
BN1-3
Am^2/T^2
20
0
-20
-40
-60

Figure 4: Selected slope corrected hysteresis loops and FORC diagram (plotted with FORCINEL (Harrison and Feinberg, 2008, smoothing factor 13). All measurements performed are available in supplementary file SF2.

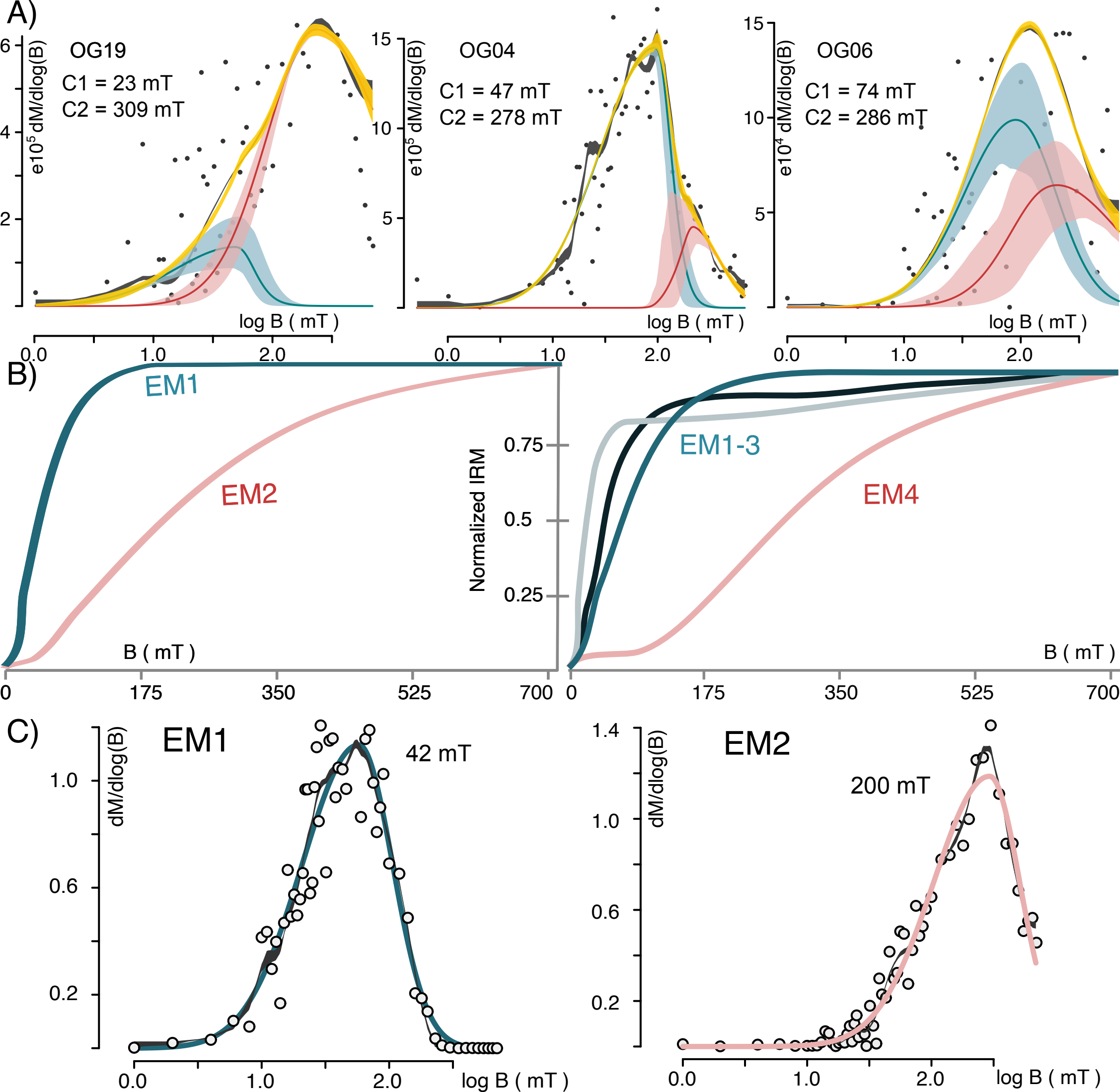

A)
OG19
C1 = 23 mT
C2 = 309 mT
e10^5 dM/dlog(B)
6
4
2
log B (mT)
0.0
1.0
2.0
OG04
C1 = 47 mT
C2 = 278 mT
e10^5 dM/dlog(B)
15
10
5
log B (mT)
0.0
1.0
2.0
OG06
C1 = 74 mT
C2 = 286 mT
e10^4 dM/dlog(B)
15
10
5
log B (mT)
0.0
1.0
2.0
B)
EM1
EM2
B (mT)
0
175
350
525
700
Normalized IRM
0.75
0.5
0.25
EM1-3
EM4
B (mT)
0
175
350
525
700
C)
EM1
42 mT
dM/dlog(B)
1.0
0.6
0.2
0.0
1.0
2.0
log B (mT)
EM2
200 mT
dM/dlog(B)
1.4
1.0
0.6
0.2
0.0
1.0
2.0
log B (mT)

Figure 5: A) Unmixing of IRM acquisition curve for three samples showing different proportions of a ‘soft’ mineral that saturates below 75 mT (magnetite) and a ‘harder’ one that saturates over 200 mT (pyrrhotite). B) Results from the endmember modeling showing our preferred two end-member solution (left) and the four end-member solution (right). C) Unmixing of the IRM synthetic acquisition curve from the two end members showing the same two magnetic mineralogies as in the forward modeling of different samples: magnetite to the left and pyrrhotite to the right.

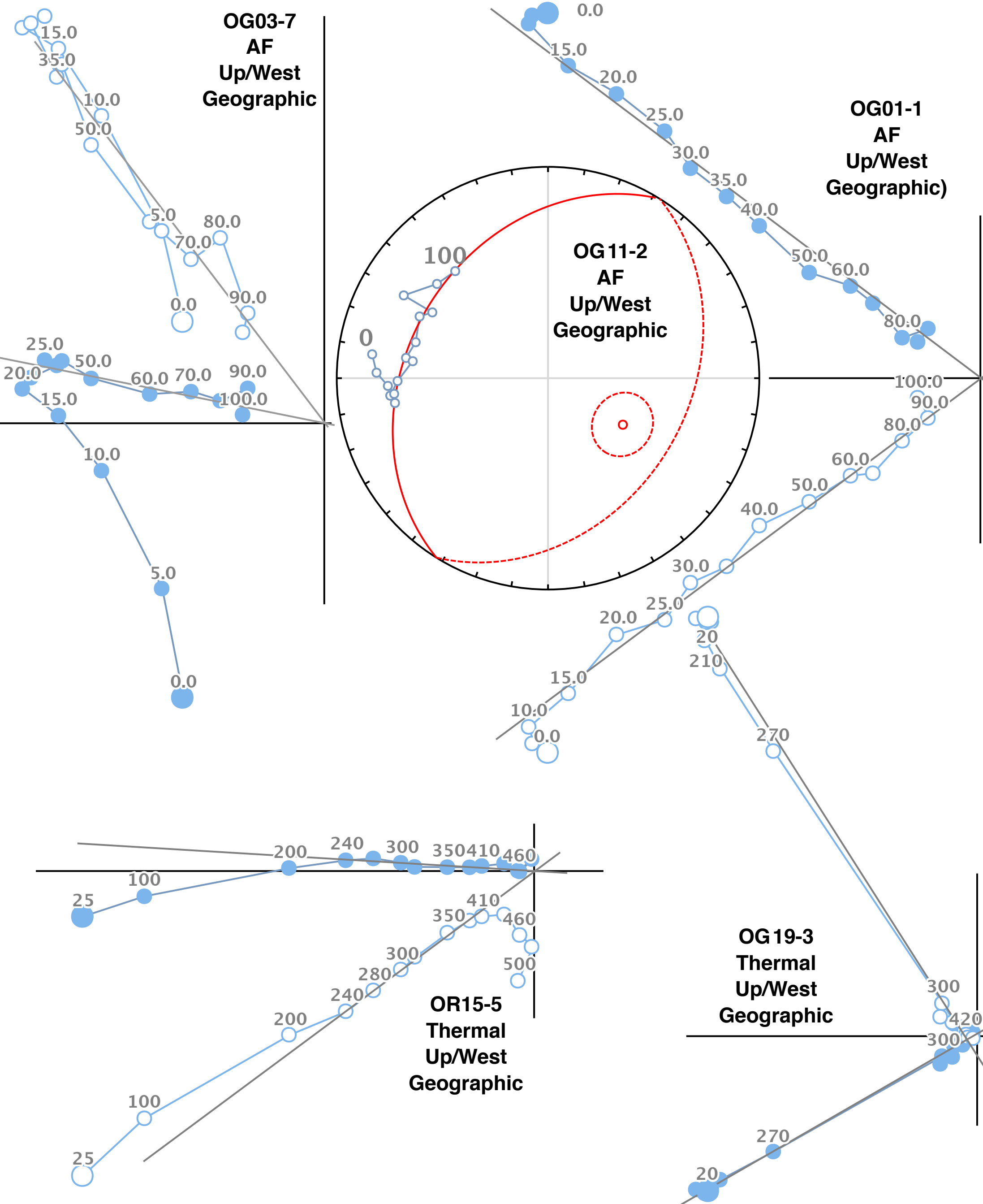

OG03-7
AF
Up/West
Geographic
15.0
35.0
10.0
50.0
5.0
80.0
70.0
90.0
0.0
25.0
50.0
20.0
60.0
70.0
90.0
15.0
100.0
10.0
5.0
0.0
0.0
15.0
20.0
25.0
30.0
35.0
40.0
50.0
60.0
80.0
OG01-1
AF
Up/West
Geographic)
100.0
90.0
80.0
60.0
50.0
40.0
30.0
25.0
20.0
15.0
10.0
0.0
100
0
OG11-2
AF
Up/West
Geographic
20
210
270
300
420
300
270
20
OG19-3
Thermal
Up/West
Geographic
25
100
200
240
300
350
410
460
410
350
460
300
280
240
500
200
100
25
OR15-5
Thermal
Up/West
Geographic

Figure 6: Examples of ‘Zijderveld’ vector-end point plots (Zijderveld, 1967) for selected samples. All samples plotted in geographic coordinates. Close-open circles represent declination and inclination projections respectively. Complete analyses are available in paleomagnetism.org through the Persistent identifyer PID given in the acknowledgements.

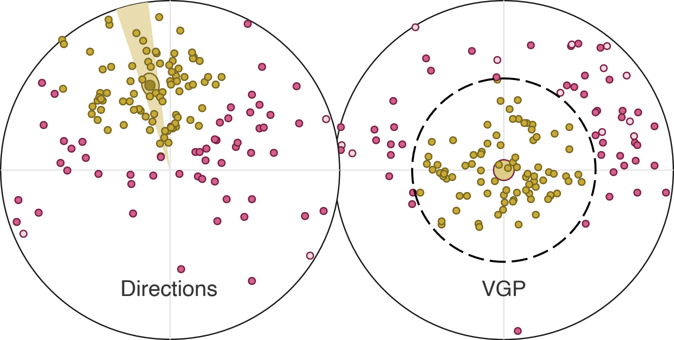

Directions
VGP

Figure 7: Viscous remanent magnetization (VRM) from all samples is compatible with the present-day field (geographic coordinates). Red dots are those that fall outside of the 45° cut-off. Uncertainty envelope is in both cases VGP A95. The rather large scattering is likely due to the small number of demagnetization levels containing the VRM (3-4) and the possible migration of the VRM during transport, storage and analysis.

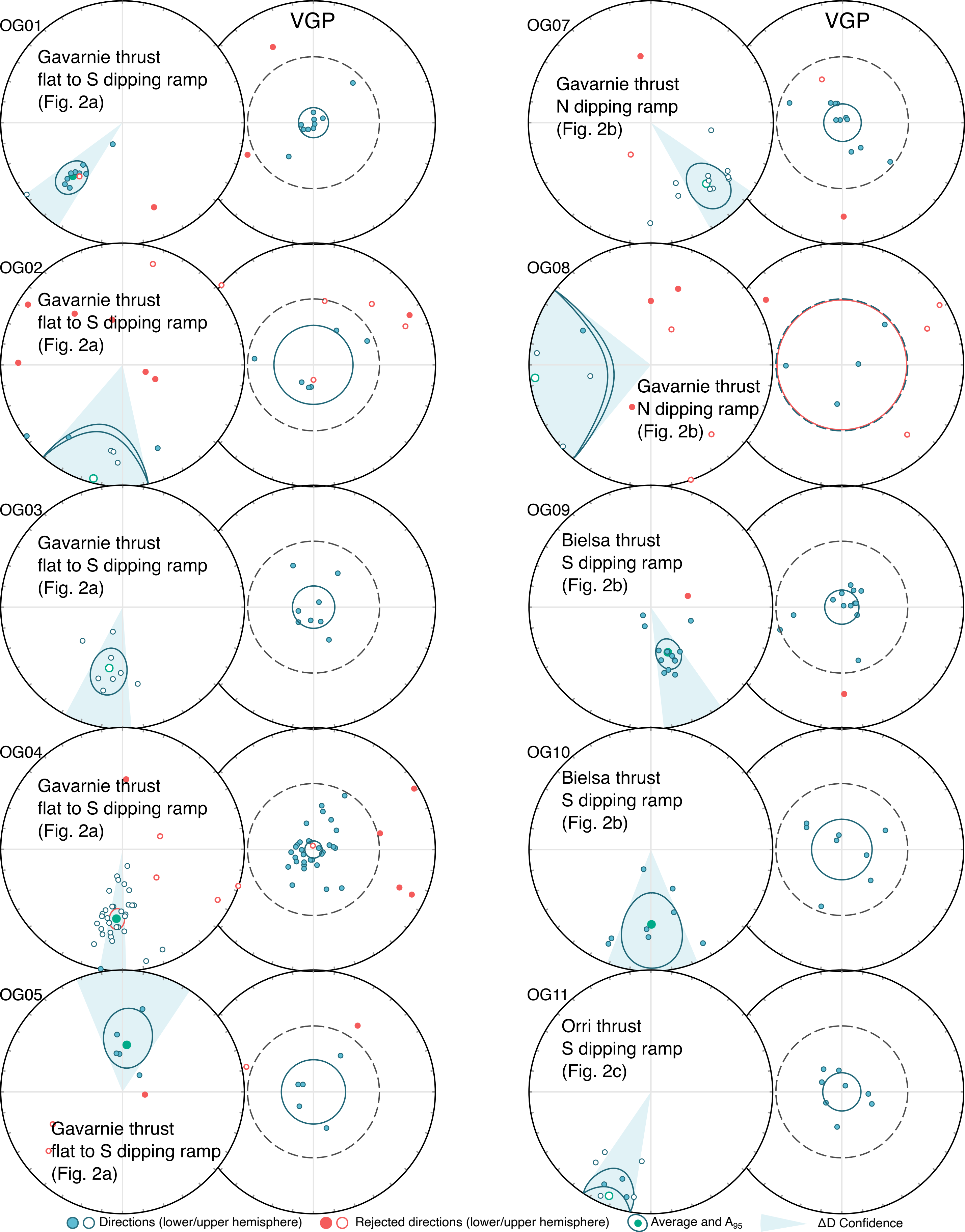

OG01
Gavarnie thrust
flat to S dipping ramp
(Fig. 2a)
VGP
OG02
Gavarnie thrust
flat to S dipping ramp
(Fig. 2a)
OG03
Gavarnie thrust
flat to S dipping ramp
(Fig. 2a)
OG04
Gavarnie thrust
flat to S dipping ramp
(Fig. 2a)
OG05
Gavarnie thrust
flat to S dipping ramp
(Fig. 2a)
OG07
Gavarnie thrust
N dipping ramp
(Fig. 2b)
VGP
OG08
Gavarnie thrust
N dipping ramp
(Fig. 2b)
OG09
Bielsa thrust
S dipping ramp
(Fig. 2b)
OG10
Bielsa thrust
S dipping ramp
(Fig. 2b)
OG11
Orri thrust
S dipping ramp
(Fig. 2c)
Directions (lower/upper hemisphere)
Rejected directions (lower/upper hemisphere)
Average and A_{95}
ΔD Confidence

Figure 8: Directional and VGP results in geographic coordinates of sites OG01 to OG11. Uncertainty envelope is in both cases VGP A95. Red dots are those that fall outside of the 45° cut-off. Sites OG02, 05 and 08 did not provide statistically meaningful results and were not interpreted.

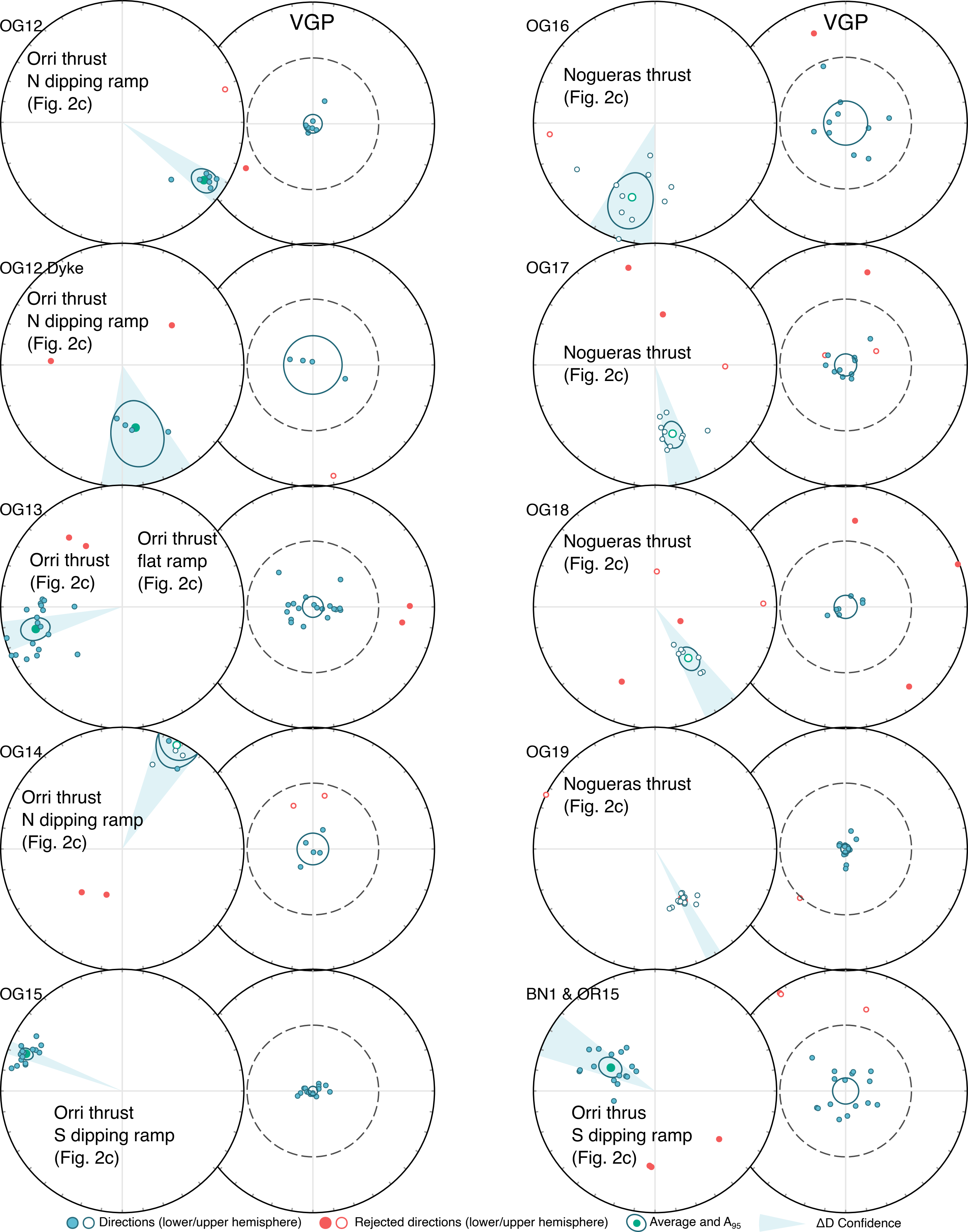

OG12
Orri thrust
N dipping ramp
(Fig. 2c)
VGP
OG16
Nogueras thrust
(Fig. 2c)
VGP
OG12 Dyke
Orri thrust
N dipping ramp
(Fig. 2c)
OG17
Nogueras thrust
(Fig. 2c)
OG13
Orri thrust
(Fig. 2c)
Orri thrust
flat ramp
(Fig. 2c)
OG18
Nogueras thrust
(Fig. 2c)
OG14
Orri thrust
N dipping ramp
(Fig. 2c)
OG19
Nogueras thrust
(Fig. 2c)
OG15
Orri thrust
S dipping ramp
(Fig. 2c)
BN1 & OR15
Orri thrus
S dipping ramp
(Fig. 2c)
Directions (lower/upper hemisphere)
Rejected directions (lower/upper hemisphere)
Average and A_{95}
ΔD Confidence

Figure 9: Directional and VGP results in geographic coordinates of sites OG12 to OG19 and BN1-OR15. Uncertainty envelope is in both cases VGP A95. Red dots are those that fall outside of the 45° cut-off. Sites OG02, 05 and 08 did not provide statistically meaningful results and were not interpreted.

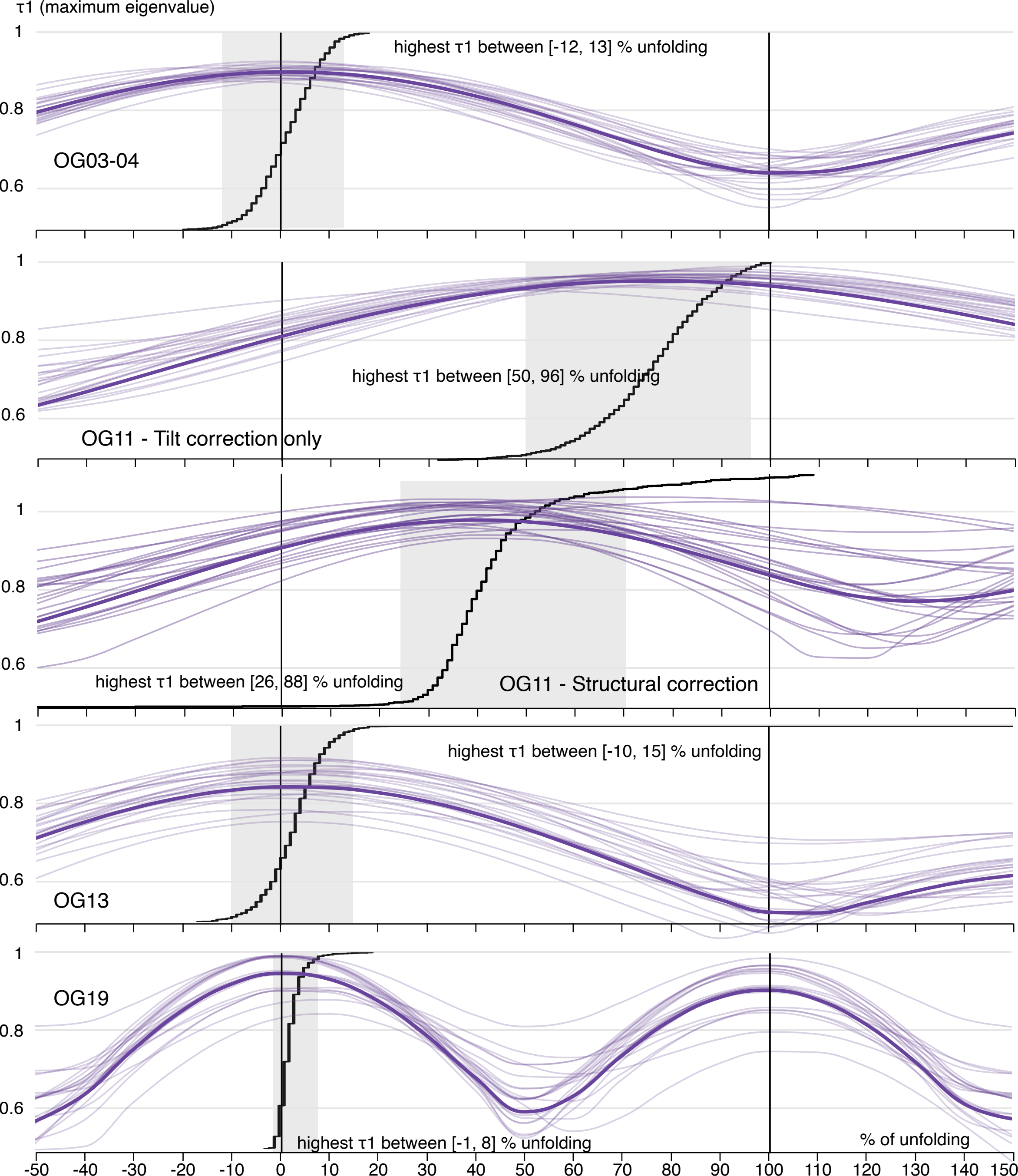

τ1 (maximum eigenvalue)
1
0.8
0.6
OG03-04
highest τ1 between [-12, 13] % unfolding
1
0.8
0.6
OG11 - Tilt correction only
highest τ1 between [50, 96] % unfolding
1
0.8
0.6
highest τ1 between [26, 88] % unfolding
OG11 - Structural correction
1
0.8
0.6
OG13
highest τ1 between [-10, 15] % unfolding
1
0.8
0.6
OG19
highest τ1 between [-1, 8] % unfolding
% of unfolding
-50
-40
-30
-20
-10
0
10
20
30
40
50
60
70
80
90
100
110
120
130
140
150

Figure 10: Within-site fold tests. All are negative but OG11, which is inconclusive.

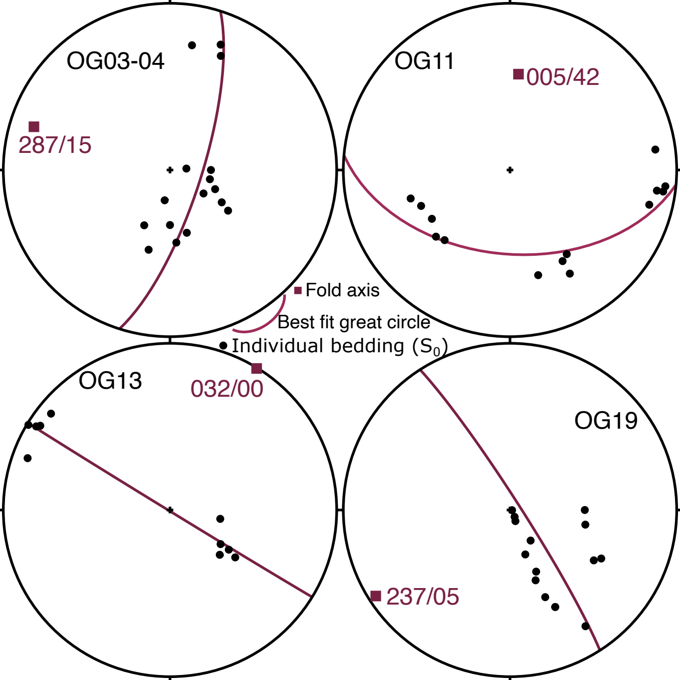

OG03-04
287/15
OG11
005/42
Fold axis
Best fit great circle
Individual bedding (S_0)
OG13
032/00
OG19
237/05

Figure 11: Pi diagrams for the studied folds. Only OG11 shows a steeply plunging axis.

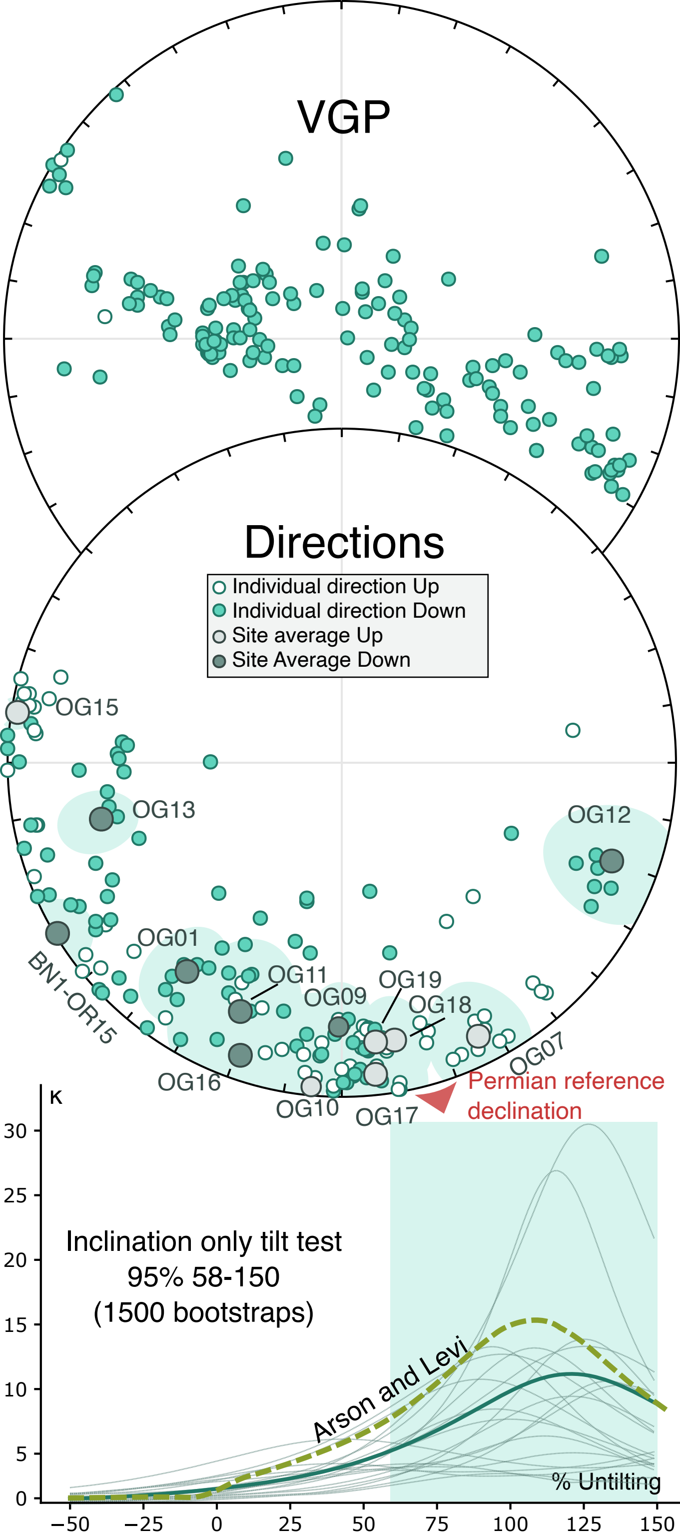

VGP
Directions
Individual direction Up
Individual direction Down
Site average Up
Site Average Down
OG15
OG13
OG12
OG01
BN1-OR15
OG11
OG19
OG09
OG18
OG16
OG10
OG17
OG07
Permian reference declination
κ
30
25
20
15
10
5
0
Inclination only tilt test
95% 58-150
(1500 bootstraps)
Arson and Levi
% Untilting
−50
−25
0
25
50
75
100
125
150

Figure 12: Results after the inferred Alpine tilt correction. Permian reference declination for Iberia is after Weil et al. (2010). The results show a positive inclination only tilt test following the methodology of Enkin and Watson (1996) (selected bootstraps in thin gray lines) and Arson and Levi (2010) approach (dashed line).

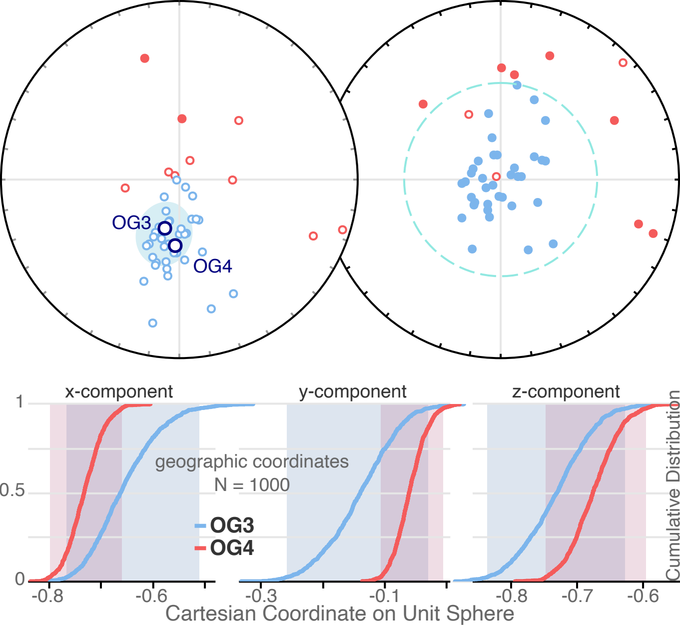

OG3
OG4
x-component
y-component
z-component
1
0.5
0
Cumulative Distribution
geographic coordinates
N = 1000
OG3
OG4
-0.8
-0.6
-0.3
-0.1
-0.8
-0.7
-0.6
Cartesian Coordinate on Unit Sphere

Figure 13: OG03 and OG04 show a common true mean bootstrapped direction (after Tauxe, 2010) in geographic coordinates (and Alpine tilt corrected, since it is the same for both).

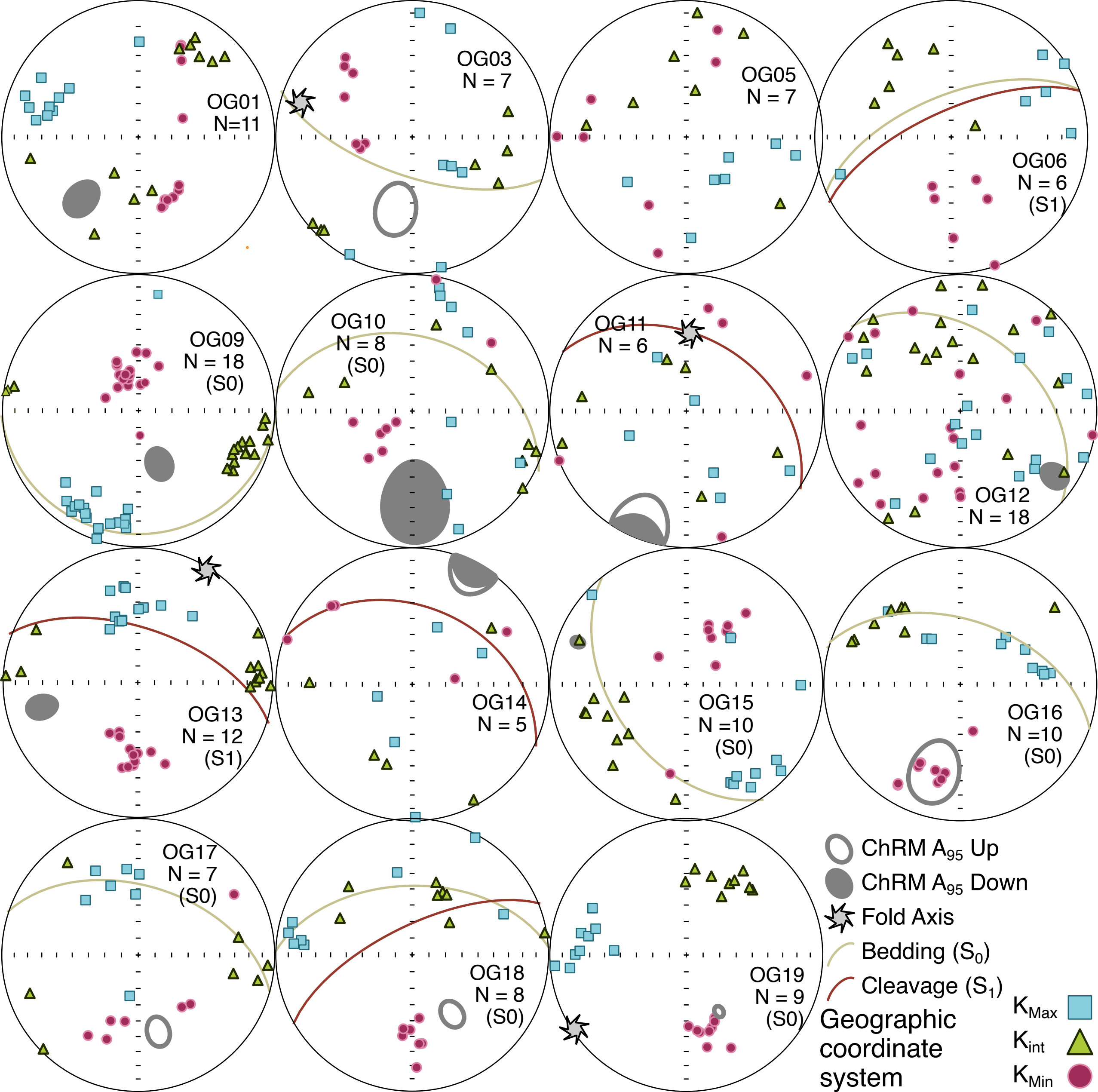

OG01
N=11
OG03
N = 7
OG05
N = 7
OG06
N = 6
(S1)
OG09
N = 18
(S0)
OG10
N = 8
(S0)
OG11
N = 6
OG12
N = 18
OG13
N = 12
(S1)
OG14
N = 5
OG15
N =10
(S0)
OG16
N =10
(S0)
OG17
N = 7
(S0)
OG18
N = 8
(S0)
OG19
N = 9
(S0)
ChRM A_{95} Up
ChRM A_{95} Down
Fold Axis
Bedding (S_0)
Cleavage (S_1)
Geographic coordinate system
K_{Max}
K_{int}
K_{Min}

Figure 14: Results from the anisotropy of the magnetic susceptibility analyses. Magnetic fabrics represent bedding or S1 cleavage. Magnetic fabric directions do not coincide with the paleomagnetic directions, which allows us to discard an internal deformation control of the paleomagnetic remanence.

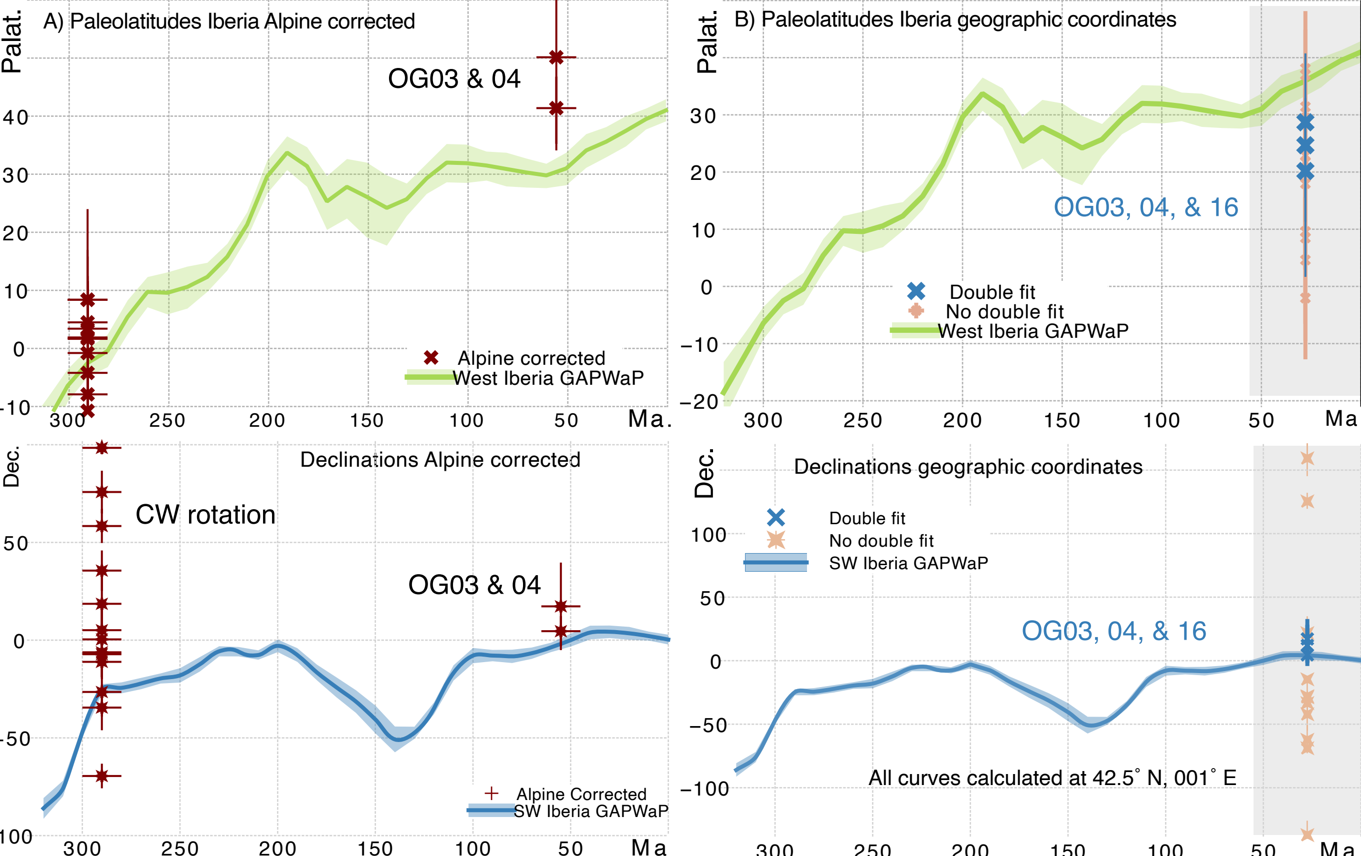

A) Paleolatitudes Iberia Alpine corrected
Palat.
OG03 & 04
Alpine corrected
West Iberia GAPWaP
40
30
20
10
0
-10
300
250
200
150
100
50
Ma.
B) Paleolatitudes Iberia geographic coordinates
Palat.
OG03, 04, & 16
Double fit
No double fit
West Iberia GAPWaP
30
20
10
0
−10
−20
300
250
200
150
100
50
Ma
Declinations Alpine corrected
Dec.
CW rotation
OG03 & 04
Alpine Corrected
SW Iberia GAPWaP
50
0
-50
100
300
250
200
150
100
50
Ma
Declinations geographic coordinates
Dec.
Double fit
No double fit
SW Iberia GAPWaP
OG03, 04, & 16
All curves calculated at 42.5° N, 001° E
100
50
0
−50
−100
300
250
200
150
100
50
Ma

Figure 15: Figure showing paleolatitudes (up) and declinations (down) of OG sites compared to the GAPWaP of Torsvik et al. (2012) rotated to Iberia (following Pastor-Galán et al., 2018). A) After Alpine Tilt B) Geographic coordinates.

Structure	S0 (Bedding)		Fold Axis		S1		Inferred Alpine tilt			Location	
	Dip Direction	Dip	Trend	Plunge	Dip direction	Dip	Basement thrust unit	Dip Direction	Dip	Latitude	Longitude
OG01	-	-	-	-	-	-	Gavarnie thrust	185	20	42.7473	-0.24497
OG02*	Fold		191	14	-	-	Gavarnie thrust	185	20	42.7361	-0.25841
OG03	199	66	287	15	-	-	Gavarnie thrust	185	20	42.7249	-0.28900
OG04	Fold				20	60	Gavarnie thrust	185	20	42.7259	-0.28300
OG05*	299	17	-	-	-	-	Gavarnie thrust	180	20	42.7023	-0.12022
OG06*	358	62	-	-	256	78	Gavarnie thrust	180	20	42.7064	-0.12464
OG07	36	42	-	-	35	55	Gavarnie thrust	25	45	42.7771	0.19758
OG08*	10	36	-	-	-	-	Gavarnie thrust	25	45	42.7759	0.19858
OG09	89	13	-	-	-	-	Bielsa thrust	205	45	42.5673	0.46444
OG10	17	31	-	-	-	-	Bielsa thrust	205	45	42.5824	0.47447
OG11	Fold		5	42	-	-	Orri thrust	25	30	42.5222	0.65244
OG12	40	39	-	-	-	-	Orri thrust	25	30	42.5167	0.65100
OG13	Fold		32	0	-	-	Orri thrust	25	0	42.5028	0.65228
OG14*	Fold		292	19	-	-	Orri thrust	25	30	42.4752	0.77733
OG15	234	48	-	-	-	-	Nogueras thrust units	205	90	42.3380	1.06883
OG16	16	51	-	-	-	-	Orri thrust	25	45	42.5339	1.17550
OG17	17	50	-	-	-	-	Orri thrust	25	45	42.5048	1.20275
OG18	359	51	-	-	254	67	Gavarnie thrust	25	45	42.5689	1.59106
OG19	Fold		237	5	250	60	Gavarnie thrust	25	45	42.5682	1.58550
BN1 & OI	185	25	-	-	-	-	Nogueras thrust units	205	90	42.3220	1.10466

*Unrealiable paleomagnetic result

Table 1

Table 1: Site Location and key structural data from each site.

Geographic	N	Ns	Cutoff	S	Dec	Inc	R	k	a95	K	A95	A95min	A95max	ΔDx	ΔIx	λ
OG01	10	12	45	16.71	222.7	40.19	9.48	17.3	12	24.19	10	4.78	19.22	10.89	13.79	22.9
OG02*	6	13	45	30.38	194.24	-4.41	4.97	4.86	33.9	7.34	26.5	5.86	26.52	26.52	52.76	-2.21
OG03	8	8	45	20.23	191.98	-47.6	7.66	20.7	12.5	16.32	14.1	5.22	22.12	16.16	16.7	-28.7
OG04	32	37	45	18.08	184.57	-42.53	30.18	17	6.34	20.49	5.75	3	9.24	6.33	7.56	-24.6
OG05*	5	8	45	22.16	5.71	58.01	4.78	17.9	18.6	13.61	21.5	6.3	29.75	28.01	19.81	38.7
OG06*	2	3	45	24.06	248.92	-20.54	1.88	8.48	101	11.42	82.5	9.09	52.99	NaN	149.8	-10.6
OG07	11	13	45	21.77	137.77	-32.97	10.37	15.8	11.8	14.29	12.5	4.6	18.1	13.16	19.46	-18
OG08*	4	10	45	35.85	263.68	-5.38	3.11	3.36	59.5	5.28	44.3	6.89	34.24	44.31	87.91	-2.7
OG09	13	14	45	21.87	159.34	57.48	12.55	26.7	8.18	14.19	11.4	4.3	16.29	14.55	10.64	38.1
OG10	8	8	45	28.26	179.3	38.56	7.17	8.42	20.3	8.45	20.2	5.22	22.12	21.86	28.68	21.7
OG11	8	8	45	18.26	201.72	-9.18	7.18	8.58	20.1	19.89	12.7	5.22	22.12	12.77	24.98	-4.62
OG12	7	8	45	8.33	125.38	18.71	6.89	53	8.36	95.15	6.22	5.51	24.07	6.31	11.48	9.61
OG12-Dyke	4	6	45	16.99	167.95	46.44	3.9	28.8	17.4	23.01	19.6	6.89	34.24	22.23	23.72	27.7
OG13	19	21	45	16.57	255.81	27.7	18.01	18.2	8.09	24.22	6.96	3.7	12.83	7.2	11.67	14.7
OG14*	5	7	45	11.11	27.96	-4.07	4.79	19.1	18	53.41	10.6	6.3	29.75	10.57	21.05	-2.04
OG15	15	15	45	6.56	291.06	16.61	14.87	107	3.71	152.9	3.1	4.06	14.89	3.14	5.82	8.48
OG16	10	11	45	23.96	197.11	-36.31	9.14	10.5	15.7	11.72	14.7	4.78	19.22	15.71	21.7	-20.2
OG17	10	13	45	12.28	165.55	-41.26	9.79	42	7.54	43.86	7.38	4.78	19.22	8.06	9.94	-23.7
OG18	7	11	45	10.29	146.65	-48.47	6.88	49.7	8.64	62.26	7.71	5.51	24.07	8.86	8.94	-29.5
OG19	15	16	45	6.98	151.24	-50.69	14.93	188	2.8	135.3	3.3	4.06	14.89	3.87	3.64	-31.4
BN1 & OR15	16	19	45	18.76	297.88	56.61	15.43	26.1	7.35	18.89	8.71	3.96	14.3	10.96	8.32	37.2
Tilt corrected																
OG01	10	12	45	16.71	222.7	40.19	9.48	17.3	12	24.19	10	4.78	19.22	10.89	13.79	22.9
OG02*	5	13	45	28.84	290.83	0.19	4.02	4.07	43.3	8.09	28.6	6.3	29.75	28.63	57.26	0.1
OG03	8	8	45	24.81	30.57	-65.75	7.66	20.7	12.5	10.9	17.6	5.22	22.12	26.79	13.23	-48
OG04	32	37	45	18.27	179.62	-19.66	29.27	11.4	7.89	19.99	5.83	3	9.24	5.92	10.66	-10.1
OG05*	6	8	45	27.91	19.38	45.79	5.54	10.9	21.2	8.67	24.1	5.86	26.52	27.35	29.65	27.2
OG06*	2	3	45	16.59	230.08	-10.25	1.88	8.48	101	23.93	53.5	9.09	52.99	53.83	104.5	-5.17
OG07	11	13	45	16.8	156.89	-16.86	10.37	15.8	11.8	23.56	9.61	4.6	18.1	9.72	18	-8.62
OG08*	3	10	45	26.69	247.31	-2.42	2.63	5.46	58.9	9.34	42.9	7.73	41.04	42.86	85.58	-1.21
OG09	13	14	45	17.61	164.17	45.05	12.55	26.7	8.18	21.64	9.12	4.3	16.29	10.21	11.39	26.6

OG10	6	8	45	30.89	169.06	56.53	5.52	10.5	21.7	7.09	27	5.86	26.52	34.74	25.85	37.1
OG11	8	8	45	11.72	200.07	-4.48	7.75	27.9	10.7	48.06	8.07	5.22	22.12	8.08	16.07	-2.24
OG12	8	8	45	16.16	111.8	5.44	7.43	12.3	16.5	25.83	11.1	5.22	22.12	11.12	22.06	2.72
OG12-Dyke	4	6	45	19.2	127.4	56.64	3.85	20.5	20.8	18.07	22.2	6.89	34.24	28.35	21.19	37.2
OG13	11	21	45	19.66	183.15	41.74	10.46	18.4	10.9	17.25	11.3	4.6	18.1	12.4	15.1	24
OG14*	5	7	45	21.6	46.47	-55.84	4.79	19.1	18	14.34	20.9	6.3	29.75	26.32	20.34	-36.4
OG15	15	15	45	5.25	289.1	-11.31	14.87	107	3.71	238.1	2.48	4.06	14.89	2.5	4.82	-5.71
OG16	10	11	45	20.18	15.07	-14.68	9.14	10.5	15.7	16.44	12.3	4.78	19.22	12.38	23.37	-7.46
OG17	11	13	45	16.57	171.02	0.43	10.41	16.9	11.5	24.55	9.4	4.6	18.1	9.4	18.8	0.21
OG18	8	11	45	17.99	159.26	-8.83	7.4	11.7	16.9	20.94	12.4	5.22	22.12	12.43	24.35	-4.44
OG19	9	16	45	4.48	155.85	10.2	8.96	217	3.5	327.9	2.85	4.98	20.54	2.86	5.56	5.14
BN1 & OR15	14	19	45	21.46	257.17	56	13.43	22.9	8.49	14.47	10.8	4.18	15.55	13.51	10.49	36.6

* Less than n = 7 was not considered

Table 2

Table 2: Paleomagnetic results for ChRM component for all sites in geographic and tilt (bedding) corrected coordinates.

ALPINE TILT CORRECTION

	Dec	Inc
OG01	215.64	23.55
OG03	197.28	-67.34
OG04	184.57	-60.42
OG07	153.46	-8.93
OG09	145.47	21.97
OG10	180.36	-3.3
OG11	185.14	20.77
OG12	110.54	15.53
OG13	255.81	27.7
OG15	278.36	-3.78
OG16	198.58	8.37
OG17	173.76	-6.78
OG18	168.95	-16.46
OG19	172.82	-16.4
BN1 & OR15	238.36	1.58

Table 3

Table 3: Paleomagnetic results (ChRM) after the Alpine tilt correction (tilt associated to the emplacement of the thrusts).

Inclination only statistics

Geographic	k	Inclination	α95
Site means avg.	0	0	90
Bedding corrected			
Site means avg.	2.17	18.25	38.91
Site means avg. but OG03 & 04	3.93	22.67	25.44
Alpine tilt corrected			
Site means avg.	4.19	-3.4	21.25
Site means avg. but OG03 & 04	14.42	5.08	11.3

Table 4

Table 4: Concentration parameters of inclination only statistics from the mean values of the sites.