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Comparative geochemical study on Furongian–earliest Ordovician (Toledanian) and Ordovician (Sardic) felsic magmatic events in south-western Europe: underplating of hot mafic magmas linked to the opening of the Rheic Ocean

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Abstract. A geochemical comparison of early Palaeozoic felsic magmatic episodes throughout the south-western European margin of Gondwana is made and includes (i) Furongian–Early Ordovician (Toledanian) activities recorded in the Central Iberian and Galicia–Trás-os-Montes zones of the Iberian Massif, and (ii) Early–Late Ordovician (Sardic) activities in the Eastern Pyrenees, Occitan Domain (Albigeois, Montagne Noire and Mouthoumet massifs) and Sardinia. Both phases are related to uplift and denudation of an inherited palaeorelief, and stratigraphically preserved as distinct angular discordances and paraconformities involving gaps of up to 22 million years. The geochemical features of the predominantly felsic Toledanian and Sardic activities point to a predominance of magmatic byproducts derived from the melting of metasedimentary rocks, rich in SiO_2 and K_2O and with a peraluminous character. Zr / TiO_2, Zr / Nb, Nb / Y and Zr vs. Ga / Al ratios, and rare-earth element (REE) and $_{\varepsilon}Nd_{(t)}$ values suggest the contemporaneity, for both phases, of two geochemical scenarios characterized by arc and extensional features evolving to distinct extensional and rifting conditions associated with the final outpouring of mafic tholeiite-dominant lava flows. The Toledanian and Sardic magmatic phases are linked to neither metamorphism nor penetrative deformation; on the contrary, their unconformities are associated with foliation-free open folds subsequently affected by the Variscan deformation. The geochemical and structural framework precludes subduction-generated melts reaching the crust in a magmatic arc-to-back-arc setting and favours partial melting of sediments and/or granitoids in the lower continental crust triggered by the underplating of hot mafic magmas related to the opening of the Rheic Ocean.

1 Introduction

A succession of early Palaeozoic felsic magmatic episodes, ranging in age from Furongian (formerly "late Cambrian") to Late Ordovician, are widespread along the south-western European margin of Gondwana. Magmatic pulses are characterized by preferential development in different palaeogeographic areas and linked to the development of stratigraphic unconformities, but they are related to neither metamorphism nor penetrative deformation (Gutiérrez Marco et al., 2002; Montero et al., 2007). In the Central Iberian Zone of the

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Iberian Massif (representing the western branch of the Ibero-Armorican Arc; Fig. 1a–b), this magmatism is mainly represented by the Ollo de Sapo Formation, which has long been recognized as a Furongian–Early Ordovician (495–470 Ma) assemblage of predominantly felsic volcanic, subvolcanic and plutonic igneous rocks. This magmatic activity is contemporaneous with the development of the Toledanian phase, which places Lower Ordovician (upper Tremadocian–Floian) rocks onlapping an inherited palaeorelief formed by Ediacaran–Cambrian rocks and involving a sedimentary gap of ca. 22 million years. This unconformity can be correlated with the "Furongian gap" identified in the Ossa-Morena Zone of the Iberian Massif and the Anti-Atlas of Morocco (Álvaro et al., 2007, 2018; Álvaro and Vizcaïno, 2018; Sánchez-García et al., 2019), and with the "lacaune normande" in the central and North Armorican domains (Le Corre et al., 1991).

Another predominantly felsic magmatic event, although younger (Early–Late Ordovician) in age, has been recognized in some massifs situated along the eastern branch of the Variscan Ibero-Armorican Arc, such as the Pyrenees, the Occitan Domain and Sardinia (Fig. 1a, c–e). This magmatism is related to the Sardic unconformity, where Furongian–Lower Ordovician rocks are unconformably overlain by those attributed to the Sandbian–lower Katian (formerly Caradoc). The Sardic phase is related to both (i) a sedimentary gap of ca. 16–20 million years, along with an unconformity that geometrically ranges from 90° (angular discordance) to 0° (paraconformity) (Barca and Cherchi, 2004; Funneda and Oggiano, 2009; Álvaro et al., 2016, 2018; Casas et al., 2019), and (ii) a Middle Ordovician development of cleavage-free folds lacking any contemporaneous metamorphism (for an updated revision, see Casas et al., 2019). The associated magmatic activity took place during a time span of about 25–30 million years (from 475 to 445 Ma), so broadly contemporaneous with the sedimentary gap.

Although a general consensus exists associating this Furongian–Ordovician magmatism with the opening of the Rheic Ocean and the drift of Avalonia from north-western Gondwana (Díez Montes et al., 2010; Nance et al., 2010; Thomson et al., 2010; Álvaro et al., 2014a), the origin of this magmatism has received different interpretations. In the Central Iberian Zone, for instance, several geodynamic models have been proposed, such as (i) subduction-related melts reaching the crust in a magmatic arc-to-back-arc setting (Valverde-Vaquero and Dunning, 2000; Castro et al., 2009), (ii) partial melting of sediments or granitoids in the lower continental crust (LCC) affected by the underplating of hot mafic magmas during an extensional regime (Bea et al., 2007; Montero et al., 2009; Díez Montes et al., 2010) and (iii) post-collisional decompression melting of an earlier thickened continental crust without significant mantle involvement (Villaseca et al., 2016). In the Occitan Domain (southern French Massif Central and Mouthoumet massifs) and the Pyrenees, Marini (1988), Pouclet et al. (2017) and Puddu et al. (2019) have suggested a link to mantle thermal anomalies. Navidad et al. (2018) proposed that the Pyrenean magmatism was induced by progressive crustal thinning and uplift of lithospheric mantle isotherms. In Sardinia, Oggiano et al. (2010), Carmignani et al. (2001), Gaggero et al. (2012) and Cruciani et al. (2018) have suggested that a subduction scenario, mirroring an Andean-type active margin, caused the main Mid-Ordovician magmatic activity. In the Alps, the Sardic counterpart is also interpreted as a result of the collision of the so-called Qaidam Arc with the Gondwanan margin, followed by the accretion of the Qilian Block (Von Raumer and Stampfli, 2008; Von Raumer et al., 2013, 2015). This geodynamic interpretation is mainly suggested for the Alpine Briançonnais–Austroalpine basement, where the volcanosedimentary complexes postdating the Sardic tectonic inversion and folding stage portray a younger arc–arc oblique collision (450 Ma) of the eastern tail of the internal Alpine margin with the Hun terrane, succeeded by conspicuous exhumation in a transform margin setting (430 Ma) (Zurbriggen et al., 1997; Schaltegger et al., 2003; Franz and Romer, 2007; Von Raumer and Stampfli, 2008; Von Raumer et al., 2013; Zurbriggen, 2015, 2017).

Until now the Toledanian and Sardic magmatic events had been studied in different areas and interpreted separately, without taking into account their similarities and differences. In this work, the geochemical affinities of the Furongian–Early Ordovician (Toledanian) and Early–Late Ordovician (Sardic) felsic magmatic activities recorded in the Central Iberian and Galicia–Trás-os-Montes zones, the Pyrenees, the Occitan Domain and Sardinia are compared. The re-appraisal is based on 17 new samples from the Pyrenees, Montagne Noire and Sardinia, completing the absence of analysis in these areas, and a wide-ranging dataset of 93 previously published geochemical analyses throughout the study region in south-western Europe. This comparison may contribute to a better understanding of the meaning and origin of this felsic magmatism and, thus, to a discussion on the geodynamic scenario of this Gondwana margin (Fig. 1a) during Cambrian–Ordovician times, bracketed between the Cadomian and Variscan orogenies.

2 Emplacement and age of magmatic events

This section documents the emplacement (summarized in Fig. 2) and age (Fig. 3) of the Toledanian and Sardic magmatic events throughout the south-western basement European Variscan Belt, in the north-western margin of Gondwana during Cambro-Ordovician times.

2.1 Iberian Massif

In the Ossa-Morena and southern Central Iberian zones of the Iberian Massif (Fig. 1a–b), the so-called Toledanian phase is recognized as an angular discordance that separates variably tilted Ediacaran–Cambrian Series 2 rifting volcanosedimen-

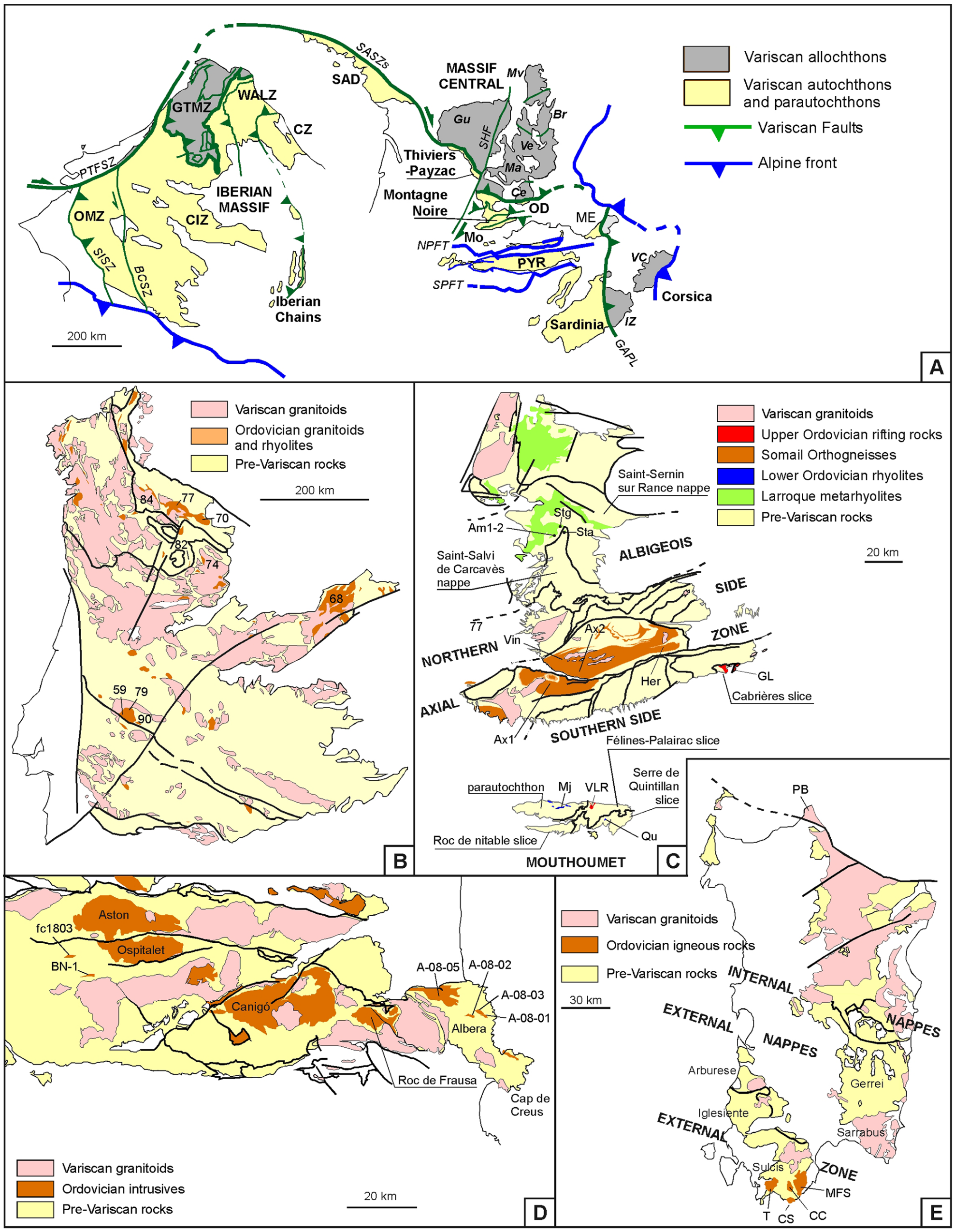


Figure 1. **(a)** Pre-Variscan reconstruction of the Variscan tectonostratigraphic units bearing Cambro-Ordovician exposures reported in this work, from the south-western European margin of Gondwana; based on Pouclet et al. (2017) and Álvaro et al. (2020). **(b)** Setting of samples in the Central Iberian and Galicia–Trás-os-Montes zones; 59 – Carrascal, 68 – Guadarrama, 70 – Sanabria, 74 – Miranda do Douro, 77 – Ollo de Sapo, 79 – Portalegre, 82 – Saldanha, 84 – San Sebastián, 90 – Urra, BCSZ – Badajoz-Córdoba Shear Zone, Ce – Cévennes Massif, CIZ – Central Iberian Zone, CZ – Cantabrian Zone, GAPL – Grimaud–Asinara–Posada Line, GTMZ – Galicia–Trás-os-Montes Zone, ME – Maures-Estérel Massif, Mo – Mouthoumet Massif, NPFT – North Pyrenean Fault thrust, OD – Occitan Domain, OMZ – Ossa-Morena Zone, PTFSZ – Pyrenean Domain of the Porto–Tomar–Ferreira do Alentejo Shear Zone, Sa – Sanabria, SAD – South Armorican Domain, SASZs – South Armorican Shear Zone southern branch, SHF – Sillon Houiller Fault, SISZ – South-Iberian Shear Zone, SPFT – South Pyrenean Fault thrust, WALZ – West Asturian–Leonese Zone; modified from Sánchez-García et al. (2019). **(c)** Setting of samples in the Montagne Noire and Mouthoumet massifs; Am1-2 Larroque hamlet (Ambialet), Stg – St-Géraud, Sta – St-André, Mj – Montjoi, Qu – Quintillan, GL – Roque de Bandies, VLR – Villerouge-Termenès, VIN – Le Vintrou, HER – Gorges d’Héric (Caroux Massif), Ax1 – southern Mazamet (Nore Massif), Ax2 – (Rou) S Rouairoux (Agout Massif); modified from Álvaro et al. (2016). **(d)** Setting of Pyrenean samples; AB-08-01, 02, 03 – Albera metavolcanics, AB-08-05 – Albera orthogneisses, BN-1 – Andorra rhyolites, fc-1803 – Pallaresa rhyolites; modified from Casas et al. (2019). **(e)** Setting of Sardinian samples; CS – Capo Spartivento, T2 – Tuerredda, CC5 – Cuile Culurgioni, MF1 – Monte Filau, MFS1 – Monte Setti Ballas, PB – Punta Bianca; modified from Oggiano et al. (2010).

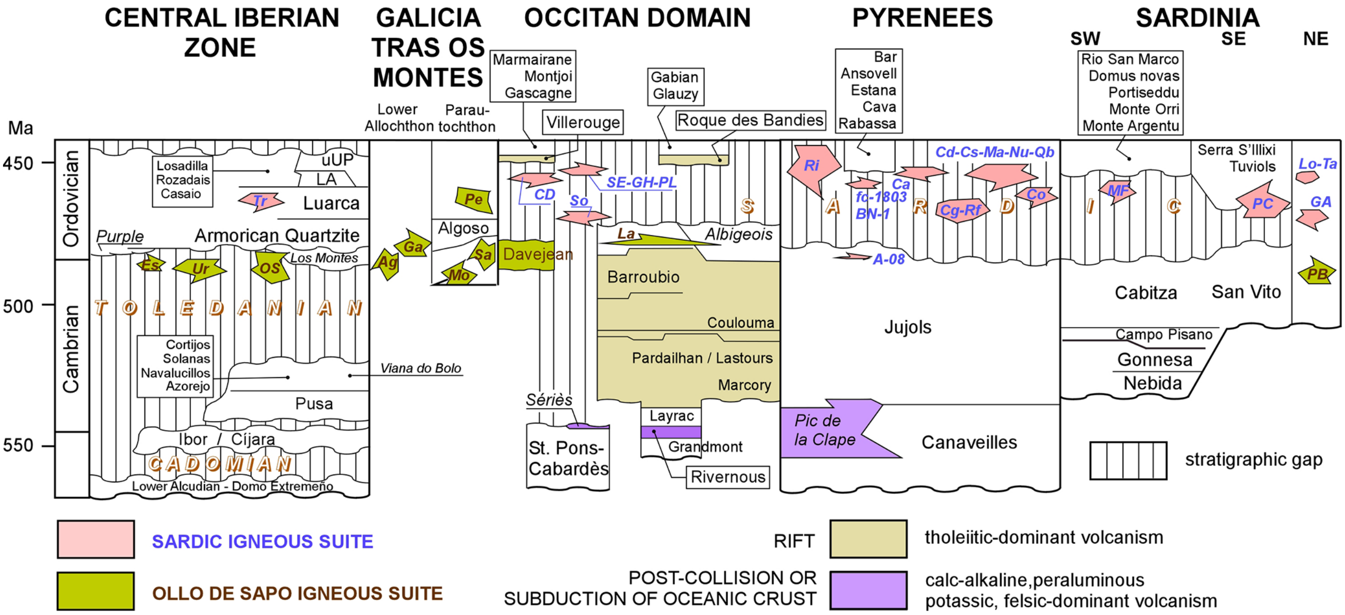


Figure 2. Cambro-Ordovician lithostratigraphic chart of the areas studied in this work from the Central Iberian Zone, Galicia–Trás-os-Montes Zone, Occitan Domain, Eastern Pyrenees and Sardinia; modified from Álvaro et al. (2014b, 2016, 2018), Pouclet et al. (2017) and Sánchez-García et al. (2019); other lithostratigraphic sketches – such as those of the Ciudad Rodrigo–Hurdes–Sierra de Gata Domain (Díez Balda et al., 1990) and the Portuguese sector (Medina et al., 1998; Meireles et al., 2013) in the Central Iberian Zone, the Central Pyrenees (Zwart, 1979; Laumonier et al., 1996), the Albigeois Mountains in the Occitan Domain (Guérangé-Lozes and Alsac, 1986; Pouclet et al., 2017) and northern Sardinia (Elter et al., 1986) – are not included here; abbreviations: A-08 – Albera orthogneisses and metavolcanics (ca. 465–472 Ma; Liesa et al., 2011), Ag – Agualada, BN-1 – Andorra rhyolites, Ca – Campelles ignimbrites (ca. 455 Ma; Martí et al., 2019), CD – Cadí gneiss (456 ± 5 Ma; Casas et al., 2010), Cg – Canigó gneiss (472–462 Ma; Cocherie et al., 2005; Navidad et al., 2018), Co – Cortalets metabasite (460 ± 3 Ma; Navidad et al., 2018), Cs – Casemí gneiss (446 ± 5 and 452 ± 5 Ma; Casas et al., 2010), Es – Estremoz rhyolites (499 Ma; Pereira et al., 2012), fc-1803 – Pallaresa rhyolites (ca. 453 Ma; Clariana et al., 2018), Ga – Galiñero, GA – Golfo Aranci orthogneiss (469 ± 3.7 Ma; Giacomini et al., 2006), GH – Gorges d'Héric orthogneiss (450 ± 6 Ma; Roger et al., 2004), La – Larroque volcanic complex, LA – La Aquiana limestone, Ma – Marialles microdiorite (453 ± 4 Ma; Casas et al., 2010), Lo – Lodè orthogneiss (456 ± 14 Ma; Helbing and Tiepolo, 2005), MF – Monte Filau–Capo Spartivento orthogneiss (449 ± 6 Ma according to Ludwing and Turi, 1989; 457.5 ± 0.3 and 458.2 ± 0.3 Ma according to Pavanetto et al., 2012), Mo – Mora (493.5 ± 2 Ma; Dias Da Silva et al., 2014), Nu – Núria gneiss (457 ± 4 Ma; Martínez et al., 2011), OS – Ollo de Sapo rhyolites and ash-fall tuff beds (ca. 477 Ma; Gutiérrez-Alonso et al., 2016), Pe – Peso volcanic complex, PL – Pont-de-Larn orthogneiss (456 ± 3 Ma; Roger et al., 2004), Qb – Queralbs gneiss (457 ± 5 Ma; Martínez et al., 2011), PB – Punta Bianca orthogneiss (broadly Furongian–Tremadocian in age), PC – Porto Corallo dacites (465.4 ± 1.9 and 464 ± 1 Ma; Giacomini et al., 2006; Oggiano et al., 2010), Ri – Ribes granophyre (458 ± 3 Ma; Martínez et al., 2011), Rf – Roc de Frausa gneiss (477 ± 4, 476 ± 5 Ma; Cocherie et al., 2005; Castiñeiras et al., 2008a), So – Somail orthogneiss (471 ± 4 Ma; Cocherie et al., 2005), Sa – Saldanha (483.7 ± 1.5; Dias da Silva, 2014), SE – Saint Eutrope gneiss (455 ± 2 Ma; Pitra et al., 2012), Ta – Tanaunella orthogneiss (458 ± 7 Ma; Helbing and Tiepolo, 2005), Tr – Truchas, Ur – Urra rhyolites, and uUP – undifferentiated Upper Ordovician.

tary packages from overlying passive-margin successions. The Toledanian gap comprises, at least, most of the Furongian and basal Ordovician, but the involved erosion can incise the entire Cambrian and the upper Ediacaran Cadomian basement (Gutiérrez-Marco et al., 2019; Álvaro et al., 2019; Sánchez-García et al., 2019). Recently, Sánchez-García et al. (2019) have interpreted the Toledanian phase as a break-up (or rift/drift) unconformity with the Armorican Quartzite (including the Purple Series and Los Montes beds; McDougall et al., 1987; Gutiérrez-Alonso et al., 2007; Shaw et al., 2012, 2014) sealing an inherited Toledanian palaeorelief (Fig. 2).

The phase of uplift and denudation of an inherited palaeorelief composed of upper Ediacaran–Cambrian rocks is associated with the massive outpouring of predominantly felsic calc-alkaline magmatic episodes related to neither metamorphic nor cleavage features. This magmatic activity is widely distributed throughout several areas of the Iberian Massif, such as the Cantabrian Zone and the easternmost flank of the West Asturian–Leonese Zone, where sills and rhyolitic lava flows and volcaniclastics mark the base of the Armorican Quartzite (dated at ca. 477.5 Ma; Gutiérrez-Alonso et al., 2007, 2016), and the lower Tremadocian Borrachón Formation of the Iberian Chains (Álvaro et al., 2008). Similar ages have been reported from igneous rocks of the basal al-

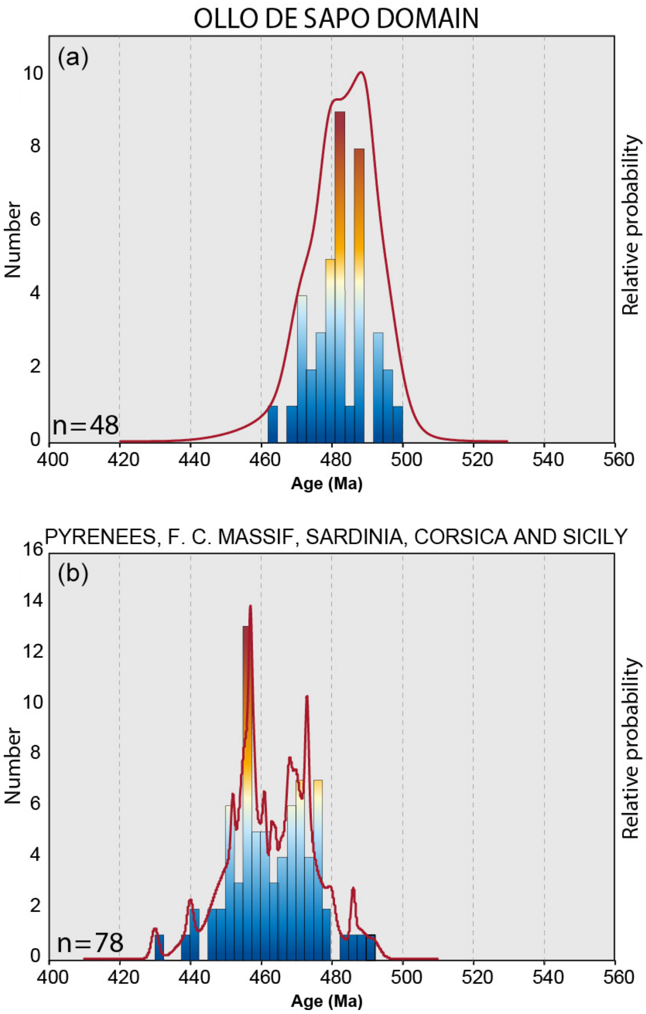


Figure 3. Relative probability plots of the age of the Cambrian–Ordovician magmatism for **(a)** the Ollo de Sapo domain from the Central Iberian Zone and **(b)** the Pyrenees (Guilleries and Gavarres massifs), French Central Massif (including Montagne Noire), Sardinia, Corsica and Sicily (n: number of analyses). Data obtained from references cited in the text.

lochthonous units and the Schistose Domain in the Galicia–Trás-os-Montes Zone (500–462 Ma; Valverde-Vaquero et al., 2005, 2007; Montero et al., 2009; Talavera et al., 2008, 2013; Dias da Silva et al., 2012, 2014; Díez Fernández et al., 2012; Farias et al., 2014) and different areas of the Central Iberian Zone, including the contact between the Central Iberian and Ossa-Morena zones, where the Carrascal and Portalegre batholiths are intruded and the felsic volcanosedimentary Urra Formation marks the unconformity that separates Cambrian and Ordovician strata (494–470 Ma; Solá et al., 2008; Antunes et al., 2009; Neiva et al., 2009; Romaõ et al., 2010; Rubio-Ordóñez et al., 2012; Villaseca et al., 2013) (Fig. 1b).

The most voluminous Toledanian-related volcanic episode is represented by the Ollo de Sapo Formation, which crops out throughout the north-eastern Central Iberian Zone. It mainly consists of felsic volcanosedimentary and volcanic rocks, interbedded at the base of the Lower Ordovician strata and plutonic bodies. The volcanosedimentary Ollo de Sapo Formation has long been recognized as an enigmatic Furongian–Early Ordovician (495–470 Ma) magmatic event exposed along the core of a 600 km long antiform (labelled as 77 in Fig. 1b) (Valverde-Vaquero and Dunning, 2000; Bea et al., 2006; Montero et al., 2007, 2009; Zeck et al., 2007; Castiñeiras et al., 2008a; Díez Montes et al., 2010; Navidad and Castiñeiras, 2011; Talavera et al., 2013; López-Sánchez et al., 2015; Díaz-Alvarado el al., 2016; Villaseca et al., 2016; García-Arias et al., 2018). The peak of magmatic activity was reached at ca. 490–485 Ma, and its most recognizable characteristic is the presence of abundant megacrysts of K-feldspar, plagioclase and blue quartz. There is no evident space–time relationship in its distribution (for a discussion, see López-Sánchez et al., 2015), and, collectively, the Ollo de Sapo Formation rocks record a major tectonothermal event whose expression can be found in most of the Variscan massifs of continental Europe, including the Armorican and Bohemian massifs (e.g. von Quadt, 1997; Kröner and Willmer, 1998; Linnemann et al., 2000; Tichomirowa et al., 2001; Friedl et al., 2004; Mingram et al., 2004; Teipel et al., 2004; Ballèvre et al., 2012; El Korh et al., 2012; Tichomirowa et al., 2012; for a summary, see Casas and Murphy, 2018). The large volume of magmatic rocks located in the European Variscan Belt has led some authors to propose the existence of a siliceous large igneous province (LIP) (Díez Montes et al., 2010; Gutiérrez-Alonso et al., 2016), named Ibero-Armorican LIP by García-Arias et al. (2018).

The Sardic phase has been proposed, marking a stratigraphic discontinuity close to the Middle–Upper Ordovician boundary interval in some areas of the Central Iberian Zone (e.g. Buçaco and Truchas Syncline; Martínez Catalán et al., 1992; Días da Silva et al., 2016) and the Morais allochthonous complex of the Galicia–Trás-os-Montes Zone (Días da Silva, 2014; Días da Silva et al., 2014, 2016). In the Truchas Syncline, the significance of the discontinuity (or discontinuities) was questioned by a biostratigraphic study of conodonts and the re-interpretation of some of these scouring surfaces as the result of Hirnantian glaciogenic incisions (Sarmiento et al., 1999). The pre-Hirnantian discontinuities have been interpreted as linked to the development of "horsts and half-grabens of local extent", as a result of which "tilting and gentle folding of the Lower–Middle Ordovician strata, due to the rotation of individual half-grabens and horsts, create the Sardic unconformity in Iberia" (Dias da Silva et al., 2016: p. 1131 and p. 1143). However, the presence of synsedimentary listric faults associated with local outpouring of a basic volcanism, related to extensional pulses in the Ordovician passive-margin platform fringing north-western Gondwana, cannot be associated with the Sardic phase. As summarized in this work, the Sardic phase is characterized by generalized cortical uplift, denudation of exposed uplifted areas under subaerial exposure, stratigraphic gaps of about 25–30 million years, broad intrusion of felsic granitic plutons (now orthogneisses after Variscan deformation and metamorphism) with calc-alkaline affinity and a record of alluvial-to-fluvial deposits onlapping the unconformity. These are the features that characterize the Ordovician Sardic phase, not the record of Ordovician volcanism associated with local listric faults (e.g. Casas et al., 2010, 2019; Álvaro et al., 2016).

In contrast, the Sardic aftermath is represented by predominantly basic volcanic activity, mainly of tholeiitic affinity, and lining rift branches highlighting the onset of listric-fault networks; this event could be geodynamically compared with some processes recorded in the Central Iberian and the Galicia–Trás-os-Montes zones, but not with the Sardic phase. Therefore, the presence of the Sardic phase in the Iberian Massif has already been ruled out by the information published during the last 2 decades and should not be maintained unless the above-reported tectonothermal events are really found. The presence of an Ordovician volcanism associated with listric faults is not an argument in support of the record of the Sardic phase.

2.2 Central and Eastern Pyrenees

In the Central and Eastern Pyrenees (Fig. 1d), earliest Ordovician volcanic-free passive-margin conditions, represented by the Jujols Group (Padel et al., 2018), were succeeded by a late Early–Mid-Ordovician phase of uplift and erosion that led to the onset of the Sardic unconformity (Fig. 2). Uplift was associated with magmatic activity, which continued until Late Ordovician times. An extensional interval took place then, developing normal faults that controlled the sedimentation of post-Sardic siliciclastic deposits infilling palaeorelief depressions. Acritarchs recovered in the uppermost part of the Jujols Group suggest a broad Furongian–earliest Ordovician age (Casas and Palacios, 2012), conterminous with a maximum depositional age of ca. 475 Ma, based on the age of the youngest detrital zircon populations (Margalef et al., 2016). On the other hand, a ca. 459 Ma U–Pb age for the Upper Ordovician volcanic rocks overlying the Sardic unconformity has been proposed in the Eastern Pyrenees (Martí et al., 2019), and ca. 452–455 Ma in the neighbouring Catalan Coastal Range, which represents the southern prolongation of the Pyrenees (Navidad et al., 2010; Martínez et al., 2011). Thus, a time gap of about 16–23 million years can be related to the Sardic phase in the Eastern Pyrenees and the neighbouring Catalan Coastal Range.

Coeval with the late Early–Mid-Ordovician phase of generalized uplift and denudation, key magmatic activity led to the intrusion of voluminous granitoids, about 500 to 3000 m thick and encased in strata of the Ediacaran–Lower Cambrian Canaveilles Group (Fig. 2). These granitoids constitute the protoliths of the large orthogneissic laccoliths that punctuate the backbone of the Central and Eastern Pyrenees. These are, from west to east (Fig. 1d), the Aston (467–470 Ma; Denèle et al., 2009; Mezger and Gerdes, 2016), Hospitalet (about 472 Ma; Denèle et al., 2009), Canigó (472–462 Ma; Cocherie et al., 2005; Navidad et al., 2018), Roc de Frausa (477–476 Ma; Cocherie et al., 2005; Castiñeiras et al., 2008b) and Albera (about 470 Ma; Liesa et al., 2011) massifs, which comprise a dominant Floian–Dapingian age. It is noticeable that only a minor representation of coeval basic magmatic rocks are outcropped. The acidic volcanic equivalents have been documented in the Albera Massif, where subvolcanic rhyolitic porphyroid rocks have yielded similar ages to those of the main gneissic bodies at about 474–465 Ma (Liesa et al., 2011). Similar acidic byproducts are represented by the rhyolitic sills of Pierrefitte (Calvet et al., 1988).

The late Early–Mid-Ordovician ("Sardic") phase of uplift was succeeded by a Late Ordovician extensional interval responsible for the opening of (half-)grabens infilled with the basal Upper Ordovician alluvial-to-fluvial conglomerates (La Rabassa Conglomerate Formation). At map scale, a set of NE–SW-trending normal faults abruptly controlling the thickness of the basal Upper Ordovician formations can be recognized in the La Cerdanya area (Casas and Fernández, 2007; Casas, 2010). Sharp variations in the thickness of the Upper Ordovician strata have been documented by Hartevelt (1970) and Casas and Fernández (2007). Drastic variations in grain size and thickness can be attributed to the development of palaeotopographies controlled by faults and subsequent erosion of uplifted palaeoreliefs, with subsequent infill of depressed areas by alluvial fan and fluvial deposits, finally sealed by Silurian sediments (Puddu et al., 2019). A Late Ordovician magmatic pulse contemporaneously yielded a varied set of magmatic rocks. Small granitic bodies are encased in the Canaveilles strata of the Canigó Massif. They constitute the protoliths of the Cadí (about 456 Ma; Casas et al., 2010), Casemí (446 to 452 Ma; Casas et al., 2010), Núria (ca. 457 Ma; Martínez et al., 2011) and Canigó G1-type (ca. 457 Ma; Navidad et al., 2018) gneisses.

The lowermost part of the Canaveilles Group (the so-called Balaig Series) host metre-scale thick bodies of metadiorite sills related to an Upper Ordovician protolith, (ca. 453 Ma, SHRIMP U–Pb in zircon; Casas et al., 2010). Coeval calc-alkaline ignimbrites, andesites and volcaniclastic rocks are interbedded in the Upper Ordovician succession of the Bruguera and Ribes de Freser areas (Robert and Thiebaut, 1976; Ayora, 1980; Robert, 1980; Martí et al., 1986, 2019). In the Ribes area, a granitic body with granophyric texture, dated at ca. 458 Ma by Martínez et al. (2011), intruded at the base of the Upper Ordovician succession. In the La Pallaresa dome, some metre-scale rhyodacitic-to-dacitic subvolcanic sills, Late Ordovician in age (ca. 453 Ma; Clariana et al., 2018), occur interbedded within the pre-unconformity strata and close to the base of the Upper Ordovician.

2.3 Occitan Domain: Albigeois, Montagne Noire and Mouthoumet massifs

The parautochthonous framework of the southern French Massif Central, named Occitan Domain by Pouclet et al. (2017), includes, among others, from south to north, the Mouthoumet, Montagne Noire and Albigeois massifs. The domain represents the south-eastern prolongation of the Variscan South Armorican Zone (including south-western Brittany and Vendée). Since Gèze (1949) and Arthaud (1970), the southern edge of the French Massif Central has been traditionally subdivided, from north to south, into the northern, axial and southern Montagne Noire (Fig. 1c). The Palaeozoic succession of the northern and southern sides includes sediments ranging from late Ediacaran to Silurian and from Terreneuvian (Cambrian) to Visean in age, respectively. These successions are affected by large-scale, south-verging recumbent folds that display a low-to-moderate metamorphic grade. Their emplacement took place in late Visean to Namurian times (Engel et al., 1980; Feist and Galtier, 1985; Echtler and Malavieille, 1990). The axial zone consists of plutonic, migmatitic and metamorphic rocks forming a regional ENE–WSW-oriented dome (Fig. 1c), where four principal lithological units can be recognized: (i) schists and mica schists, (ii) migmatitic orthogneisses, (iii) metapelitic metatexites, and (iv) diatexites and granites (Cocherie, 2003; Faure et al., 2004; Roger et al., 2004, 2015; Bé Mézème, 2005; Charles et al., 2009; Rabin et al., 2015). The Rosis mica schist synform subdivides the eastern axial zone into the Espinouse and Caroux sub-domes, whereas the south-western edge of the axial zone comprises the Nore Massif.

In the Occitan Domain, two main Cambro-Ordovician felsic events can be identified giving rise to the protoliths of (i) the Larroque metarhyolites in the northern Montagne Noire and Albigeois Mountains, thrusted southward from Rouergue, and (ii) the migmatitic orthogneisses that form the axial zone of the Montagne Noire (Fig. 2).

i. The Larroque volcanosedimentary complex is a thick (500–1000 m) package of porphyroclastic metarhyolites located on the northern Montagne Noire (Lacaune Mountains), Albigeois Mountains (St-Salvi-de-Carcavès and St-Sernin-sur-Rance nappes) and Rouergue; the Variscan setting of the formation is allochthonous in the Albigeois Mountains and parautochthonous in the rest. This volcanism is encased in the so-called "Série schisto-gréseuse verte" (see Guérangé-Lozes et al., 1996; Guérangé-Lozes and Alabouvette, 1999; Pouclet et al., 2017) (Fig. 2). The Larroque volcanic rocks consist of deformed porphyroclastic rhyolites rich in largely fragmented, lacunous (rhyolitic) quartz and alkali feldspar phenocrysts. The metarhyolites occur as porphyritic lava flows; sills; and other associated facies, such as aphyric lava flows, porphyritic and aphyric pyroclastic flows of welded or unwelded ignimbritic types, fine-to-coarse tephra deposits, and epiclastic and volcaniclastic deposits. These rocks are named "augen gneiss" or augengneiss and do not display a high-grade gneiss paragenesis but a general lower-grade metamorphic mineralogy. The Occitan augengneisses mimic the Ollo de Sapo facies from the Central Iberian Zone because of their large bluish quartz phenocrysts. Based on geochemical similarities and contemporaneous emplacement, Pouclet et al. (2017) suggested that this event also supplied the Davejean acidic volcanic rocks in the Mouthoumet Massif, which represent the southern prolongation of the Montagne Noire (Fig. 2), and the Génis rhyolitic unit of the western Limousin sector.

ii. Some migmatitic orthogneisses make up the southern axial zone, from the western Cabardès to the eastern Caroux domes. The orthogneisses, derived from Ordovician metagranites bearing large K-feldspar phenocrysts, were emplaced at about 471 Ma (Somail orthogneiss; Cocherie et al., 2005), 456 to 450 Ma (Pont-de-Larn and Gorges d'Héric gneisses, Roger et al., 2004) and ca. 455 Ma (Saint Eutrope gneiss; Pitra et al., 2012). They intruded a metasedimentary pile, traditionally known as "Schistes X" and formally named St-Pons–Cabardès Group (Fig. 2). The latter consists of schists, greywackes, quartzites, and subsidiary volcanic tuffs and marbles (Demange et al., 1996; Demange, 1999; Roger et al., 2004; Cocherie et al., 2005). The group is topped by the Sériès tuff, dated at about 545 Ma (Lescuyer and Cocherie, 1992), which represents a contemporaneous equivalent of the Cadomian Rivernous rhyolitic tuff (542.5 to 537.1 Ma) from the Lodève inlier of the northern Montagne Noire (Álvaro et al., 2014b, 2018; Padel et al., 2017). The age of migmatization has been inferred from U–Pb dates on monazite from migmatites and anatectic granites at 333 to 327 Ma (Bé Mézème, 2005; Charles et al., 2008); as a result, the 330–325 Ma time interval can represent a Variscan crustal melting event in the axial zone.

As in the Pyrenees, the Middle Ordovician is absent in the Occitan Domain. Its gap allows distinction between a Lower Ordovician pre-unconformity sedimentary package para- to unconformably overlain by an Upper Ordovician–Silurian succession (Álvaro et al., 2016; Pouclet et al., 2017).

2.4 Sardinia

In Sardinia the Cambro-Ordovician magmatism is well represented in the external (southern) and internal (northern) nappe zones of the exposed Variscan Belt (Fig. 1e), and ranges in age from late Furongian to Late Ordovician. Furongian–Tremadocian (ca. 491–480 Ma) magmatic activity, predating the Sardic phase, is mostly represented by felsic volcanic and subvolcanic rocks encased in the sandy San Vito Formation. The Sardic-related volcanic products differ from one nappe to another: intermediate and basic (mostly metandesites and andesitic basalts) are common in the nappe stacking of the central part of the island (Barbagia and Goceano), whereas felsic metavolcanites prevail in the south-eastern units. Their age is bracketed between 465 and 455 Ma (Giacomini et al., 2006; Oggiano et al., 2010; Pavanetto et al., 2012; Cruciani et al., 2018) and matches the Sardic gap based on biostratigraphy (Barca et al., 1988).

Teichmüller (1931) and Stille (1939) were the first to recognize in south-western Sardinia an intra-Ordovician stratigraphic hiatus. Its linked erosive unconformity is supported by a correlatable strong angular discordance in the Palaeozoic basement of the Iglesiente–Sulcis area, external zone (Carmignani et al., 2001). This major discontinuity separates the Cambrian–Lower Ordovician Nebida, Gonnesa and Iglesias groups (Pillola et al., 1998) from the overlying coarse-grained ("puddinga") Monte Argentu metasediments (Leone et al., 1991, 2002; Laske et al., 1994). The gap comprises a chronostratigraphically constrained minimum gap of about 18 million years that includes the Floian and Dapingian (Barca et al., 1987, 1988; Pillola et al., 1998; Barca and Cherchi, 2004) (Fig. 2). The hiatus is related to neither metamorphism nor cleavage, though some E–W folds have been documented in the Gonnesa Anticline and the Iglesias Syncline (Cocco et al., 2018), which are overstepped by the puddinga metaconglomerates. Both the E–W folds and the overlying metaconglomerates were subsequently affected by Variscan N–S folds (Cocco and Funneda, 2011, 2017). Sardic-related volcanic rocks are not involved in this area, but Sardic-inherited palaeoreliefs are lined with breccia slides that include metre-to-decametre-scale carbonate boulders ("olistoliti"), some of them hosting synsedimentary faults contemporaneously mineralized with ore bodies (Boni and Koeppel, 1985; Boni, 1986; Barca, 1991; Caron et al., 1997). The lower part of the unconformably overlying Monte Argentu Formation was deposited in alluvial to fluvial environments (Martini et al., 1991; Loi et al., 1992; Loi and Dabard, 1997).

A similar gap was reported by Calvino (1972) in the Sarrabus-Gerrei units of the external nappe zone. The so-called "Sarrabese phase" is related to the onset of thick (up to 500 m thick) volcanosedimentary complexes and volcanites (Barca et al., 1996; Di Pisa et al., 1992) with a Darriwilian age for the protoliths of the metavolcanic rocks (465.4 to 464 Ma; Giacomini et al., 2006; Oggiano et al., 2010). In the Iglesiente–Sulcis region (Fig. 1e), Carmignani et al. (1986, 1992, 1994, 2001) suggested that the "Sardic-Sarrabese phase" should be associated with the compression of a Cambro-Ordovician back-arc basin that originated the migration of the Ordovician volcanic arc toward the Gondwanan margin.

Some gneissic bodies, interpreted as the plutonic counterpart of metavolcanic rocks, are located in the Bithia unit (e.g. the Monte Filau area, 458 to 457 Ma, surrounded by a Middle Ordovician andalusite thermal aureole; Pavanetto et al., 2012; Costamagna et al., 2016) and in the internal units (Lodè orthogneiss, ca. 456 Ma; Tanaunella orthogneiss, ca. 458 Ma, Helbing and Tiepolo, 2005; Golfo Aranci orthogneiss, ca. 469 Ma, Giacomini et al., 2006).

The Sardic palaeorelief is sealed by Upper Ordovician transgressive deposits. The sedimentary facies show high variability, but the – mostly terrigenous – sediments vary from fine-to-medium-sized grey sandstones to muddy sandstones and claystones. They are referred to as the Katian Punta Serpeddì and Orroeledu formations (Pistis et al., 2016). This post-Sardic sedimentary succession is coeval with a new magmatic pulsation represented by alkaline to tholeiitic within-plate basalts (Di Pisa et al., 1992; Gaggero et al., 2012).

3 Geochemical data

3.1 Materials and methods

The rocks selected for geochemical analysis (231 samples; see tectonostratigraphic location in Fig. 1 and stratigraphic emplacement in Fig. 2) have recorded different degrees of hydrothermalism and metamorphism, as a result of which only the most immobile elements have been considered. The geochemical calculations, in which the major elements take part, have been made with values recalculated to 100 in volatility-free compositions; Fe is reported as FeO_t.

The geochemical dataset of the Central Iberian Zone includes 152 published geochemical data, from which 85 are plutonic and 67 are volcanic and volcaniclastic rocks from the Ollo de Sapo Formation (Galicia, Sanabria and Guadarrama areas), and the contact between the Central Iberian and Ossa-Morena zones (Urra Formation and Portalegre and Carrascal granites). Other data were yielded from six volcanic rocks of the Galicia–Trás-os-Montes Zone (Saldanha area) (Supplement).

The dataset of the Eastern Pyrenees consists of 38 samples, of which 6 are upper Lower Ordovician volcanic rocks and 7 are upper Lower Ordovician plutonic rocks, together with 9 Upper Ordovician volcanic and 14 Upper Ordovician plutonic rocks (Supplement). New data reported below include two samples of subvolcanic sills intercalated in the pre-Sardic unconformity succession (Clariana et al., 2018; Margalef, unpublished, Table 1).

The study samples from the Occitan Domain comprise six metavolcanic rocks, four from the Larroque volcanosedimentary complex in the Albigeois Mountains and northern Montagne Noire and two from the Mouthoumet Massif (Pouclet et al., 2017) (Supplement), and four new samples for the axial zone gneisses (Table 1).

Table 1. Chemical analyses of magmatic rocks. ICP and ICP-MS methods at ACME Laboratories in Canada.

	Pyrenees			Montagne Noire				Sardinia									
	Albera	Pallaresa	Andorra		Axial	zone					External	zone				Inner	zone
Sample	A-08-03	fC1803	BN 1	Ax – 1	Ax – 2	HER	VIN	CC 5	CS 2	CS 3	CS 5	CS 8	MF 1	MFS 1	T 2	PB50	PB100
Long (E) Lat (N)	3°7′39″ 42°25′2″	1°27′43″ 42°36′1″	1°33′29″ 42°32′30″	2°13′50″ 43°34′32″	2°33′58″ 43°29′3″	2°57′58″ 43°34′32″	2°13′50″ 43°17′45″	8°50′37″ 38°54′16″	8°50′35″ 38°52′38″	8°50′35″ 38°52′38″	8°50′40″ 38°52′36″	8°50′35″ 38°52′39″	8°50′47″ 38°54′58″	8°52′02″ 38°53′57″	8°48′54″ 38°53′57″	9°09′32″ 41°11″	9°09′32″ 41°11′04″
SiO_2	68.38	71.67	69.18	70.38	67.43	68.31	73.97	76.43	75.14	76.52	76.61	76.36	72.13	75.94	75.55	68.93	67.24
TiO_2	0.57	0.63	0.61	0.36	0.64	0.61	0.20	0.08	0.08	0.09	0.04	0.06	0.31	0.13	0.18	0.41	0.46
Al_2O_3	15.68	14.24	15.05	14.90	15.76	15.39	13.82	13.28	12.81	11.80	12.71	12.63	13.80	13.16	12.94	16.32	15.79
Fe_2O_3	4.09	4.54	4.20	3.04	4.11	4.19	2.05	0.69	1.39	1.44	1.28	1.35	2.96	1.55	1.62	3.19	4.78
MnO	0.07	0.06	0.05	0.04	0.04	0.04	0.04	0.01	0.01	0.01	0.01	0.01	0.02	0.03	0.04	0.08	0.08
MgO	1.35	0.78	1.16	0.78	1.33	1.34	0.43	0.08	0.15	0.16	0.06	0.05	0.36	0.19	0.08	1.15	1.58
CaO	0.21	0.53	1.78	1.22	1.44	1.58	0.62	0.32	0.25	0.15	0.20	0.35	0.61	0.38	0.17	3.05	2.70
Na_2O	4.07	1.67	3.40	3.33	2.78	2.93	2.87	3.04	1.71	1.58	2.91	3.35	2.89	2.57	2.53	3.85	3.43
K_2O	2.84	2.91	2.71	4.35	4.68	4.03	4.55	4.79	7.84	7.43	5.16	4.91	5.47	4.94	5.36	2.26	2.96
P_2O_5	0.17	0.24	0.20	0.21	0.2	0.19	0.18	0.15	0.05	0.05	0.03	0.04	0.12	0.11	0.07	0.15	0.14
L.O.I.	2.03	2.60	1.50	1.2	1.3	1.2	1.2	1.1	0.4	0.7	0.9	0.8	1.1	0.9	1.4	0.90	0.70
Total	99.05	99.42	99.42	99.51	99.30	99.39	99.73	99.90	99.69	99.79	99.78	99.78	99.47	99.75	99.78	99.97	99.37
As	77.20	1.70	6.80	2.50	6.00	1.80	1.90	0.70	1.00	0.50	2.80	1.10	1.80	101.10	4.00	5.00	5.00
Ba	742.50	388.00	398.00	499	1050	767	256	60	467	109	21	27	784	194	192	689.00	600.00
Be	2.44	3.00	2.00	4.00	2.00	5.00	3.00	6.00	3.00	1.00	9.00	2.00	7.00	3.00	7.00	3.00	5.00
Bi	0.30	0.20	0.10	0.20	0.20	0.20	0.40	0.30	0.10	0.10	0.10	0.10	0.10	0.70	0.40	4.00	4.00
Cd	0.18	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10		
Co	5.84	4.60	6.20	5.20	5.20	5.40	2.70	0.50	1.60	1.00	0.80	0.60	2.30	1.50	1.20	5.00	14.00
Cs	9.79	5.60	4.90	14.30	7.10	6.80	7.30	4.20	3.40	1.60	4.50	4.60	6.40	3.90	4.10	4.20	9.40
Cu	16.34	13.20	10.30	7.20	7.40	10.10	8.70	4.70	4.60	8.20	26.80	2.50	5.00	5.50	5.00	10.00	60.00
Ga	21.03	19.80	18.80	19.10	19.20	18.90	16.70	19.30	14.90	15.30	19.40	19.20	20.70	19.00	19.90	17.00	18.00
Hf	6.40	7.30	6.40	5.00	6.90	5.70	3.10	3.10	4.10	4.30	3.50	3.80	8.80	3.70	5.80	5.90	5.30
Mo	1.20	0.90	1.00	0.60	0.90	0.60	0.30	0.70	0.70	0.70	0.80	0.50	1.70	0.80	1.60	2.00	2.00
Nb	10.49	11.30	11.30	9.60	12.40	11.90	7.90	10.30	7.70	12.10	13.20	13.30	20.20	9.10	20.60	9.00	11.00
Ni	16.56	8.00	7.70	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00	80.00
Pb	7.94	9.80	22.90	3.50	4.60	5.10	3.60	2.90	7.40	8.60	4.50	5.50	5.10	6.30	5.50	21.00	24.00
Rb	124.40	123.70	137.20	204.6	161.6	142.2	188.2	289.9	206.1	187.4	294.1	275.1	208.7	256.4	227.1	85.00	118.00
Sb	2.27	0.10	0.30	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	5.00	5.00
Sc		10.00	10.00	6.00	9.00	9.00	4.00	3.00	3.00	4.00	4.00	4.00	15.00	4.00	8.00	9.00	12.00
Sn	2.11	5.00	5.00	9.00	3.00	3.00	7.00	9.00	4.00	3.00	13.00	15.00	7.00	15.00	12.00	3.00	3.00
Sr	158.00	201.80	83.70	91.20	160.30	150.10	68.70	30.70	73.90	25.20	7.90	8.10	59.90	45.60	25.00	217.00	167.00
Ta	1.07	1.10	1.10	0.80	1.00	0.80	0.70	2.10	0.90	1.10	3.40	1.70	1.60	1.70	2.30	1.00	1.20
Th	11.90	15.70	13.50	11.10	14.40	14.30	5.90	9.10	14.10	17.00	13.50	13.10	22.80	10.20	26.90	13.30	11.50
U	3.70	5.10	4.60	4.10	3.60	3.20	4.80	3.30	2.90	3.20	3.50	3.50	4.60	8.10	4.90	4.50	2.20
V	44.49	49.00	36.00	36.00	63.00	68.00	22.00	8.00	8.00	8.00	8.00	8.00	15.00	8.00	10.00	62.00	53.00
W	1.80	1.90	2.50	3.20	2.60	1.60	3.00	5.60	0.90	2.10	5.20	3.00	2.40	4.40	3.50	1.00	20.00
Y	29.29	43.90	50.60	28.30	38.40	36.20	27.80	28.00	60.10	53.60	44.40	46.00	61.60	31.80	55.80	29.00	24.00
Zn	63.71	52.00	70.00	55.00	71.00	78.00	46.00	7.00	35.00	39.00	15.00	24.00	37.00	30.00	22.00	70.00	70.00
Zr	233.30	263.20	237.10	174.40	249.20	219.10	93.70	73.50	93.80	105.10	62.20	74.50	311.80	108.10	161.90	245.00	214.00
La	27.90	45.30	38.00	29.60	39.50	38.70	13.60	10.50	22.70	19.50	12.10	13.40	54.20	17.90	31.30	26.90	34.30
Ce	59.00	86.90	75.50	58.10	77.00	78.20	26.70	21.60	42.10	39.70	26.20	29.90	109.80	37.40	97.60	53.20	70.50
Pr	7.26	9.80	8.47	6.99	9.41	9.55	3.36	2.36	4.73	4.85	3.00	3.24	11.94	4.07	6.86	5.88	8.20
Nd	27.83	35.60	31.20	26.00	36.40	36.40	12.60	8.40	16.60	17.10	10.50	10.90	44.70	15.00	24.00	21.60	29.40
Sm	5.80	7.69	7.16	5.70	7.55	7.63	3.15	2.43	4.10	4.41	3.28	3.44	9.37	3.88	4.93	4.70	6.00
Eu	0.98	1.05	1.03	0.87	1.27	1.15	0.41	0.14	0.43	0.13	0.06	0.09	1.17	0.30	0.19	0.95	0.93
Gd	5.22	8.32	7.89	5.59	7.28	7.05	3.38	3.20	5.60	5.50	4.42	4.69	10.60	4.50	6.34	4.00	5.10
Tb	0.87	1.26	1.27	0.89	1.17	1.10	0.67	0.69	1.13	1.18	1.03	1.07	1.70	0.82	1.27	0.70	0.80
Dy	5.30	6.68	8.00	5.09	6.89	6.39	4.59	4.30	7.69	8.23	7.31	7.66	10.28	5.24	9.00	3.70	4.30
Ho	1.06	1.52	1.73	0.99	1.42	1.30	0.98	0.91	1.91	1.91	1.59	1.65	2.13	1.12	2.01	0.70	0.80
Er	2.98	4.52	4.96	2.64	3.92	3.56	3.07	2.85	5.80	6.46	5.35	5.38	6.25	3.64	6.17	2.20	2.10
Tm	0.46	0.60	0.73	0.38	0.57	0.50	0.44	0.43	0.91	1.00	0.85	0.85	0.89	0.52	0.92	0.35	0.32
Yb	3.00	3.98	4.72	2.33	3.56	3.11	2.83	2.95	5.81	6.60	6.10	6.16	5.53	3.70	6.04	2.50	2.20
Lu	0.44	0.58	0.69	0.33	0.53	0.45	0.39	0.44	0.90	0.94	0.92	0.94	0.86	0.56	0.90	0.41	0.36

In the Sardinian dataset, 25 published analyses are selected: 5 correspond to the Golfo Aranci orthogneiss (Giacomini et al., 2006), 6 to metavolcanics from the central part of the island (Giacomini et al., 2006; Cruciani et al., 2013), and 5 to metavolcanics and 1 to gneisses from the Bithia unit (Cruciani et al., 2018) (Supplement). Ten new analyses are added from the Monte Filau and Capo Spartivento gneisses of the Bithia unit, and from the Punta Bianca gneisses embedded within the migmatites of the high-grade metamorphic complex from the inner zone (Table 1).

Whole-rock major and trace elements and rare-earth element (REE) compositions were determined at ACME Laboratories, Vancouver, Canada. $LiBO_2$ fusion followed by X-ray fluorescence spectroscopy (XRF) analysis was used to determine major elements. Rare-earth and refractory elements were measured by inductively coupled plasma–mass spectrometry (ICP-MS) following lithium metaborate–tetraborate fusion and nitric acid digestion on a 0.2 g sample. For base metals, a 0.5 g sample was digested in aqua regia at 95 °C and analysed by inductively coupled plasma–atomic emission spectrometry (ICP-AES). Analyses of standards and duplicate samples indicate precision of better than 1 % for major oxides and 3–10 % for minor and trace elements.

Additional Sm–Nd isotopic analyses were performed at the Centro de Geocronología y Geoquímica Isotópica of the Complutense University in Madrid. They were carried out in whole-rock powders using a $^{150}Nd / ^{149}Sm$ tracer by isotope dilution–thermal ionization mass spectrometry (ID-TIMS). The samples were first dissolved through oven digestion in sealed Teflon bombs with ultra-pure reagents to perform two-stage conventional cation-exchange chromatography for separation of Sm and Nd (Strelow, 1960; Winchester, 1963), and subsequently analysed using a VG Micromass Sector 54 multicollector spectrometer. The measured $^{143}Nd / ^{144}Nd$ isotopic ratios were corrected for possible isobaric interferences from ^{142}Ce and ^{144}Sm (only for samples with $^{147}Sm / ^{144}Sm < 0.0001$) and normalized to $^{146}Nd / ^{144}Nd = 0.7219$ to correct for mass fractionation. The La Jolla Nd international isotopic standard was analysed during sample measurement and gave an average value of $^{143}Nd / ^{144}Nd = 0.5114840$ for nine replicas, with an internal precision of ± 0.000032 (2σ). These values were used to correct the measured ratios for possible sample drift. The estimated error for the $^{147}Sm / ^{144}Nd$ ratio is 0.1 %.

A general classification of the analysed samples, following Winchester and Floyd (1977), can be seen in Fig. 4a–b, and the geographical coordinates of the new samples in Table 1. For geochemical comparison (summarized in Table 2), two large groups or suites are differentiated in order to check the similarities and differences between the magmatic rocks, and to infer a possible geochemical trend following a palaeogeographic SW–NE transect. The description reported below follows the same palaeogeographic and chronological order.

Table 2. Summarized geochemical features of the Furongian and Ordovician felsic episodes described in the text; data from Lancelot et al. (1985), Calvet et al. (1988), Valverde-Vaquero and Dunning (2000), Roger et al. (2004), Vilà et al. (2005), Giacomini et al. (2006), Díez-Montes (2007), Montero et al. (2007, 2009), Solá (2007), Zeck et al. (2007), Castiñeiras et al. (2008b), Talavera (2009), Casas et al. (2010), Navidad et al. (2010, 2018), Liesa et al. (2011), Martínez et al. (2011, 2019), Navidad and Castiñeiras (2011), Gaggero et al. (2012), Talavera et al. (2013), Villaseca et al. (2016), Pouclet et al. (2017), Cruciani et al. (2018) and this work. Abbreviations: CIZ – Central Iberian Zone, GTOMZ – Galicia–Trás-os-Montes Zone, OCC – Occitan Domain, PYR – Pyrenees, and SAR – Sardinia. * sensu Guitard (1970). A / CNK ratio is always peraluminous.

Orthogneiss Facies	Code	Composition	SiO_2 wt %	Na_2O wt %	K_{20} wt %	A / CNK ratio	$_{\varepsilon}Nd$	TDM (Ga)	$^{147}Sm / ^{144}Nd$	Area
1. Furongian–Middle Ordovician suite										
CIZ – Ollo de Sapo orthogneiss	OG	K-rich dacite to rhyolite	75–60.3	3.9–0.1	5.9–3.4	3.1–1.0	−5.1 to −1.8	1.8–1.1	0.15–0.09	Sanabria (ca. 472 Ma) and Guadarrama (ca. 488-473 Ma)
CIZ – leucogneiss	LG	K-rich dacite to rhyolite	75.9–73.6	3.1–2.7	5.3–4.2	1.3–1.1	−5.1 to −4.9	4.1	0.22–0.18	Guadarrama
CIZ – metagranite	GRA	K-rich dacite to rhyolite	77–64.6	4.8–0.5	6.3–2.5	1.8–1.0	−5.2 to +2.6	3.6–0.9	0.19–0.09	NE central system, Sanabria, Miranda do Douro (ca. 496–473 Ma), CIZ (496–471 Ma for Carrascal, Fermoselle, Ledesma, Portalegre and Vitigudino granites)
CIZ/GTMZ – volcanic rocks	VOL	andesite to rhyolite	79.3–64.6	3.2–0.1	6.3–2.2	2.7–1.1	−5.5 to −1.6	1.7–1.3	0.15–0.13	Saldanha Fm. in GTMZ, Ollo de Sapo Fm. in Sanabria, and Urra Fm.
CIZ – San Sebastián orthogneiss	OSS	rhyolite	75.4–73.8	3.1–2.5	5.4–4.9	1.2–1.1	−4.0 to 0	1.6–1.2	0.14–0.14	Sanabria (ca. 470–465 Ma)
PYR – augengneiss	G2*	dacite to rhyolite	73.6–68.3	3.9–3.2	4.4–2.5	1.2–1.1	−4.4 to −3.0	1.4–1.2	0.14–0.13	ca. 476–462 Ma
PYR – orthogneiss	G3*	K-rich dacite	73.5–68.4	2.9–2.4	4.4	1.2	−4.2	1.33	0.13	ca. 463 Ma
PYR – volcanic rocks	V1	Na-rich rhyolite	73.5–68.4	7.8–2.4	3.2–1.3	2.0–1.1	−5.1 to −2.6	1.7–1.6	0.19–0.13	Pierrefite Fm. and Albera Massif (ca. 472–465 Ma)
OCC – volcanic rocks	VOL-OD	K-rich dacite to rhyolite	75.6–66.7	3.7–0.6	9.3–2.3	2.4–1.3				Saint-Sernin-sur-Rance and Saint-Salvi-de-Carcavès nappes
SAR – orthogneiss	OG-SMO	K-rich rhyolite	74–67.2	3.8–2.6	5.8–2.3	1.3–1.1				ca. 469 Ma
SAR – volcanic rocks	VOL-SMO	K-rich dacite to rhyolite	76.7–67.6	4.7–1.9	5.4–2.9	2.0–1.2			0.16	ca. 464–462 Ma
2. Upper Ordovician suite										
PYR – orthogneiss	G1*	K-rich dacite to rhyodacite	76.4–73.4	3.1–2.6	5.3–4.7	1.2–1.1	−5.3 to −3.1	2.7–1.5	0.17–0.12	ca. 457 Ma
PYR – orthogneiss	CADí	K-rich dacite to rhyodacite	69.4	3	4	1.2	−4.1	1.5	0.13	Cadí Massif (ca. 456 Ma)
PYR – orthogneiss	CASEMí	K-rich dacite to rhyodacite	76–71.9	4–1.8	6.3–3.2	1.2–0.9	−3.6 to −1.3	2.6–1.3	0.17–0.13	Casemí Massif (ca. 451–446)
PYR – volcanic rocks	V2	andesite to rhyodacite	86.1–63	6–0	4.3–0.6	3.6–1	−5.1 to −2.6	1.7–1.6	0.14–0.14	Ribes de Freser, Andorra (ca. 457 Ma), Pallaresa (ca. 453 Ma), Els Metges (ca. 455.2 Ma)
OCC – orthogneiss	OG-OD	K-rich dacite to rhyolite	73.9–67.4	3.3–2.8	4.7–4	1.3–1.2	−4 to −3.5	1.8–1.4	0.15–0.13	Gorges d'Héric (ca. 450 Ma; Caroux), S Mazamet (Nore), S Rouairoux (Agout), Le Vintrou
SAR – external zone orthogneiss	OG-SUO	K-rich dacite to rhyolite	76.6–72.1	3.3–1.6	7.8–4.8	1.3–1.1	−3.3 to −1.6	4.2–1.2	0.19–0.12	Capo Spartivento, Cuile Culurgioni, Tuerredda, Monte Filau, Monte Setti Ballas (ca. 458–457 Ma)
SAR – nappe zone volcanic rocks	VOL-SUD	K-rich dacite to rhyodacite	76.7–70.7	3.3–1.6	7.8–4.8	1.3–1.1				Truzzulla Fm. at Monte Grighini

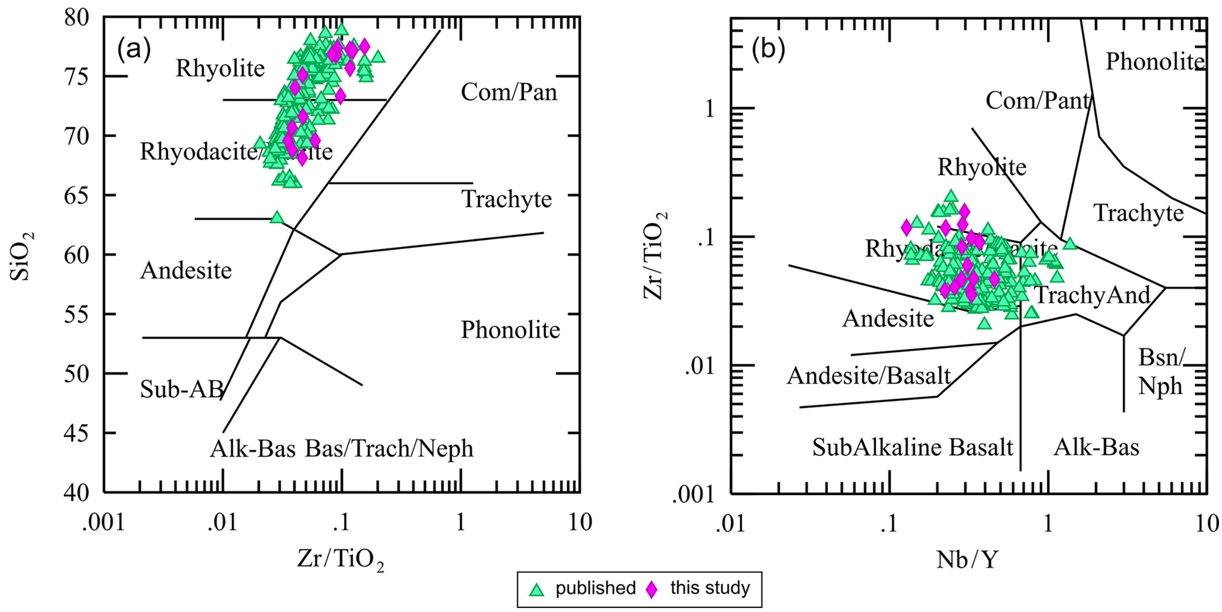


Figure 4. **(a)** SiO_2 vs. Zr / TiO_2 and **(b)** Zr / TiO_2 vs. Nb / Y plots (Winchester and Floyd, 1977) showing the composition of new samples (purple diamonds) and those taken from the literature (green triangles).

3.2 Furongian–Middle Ordovician suite

In the Central Iberian and Galicia–Trás-os-Montes zones, the Furongian-to-Middle Ordovician magmatic activity is pervasive. Their main representative is the Ollo de Sapo Formation, which includes volcanic and subvolcanic rocks (67 samples) as well as plutonic rocks (85 samples) (data from Murphy et al., 2006; Díez-Montes, 2007; Montero et al., 2007, 2009; Solá, 2007; Solá et al., 2008; Talavera, 2009; Villaseca et al., 2016). From the parautochthonous Schistose Domain from the Galicia–Trás-os Montes Zone, six samples of rhyolite tuffs of the Saldanha Formation (Dias da Silva et al., 2014) are selected, which share geochemical features with the Ollo de Sapo Formation. In summary, five facies are differentiated in the Central Iberian and Galicia–Trás-os Montes zones: the Ollo de Sapo orthogneisses, some leucogneisses, metagranites and volcanic rocks, and the San Sebastián orthogneiss (for a geochemical characterization, see Table 2).

In the Central and Eastern Pyrenees, Early–Mid-Ordovician magmatic activity gave rise to the intrusion of voluminous (about 500–3000 m in size) aluminous granitic bodies, encased in the Canaveilles beds (Álvaro et al., 2018; Casas et al., 2019). They constitute the protoliths of the large orthogneissic laccoliths that form the core of the domal massifs scattered throughout the backbone of the Pyrenees. Rocks of the Canigó, Roc de Frausa and Albera massifs have been taken into account in this work, in which volcanic rocks of the Pierrefite and Albera massifs, and the so-called G2 and G3 orthogneisses by Guitard (1970) are also included. All subgroups vary compositionally from subalkaline andesite to rhyolite, as illustrated in the Pearce (1996) diagram of Fig. 5 (data compiled from Vilà et al., 2005; Castiñeiras et al., 2008b; Liesa et al., 2011; Navidad et al., 2018).

Although most rocks in this area are acidic, the presence of minor mafic bodies is remarkable (Cortalet and Marialles metabasites, not studied in this work), which could indicate a mantle connection with parental magmas during the Mid and Late Ordovician. Additionally, it should be noted that there are no andesitic rocks in the area.

In the Occitan Domain, six samples of the Larroque volcanosedimentary complex (early Tremadocian in age) represent basin floors and subaerial explosive and effusive rhyolites (Pouclet et al., 2017). The porphyroclastic rocks of the Larroque metarhyolites were sampled in the Saint-Géraud and Larroque areas from the Saint-Sernin-sur-Rance nappe and the Saint-André klippe above the Saint-Salvi-de-Carcavès nappe (Pouclet et al., 2017).

In the Middle Ordovician rocks of Sardinia, 11 samples are selected, 5 of which correspond to orthogneisses of the Aranci Gulf, in the inner zone of the NE island (Giacomini et al., 2006), completed with 6 volcanic rocks of the external zone (Giacomini et al., 2006; Cruciani et al., 2018) (Table 2).

3.3 Upper Ordovician suite

In the Central and Eastern Pyrenees, four Upper Ordovician subgroups are distinguished based on their field occurrence and geochemical and geochronological features: the G1-type orthogneisses (sensu Guitard, 1970), the Cadí and Casemí orthogneisses and the metavolcanic rocks that include the Ribes de Freser rhyolites, the Els Metges volcanic tuffs, and the rhyolites from Andorra and Pallaresa areas (the latter

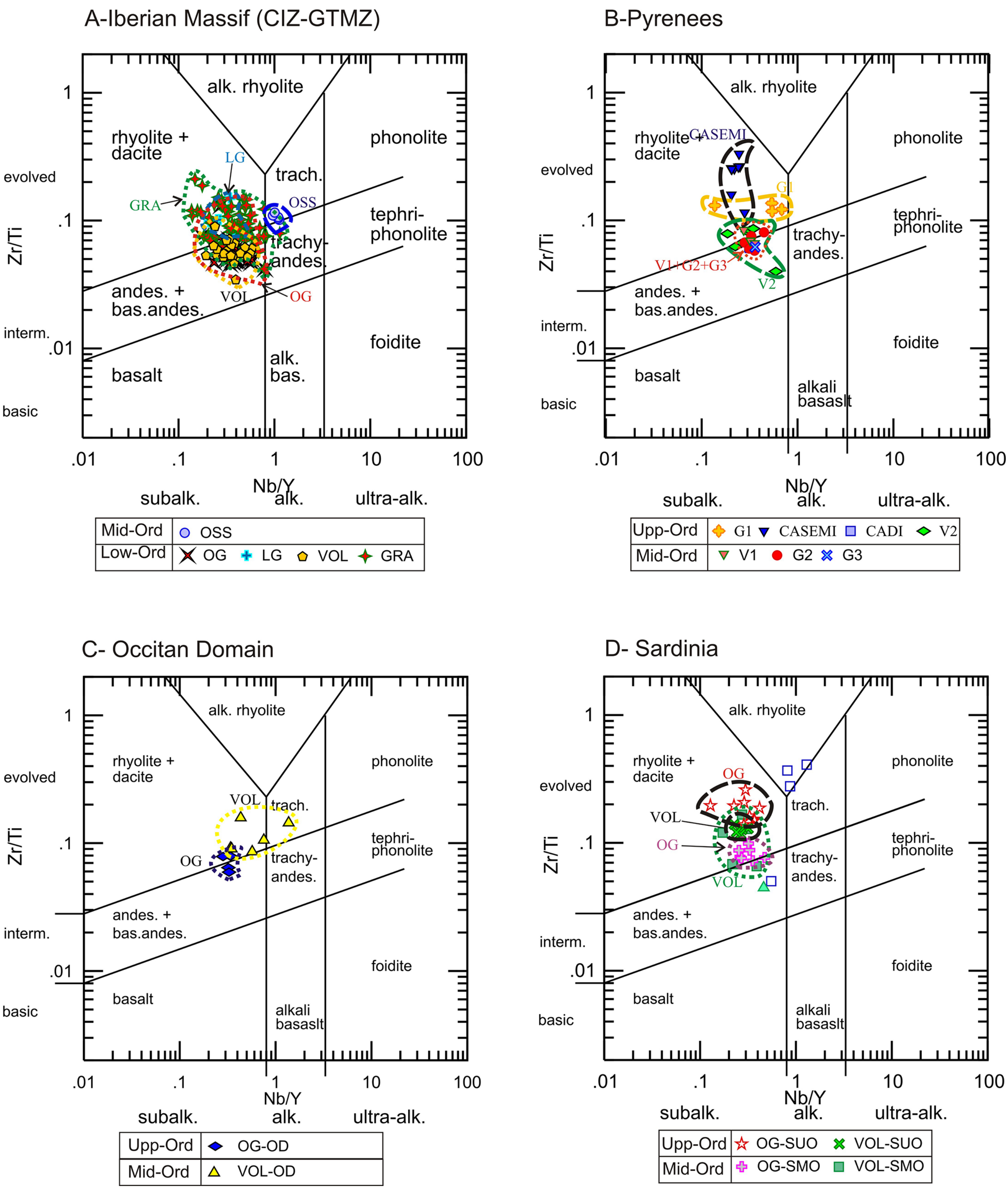


Figure 5. Zr / Ti vs. Nb / Y discrimination diagram (after Winchester and Floyd, 1977; Pearce, 1996). **(a)** Lower–Middle Ordovician rocks of the Iberian Massif (Central Iberian and Galicia–Trás-os-Montes zones). **(b)** Middle–Upper Ordovician rocks of the Eastern Pyrenees. **(c)** Middle Ordovician rocks of the Occitan Domain. **(c–d)** Middle–Upper Ordovician rocks of Sardinia.

dated at ca. 453 Ma; Clariana et al., 2018) (Table 2). The suite is completed with the Somail orthogneisses of the axial Montagne Noire (dated at ca. 450 Ma at Gorges d'Héric; Roger et al., 2004) and the orthogneisses from the Sardinian external zone (dated at ca. 458–457 Ma at Monte Filau; Pavanetto et al., 2012) and the volcanic rocks from the Sardinian nappe zone (Table 2).

4 Geochemical framework

A geochemical comparison between the Furongian–Ordovician felsic rocks of all the above-reported groups offers the opportunity to characterize the successive sources of crust-derived melts along the south-western European margin of Gondwana.

The geochemical features point to a predominance of materials derived from the melting of metasedimentary rocks, rich in SiO_2 and K_2O (average $K_2O / Na_2O = 2.25$) and peraluminous ($0.4 < C_{norm} < 4.5$ and $0.94 < A / CNK > 3.12$), with only three samples with $A / CNK < 1$ (samples 10 0786 of the Casemí subgroup, and T26 and T27 of the San Sebastián subgroup).

The result of plotting the REE content vs. average values of continental crust (Rudnick and Gao, 2003; Fig. 6) yields a flat spectrum and a base level shared by most of the considered groups. The total content in REE is moderate to high (average REE = 176 ppm, ranging between 482.2 and 26.0 ppm; Fig. 7), with a maximum in the subgroup of the Middle Ordovician volcanic rocks from Sardinia (average REE = 335 ppm, VOL-SMO), and with light rare-earth element (LREE) values more fractionated than heavy rare-earth element (HREE) ones, and negative anomalies of Eu, which would indicate a characteristic process of magmatic evolution with plagioclase fractionation. These features are common in peraluminous granitoids.

All subgroups display similar chondrite-normalized REE patterns (Fig. 7), with an enrichment in LREE relative to HREE, which should indicate the involvement of crustal materials in their parental magmas. Nevertheless, some variations can be highlighted, such as the lesser fractionation in REE content of some subgroups. These are the leucogneisses from the Iberian Massif (LG, $La / Yb_n = 2.01$), the Upper Ordovician orthogneisses from Sardinia (OG-SUO, $La / Yb_n = 2.94$), the Casemí orthogneisses ($La / Yb_n = 4.42$) and the Middle Ordovician volcanic rocks from Sardinia (OG-SUO, $La / Yb_n = 2.94$). This may be interpreted as a greater degree of partial fusion in the origin of their parental magmas (Rollinson, 1993).

There are three geochemical groups displaying $(Gd / Yb)_n$ values > 2 and $(La / Yb)_n$ values ≥ 9. These groups are OSS (Central Iberian Zone), VOL-OD (Occitan Domain) and G1 (Pyrenees), and they share higher-alkalinity features.

Some V1 rocks from the Pyrenees (Pierrefite Formation) show no negative anomalies in Eu. Their parental magmas

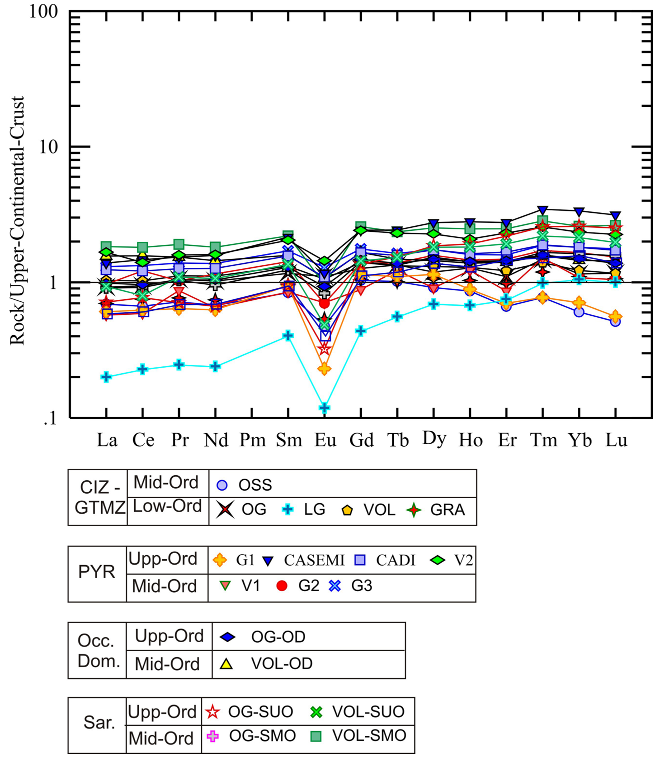
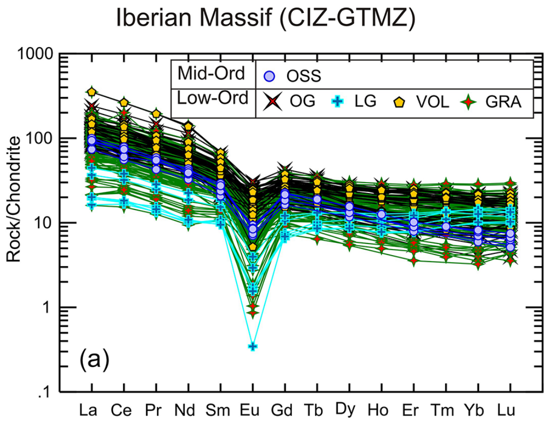


Figure 6. Upper-crust-normalized REE patterns (Rudnick and Gao, 2003) with average values for all distinguished groups; symbols as in Fig. 4.

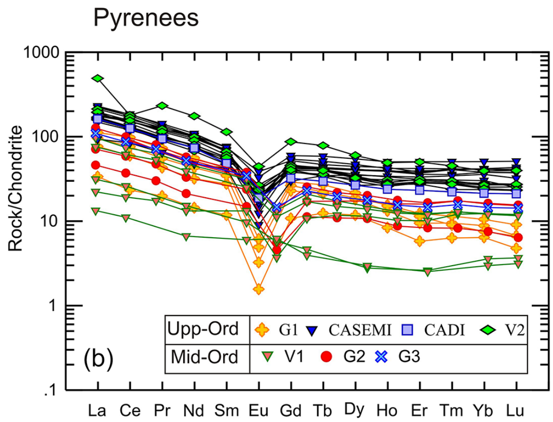
could have been derived from deeper origins and related to residual materials of the lower continental crust, in areas generating K-rich granites (Taylor and McLenan, 1985).

The spider diagrams (Fig. 8), however, exhibit strong negative anomalies in Nb, Sr and Ti, which indicate a distinct crustal affiliation (Díez-Montes, 2007). Only the San Sebastián orthogneisses (OSS) show distinct discrepancies with respect to the remaining samples from the Ollo de Sapo Formation. They display lower negative anomalies in Nb and a more alkaline character by comparison with the rest of the Ollo de Sapo rocks, which point to alkaline affinities and greater negative anomalies in Nb.

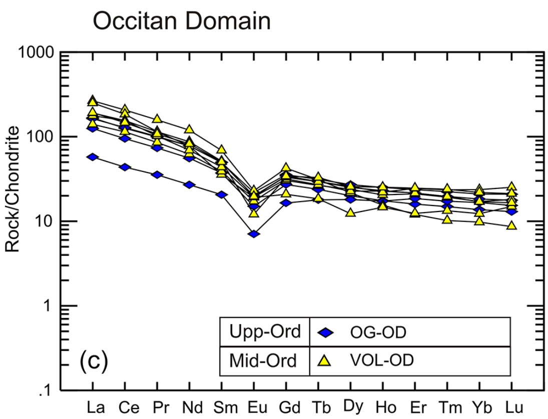
Despite some small differences in the chemical ranges of some major elements, most felsic Ordovician rocks from the Iberian Massif (Central Iberian and Galicia–Trás-os Montes zones), the Eastern Pyrenees, the Occitan Domain and Sardinia share a common chemical pattern. The Lower–Middle Ordovician rocks of the Eastern Pyrenees show less variation in the content of Zr and Nb (Fig. 8b). The volcanic rocks of these groups show a different REE behaviour, which would indicate different sources. Two groups are distinguished in Fig. 7, one with greater enrichment in REE and a negative Eu anomaly, and another with less HREE content and without Eu negative anomalies.



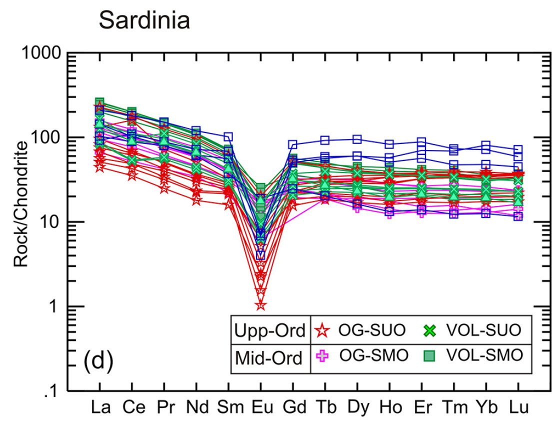
Iberian Massif	$(La/Yb)_n$	$(La/Sm)_n$	Eu/Eu*	$(Gd/Yb)_n$	REE
OSS (n=5)	12.09	3.41	0.36	2.78	110.1
OG (n=47)	9.34	3.35	0.66	1.93	176.4
LG (n=6)	2.01	2.01	0.19	0.68	57.1
GRA (n=71)	8.07	2.84	0.36	1.79	141.7
V1 (n=29)	10.24	3.18	0.55	2.02	191.8



Pyrenees	$(La/Yb)_n$	$(La/Sm)_n$	Eu/Eu*	$(Gd/Yb)_n$	REE
V1 (n=6)	5.85	2.65	0.68	1.32	110.4
G2 (n=6)	6.14	3.14	0.35	1.38	190.1
G3 (n=1)	7.54	3.17	0.51	1.58	242.9
CADÍ (n=2)	7.15	3.28	0.47	1.48	230.2
G1 (n=4)	8.92	2.70	0.15	2.60	120.7
CASEMÍ (n=8)	4.42	2.65	0.35	1.18	301.3
V2 (n=11)	7.18	3.29	0.45	1.64	287.7



Occitain D.	$(La/Yb)_n$	$(La/Sm)_n$	Eu/Eu*	$(Gd/Yb)_n$	REE
OG-OD (n=4)	6.93	3.12	0.46	1.60	186.0
VOL-OD (n=6)	13.04	4.24	0.48	2.06	254.4



Sardinia	$(La/Yb)_n$	$(La/Sm)_n$	Eu/Eu*	$(Gd/Yb)_n$	REE
SUO-OG (n=9)	2.94	3.00	0.18	0.87	170.2
SUO-VOL (n=5)	4.52	2.84	0.24	1.08	188.1
SMO-OG (n=7)	6.21	3.07	0.48	1.46	190.4
SMO-VOL (n=6)	7.62	3.46	0.29	1.63	334.6
SLO-VOL (n=5)	4.72	2.44	0.27	1.28	271.8

Figure 7. Chondrite-normalized REE patterns (Sun and McDonough, 1989) for all study samples.

Figure 9 illustrates how the average of all the considered groups approximates the mean values of the upper continental crust (UCC) of Rudnick and Gao (2003). In this figure, small deviations can be observed, some of them toward lower continental crust (LCC) values and others toward bulk continental crust (BCC), indicating variations in their parental magmas but with quite similar spectra. Overall chondrite-normalized patterns are close to the values that represent the upper continental crust, with slight enrichments in the Th / Nb, Th / La and Th / Yb ratios.

Finally, in the Occitan volcanic rocks (VOL-OD) the rare-earth elements are enriched and fractionated (33.2 ppm < La < 45.6 ppm; 11.2 < La / Yb < 14.5). The upper-continental-crust-normalized diagram exhibits nega-

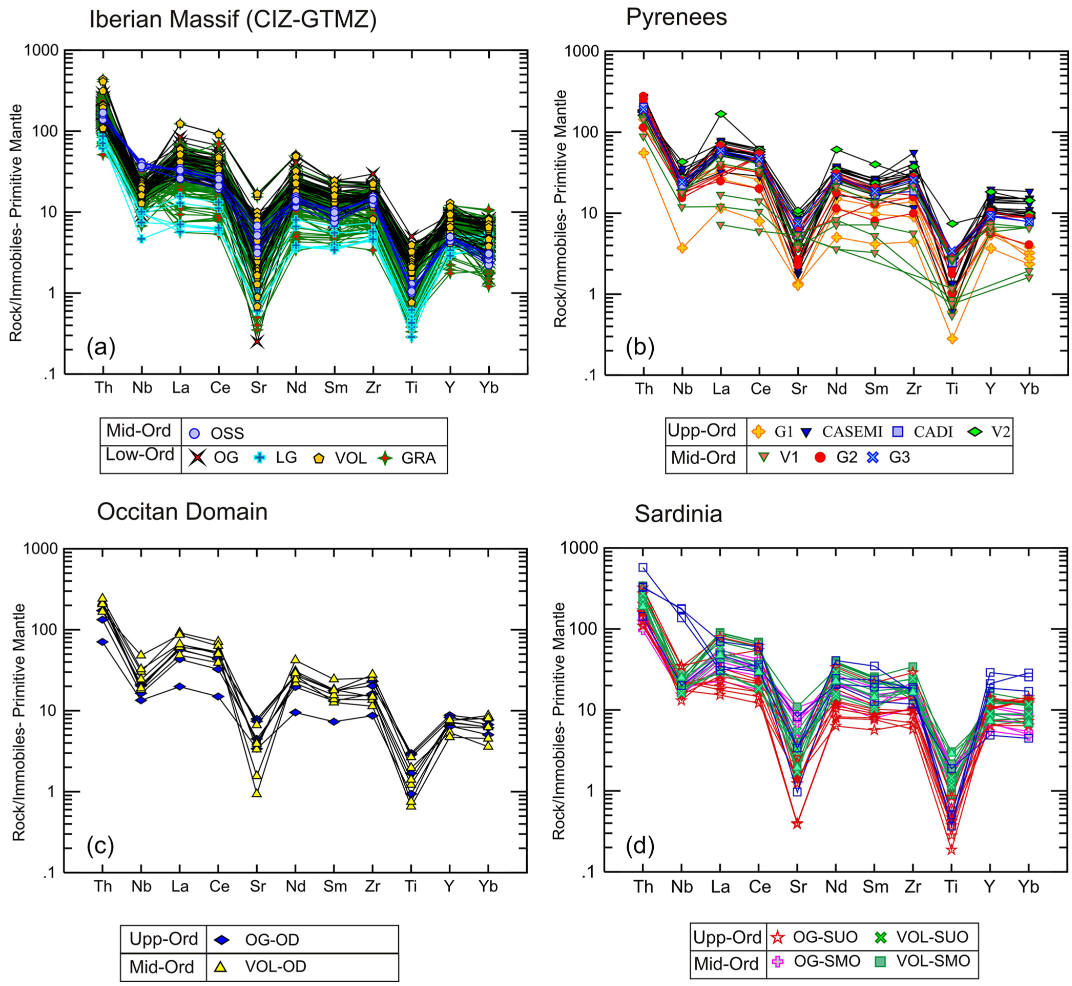


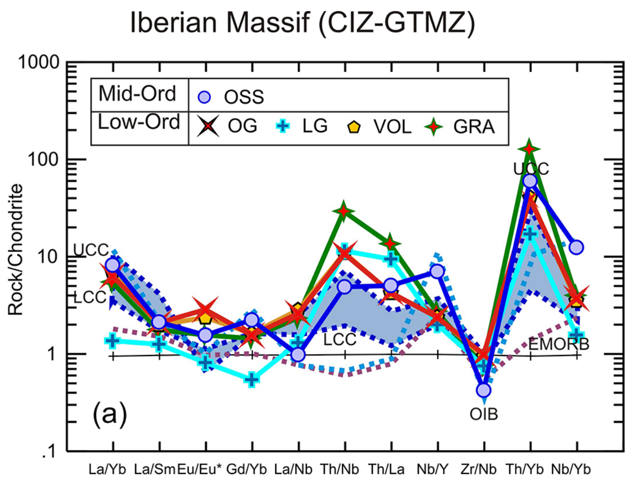
Figure 8. Multi-element diagram normalized to the primitive mantle of Palme and O'Neill (2004) for all study samples.

tive anomalies of Ti, V, Cr, Mn and Fe associated with oxide fractionation; of Zr and Hf linked to zircon fractionation; and of Eu related to plagioclase fractionation. The profiles are comparable to the Vendean Saint-Gilles rhyolitic ones. The Th vs. Rb / Ba features are also similar to those of the Saint-Gilles rhyolites, and the Iberian Ollo de Sapo and Urra rhyolites (Solá et al., 2008; Díez Montes et al., 2010).

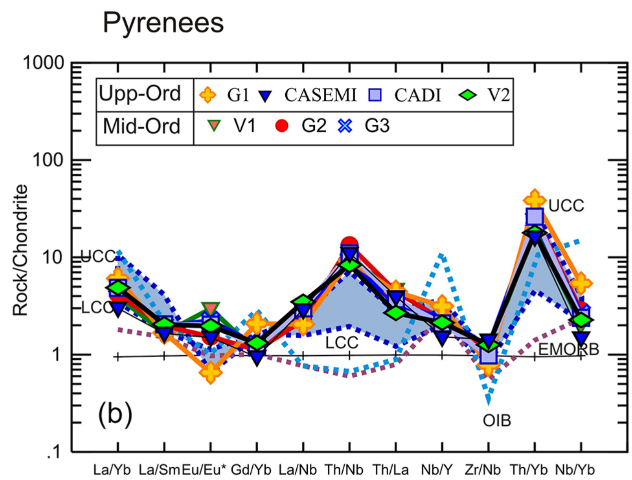
5 Discussion

5.1 Inferred tectonic settings

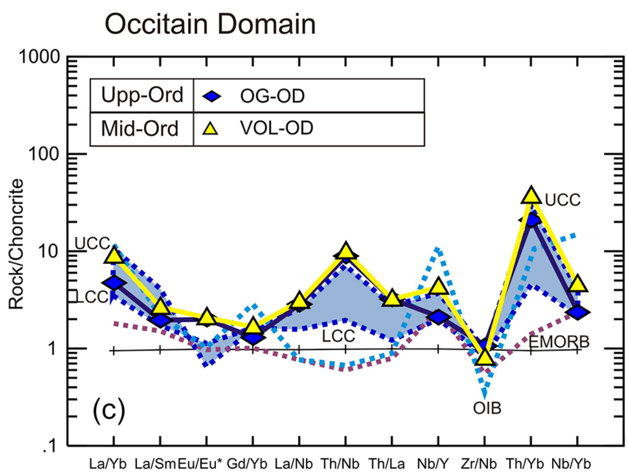
In order to clarify the evolution of geotectonic environments, the data have been represented in different discrimination diagrams. The Zr / TiO_2 ratio (Lentz, 1996; Syme, 1998) is a key index of compositional evolution for intermediate and felsic rocks. In the Syme diagram (Fig. 10), most rocks from the Central Iberian Zone represent a characteristic arc association, although there are some contemporaneous samples characterized by extension-related values (Zr / Ti = 0.10, LG). The rocks of the Middle–Ordovician San Sebastián orthogneisses (OSS) show values of Zr / Ti = 0.08, intermediate between extensional and arc conditions. This could be interpreted as a sharp change in geotectonic conditions toward the Mid-Ordovician (Fig. 10a). For a better comparison, the samples of the San Sebastián orthogneisses (OSS) and the granites (GRA) have been distinguished with a shaded area in all the diagrams, since they have slightly different characteristics to the rest of the samples from the Ollo de Sapo Group. The samples G1 (Pyrenees) and VOL (Central Iberian Zone) broadly share similar values, as a result of which the three latter groups (OSS, G1 and VOL) are arranged following a good-correlation line. The same trend seems to be inferred in the Eastern Pyrenees (Fig. 10b), where the Middle Ordovician subgroups display arc features, but half of the Upper Ordovician subgroups show extensional affinities (G1 and Casemí orthogneisses). In the case of the Occitan orthogneisses (Fig. 10c), they show arc characteristics, which contrast with the contemporaneous volcanic rocks displaying extensional values of Zr / Ti = 0.10. This disparity



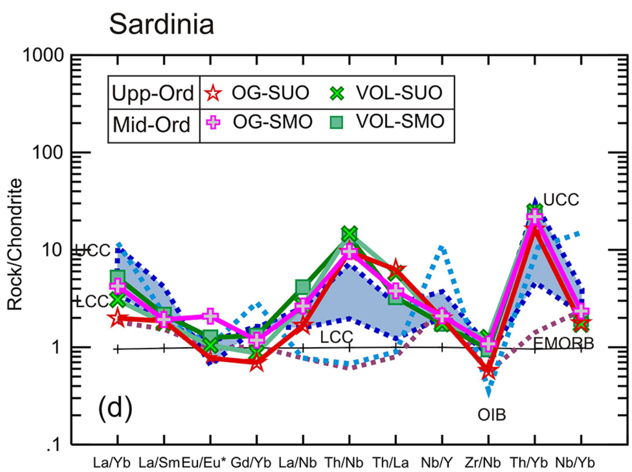
Iberian M.	$(La/Yb)_n$	$(La/Sm)_n$	Eu/Eu*	$(Gd/Yb)_n$	La/Nb	Th/Nb	Th/La	Nb/Y	Zr/Nb	Th/Yb	Nb/Yb
OSS (n=5)	12.09	3.41	0.36	2.78	0.97	0.59	0.61	1.06	6.75	10,82	18.58
OG (n=47)	9.34	3.35	0.66	1.93	2.59	1.29	0.51	0.36	15.74	7.05	5.56
LG (n=6)	2.01	2.01	0.19	0.68	1.30	1.38	1.14	0.30	12.03	3.09	2.34
GRA (n=71)	8.07	2.84	0.36	1.79	2.31	3.51	1.63	0.41	12.08	22.96	5.96
V1 (n=29)	10.24	3.18	0.55	2.02	2.81	1.32	0.51	0.36	15.05	7.32	5.27



Pyrenees	$(La/Yb)_n$	$(La/Sm)_n$	Eu/Eu*	$(Gd/Yb)_n$	La/Nb	Th/Nb	Th/La	Nb/Y	Zr/Nb	Th/Yb	Nb/Yb
V1 (n=6)	5.85	2.65	0.68	1.32	2.43	1.13	0.56	0.32	17.66	3.50	3.06
G2 (n=6)	6.14	3.14	0.35	1.38	2.23	1.59	0.53	0.34	17.69	4.72	4.25
G3 (n=1)	7.54	3.17	0.51	1.58	2.83	1.11	0.39	0.36	19.35	4.37	3.94
CADÍ (n=2)	7.15	3.28	0.47	1.48	2.96	1.32	0.45	0.31	15.71	4.72	3.57
G1 (n=4)	8.92	2.70	0.15	2.60	2.04	1.11	0.53	0.47	12.50	6.94	8.07
CASEMÍ (n=8)	4.42	2.65	0.35	1.18	2.91	1.33	0.48	0.23	22.79	2.95	2.25
V2 (n=11)	7.18	3.29	0.45	1.64	3.46	1.01	0.32	0.32	20.17	3.23	3.40



Occitain D.	$(La/Yb)_n$	$(La/Sm)_n$	Eu/Eu*	$(Gd/Yb)_n$	La/Nb	Th/Nb	Th/La	Nb/Y	Zr/Nb	Th/Yb	Nb/Yb
OG-OD (n=4)	6.93	3.12	0.46	1.60	2.81	1.07	0.39	0.32	17.13	3.87	3.56
VOL-OD (n=6)	13.04	4.24	0.48	2.06	3.02	1.18	0.38	0.64	12.61	6.59	6.74



SUO-OG (n=9)	2.94	3.00	0.18	0.87	1.67	1.16	0.76	0.30	9.23	2.94	2.66
SUO-VOL (n=5)	4.52	2.84	0.24	1.08	2.67	1.75	0.69	0.26	20.07	4.46	2.55
SMO-OG (n=7)	6.21	3.07	0.48	1.46	2.62	1.15	0.45	0.31	17.18	3.94	3.50
SMO-VOL (n=6)	7.62	3.46	0.29	1.63	4.09	1.63	0.39	0.26	15.06	4.33	2.89

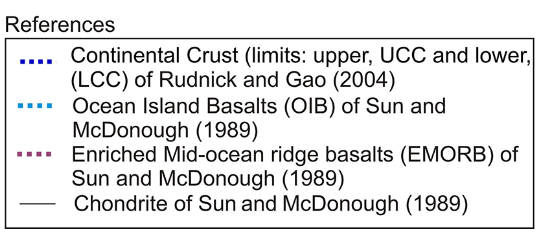


Figure 9. Chondrite-normalized isotope ratio patterns (Sun and McDonough, 1989) for standard comparison for all study samples. Blue area: limits of continental crust values (lower and upper) of Rudnick and Gao (2003).

between plutonic and volcanic rocks could be interpreted as different conditions for the origin of these magmas. In Sardinia (Fig. 10d), the same evolution from arc to extensional conditions is highlighted for the Upper Ordovician samples, although some Middle Ordovician volcanic rocks already shared extensional patterns (Zr / Ti = 0.09). In summary, there seems to be a geochemical evolution in the Ordovician magmas grading from arc to extensional environments.

In the Nb–Y tectonic discrimination diagram of Pearce et al. (1984) (Fig. 11), most samples are plotted in the volcanic-arc type, though some subgroups are projected in the within-plate and anomalous ocean ridge granites (ORG). The majority of samples display very similar Zr / Nb and Nb / Y ratios, typical of island arc or active continental-margin rhyolites (Díez-Montes et al., 2010). Only some samples are plotted separately: OSS samples with the highest Nb contents (> 20 ppm) and some volcanic rocks of the Occitan Domain (average Nb = 16.87 ppm). In the Eastern Pyrenees, the Middle Ordovician rocks are plotted in the volcanic-arc field, whereas the Upper Ordovician ones point to the ORG type, except the Casemí samples. This progress of magmatic sources agrees with the evolution seen in Fig. 10. In the Occitan Domain, VOL-OD samples share values with those of the San Sebastián orthogneiss, while OG-OD shares values with those of OG from the Central Iberian Zone.

The Zr–Nb diagram (Leat et al., 1986; modified by Piercey, 2011) (Fig. 13) illustrates how magmas evolved toward richer values in Zr and Nb, which is consistent with what is observed in the Syme diagram (Fig. 10). Figure 12a documents how most samples show a generally positive correlation. These different groups correspond to the OSS and Portalegre granites, highlighted in the figure. The two groups indicate a tendency toward alkaline magmas. Some samples – such as the Pyrenean G1, some Occitan VOL-OD samples and some Sardinian OG-SUO samples – share the same affinity, clearly distinguished from the general geochemical trend exhibited by the Central Iberian Zone.

On a Zr vs. Ga / Al diagram (Whalen et al., 1987) (Fig. 13), the samples depict an intermediate character between anorogenic or alkaline (A type) and orogenic (I and S type). In the Central Iberian Zone, samples from the San Sebastián orthogneisses and Portalegre granites show characters of A-type granites, while the remaining samples display affinities of I- and S-type granites. For the Central Iberian Zone, a clear magmatic shift toward more extensional geotectonic environments is characterized. For the Eastern Pyrenees, we find the same situation as in the Central Iberian Zone, with a magmatic evolution toward A-type granite characteristics, indicating more extensional geotectonic environments. In the Occitan Domain, the samples show a clear I- and S-type character. In the Sardinian case, the same seems to happen as in the Central Iberian Zone: the Upper Ordovician orthogneisses suggest a more extensional character.

In summary, all the reported diagrams point to a magmatic evolution through time, grading from arc to extensional geotectonic environments (with increased Zr / Ti ratios) and to A-type granite characteristics. This geotectonic framework is consistent with that illustrated in Fig. 10. The geochemical characters of these rocks show a rhyodacite-to-dacite composition, peraluminous and calc-alkaline K-rich character, and a volcanic-arc affinity for most of samples but without intermediate rocks associated with andesitic types. Hence a change in time is documented toward more alkaline magmas.

5.2 Interpretation of $_{\varepsilon}$Nd values

$_{\varepsilon}\mathrm{Nd}_{(t)}$ values are useful for interpreting the nature of magmatic sources. Most samples of the above-reported groups show no significant differences in isotopic $_{\varepsilon}\mathrm{Nd}_{(t)}$ values and $\mathrm{Nd}_{\mathrm{CHUR}}$ model ages (Fig. 14). Some exceptions are related to granites from the southern Central Iberian Zone, which display positive values (from +2.6 to −2.4) and T_{DM} values from 0.90 to 3.46 Ga. These granites, space-related to calc-alkaline diorites and gabbros, were interpreted by Solá et al. (2008) as the result of underplating and temporal storage of mantle-derived magmas as a potential source for the intrusive "orogenic melts" during early Palaeozoic extension.

Some samples from (i) the Central Iberian Zone, such as VI-3 (Leucogneiss subgroup) and PORT2 and PORT15 (Granite subgroup); (ii) the Eastern Pyrenees, such as samples 99338 (G1 subgroup) and 100786 (Casemí subgroup); and (iii) the Sardinian CS5, CS8 and CC5 samples (Upper Ordovician orthogneiss subgroup) display anomalous T_{DM} values and $^{147}\mathrm{Sm}/^{144}\mathrm{Nd}$ ratios > 0.17 (Table 2; Fig. 14), a characteristic relatively common in some felsic rocks (DePaolo, 1988; Martínez et al., 2011). According to Stern (2002), these values should not be considered, but a possible explanation for these high ratios may be related to the M-type tetrad effect (e.g. Irber, 1999; Monecke et al., 2007; Ibrahim et al., 2015), which affects REE fractionation in highly evolved felsic rocks due to the interaction with hydrothermal fluids. This process can be reflected as an enrichment of Sm related to Nd. Other authors, however, explain this enrichment as a result of both magmatic evolution (e.g. McLennan, 1994; Pan, 1997) and weathering processes after exhumation (e.g. Masuda and Akagi, 1989; Takahashi et al., 2002).

In the granites of the southern Central Iberian Zone and the volcanic rocks of Sardinia, positive values in $_{\varepsilon}\mathrm{Nd}_{(t)}$ could be interpreted as a more primitive nature of their parental magmas, even though the samples with highest T_{DM} values are those that display higher $^{147}\mathrm{Sm}/^{144}\mathrm{Nd}$ ratios (> 0.17; Table 2).

The volcanic rocks of the Central Iberian Zone display some differences following a N–S transect, with $_{\varepsilon}\mathrm{Nd}_{(t)}$ values being less variable in the north ($_{\varepsilon}\mathrm{Nd}_{(t)}$: −4.0 to −5.0) than in the south ($_{\varepsilon}\mathrm{Nd}_{(t)}$: −1.6 to −5.5). The isotopic signature of the volcaniclastic Urra rocks is compatible with mag-

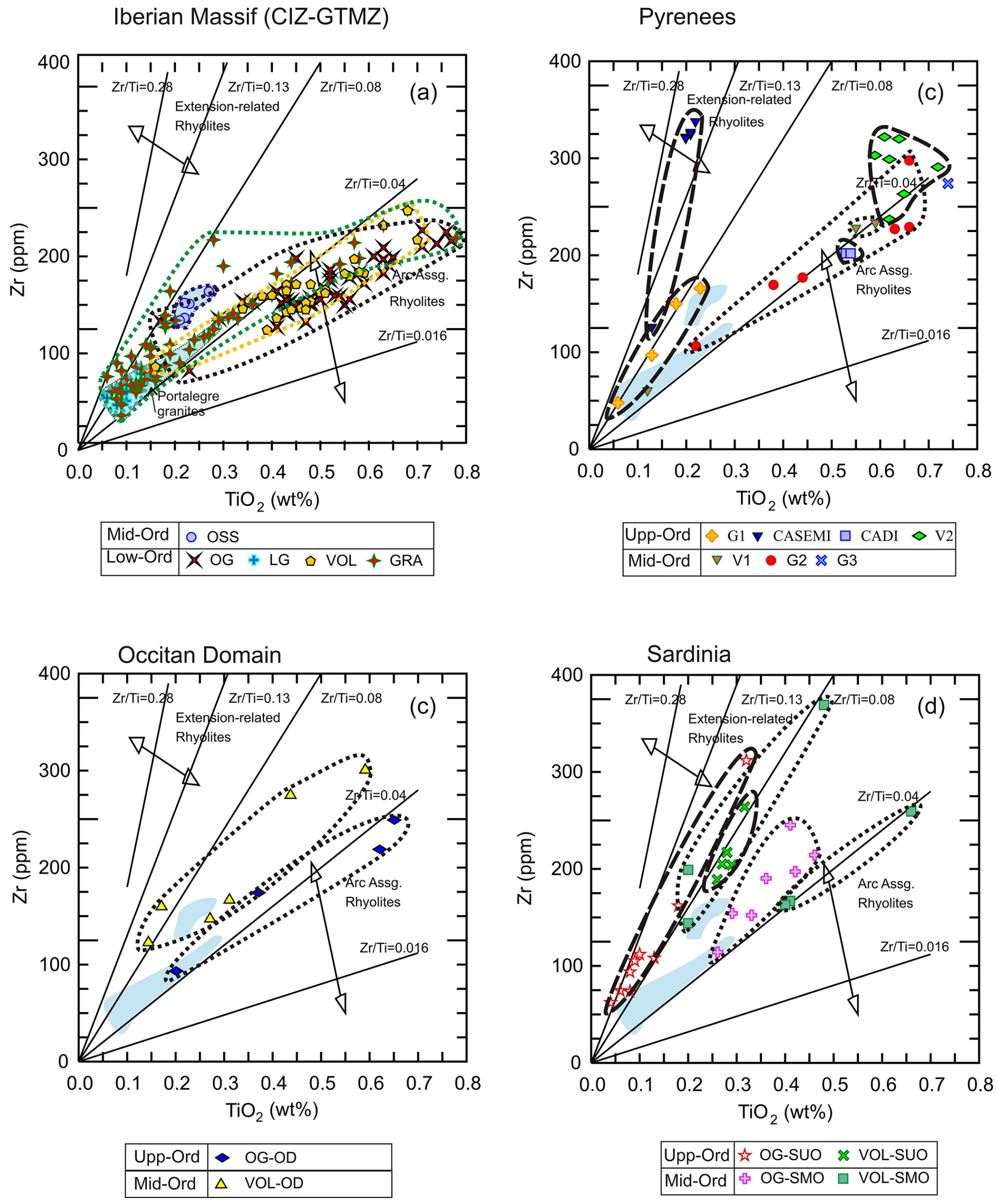


Figure 10. Tectonic discrimination diagram of Zr vs. TiO_2 (Syme, 1998) for all study samples. Double-sided arrows indicate ranging of different fields: rhyolites in tholeiitic and calc-alkaline arc suites have Zr / TiO_2 ratios ranging from about 0.016 to 0.04, and extension-related rhyolites from about 0.13 to 0.28 (Syme, 1998).

mas derived from young crustal rocks, with intermediate-to-felsic igneous compositions (Solá et al., 2008). The volcanic rocks of the northern Central Iberian Zone could be derived from older crustal rocks (Montero et al., 2007). The isotopic composition of the granitoids from the southern Central Iberian Zone has more primitive characteristics than those of the northern Central Iberian Zone, suggesting different sources for both sides (Talavera et al., 2013). OSS shows lower inheritance patterns, more primitive Sr–Nd isotopic composition than other rocks of the Ollo de Sapo suite, and an age some 15 million years younger than most meta-igneous rocks of the Sanabria region (Montero et al., 2009), likely reflecting a greater mantle involvement in its genesis (Díez-Montes, 2007).

According to Talavera et al. (2013), the Cambro-Ordovician rocks of the Galicia–Trás-os-Montes Zone schistose area and the magmatic rocks of the northern Central Iberian Zone are contemporary. Both metavolcanic and metagranitic rocks almost share the same isotopic compositions.

The Upper Ordovician orthogneisses from the Occitan Domain show very little variation in $_{\varepsilon}Nd_{(t)}$ values (−3.5 to −4.0), typical of magmas derived from young crustal rocks. The variation in depleted mantle model age (TDM) values is

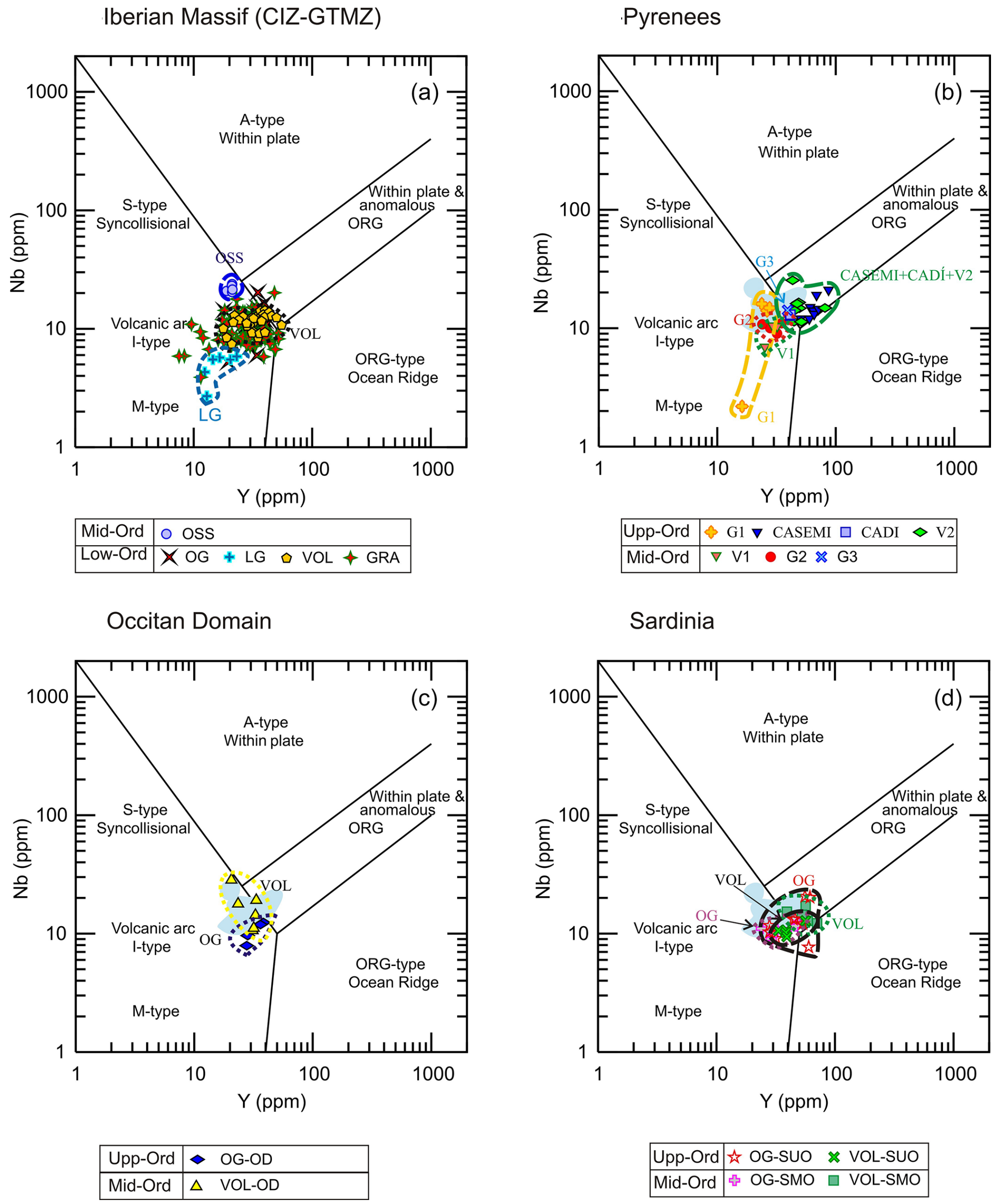


Figure 11. Tectonic discrimination diagram of Y vs. Nb (Pearce et al., 1984) for all study samples.

also small (1.4 to 1.8 Ga), indicating similar crustal residence times to other rock groups.

In Sardinia, $_{\varepsilon}Nd_{(t)}$ values present a greater variation (−1.6 to −3.3), but they are also included in the typical continental crust range. As noted above, anormal TDM values (between 1.2 and 4.5 Ga) may be due to post-magmatic hydrothermal alteration processes.

6 Geodynamic setting

In the Iberian Massif, the Ediacaran–Cambrian transition was marked by paraconformities and angular discordances indicating the passage from Cadomian volcanic arc to rifting conditions. The axis of the so-called Ossa-Morena Rift lies along the homonymous zone (Quesada, 1991; Sánchez-García et al., 2003, 2008, 2010) close to the remains of the Cadomian suture (Murphy et al., 2006). Rifting conditions were accompanied by a voluminous magmatism that changed from peraluminous acid to bimodal (Sánchez-García et al., 2003, 2008, 2016, 2019). Some authors (Álvaro et al., 2014a; Sánchez-García et al., 2019) propose that this rift resulted from a SW-to-NE inward migration, toward innermost parts of Gondwana, of rifting axes from the Anti-Atlas in Morocco to the Ossa-Morena Zone in the Iberian Massif. According to this proposal the rift developed later (in Cambro-Ordovician times) in the Iberian, Armorican and Bohemian massifs.

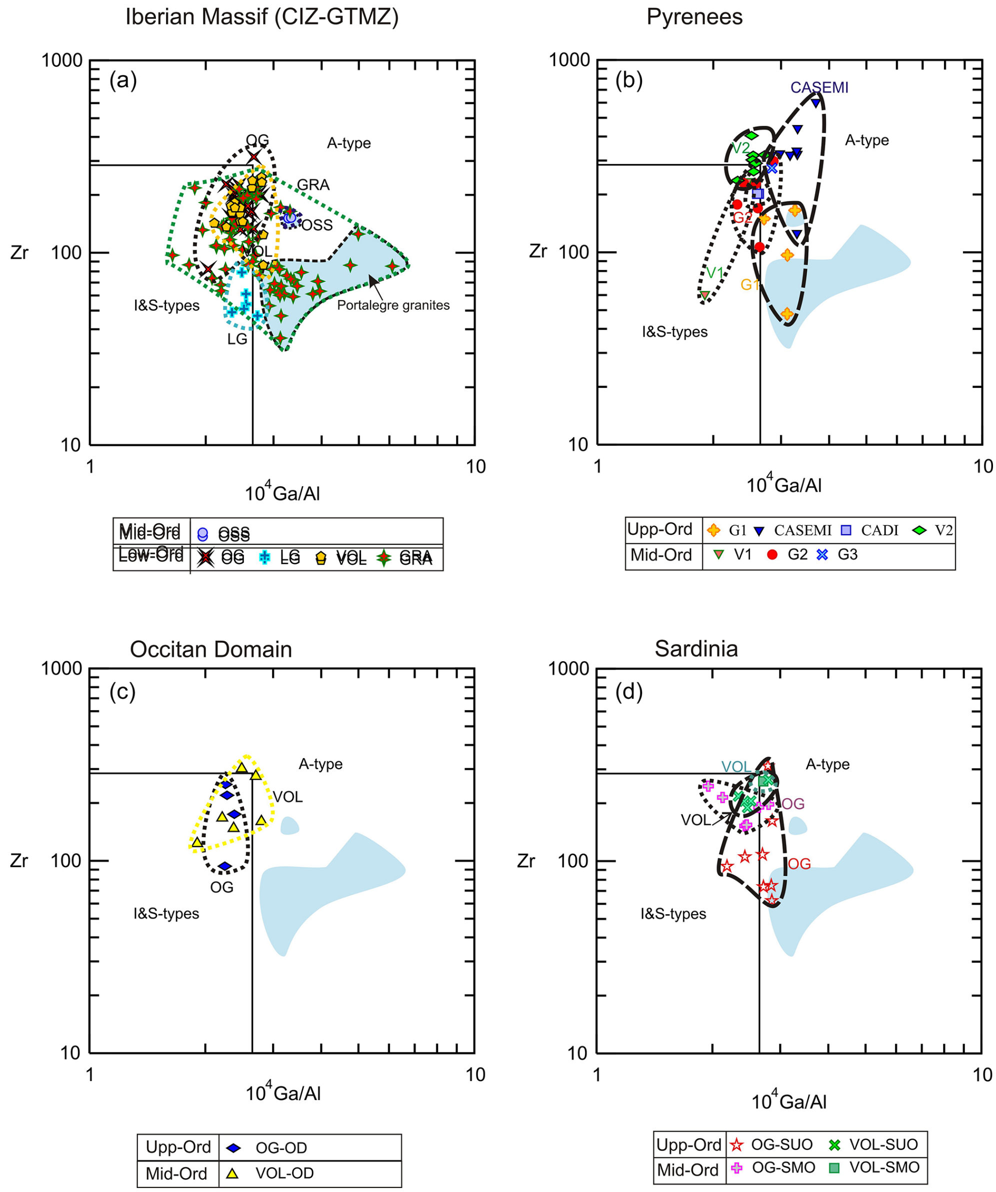


Figure 12. Zr vs. 10^4 Ga / Al discrimination diagram (Whalen et al., 1987) for all study samples.

The Furongian–Ordovician transition to drifting conditions is associated – in the Iberian Massif, the Occitan Domain, the Pyrenees and Sardinia – with stepwise magmatic activity contemporaneous with the record of the Toledanian and Sardic unconformities. These, related to neither metamorphism nor penetrative deformations, are linked to uplift, erosion and irregularly distributed mesoscale deformation that gave rise to angular unconformities up to 90°. The time span involved in these gaps is similar (22 million years in the Iberian Massif, 16–23 million years in the Pyrenees and 18 million years in Sardinia). This contrasts with the greater time span displayed by the magmatic activity (30–45 million years), which started before the unconformity formation (early Furongian in the Central Iberian Zone vs. Floian in the Pyrenees, Occitan Domain and Sardinia), continued during the unconformity formation (Furongian and early Tremadocian in the Central Iberian Zone vs. Floian–Darriwilian in the Pyrenees, Occitan Domain and Sardinia) and ended during the sealing of the uplifted and eroded palaeorelief (Tremadocian–Floian volcaniclastic rocks at the base of the Armorican Quartzite in the Central Iberian Zone vs. Sandbian–Katian volcanic rocks at the lowermost part of the Upper Ordovician successions in the Pyrenees, Occitan Domain and Sardinia; Gutiérrez-Alonso et al., 2007, 2016; Navidad et al., 2010; Martínez et al., 2011; Álvaro et al., 2016; Martí et al., 2019). In the Pyrenees, Upper Ordovician magmatism and sedimentation coexist with normal faults controlling marked thickness changes of the basal Upper Or-

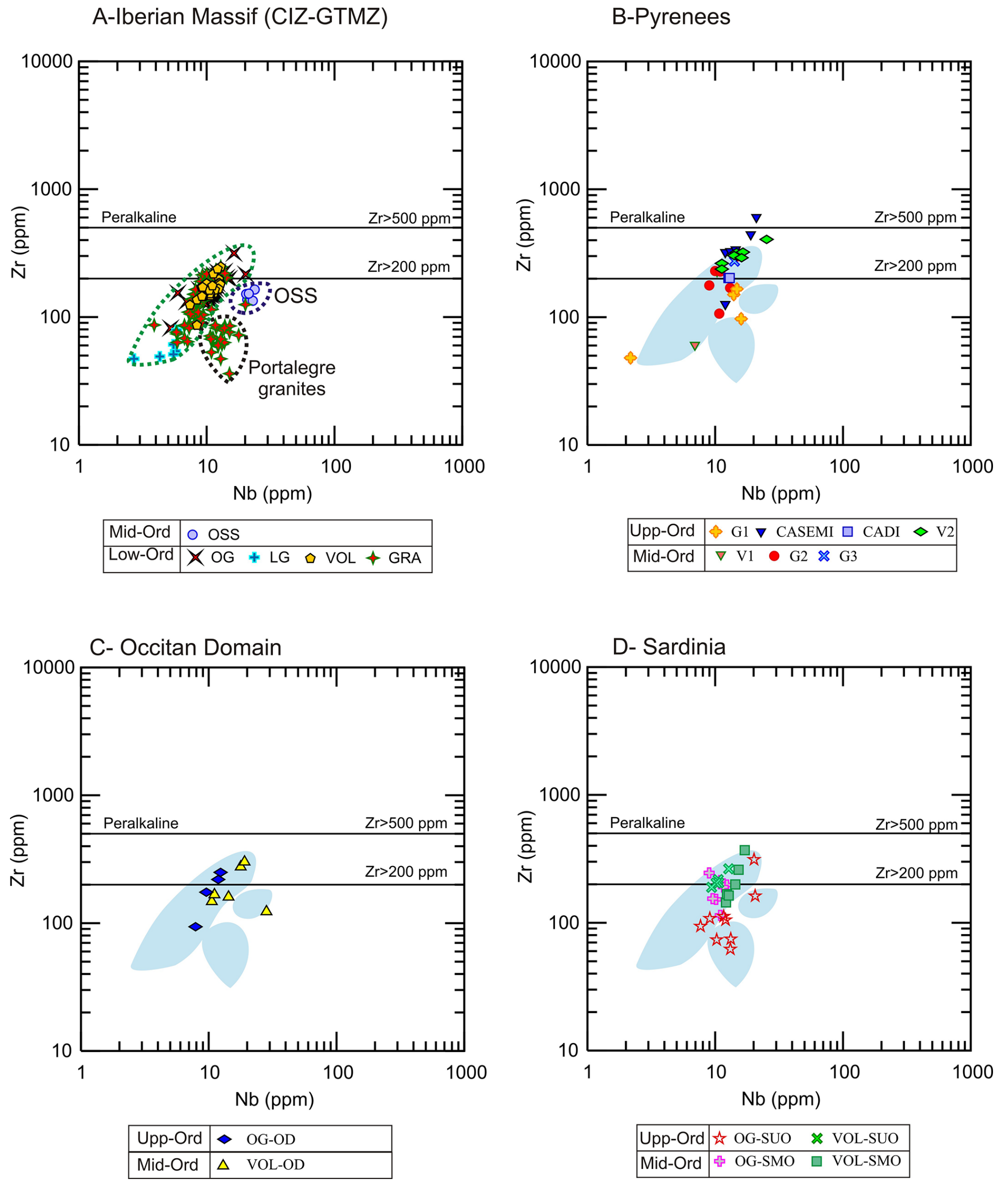


Figure 13. Zr–Nb plot diagram (Leat et al.,1986; modified by Piercey, 2011) for all study samples.

dovician succession and cutting the lower part of this succession, the Sardic unconformity and the underlying Cambro-Ordovician sequence (Puddu et al., 2018, 2019).

Although the Toledanian and Sardic phases reflect similar geodynamic conditions in two distinct palaeogeographic areas, at present forming the western and eastern branches of the Variscan Ibero-Armorican Arc, they display different peaks in magmatic activity with minor chronological overlapping (Fig. 3). This may reflect a SW-to-NE "zip-like" propagation of the latest Ediacaran–Terreneuvian rift axes in the so-called Atlas–Ossa-Morena Rift.

6.1 Toledanian phase

The Early Ordovician (Toledanian) magmatism of the Central Iberian Zone evolved to a typical passive-margin setting, with geochemical features dominated by acidic rocks, peraluminous and rich in K, and lacking any association with basic or intermediate rocks. Some of the orthogneisses of the Galicia–Trás-os-Montes Zone basal and allochthonous complex units share these same patterns. This fact has been interpreted by some authors as a basin environment subject to important episodes of crustal extension (Martínez-Catalán et al., 2007; Díez-Montes et al., 2010). In contrast, Villaseca et al. (2016) interpreted this absence as evidence against rifting conditions, though the absence of contemporary basic mag-

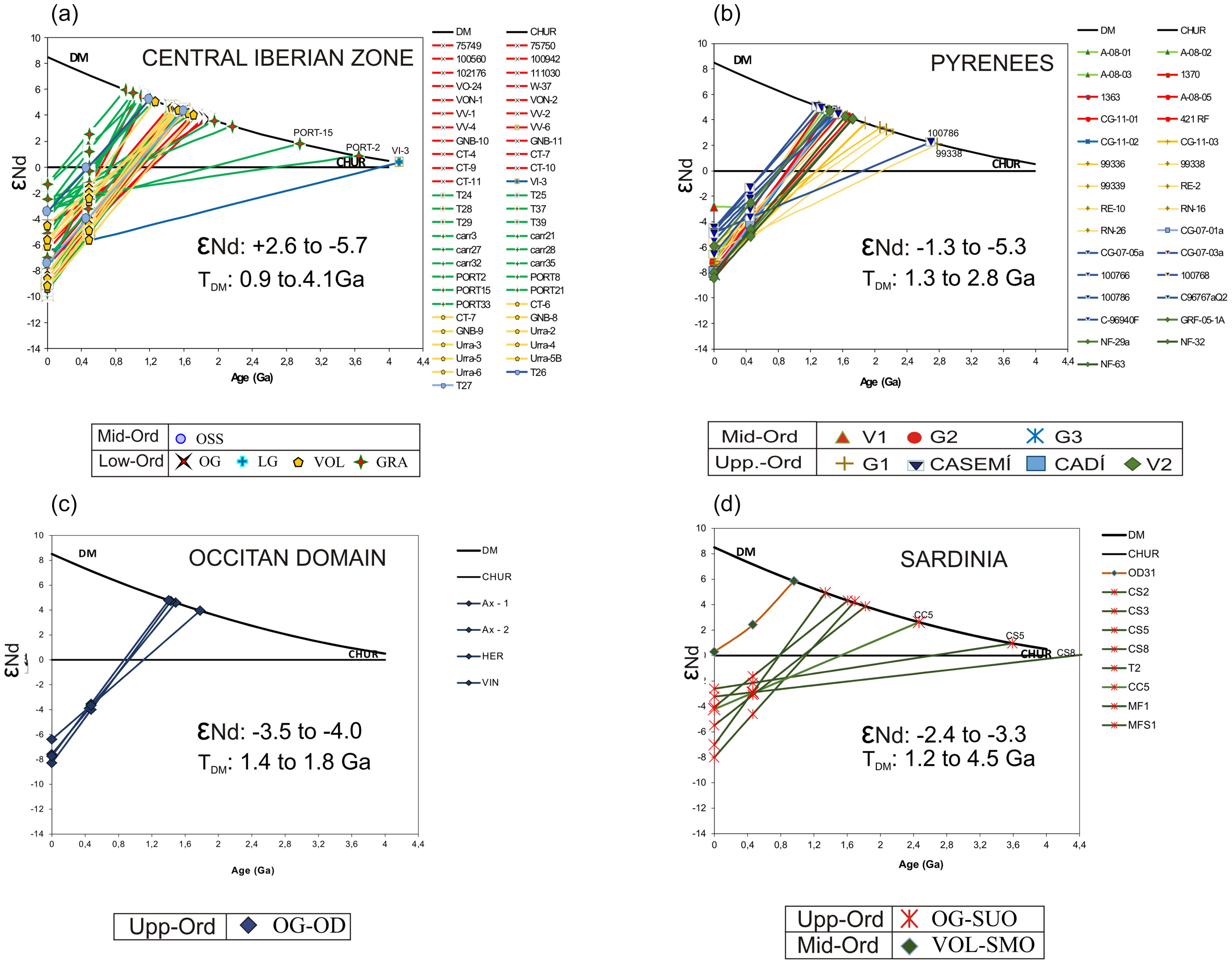


Figure 14. $_{\varepsilon}\mathrm{Nd}_{(t)}$–age diagram (DePaolo and Wasserburg, 1976; DePaolo, 1981) for all study samples. **(a)** Central Iberian and Galicia–Trás-os-Montes zones. **(b)** Eastern Pyrenees. **(c)** Occitan Domain. **(d)** Sardinia. See references in the text.

matism may be explained by the partial fusion of a thickened crust, through recycling of Neoproterozoic crustal materials. The thrust of a large metasedimentary sequence could generate dehydration and metasomatism of the rocks above this sequence, triggering partial fusion at different levels, although the increase in peraluminosity with the basicity of the orthogneisses is against any assimilation-fractional crystallization process involving mantle materials. However, this increase in peraluminosity with the basicity has not been revealed in the samples studied above. Following the model of Villaseca et al. (2016), a flat subduction of the southern part of the Central Iberian Zone would have taken place under its northern prolongation, whereas the reflection of such a subduction is not evident in the field. The calc-alkaline signature of this magmatism has also been taken into account as proof of its relationship with volcanic-arc environments (Valverde-Vaquero and Dunning, 2000). However, calc-alkaline features may also be interpreted as a result of variable degrees of continental crust contamination and/or a previously enriched mantle source (Sánchez-García et al., 2003, 2008, 2016, 2019; Díez-Montes et al., 2010). Finally, other granites not considered here of Tremadocian age have been reported in the southern Central Iberian Zone, such as the Oledo Massif and the Beira Baixa–central Extremadura, which display an I-type affinity (Antunes et al., 2009; Rubio Ordóñez et al., 2012). These granites could represent different sources for the Ordovician magmatism in the Central Iberian Zone.

Sánchez-García et al. (2019) have proposed that the anomaly that produced the large magmatism throughout the Iberian Massif could have migrated from the rifting axis to inward zones, and the acidic, peraluminous, K-rich rocks of Mid Ordovician age should represent the initial stages of a new rifting pulse, resembling the peraluminous rocks of the

Early Rift Event (sensu Sánchez-García et al., 2003) from the Cambrian Epoch 2 of the Ossa-Morena Rift.

In the parautochthon of the Galicia–Trás-os-Montes Zone, the appearance of tholeiitic and alkaline–peralkaline magmatism in the Middle Ordovician would signal the first steps toward extensional conditions (Díez Fernández et al., 2012; Dias da Silva et al., 2016). In the Montagne Noire and the Mouthoumet massifs, contemporaneous tholeiitic lava indicates a similar change in the tectonic regimen (Álvaro et al., 2016). This gradual change in geodynamic conditions is also marked by the appearance of rocks with extensional characteristics in some of the subgroups considered here, such as the Central Iberian Zone (San Sebastián orthogneisses), Eastern Pyrenees (Casemí orthogneisses and G1), volcanic rocks of the Occitan Domain, and the orthogneisses and volcanic rocks from Sardinia.

6.2 Sardic phase

In the Eastern Pyrenees, two peaks of Ordovician magmatic activity are observed (Casas et al., 2019). Large Lower–Middle Ordovician peraluminous granite bodies are known, representing the protoliths of numerous gneissic bodies with laccolithic morphologies. In the Canigó Massif, the Upper Ordovician granite bodies (protoliths of Cadí and Casemí, G1) are encased in sediments of the Canaveilles and Jujols groups. During this time span, there was generalized uplift and erosion that culminated with the onset of the Sardic unconformity. The Sardic phase was succeeded by an extensional interval related to the formation of normal faults affecting the pre-unconformity strata (Puddu et al., 2018, 2019). The volcanic-arc signature can be explained by crustal recycling (Navidad et al., 2010; Casas et al., 2010; Martínez et al., 2011), as in the case of the Toledanian phase in the Central Iberian Zone, although, according to Casas et al. (2019), the Pyrenees and the Catalan Coastal Range were probably fringing the Gondwana margin in a different position than that occupied by the Iberian Massif. As a whole, the Ordovician magmatism in the Pyrenees lasted about 30 million years, from ca. 477 to 446 Ma, in a time span contemporaneous with the formation of the Sardic unconformity (Fig. 2). Recently, Puddu et al. (2019) proposed that a thermal doming, bracketed between 475 and 450 Ma, could have stretched the Ordovician lithosphere. The emersion and denudation of the inherited Cambrian–Ordovician palaeorelief would have given rise to the onset of the Sardic unconformity. According to these authors, thermal doming triggered by hot mafic magma underplating may also be responsible for the late Early–Late Ordovician coeval magmatic activity.

In the Occitan Domain, there was a dramatic volcanic event in early Tremadocian times, with the uprising of basin floors and the subsequent effusion of abundant rhyolitic activities under subaerial explosive conditions (Larroque volcanosedimentary complex in the Montagne Noire and Davejean acidic volcanic counterpart in the Mouthoumet Massif). Pouclet el al. (2017) interpreted this as a delayed Ollo de Sapo-style outpouring where a massive crustal melting required a rather significant heat supply. Asthenospheric upwelling leading to the interplay of lithospheric doming, continental break-up and decompression-driven mantle melting can explain such a great thermal anomaly. The magmatic products accumulated on the mantle–crust contact would provide enough heat transfer for crustal melting (Huppert and Sparks, 1988). Subsequently, a post-Sardic reactivation of rifting conditions is documented in the Cabrières klippes (southern Montagne Noire) and the Mouthoumet Massif. There, a Late Ordovician fault-controlled subsidence linked to the record of rift-related tholeiites (Roque de Bandies and Villerouge formations) was contemporaneous with the record of the Hirnantian glaciation (Álvaro et al., 2016). Re-opening of rifting branches (Montagne Noire and Mouthoumet massifs) was geometrically recorded as onlapping patterns and final sealing of Sardic palaeoreliefs by Silurian and Lower Devonian strata.

Sardinia illustrates an almost complete record of the Variscan Belt (Carmignani et al., 1994; Rossi et al., 2009). Some plutonic orthogneisses of the inner zone belong to this cycle, such as the orthogneisses of Golfo Aranci (Giacomini et al., 2006). Gaggero et al. (2012) described three magmatic cycles. The first cycle is well represented in the Sarrabus unit by Furongian–Tremadocian volcanic and subvolcanic interbeds within a terrigenous succession (San Vito Formation) which is topped by the Sardic unconformity. Some plutonic orthogneisses of the inner zone belong to this cycle, such as the orthogneisses of Golfo Aranci (Giacomini et al., 2006) and the orthogneiss of Punta Bianca (PB). The second Middle Ordovician cycle, postdating the previous cycle by about 50 million years, is of a volcanic-arc type with calc-alkaline affinity and acidic-to-intermediate composition. The acidic metavolcanites are referred to in the literature as "porphyroids", which crop out in the external nappe zone and some localities of the inner zone. The intermediate-to-basic byproducts are widespread in central Sardinia (Serra Tonnai Formation). Some plutonic rocks (Mt Filau orthogneisses and Capo Spartivento) of the second cycle are discussed above. The third cycle consists of alkalic meta-epiclastites interbedded in post-Sandbian strata and metabasites marking the Ordovician–Silurian contact and reflecting rifting conditions. In this work only the first two cycles are considered. Giacomini et al. (2006) cite coeval mafic rocks of felsic magmatism of Mid-Ordovician age (Cortesogno et al., 2004; Palmeri et al., 2004; Giacomini et al., 2005), although they interpret a subduction scenario of the Hun terrain below Corsica and Sardinia in the Mid-Ordovician.

6.3 Origin of intracrustal siliceous melts

In this scenario, the key to generating large volumes of acidic rocks in an intraplate context would be the existence of a lower–middle crust, highly hydrated, in addition to a high-heat flow, possibly caused by mafic melts (Bryan et al., 2002; Díez-Montes, 2007). This could be the scenario initiated by the arrival of a thermal anomaly in a subduction-free area (Sánchez-García et al., 2003, 2008, 2019; Álvaro et al., 2016). The formation of large volumes of intracrustal siliceous melts could act as a viscous barrier, preventing the rise of mafic magmas within volcanic environments and causing the underplating of these magmas at the contact between the lower crust and the mantle (Huppert and Sparks, 1988; Pankhurst et al., 1998; Bindeman and Valley, 2003). The cooling of these magmas could lead to crustal thickening, and in this case the volcanic-arc signature can be explained by crustal recycling (Navidad et al., 2010; Díez-Montes et al., 2010; Martínez et al., 2011).

Sánchez-García et al. (2019) have proposed that the anomaly that produced the large magmatism throughout the Iberian Massif could have migrated from the rifting axis to inward zones, and the acidic, peraluminous, K-rich rocks of Mid Ordovician age should represent the initial stages of a new rifting pulse, resembling the peraluminous rocks of the Early Rift Event (sensu Sánchez-García et al., 2003) from the Cambrian Epoch 2 of the Ossa-Morena Rift. In the parautochthon of the Galicia–Trás-os-Montes Zone, the appearance of tholeiitic and alkaline–peralkaline magmatism in the Middle Ordovician would signal the first steps toward extensional conditions (Díez Fernández et al., 2012; Dias da Silva et al., 2016). In the Montagne Noire and the Mouthoumet massifs, contemporaneous tholeiitic lava indicates a similar change in the tectonic regimen (Álvaro et al., 2016). This change in geodynamic conditions is also marked by the appearance of rocks with extensional characteristics in some of the subgroups considered here, such as the Central Iberian Zone (San Sebastián orthogneisses), Eastern Pyrenees (Casemí orthogneisses and G1), volcanic rocks of the Occitan Domain, and the orthogneisses and volcanic rocks from Sardinia. In the Pyrenees, Puddu et al. (2019) proposed that a thermal doming, between 475 and 450 Ma, should have stretched the Ordovician lithosphere, leading to emersion and denudation of a Cambrian–Ordovician palaeorelief and giving rise to the onset of the Sardic unconformity. According to these authors, thermal doming triggered by hot mafic magma underplating may also be responsible for the late Early–Late Ordovician coeval magmatic activity

A major continental break-up, leading to the so-called Tremadocian Tectonic Belt, was suggested by Pouclet et al. (2017), which was initiated by upwelling of the asthenosphere and tectonic thinning of the lithosphere. Mantle-derived mafic magmas were underplated at the mantle–crust transition zone and intruded the crust. These magmas provided heat for crustal melting, which supplied the rhyolitic volcanism. After emptying the rhyolitic crustal reservoirs, the underlying mafic magmas finally rose and reached the surface. According to Pouclet et al. (2017), the acidic magmatic output associated with the onset of the Larroque metarhyolites resulted in massive crustal melting requiring a rather important heat supply. Asthenospheric upwelling leading to lithospheric doming, continental break-up, and decompression-driven mantle melting can explain such a great thermal anomaly. Magmatic products accumulated on the mantle–crust contact, providing enough heat transfer for crustal melting.

7 Conclusions

A geochemical comparison of 231 plutonic and volcanic samples of 2 major suites, Furongian–Mid Ordovician and Late Ordovician in age, from the Central Iberian and Galicia–Trás-os-Montes zones of the Iberian Massif and in the Eastern Pyrenees, the Occitan Domain (Albigeois, Montagne Noire and Mouthoumet massifs) and Sardinia points to a predominance of materials derived from the melting of metasedimentary rocks, peraluminous and rich in SiO_2 and K_2O. The total content in REE is moderate to high. Most felsic rocks display similar chondrite-normalized REE patterns, with an enrichment of LREE relative to HREE, which should indicate the involvement of crustal materials in their parental magmas.

Zr / TiO_2, Zr / Nb, Nb / Y and Zr vs. Ga / Al ratios, and REE and $_\varepsilon$Nd values reflect contemporaneous arc and extensional scenarios, which progressed to distinct extensional conditions finally associated with outpouring of mafic tholeiite-dominant rifting lava flows. Magmatic events are contemporaneous with the formation of the Toledanian (Furongian–Early Ordovician) and Sardic (Early–Late Ordovician) unconformities, related to neither metamorphism nor penetrative deformation. The geochemical and structural framework precludes subduction-generated melts reaching the crust in a magmatic arc-to-back-arc setting. On the contrary, it favours partial melting of sediments and/or granitoids in a lower continental crust triggered by the underplating of hot mafic magmas related to the opening of the Rheic Ocean as a result of asthenospheric upwelling.

Data availability. All data are included in the paper and in the Supplement.

Supplement. The supplement related to this article is available online at: https://doi.org/10.5194/se-11-2377-2020-supplement.

Author contributions. JJA, TSG and JMC led the methodology and wrote, reviewed and edited the original draft. CP, ADM, ML and GO supported the same processes.

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