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Birth and demise of the Rheic Ocean magmatic arc(s): Combined U–Pb and Hf isotope analyses in detrital zircon from SW Iberia siliciclastic strata

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ABSTRACT

Paleozoic continental reconstructions indicate that subduction of Rheic oceanic lithosphere led to collision between Laurussia and Gondwana which was a major event in the formation of the Ouachita-Appalachian-Variscan orogenic belt and the amalgamation of Pangea. However, arc systems which record Rheic Ocean subduction are poorly preserved. The preservation of Devonian detrital zircon in Late Devonian–Early Carboniferous siliciclastic rocks of SW Iberia, rather than arc-related igneous rocks indicates that direct evidence of the arc system may have been largely destroyed by erosion. Here we report in-situ detrital zircon U–Pb isotopic analyses of Late Devonian–Early Carboniferous siliciclastic rocks from the Pulo do Lobo Zone, which is a reworked Late Paleozoic suture zone located between Laurussia and Gondwana. Detrital zircon age spectra from the Pulo do Lobo Zone Frasnian formations show striking similarities, revealing a wide range of ages dominated by Neoproterozoic and Paleoproterozoic grains sourced from rocks typical of peri-Gondwanan terranes, such as Avalonia, the Meguma terrane and the Ossa-Morena Zone. Pulo do Lobo rocks also include representative populations of Mesoproterozoic and Early Silurian zircons that are typical of Avalonia and the Meguma terrane which are absent in the Ossa-Morena Zone. The Famennian–Tournaisian formations from the Pulo do Lobo Zone, however, contain more abundant Middle–Late Devonian zircon indicating the contribution from a previously unrecognized source probably related to the Rheic Ocean magmatic arc(s). The Middle–Late Devonian to Early Carboniferous zircon ages from the siliciclastic rocks of SW Iberia (South Portuguese, Pulo do Lobo and Ossa-Morena zones) have a wide range in εHf_T values (−8.2 to +8.3) indicating the likely crystallization from magmas formed in a convergent setting. The missing Rheic Ocean arc was probably built on a Meguma/Avalonia type basement. We propose for the Pulo do Lobo Zone that the Frasnian sedimentation occurred through the opening of a back-arc basin formed along the Laurussian active margin during Rheic Ocean subduction, as has been recently proposed for the Rhenohercynian Zone in Central Europe. Detrital zircon ages in the Frasnian siliciclastic rocks indicate provenance in the Meguma terrane, Avalonia and Devonian Rheic Ocean arc(s). As a result of back-arc basin inversion, the Frasnian formations underwent deformation, metamorphism and denudation and were unconformably overlain by Famennian to Visean siliciclastic strata (including the Phyllite-Quartzite Formation of the South Portuguese Zone). The Latest Devonian–Early Carboniferous detritus were probably shed to the Pulo do Lobo Zone (Represa and Santa Iria formations) by recycling of Devonian siliciclastic rocks, from the South Portuguese Zone (Meguma terrane) and from a new distinct source with Baltica/Laurentia derivation (preserved in the Horta da Torre Formation and Alajar Mélange).

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1. Introduction

Continental collision between Laurussia and Gondwana resulted in the Ouachita–Alleghanian–Variscan orogen and was a major event in the amalgamation of Pangea (Murphy and Nance, 2013). Regional syntheses suggest that subduction within the Rheic Ocean, which separated Laurussia and Gondwana prior to their collision, began after the Early Devonian closure of the Iapetus Ocean (e.g. van Staal et al., 1998, 2009). Although the destruction of the Rheic Ocean by the end of the Devonian is indicated by a wealth of faunal, petrological and structural data, documentation of Devonian magmatic arc systems that record the history of convergence prior to collision is fragmentary (Nance et al., 2012; Stampfli and Kozur, 2006).

Along Cenozoic convergent margins, the paucity of arc assemblages is commonly attributed to either removal by strike-slip faults inboard of the arc, by subduction erosion in which the arc is destroyed by progressive or piecemeal subduction erosion, or by progressive erosion (e.g., Boston et al., 2017; Clift and Vannucchi, 2004; Draut and Clift, 2013; Keppie et al., 2009; Paterson and Ducea, 2015; Stern, 2011; von Huene and Scholl, 1991). Although strike-slip motion is highly probable especially if convergence is oblique, this mechanism by itself, as well as subduction erosion, may explain the absence of an arc assemblage in a given region but cannot explain the overall paucity of preserved arc magmatism along both the Laurussian and Gondwanan margins during convergence of the Rheic Ocean. The timing and duration (but not necessarily the volumes) of magmatic arcs can be provided by the sedimentary record in fore-, intra-, and back-arc basins, through the study of detrital zircon grains in siliciclastic sediments derived from the erosion of arcs (e.g. Paterson and Ducea, 2015). The possibility that the Rheic Ocean arcs were largely destroyed by erosion and the recognition of the basement (Laurussian or Gondwanan) where the arcs were built can be tested by examining in detail the age and Hf isotopic composition of detrital zircon in Devonian–Carboniferous siliciclastic rocks that may have been derived from denudated portions of the arc. By analyzing siliciclastic rocks over a range of depositional ages in the suture zone that marks the closure of the Rheic Ocean, we can detect the timing of onset of arc magmatism. Furthermore, combined U–Pb and Hf isotope analyses in detrital zircon might constrain the isotopic characteristics of the source of the arc magmas at the time of zircon crystallization (including the type of basement where the arc was built), e.g., juvenile and/or evolved (e.g. Gerdes and Zeh, 2006).

In this contribution, we document for the first time Hf isotope compositions of Devonian–Carboniferous detrital zircon found in Frasnian–Visean siliciclastic strata of SW Iberia located within and on either side of the Rheic Ocean suture (Pulo do Lobo Zone; Fig. 1). We interpret the Devonian zircon to have been derived from Rheic Ocean continental magmatic arcs and arc-related basins developed during Laurussia and Gondwana convergence.

2. Geological background

The Rheic suture zone between Laurussia and Gondwana can be traced along the Appalachian–Variscan orogenic system from Central America to Western-Central Europe (Martínez-Catalán et al., 2007; Nance et al., 2010, 2012) (Fig. 1). In the northern Appalachians, terranes inferred to lie west of the Rheic suture include Avalonia and Meguma which are terranes that originated along the northern margin of Gondwana (collectively known as peri-Gondwanan terranes) but are thought to have departed from that margin as the Rheic Ocean opened (Murphy et al., 2004, 2006). These terranes simultaneously defined the northern margin of the Rheic Ocean and the southern flank of the Iapetus Ocean and accreted to Laurussia in the Early Devonian as the Iapetus Ocean closed (Murphy et al., 2004, 2006; van Staal et al., 1998, 2009, 2012). The Meguma terrane is composed of a thick lower sequence of Ediacaran to Early Ordovician siliciclastic rocks (Meguma Supergroup) overlain by an upper sequence with Late Ordovician siliciclastic rocks (Rockville Notch Group) and Early Silurian (White Rock Group; ca. 440 Ma; Keppie and Krogh, 2000; White and Barr, 2012) to Early Devonian volcanic and siliciclastic rocks containing Rhenish fauna (Murphy et al., 2004; White and Barr, 2012 and references therein). The basement to the Meguma terrane has not been identified. Paleozoic strata deposited in the Meguma terrane was deformed and metamorphosed in the Middle Devonian (Neoacadian Orogeny; van Staal et al., 2009). The Rheic suture is constrained to lie outboard of the North American Atlantic margin, to the east of the Meguma terrane. Mid–Upper Devonian granitoids (Moran et al., 2007; Tate and Clarke, 1995) are found in the Meguma terrane and are interpreted to record a rare example of subduction-related magmatism of Rheic oceanic lithosphere beneath composite Laurentia continent (Clarke et al., 1997; Murphy et al., 1999; van Staal et al., 2009). Late Famennian–Tournaisian siliciclastic rocks (Horton Group) unconformably overlie the Cambrian–Devonian rocks of the Meguma terrane (Murphy and Keppie, 1998, 2005; White and Barr, 2012).

In the suture zones of the Western European Variscan belt, the closure of the Rheic Ocean has been traditionally considered to record the obduction of ophiolites and subduction of the Gondwanan passive margin at ca. 370 Ma (Dallmeyer et al., 1997; Martínez-Catalán et al., 2007; Nutman et al., 2001; Rodríguez et al., 2003; Simancas et al., 2005, 2009). Recently, a two-stage collisional process between Laurussia and Gondwana has been proposed with the closure of the Rheic Ocean at ca. 400 Ma and of an ephemeral Devonian intra-Gondwana oceanic basin at ca. 370 Ma (Arenas et al., 2014; Díez Fernández and Arenas, 2015; Díez Fernández et al., 2016). Other authors support a "two-ocean model" with the Early Devonian closure of the Rheic Ocean predating the opening of a younger, narrow Rhenohercynian Ocean in the Laurussian margin (Eckelmann et al., 2014; Franke, 2000; Franke and Dulce, 2016; von Raumer et al., 2016). The rootless suture of the Rheic oceanic lithosphere and overlying tectonic units is represented by the allochthonous complexes transported onto the Gondwanan side which are exposed in the French Massif Central, (Lardeaux et al., 2001), the Bohemian Massif (Franke, 2000) and also in Iberia (Díez Fernández et al., 2016, 2017; Martínez-Catalán et al., 2007). The Laurussian side includes the South Portuguese Zone of southern Iberia, southern Ireland, southern England and the Rhenohercynian Zone of continental Europe, (Braid et al., 2012; Murphy et al., 2016 and references therein).

In SW Iberia, the Rheic suture was considered to be located along the complex contact between the Ossa-Morena (Gondwanan side) and the South Portuguese (Laurussian side) zones (Crespo-Blanc and Orozco, 1991). The Rheic suture zone was first considered to be marked by two tectonic units (Quesada et al., 1994): i) MORB-type greenschists to metagabbros (Beja-Acebuches ophiolitic complex) and ii) metasedimentary siliciclastic formations (Pulo do Lobo Zone) (Figs. 1 and 2). Early Carboniferous protolith ages (ca. 340–332 Ma) obtained for the Beja-Acebuches metamafic rocks suggest that this ocean lithosphere is significantly younger than both the formation and closure of the Rheic Ocean in Devonian times (Azor et al., 2008; Pérez-Cáceres et al., 2015). The Beja-Acebuches ophiolitic complex might have been formed during an intra-orogenic extensional event recorded along SW Iberia during Early Carboniferous times (Pereira et al., 2009, 2012a; Simancas et al., 2009). This interpretation implies that the Rheic suture between Gondwana and Laurussia in SW Iberia is more likely associated with the evolution of the Pulo do Lobo Zone instead of the Beja-Acebuches ophiolitic complex. Nevertheless this subject is still controversial because vestiges of the Rheic Ocean lithosphere are yet to be unequivocally identified (Pereira et al., 2012b). Another controversial issue, but not within the scope of this work, concerns the tectonic framing of Early Carboniferous calc-alkaline magmatism that transects across the Pulo do Lobo, South Portuguese and Ossa-Morena zones. Distinct models were proposed to explain the origin and the tectonic framing of these Early Carboniferous calc-alkaline plutons.

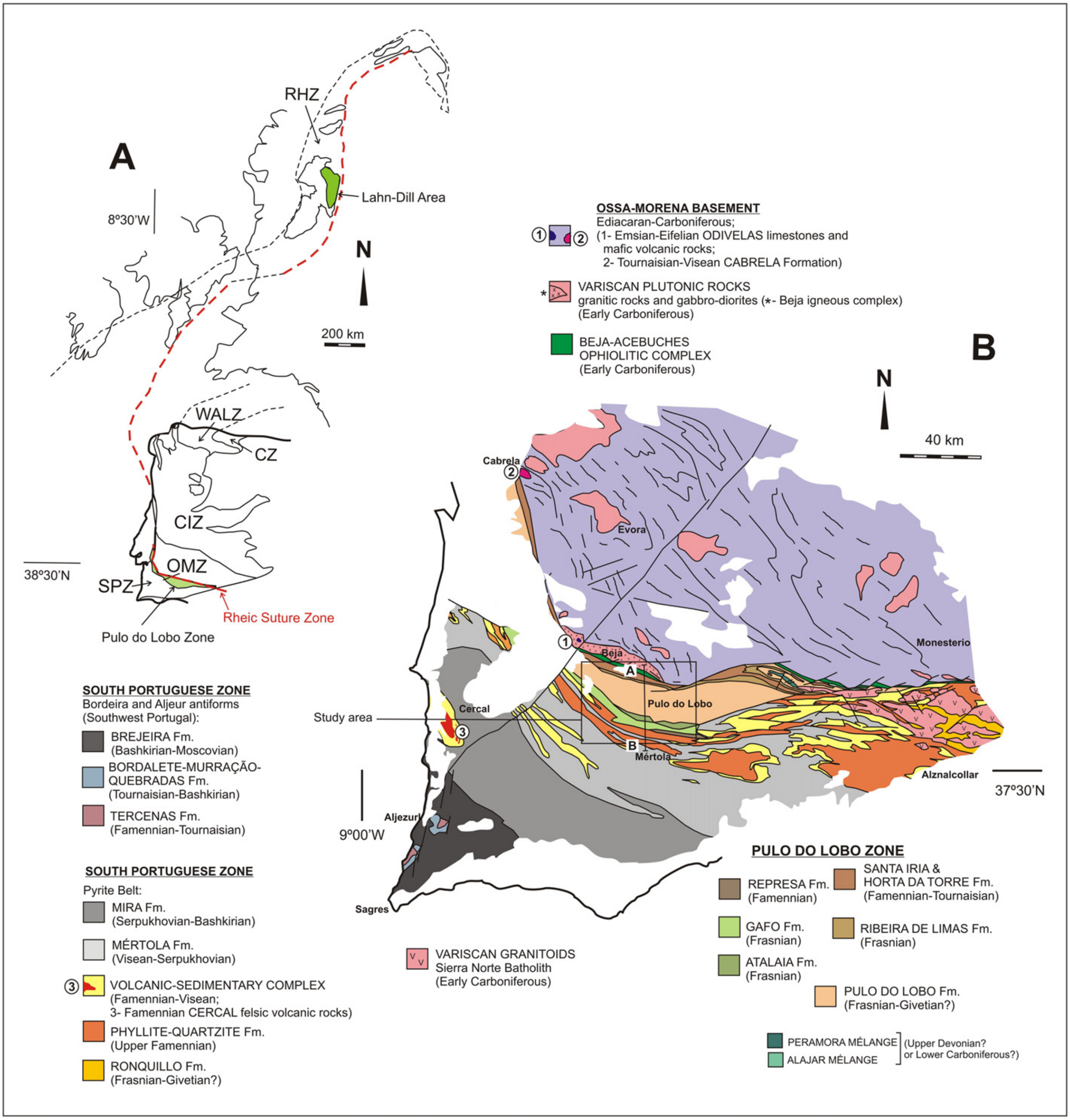

Fig. 1. A — Inset with location of the Pulo do Lobo Zone (SW Iberia) and the Lahn-Dill Area (Central Europe) in the Variscan belt (Modified from von Raumer et al., 2016): CIZ — Central Iberian Zone, CZ — Cantabrian Zone, OMZ — Ossa-Morena Zone, RHZ — Rhenohercynian Zone, SPZ — South Portuguese Zone, WALZ — West Asturian-Leonese Zone. B — Simplified geological map of SW Iberia showing the study area of the Pulo do Lobo Zone.
Modified from Oliveira, 1990; Pérez-Cáceres et al., 2017 and references therein.

They have traditionally been linked to subduction as result of the interaction of magmas derived from the lithospheric mantle with magmas developed by partial melting of a deep mafic and pelitic granulitic crust (Castro et al., 1995; de la Rosa et al., 2002 and references therein). However, recent studies have suggested that these calc-alkaline plutonic rocks were formed when subduction was inactive by interaction of mantle-derived alkaline mafic and crustal-derived peraluminous felsic magma related to intraplating of mantle-derived magma into the middle crust (Cambeses et al., 2015 and references therein).

Nowhere along the Rheic suture zone of SW Iberia, is any unequivocal evidence for the existence of a Devonian magmatic arc preserved. The only traces of Devonian magmatic rocks are: (i) the calc-alkaline mafic rocks which interbedded with the Emsian-Eifelian Odivelas reef limestones with Rhenish fauna (Machado et al., 2010; Silva et al., 2011) which are in faulted contact with Early Carboniferous calc-alkaline magmatic rocks of the Ossa-Morena Zone (Beja igneous complex; Jesus et al., 2007; Pin et al., 2008) to north of the Beja-Acebuches ophiolitic complex (Fig. 1); and ii) the Frasnian–Famennian Cercal

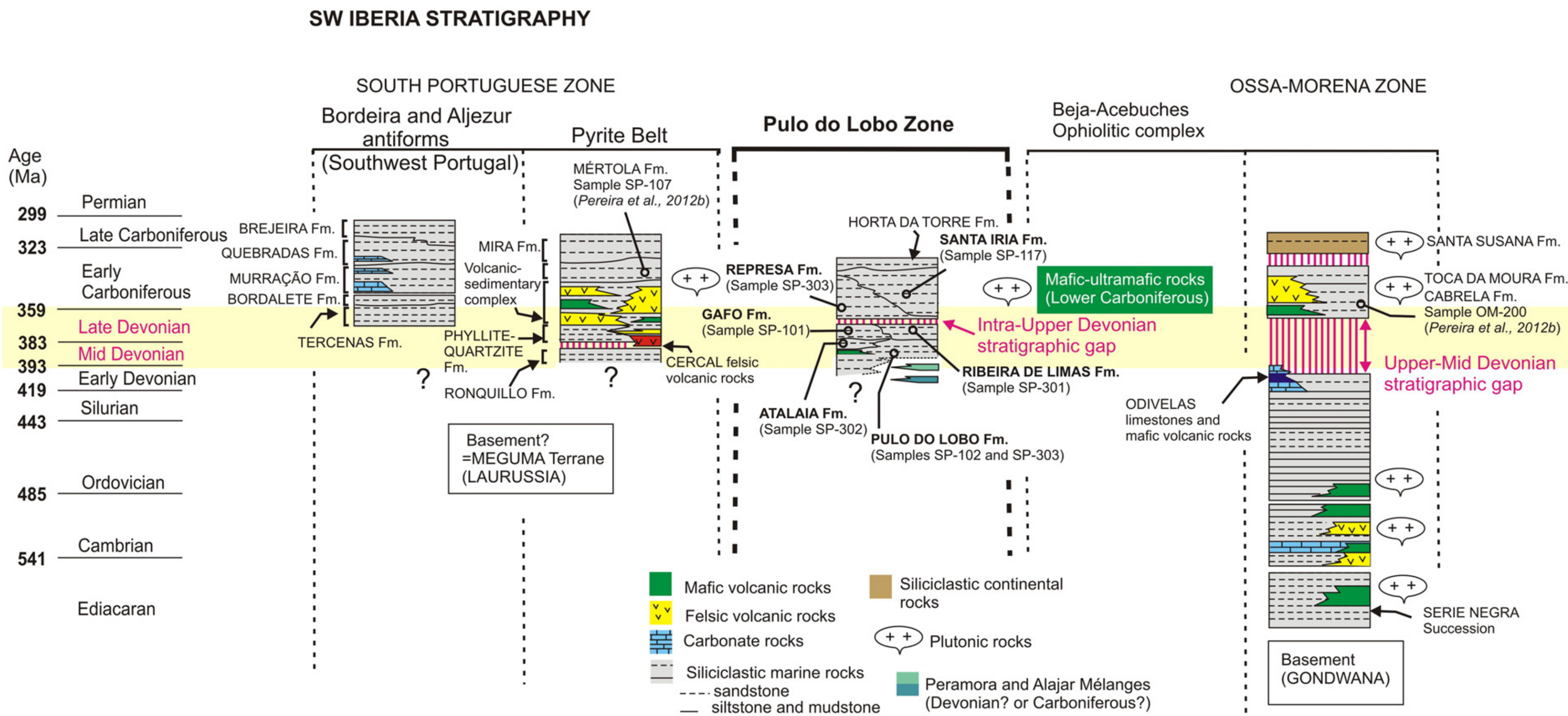

Fig. 2. Summary of the stratigraphy of the South Portuguese, Pulo do Lobo and Ossa–Morena Zones of SW Iberia with sample locations used for U–Pb and Hf analyses. Modified from Braid et al., (2010); Oliveira, (1990); Oliveira et al., (1991); Pereira et al., (2007); Pérez-Cáceres et al., (2017).

calc-alkaline felsic volcanic rocks (ca. 374–372 Ma; Rosa et al., 2008a) which occur within the Pyrite Belt of the South Portuguese Zone (Figs. 1 and 2).

2.1. Pulo do Lobo Zone: reworked suture zone

The Pulo do Lobo Zone that defines a 0.5–30 km-wide and 180 km-long curved structure at south of the Beja-Acebuches ophiolitic complex (Fig. 1), has been variously interpreted as (i) an accretionary prism (Braid et al., 2010; Eden and Andrews, 1990), (ii) a dismembered oceanic lithosphere formed during the closure of the Beja-Acebuches back-arc basin (Fonseca and Ribeiro, 1993; Quesada et al., 1994) related to a northward-dipping (present-day coordinates) Rheic Ocean subduction zone and (iii) a unit of the suture zone related to the south-dipping (present-day coordinates) subduction of the Rheic oceanic lithosphere under the Laurussian margin (Simancas et al., 2009). Irrespective, there is no agreement on the polarity of the subduction zone (or zones) south of the Ossa-Morena Zone prior to the onset of collision, and several plate tectonic models have been proposed for the Rheic Ocean closure in SW Iberia (Díaz Aspíroz et al., 2006; Onézime et al., 2003; Pérez-Cáceres et al., 2015; Quesada et al., 1994; Ribeiro et al., 2007, 2010; Rubio Pascual et al., 2013; Silva et al., 1990; Simancas et al., 2009).

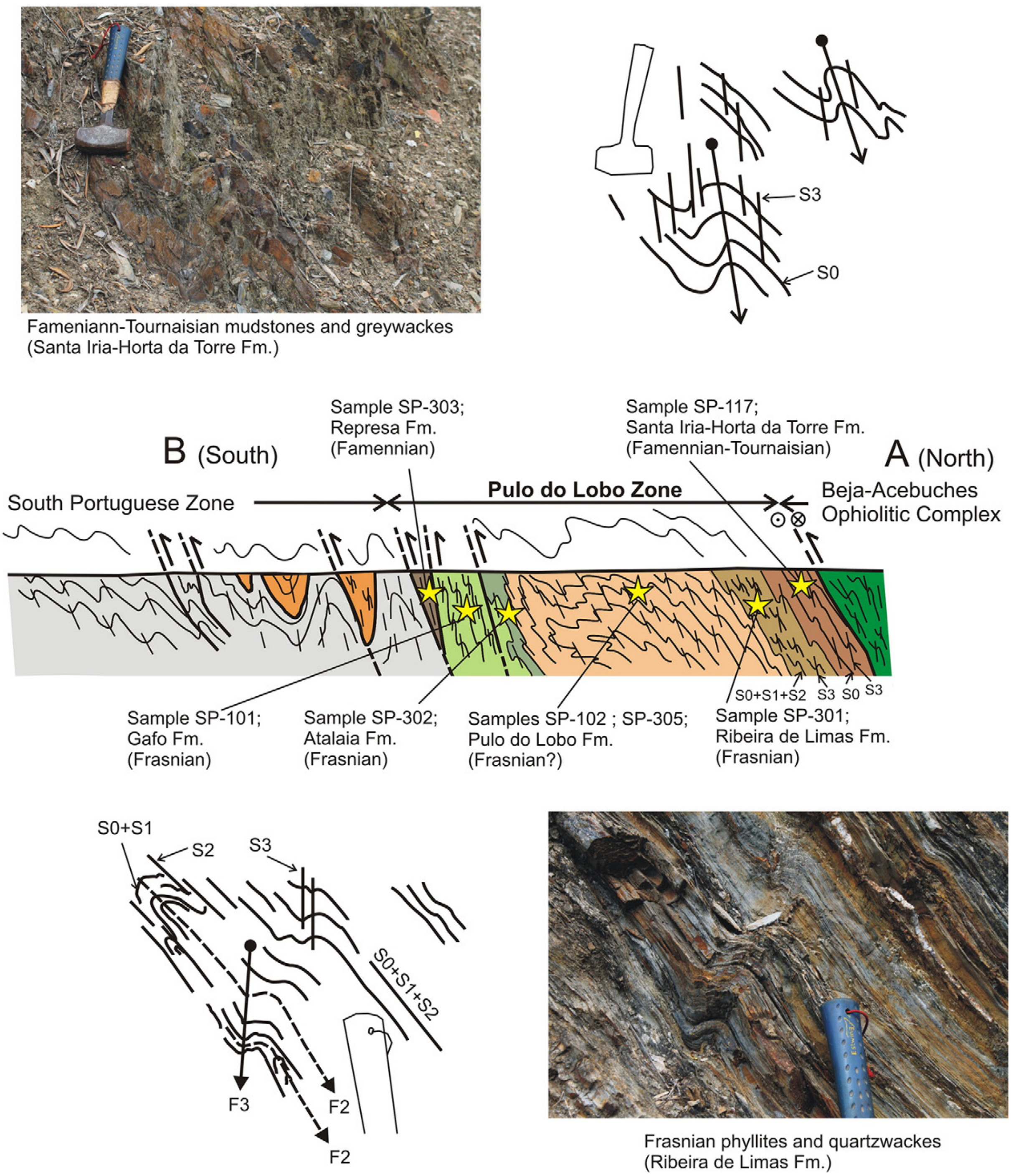

Fig. 3. A–B is a schematic cross-section through the Portuguese sector of the Pulo do Lobo Zone with sample locations used for U–Pb and Hf analyses (Modified from Pereira et al., 2012a; Silva et al., 1990). Folding and development of S_3 cleavage in mudstones and greywackes of the Santa Iria Formation (top). Superimposition of F_3 on F_2 folds, in the Ribeira de Limas phyllites and greywackes (bottom).

In the Portuguese sector of the Pulo do Lobo Zone, the original stratigraphy is preserved in a curved anticline striking northwest-southeast to east-west that verges to southwest-south (Silva et al., 1990) (Fig. 3). Although strongly deformed, primary stratigraphic relations in the Pulo do Lobo Zone formations are locally preserved. The core of the structure exposes strongly deformed phyllites, rare greywacke beds and mafic volcanic rocks (Pulo do Lobo Formation; Pfefferkorn, 1968) conformably overlain by phyllites and quartzites (Atalaia Formation; Oliveira, 1990 and references therein). The unknown age Pulo do Lobo and Atalaia formations are conformably overlain by siliciclastic strata with Frasnian biostratigraphic age (Ribeira de Limas and Gafo formations; Oliveira, 1990; Pereira et al., 2007). Distinct associations of palynomorphs reveal a sedimentary gap of about 14 My between the Famennian (Represa, Horta da Torre and Santa Iria) and the Frasnian (Gafo and Ribeira de Limas) formations (Pereira et al., 2007). The Santa Iria and Horta da Torre formations, whose contact appears gradational, unconformably overlie the older Devonian strata (Oliveira, 1990). The presence of this intra-Upper Devonian unconformity was first proposed on the basis of the contrasting style of deformation of the Frasnian and Famennian siliciclastic strata (Silva et al., 1990) (Fig. 2).

In the Spanish sector, the Pulo do Lobo Zone large-scale folds (Martínez Poza et al., 2012; Pérez-Cáceres et al., 2015, 2017) give way to fault-bounded lithotectonic units that strike east-west and dip steeply (Braid et al., 2010; Eden and Andrews, 1990; Eden, 1991), defining the strongly deformed termination of the kilometer-scale anticlinal structure. Given that the contacts between all units are tectonic, the relative age of these units is unclear (Dahn et al., 2014). LA-ICPMS detrital zircon ages obtained from a greywacke of the Santa Iria Formation, that unconformably overlie the Ribeira de Limas Formation, yielded a wide range of Devonian–Carboniferous ages, with a strong peak at ca. 347 Ma (Braid et al., 2011) interpreted as the maximum depositional age. Braid et al. (2010) defined along the southern contact of the Beja-Acebuches ophiolitic complex two lithotectonic units structurally overlain by the Pulo do Lobo phyllites and interpreted to be components of a Devonian accretionary prism (Figs. 1 and 2): i) a mélange comprised of internally deformed quartzite in a fine-grained phyllite-quartzite matrix (Alájar Mélange; Braid et al., 2011; Eden, 1991) and ii) a mélange consisting of tectonically emplaced mafic blocks in a volcaniclastic and schistose matrix (Peramora Mélange; Dahn et al., 2014; Eden, 1991). The geochronology and MORB geochemistry of the mafic rocks and matrix in the Peramora Mélange suggest that this lithotectonic unit could be a vestige of the Rheic oceanic tract between Laurussia and Gondwana, requiring the incorporation of a sedimentary component, with a maximum depositional age of ca. 343 Ma (Dahn et al., 2014) or ca. 333 Ma (Pérez-Cáceres et al., 2015). Detrital zircon obtained from a quartzwacke attributed to the Ribeira de Limas Formation and from phacoid shaped quartzite bodies and fine grained matrix of the Alájar Mélange yield a dominant population of Mesoproterozoic ages atypical of detritus derived from Gondwana (Ossa-Morena Zone and Meguma terrane) which, together with the lack of Cryogenian–Ediacaran ages, may also reflect possible Baltica/Laurentia derivation (Braid et al., 2011). Detrital zircon obtained from the Horta da Torre Formation quartzite beds (Pérez-Cáceres et al., 2017) shows remarkable similarities to the Alájar Mélange obtained by Braid et al. (2011), suggesting that this mélange is made of dismembered Late Famennian quartzite beds of the Horta da Torre Formation (Pérez-Cáceres et al., 2015, 2017).

2.2. Ossa-Morena Zone: Gondwanan side

On the Gondwanan side of the suture zone, the Ossa-Morena Zone stratigraphy (Fig. 2) records the evolution of an Ediacaran magmatic arc and of Paleozoic strata along the outermost Gondwanan passive margin of the Rheic Ocean, which was marked by significant volumes of magmatism in Cambrian–Ordovician times (Chichorro et al., 2008; Linnemann et al., 2008; Pereira et al., 2008, 2011, 2012c; Robardet and Gutiérrez-Marco, 2004; Sánchez-García et al., 2003, 2008, 2010). Detrital zircon populations of Ediacaran to Ordovician sedimentary rocks are dominated by Neoproterozoic and Paleoproterozoic ages and show a remarkable gap of Mesoproterozoic grains, indicating provenance in North-Gondwana (Cambeses et al., 2017; Linnemann et al., 2008; Pereira et al., 2008, 2011, 2012c). The Ossa-Morena Zone consists of sedimentary, plutonic and metamorphic rocks whose grade ranges from very low to catazonal. Metamorphism reached high pressure conditions during a first regional contractional event at ca. 370 Ma, followed by high temperature (up to crustal anatexis) conditions coeval with voluminous magmatism during a regional magmatic extensional event at ca. 340–330 Ma (Díez Fernández et al., 2016; Pereira et al., 2009, 2012c, 2015; Simancas et al., 2009). Tournaisian to Visean turbiditic strata (Toca da Moura and Cabrela formations; Oliveira et al., 1991; Pereira et al., 2006; Pereira et al., 2012a, 2012b; Quesada et al., 1990) occur interbedded with rhyolites and dacites and include olistoliths of Middle and Late Devonian limestones (Fig. 2).

2.3. South Portuguese Zone: Laurussian side

On the Laurussian side of the suture zone, the South Portuguese Zone also contains Devonian–Carboniferous basin strata (Figs. 1 and 2). The unexposed basement of the South Portuguese Zone has been characterized, based on indirect evidence, as part of the Meguma terrane which is unequivocally exposed only in Maritime Canada (Braid et al., 2010; Martínez-Catalán et al., 2007; Pereira et al., 2012b), although possible correlations may occur in Wales (Waldron et al., 2009) and/or Cornwall (Nance et al., 2014). The Devonian–Carboniferous stratigraphy of the South Portuguese Zone is divisible into two domains: i) the Pyrite Belt and ii) the Bordeira and Aljezur antiforms of Southwest Portugal (Oliveira, 1990; Fig. 2). The Pyrite Belt includes Late Devonian siliciclastic rocks (Phyllite–Quartzite Formation; Pereira et al., 2007 and references therein) that are overlain by the ca. 374–349 Ma volcanic–sedimentary complex (Oliveira et al., 2013; Rosa et al., 2008a, 2008b) comprising the Late Devonian Cercal volcanic rocks. In the Spanish sector, the Phyllite–Quartzite Formation unconformably overlies previously deformed and metamorphosed strata, recording high pressure metamorphism (Rubio Pascual et al., 2013). These metamorphosed siliciclastic strata (Ronquillo Formation, Simancas, 1983; or La Minilla Formation; Rubio Pascual et al., 2013) have a maximum depositional age of ca. 400 Ma (Pérez-Cáceres et al., 2017), which is compatible with a Late Givetian–Early Frasnian biostratigraphic age (González et al., 2004) (Fig. 2). The Pyrite belt volcanic–sedimentary complex is unconformably overlain by a thick succession of the Visean to Bashkirian turbidites (including the Mértola and Mira formations; Oliveira, 1990; Pereira et al., 2012a, 2012b, 2013; Rodrigues et al., 2014) (Figs. 1 and 2). The Pyrite belt volcanic–sedimentary complex and the Phyllite–Quartzite Formation can be stratigraphically correlated with quartzite and slate of the Famennian–Tournasian Tercenas Formation (Oliveira, 1990; Pereira et al., 2007) of the Bordeira and Aljezur antiforms (Figs. 1 and 2). The Tercenas Formation is conformably overlain by the Tournaisian to Moscovian turbiditic strata which include rare carbonate rocks (Bordalete, Murração, Quebradas and Brejeira formations; Oliveira, 1990; Pereira et al., 2007) (Figs. 1 and 2). Detrital zircon populations of the Ronquillo, Phyllite–Quartzite and Tercenas formation sedimentary rocks are dominated by Neoproterozoic and Paleoproterozoic ages but also include Early Silurian and Mesoproterozoic grains, indicating a most likely provenance from the Laurussian margin (Braid et al., 2011; Pereira et al., 2012b; Pérez-Cáceres et al., 2017).

3. Field relationships

In the northern limb of the Pulo do Lobo antiform (Portuguese sector), there is a gradational contact between the monotonous phyllitic sequence of the Pulo do Lobo Formation and the phyllite-quartzwacke succession of the Ribeira de Limas Formation, which together define a coarsening-upward sequence (Oliveira, 1990). In the southern limb, the Pulo do Lobo phyllites grade upwards into a sequence of phyllites and quartzites of the Atalaia Formation (Fig. 3). The Pulo do Lobo and Atalaia formations are tectonically in contact with a sequence of Frasnian greywackes, siltstones and mudstones of the Gafo Formation that is intruded by Early Carboniferous felsic rocks.

The pre-Famennian siliciclastic strata of the Pulo do Lobo, Ribeira de Limas, Atalaia and Gafo formations show three distinct phases of deformation (D_{1-3}, Silva et al., 1990; Figs. 3 and 4A-B). D_1 produced folds, S_1 penetrative cleavage and the generation of segregated quartz veins. The vergence of the first phase folds is difficult to determine due to

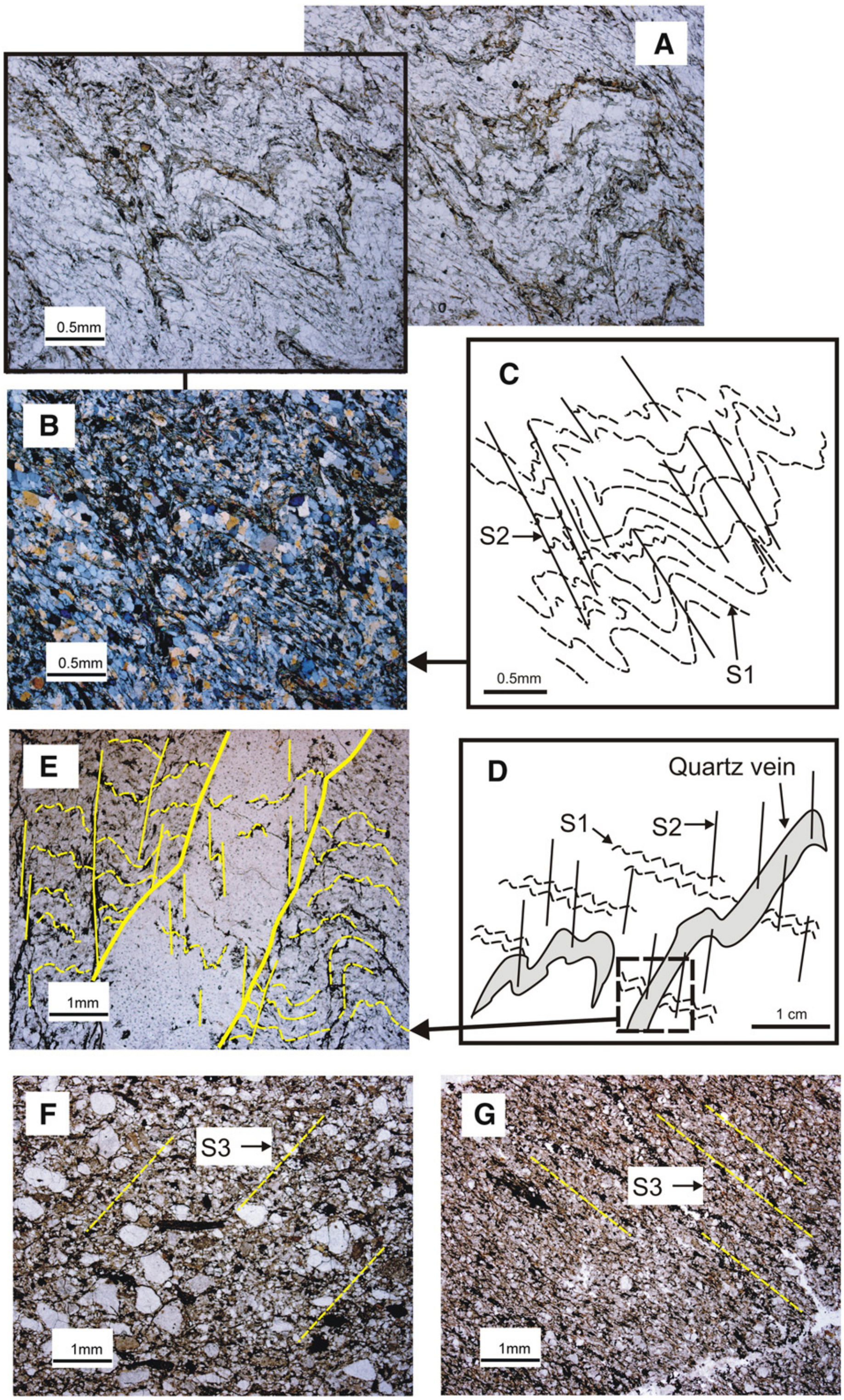

Fig. 4. Phyllite with quartz veins of the Pulo do Lobo Formation. Two deformation phases are recognized in this thin-section. The first one generated a S_1 slaty cleavage (trending upper right to lower left) and the second one produced the F_2 folds and a new spaced S_2 cleavage (trending upper left to lower right): (A) — plane polarized light, (B) — cross polarized light, (C) — schematic diagram showing S_1 and S_2. Quartzwacke with quartz veins of the Ribeira de Limas Formation. Two deformation phases are also recognized in this thin-section: (D) — schematic diagram showing quartz veins are cutting S_1 cleavage and subsequently folded by D_2, (E) — detail in plane polarized light. (F) — poorly sorted greywacke of the Represa Formation with a weak S_3 cleavage (plane polarized light). (G) — S_3 slaty cleavage in siltstone of the Santa Iria Formation (plane polarized light).

the superposition of the two subsequent deformation phases. S_1 is preserved as a relic biotite foliation in the hinges of microfolds between microlithons of the dominant second cleavage (Fig. 4A-E) that were refolded by later structures. D_1 and D_2 structures are not observed in the overlying Represa, Santa Iria and Horta da Torre formations which are respectively composed of greywackes, siltstones and mudstones. The Represa Formation is in faulted contact with the Gafo Formation. D_2 is characterized by south-southwest-verging thrusts and folds. The pre-Famennian siliciclastic strata were deformed under greenschist facies (biotite and chlorite) conditions and are characterized by the development of a S_2 penetrative cleavage, pressure solution structures, tight folds and quartz veins (Fig. 4A-E). D_3 produced near-vertical and northwest-southeast striking crenulation cleavage which crenulates the two preexisting cleavages and is observed in the pre-Famennian phyllites (Fig. 3). In the Famennian–Tournaisian flysch this deformation phase is recognized by upright cylindrical folds and a slaty/fracture cleavage (Fig. 4F-G).

4. Analytical methods

4.1. U–Pb isotopic system

A total of seven representative samples of clastic rocks were selected from the Pulo do Lobo Zone (Figs. 2 and 3). These samples are: two greywacke samples from the Pulo do Lobo Formation (SP-102 and SP-305), one sample of quartzite of the Atalaia Formation (SP-302), one sample of quartzwacke from the Ribeira de Limas Formation (SP-301), one sample of greywacke from the Gafo Formation (SP-101), one sample of quartzwacke from the Represa Formation (SP-303) and one sample of greywacke from the Santa Iria Formation (SP-117).

The separated zircon grains were mounted in epoxy and polished for CL-imaging and laser ablation ICP-MS analysis. Uranium–lead analyses were performed at Bergen Geoanalytical Facility using a 193 nm ArF excimer laser (Resonetics RESOlution M-50 LR) coupled to a Thermo-Finnigan Element II double focusing sector field ICP-MS instrument. The analytical protocol and data reduction routine were performed according to Gerdes and Zeh (2006). Spot analyses using a nominal spot diameter of 34 μm ensured good spatial resolution at feasible signal intensities. The material ablated with a repetition rate of 4 Hz at an energy density of ~2.5 J/cm^2 was transported in a He-carrier gas and mixed with Ar via a T-piece prior to entering the plasma torch. Isotopic data were acquired in peak-jumping, time resolved mode.

Raw data processing was done offline in an Excel spreadsheet program and included correction for background, instrumental mass discrimination as well as laser-induced and time-dependent elemental fractionation. Fractionation correction was done by bracketing the unknowns with GJ-1 reference zircon (608 Ma, ~430 ppm U; Jackson et al., 2004) and by applying the intercept method. ^{202}Hg and ^{204}Hg + Pb were monitored during all analyses to detect potential contributions from common Pb. In the majority of the analyzed spots, the interference- and background-corrected ^{204}Pb signal was below the detection limit, and thus a common Pb correction was not applied. However, few (<5%) of the total analyses were rejected due to too low signals, common Pb, or inconsistent fractionation patterns. Calculation of concordia ages was done using Isoplot 3.5 (Ludwig, 2003), and concordia ages are given for all analyses with a probability of concordance >0.17. Reference zircon 91,500 (1065 Ma, Wiedenbeck et al., 1995) and Plešovice (337 Ma, Sláma et al., 2008) were treated as unknowns and provided a quality control (91,500: 1065 ± 2.5 Ma, n = 70; Plešovice: 336.5 ± 0.9 Ma, n = 70).

4.2. Hf isotopic system

Hafnium isotopic compositions were analyzed using a Thermo Scientific Neptune Plus multi-collector ICP-MS and a Resonetics RESOlution M-50 excimer laser at the Geochemistry Department of the University of Tübingen. The analytical protocol largely follows that of Gerdes and Zeh (2006, 2009). The 50 μm Hf spots were drilled on top of the smaller U–Pb spots (34 μm in diameter). Data for masses ^{171}Yb, ^{172}Yb, ^{175}Lu, ^{176}Yb + Lu + Hf, ^{177}Hf, ^{178}Hf, ^{179}Hf, and ^{180}Hf were collected simultaneously in static mode during 40 s of ablation with a repetition rate of 5 Hz and a fluency of ~3.2 J/cm^2. Data were acquired during 70 cycles with an integration time of 0.5 s/cycle and processed offline in a spread-sheet based program. After background subtraction corrections for instrumental mass bias (exponential law) and for isobaric interferences of ^{176}Lu and ^{176}Yb on ^{176}Hf were done using the isotope ratios of 0.7325 for ^{179}Hf/^{177}Hf, 0.901843 for ^{176}Yb/^{171}Yb, 1.532151 for ^{172}Yb/^{171}Yb and 0.026549 for ^{176}Lu/^{175}Lu (Chu et al., 2002). During the two sessions, repeat analyses of Penglai, GJ1 and Mud Tank zircon reference materials gave time-integrated ^{176}Hf/^{177}Hf ratios of 0.282904 (±23 [2 SD], n = 35), 0.282000 (±24 [2 SD], n = 22) and 0.282504 (±27 [2 SD], n = 22), respectively identical to the published values (Penglai: 0.282906 ± 10; Li et al., 2010; GJ1: 0.281998 ± 7, Gerdes and Zeh, 2006, 2009; Mud Tank: 0.282507 ± 6, Woodhead and Hergt, 2005).

Initial ^{176}Hf/^{177}Hf and εHf were calculated using the apparent U–Pb age determined by LA-ICP-MS dating, the ^{176}Lu decay constant of 1.867×10^{-11} (Scherer et al., 2001; Soderlund et al., 2004) as well as the CHUR parameters: ^{176}Lu/^{177}Hf = 0.0336, and ^{176}Hf/^{177}Hf = 0.282785 (Bouvier et al., 2008). Two stage model ages were estimated using the measured ^{176}Lu/^{177}Hf of each spot (first stage = age of zircon), a value of 0.015 for the average continental crust (second stage; Rudnick and Gao, 2003), and a juvenile crust ^{176}Lu/^{177}Hf and ^{176}Hf/^{177}Hf of 0.0378 and 0.283158 (Dhuime et al., 2011), respectively.

5. Analytical results

5.1. Zircon U–Pb ages

Here we present zircon U–Pb ages from seven samples of the Pulo do Lobo Zone (Fig. 5; Table 1; see Electronic supplement), that were also analyzed to obtain Hf isotope compositions.

The two samples (SP-102 and SP-305, n = 169) of greywackes from the Pulo do Lobo Formation have similar detrital zircon age distribution patterns. Most grains are Cryogenian–Ediacaran (63%; ca. 840–543 Ma), although Paleoproterozoic (24%; ca. 2.3–1.6 Ga) ages are also common. Mesoproterozoic (6%; ca. 1.5–1.0 Ga), Archean (4%; ca. 3.2–2.5 Ga), Tonian (2%; ca. 895–851 Ma) and Cambrian (1%; ca. 531 Ma) grains are subordinate (Fig. 5).

The quartzwacke of the Ribeira de Limas Formation (sample SP-301; n = 80) has a similar detrital zircon age distribution to both samples of the Pulo do Lobo Formation (Fig. 5). Cryogenian–Ediacaran ages (69%; ca. 829–547 Ma) are most common, and a significant number of grains is Paleoproterozoic (23%; ca. 2.4–1.7 Ma). The remaining grains yield Mesoproterozoic (5%; ca. 1.5–1.0 Ga), Tonian (2%; ca. 875 and 874 Ma) and Archean (1%; ca. 2.6 Ga) ages. The youngest concordant grain was dated at ca. 547 Ma (Ediacaran).

The Atalaia Formation quartzite (sample SP-302; n = 90), that overlies the Pulo do Lobo Formation also yields a large proportion of Cryogenian–Ediacaran ages (68%; ca. 839–598 Ma), and a significant number of Paleoproterozoic ages (22%; ca. 2.4–1.6 Ga). Lower to Middle Devonian ages (ca. 398–384 Ma) make up ca. 5% of the spectrum. Sxubordinate Cambrian (2%; ca. 526 Ma and 486 Ma), Tonian (1%; ca. 991 Ma), Mesoproterozoic (1%; ca. 1.1 Ga) and Archean (1%; ca. 2.6 Ga) ages also occur (Fig. 5). The four concordant analyses making up the youngest peak in the age distribution pattern yield a concordia age of 390.5 ± 4.7 Ma (Middle Devonian) and provide a maximum depositional age for the Atalaia quartzite.

The detrital zircon age distribution of the greywacke of the Gafo Formation (sample SP-101; n = 81), sampled stratigraphicaly above the Atalaia quartzite, is also dominated by Cryogenian–Ediacaran ages (70%; ca. 751–552 Ma), displaying a dominant cluster at ca. 630 Ma.

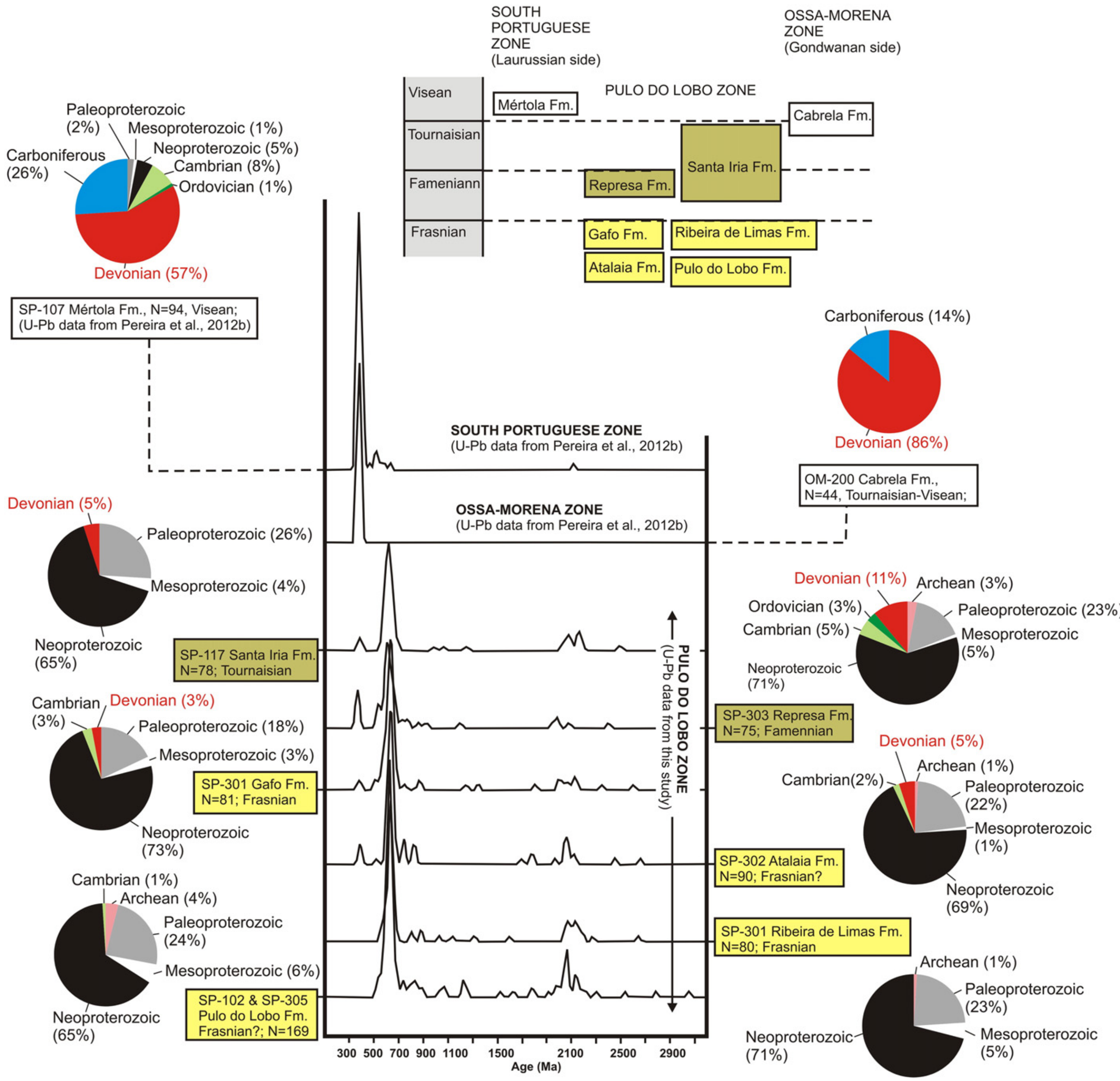

Fig. 5. Zircon age spectra and pie charts showing the age distribution of detrital zircon of samples from the Pulo do Lobo Zone (this study) and from Lower Carboniferous greywackes from the Ossa-Morena and South Portuguese zones.
(From Pereira et al., 2012b).

The remainder of the grains (ca. 30%) is dominated by Paleoproterozoic (18%; ca. 2.3–1.8 Ga) ages, with subordinate Mesoproterozoic (3%; ca. 1.3 Ga and 1.2 Ga), Tonian (3%; ca. 872 Ma and 864 Ma), Cambrian (3%; ca. 518 Ma and 515 Ma) and Middle Devonian (3%; ca. 384 Ma and 383 Ma) ages (Fig. 5). The two youngest concordant grains provide a concordia age of 384 ± 6 Ma, which is interpreted as the maximum depositional age for the sample and is compatible with the biostratigraphic age (Pereira et al., 2007).

Most of 75 zircon grains of the quartzwacke from the Represa Formation (sample SP-303), sampled in a stratigraphic position above the Gafo Formation, are Cryogenian–Ediacaran in age (59%; ca. 782–552 Ma). However, a significant number of grains have Paleoproterozoic (16%; ca. 2.4–1.7 Ga) and Upper Devonian (11%; ca. 375–367 Ma) ages (Fig. 5). The remainder of the grains yield Cambrian (5%; ca. 540–529 Ma), Archean (3%; ca. 3.1 Ga and 2.8 Ga), Tonian (2%; ca. 927 Ma and 862 Ma) and Ordovician (3%; ca. 457 Ma and 450 Ma) ages (Fig. 5). Only one Mesoproterozoic grain was found. Eight concordant grains from the youngest population provide a maximum depositional age of 372 ± 3 Ma (concordia age) for the sample, almost coincident with the biostratigraphic age (Pereira et al., 2007).

The detrital zircon age distribution of the Santa Iria Formation greywacke (sample SP-117; n = 78), which unconformably overlies the Ribeira de Limas Formation is also dominated by Ediacaran–Cryogenian ages (64%; ca. 680–559 Ma) (Fig. 5), with the remainder of the grains being dominated by Paleoproterozoic (26%; ca. 2.4–1.7 Ga) ages. Subordinate Middle Devonian (5%; ca. 380–370 Ma), Mesoproterozoic (4%; ca. 1.4–1 Ga) and Tonian (1%; ca. 986 Ma) ages occur (Fig. 5). The younger grains provide a maximum depositional age of 376 ± 4 Ma (concordia age obtained from three concordant analyses) for the sample. This age determination is in general agreement with the age interval of ca. 375–365 Ma (Late Frasnian–Famennian) obtained from the youngest detrital zircon by Pérez-Cáceres et al. (2017), but is older than the maximum depositional age of ca. 347 Ma (Tournaisian) estimated by Braid et al. (2011).

In contrast with the results from the Devonian rocks reported, results from detrital zircon ages of two Lower Carboniferous greywacke samples collected on opposite sides of the Rheic suture were reported by Pereira et al. (2012b) with significant differences. Both samples contained abundant zircon grains with Devonian–Carboniferous ages. As new Hf isotope data of those grains with Devonian–Carboniferous ages are presented in this study, we now summarize these data. Forty-four zircon grains from the Tournaisian–Visean greywacke of the Cabrela Formation (sample OM-200; Ossa-Morena Zone; Gondwanan side) yield mainly Middle to Upper Devonian ages (81.9% in the interval ca. 387–359 Ma). A maximum sedimentation age of ca. 353 Ma is inferred from the analyses corresponding to the youngest peak (n = 4) in the detrital zircon age spectrum. In this sample, pre-Devonian ages are lacking.

Of 94 detrital zircon ages obtained from a Visean greywacke of the Mértola Formation (sample SP-107, South Portuguese Zone, Laurussian side), 66% yield Mid-Devonian–Lower Carboniferous ages with Upper Devonian and Middle Devonian peaks at ca. 369 Ma and ca. 391 Ma (see Pereira et al., 2012b). Early Carboniferous zircon was dated at ca. 353–349 Ma. The youngest zircon population (n = 5) provides a maximum depositional age of ca. 343 Ma. Pre-Devonian ages range from Paleoproterozoic to Silurian, with two age peaks corresponding to Late Neoproterozoic and Early Cambrian zircon-forming events.

5.2. Hf isotope data

To better characterize the source(s) of Devonian and Carboniferous detrital zircon we analyzed 119 grains with concordant U–Pb ages between ca. 398 and 340 Ma for their Hf isotope compositions (Table 2; see Electronic supplement). Samples comprise grains from siliciclastic strata of: i) the Atalaia (SP-302), Gafo (SP-101), Represa (SP-303) and Santa Iria (SP-117) formations of the Pulo do Lobo Zone (first reported in this study), and ii) the Cabrela (OM-200) and Mértola (SP-107) formations from the Ossa-Morena (Gondwanan side) and South Portuguese (Laurussian side) zones respectively (U–Pb data reported by Pereira et al., 2012b).

The 119 analyses show εHf_T values ranging between −8.0 and +8.3 (Fig. 6; Table 2). Zircon of the Mértola Formation (sample SP-107, South Portuguese Zone) shows the most juvenile Hf isotope signatures with the majority of Devonian and Carboniferous zircon grains (90%) having positive εHf_T values ranging from +8.2 to +0.7 and model ages (T_{NC}) between 0.64 to 1.13 Ga, respectively. Only six of the 61 grains gave negative εHf_T values between −0.5 and −5.1 in combination with model ages from 1.22 to 1.49 Ga. Remarkably, Hf isotope signatures of Devonian and Carboniferous zircon grains of the Cabrela Formation (sample OM-200, Ossa-Morena Zone) show very little overlap with the Hf data of the Mértola Formation. Their εHf_T range from +2.3 to −2.7 and T_{NC} are between 1.03 and 1.34 Ga (Fig. 6). Devonian zircon grains of the Gafo, Atalaia and Represa formations (samples SP-101, SP-302 and SP-303, respectively; Pulo do Lobo Zone) are similar to those of the Cabrela Formation (Ossa Morena Zone) with regard to their Hf isotope characteristics (εHf_T + 2.6 to −2.3, T_{NC} 1.01–1.32 Ga). Devonian zircon of the Santa Iria Formation (sample SP-117, Pulo do Lobo Zone) has distinctly more negative εHf_T values (−4.2 to −8.2) and older model ages (T_{NC} 1.45–1.65 Ga) than zircon from the other units (Fig. 6).

6. Discussion

6.1. Provenance of Late Devonian–Early Carboniferous sedimentary rocks

The Pulo do Lobo Zone defines a sigmoidal tectonostratigraphic belt (Fig. 1) which at its eastern tail is predominantly composed of tectonically imbricated, polydeformed metasedimentary units whose stratigraphic relationships are debatable (Spanish sector; Braid et al., 2010; Dahn et al., 2014; Pérez-Cáceres et al., 2015). However, in the center of the sigmoidal structure, the Variscan deformation is less intense, and preserves an anticline, known as the Pulo do Lobo antiform (Fig. 3), where stratigraphic relations are locally preserved (Portuguese sector; Carvalho et al., 1976; Oliveira, 1990; Silva et al., 1990) and depositional ages are constrained by palynostratigraphic studies (Pereira et al., 2007). The youngest concordant detrital zircon grains provide an Eifelian

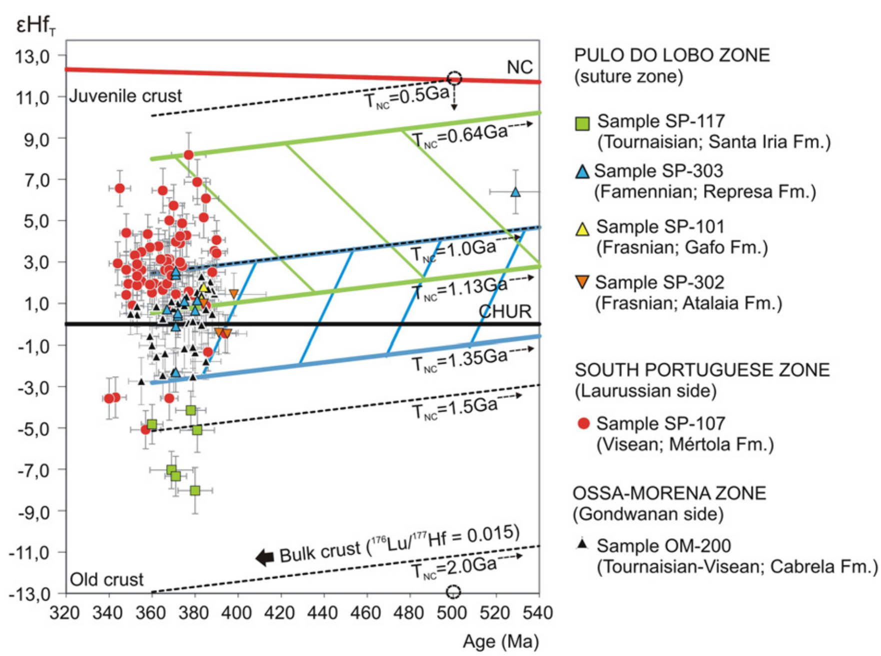

Fig. 6. Diagram of initial εHf_T values versus age showing Devonian and Carboniferous detrital zircon from the Pulo do Lobo, Ossa-Morena and South Portuguese zones. The black and red lines represent the isotope evolution of the CHUR (chondritic uniform reservoir) through time and new crust (NC; Dhuime et al., 2011), respectively.

maximum depositional age (ca. 391 Ma) for the Atalaia Formation and a Givetian–Frasnian age (ca. 384 Ma) for the Gafo Formation, the latter in agreement with the assigned biostratigraphic age. The Pulo do Lobo and Atalaia formations did not provide palynomorphs, probably due to biotite-zone greenschist facies metamorphism (Pereira et al., 2007). The similar detrital zircon age spectra of the Pulo do Lobo and Ribeira de Limas formations (Fig. 5) suggest similar source rocks, in agreement with the gradational contact observed in the field. The youngest zircon grains (Ediacaran and Cambrian) are not present in sufficient quantities to provide a statistically representative maximum depositional age. The Ribeira de Limas and Atalaia formations containing Frasnian palynomorph associations (Pereira et al., 2007) are correlatives of the Ronquillo Formation (Pyrite Belt; González et al., 2004; Fig. 2).

Pre-Devonian detrital zircon populations of the Pulo do Lobo Zone formations (excluding the Horta da Torre Formation) are dominated by Late Neoproterozoic and Paleoproterozoic ages, and contain a small number of Archean grains (Fig. 5), indicating a likely provenance in peri-Gondwanan terranes (Fernández-Suárez et al., 2002, 2014; Gutiérrez-Alonso et al., 2015; Nance et al., 2008). The Late Neoproterozoic age peak in the detrital zircon spectra are thought to reflect Cadomian–Avalonian and/or Pan-African arc magmatism in the source area (Abati et al., 2010; Drost et al., 2011; Linnemann et al., 2004, 2008; Nance et al., 2008). Abundant Late Neoproterozoic detrital zircon ages and/or igneous activity are also known from Ediacaran to Cambrian siliciclastic rocks of the Ossa-Morena Zone (Álvaro et al., 2014; Pereira et al., 2008, 2011, 2012b, 2012c), from Ediacaran to Early Devonian siliciclastic rocks of the Meguma terrane (Krogh and Keppie, 1990; Murphy et al., 2004; Waldron et al., 2011) and from Ediacaran to Ordovician siliciclastic rocks of Avalonia (Henderson et al., 2016; Pollock et al., 2015; Willner et al., 2013), suggesting either a common provenance with or derivation from these sources. The Meguma terrane and Avalonia, although of Gondwanan affinity as the Ossa-Morena Zone in the Late Neoproterozoic, resided along the southern margin of Laurussia by Devonian times, i.e. prior to Laurussia–Gondwana collision (Murphy et al., 2016). Late Ordovician to Early Devonian rocks of the Meguma terrane and coeval strata of Avalonia contain Mesoproterozoic zircon (ca. 1.0–1.4 Ga; Murphy et al., 2004) that is usually absent or very rare in Ediacaran to Cambrian siliciclastic rocks of the Ossa-Morena Zone (Cambeses et al., 2017; Linnemann et al., 2008; Pereira et al., 2008, 2011, 2012c). Mesoproterozoic zircon (<6%) found in the Pulo do Lobo Zone formations (excluding the Santa Iria and Horta da Torre formations and the Alajar Mélange; Braid et al., 2011; Pérez-Cáceres et al., 2017) suggests that potential source areas can be narrowed down to the Laurussian margin pointing to provenance either from or in common With Late Ordovician to Early Devonian siliciclastic rocks of the Meguma terrane and/or Avalonia (Braid et al., 2011; Keppie et al., 1998; Murphy et al., 2004; Nance et al., 2008). The quartzites of the Horta da Torre Formation and Alájar Mélange contain an important population of Mesoproterozoic and Paleoproterozoic zircon (ca. 1.9–1.1 Ga) which is typical of Laurentian and Baltica sources (Braid et al., 2011). These detrital zircon populations have striking similarities with those found in Early Carboniferous Kammquartzite Formation, from the Lahn-Dill area (Rhenohercynian Zone; Eckelmann et al., 2014). The Horta da Torre and the Phyllite-Quartzite (Pyrite Belt) formations also contain Early Silurian (ca. 440 Ma) detrital zircon ages suggesting a Caledonian–Appalachian provenance typical of the Laurussian margin (Pérez-Cáceres et al., 2017). Early Silurian magmatism is reported in Laurentia and in the Meguma terrane (Murphy et al., 2004) but is lacking in the Ossa-Morena Zone (Robardet and Gutiérrez-Marco, 2004) which therefore can be excluded as a possible source.

The Devonian zircon population of siliciclastic rocks from the Ribeira de Limas, Atalaia and Ronquillo formations includes Early Devonian grains (ca. 400–390 Ma; Pérez-Cáceres et al., 2017). To our knowledge, potential source rocks of the Early Devonian zircon are absent in the Ossa-Morena Zone, but grains of similar age were found in the Early Devonian siliciclastic rocks of the Meguma terrane (Murphy et al., 2004) and they probably represent detritus from eroded early magmatic arcs related to the Rheic Ocean closure (Cawood et al., 2012; Pereira et al., 2012b). The Ribeira de Limas, Atalaia and Ronquillo formations have similar detrital zircon populations to the Frasnian–Famennian siliciclastic rocks of the Gieβen and Hörre nappes, also in the Lahn-Dill area (Rhenohercynian Zone; Eckelmann et al., 2014), suggesting that they may have had similar sources.

The siliciclastic rocks of the Gafo and Represa (Frasnian to Famennian; Pulo do Lobo Zone) formations and of the Phyllite-Quartzite Formation (Famennian; Pyrite Belt) have similar pre-Devonian populations and also include Late Devonian zircon (ca. 384–366 Ma). The presence of detrital zircon with abundant ages close to the deposition age of the Devonian Pulo do Lobo Zone metasedimentary formations suggests that they were deposited in close temporal relationship with a convergent setting (Cawood et al., 2012) implying magmatism between ca. 400 Ma and 359 Ma. Exposed Devonian arc-related igneous rocks are volumetrically insignificant in SW Iberia on either side of the Rheic suture zone. The only known magmatic rocks of this age in the Ossa-Morena Zone (Gondwanan side) are the calc-alkalic mafic volcanic rocks interbedded with the allegedly Emsian–Eifelian Odivelas limestones (Machado et al., 2009, 2010; Silva et al., 2011). In the Pyrite Belt (Laurussian side), magmatic zircon from Frasnian to Famennian volcanic rocks (including the Cercal felsic volcanic rocks ca. 374–372 Ma; Rosa et al., 2008a) and inherited Givetian–Frasnian zircon (ca. 385 Ma and ca. 373 Ma; Oliveira et al., 2013) found in Tournaisian rhyodacites are also potential sources. The former large-scale presence of arc complexes, probably involving intra-oceanic arcs, is, however, indicated by detrital zircon in siliciclastic rocks derived therefrom (Pereira et al., 2012b).

The largest batholiths of the entire Appalachian orogen are Devonian and occur in the Meguma terrane, along the Laurussian margin of the Rheic Ocean (Moran et al., 2007; Tate and Clarke, 1995). Along the Laurussian side, regional deformation and metamorphism in the Meguma terrane occurred at ca. 415–390 Ma (Keppie and Dallmeyer, 1995), ca. 406–388 Ma (Hicks et al., 1999) and ca. 400–373 Ma (White and Barr, 2012), and was associated with granitoid plutonism around ca. 380–370 Ma (Clarke et al., 1997; Murphy et al., 1999). Frasnian–Famennian mafic intrusions (ca. 376–372 Ma) are probably related to the subduction of Rheic oceanic lithosphere beneath the Meguma terrane (Moran et al., 2007; Tate and Clarke, 1995), likewise the Famennian felsic volcanism of the Iberian Pyrite Belt (ca. 374–372 Ma; Cercal volcanic sedimentary complex; Rosa et al., 2008a). Moreover, detrital zircon of the same age of the Meguma terrane plutons and detrital muscovite dated at ca. 370 Ma (Dallmeyer et al., 1997) occur in the unconformably overlying Tournaisian continental clastic rocks of the Horton Group (Murphy, 2000; Murphy and Hamilton, 2000), and *P-T-t* studies of Meguma terrane rocks (e.g., Keppie and Dallmeyer, 1995) indicate ca. 10 km of rapid uplift of between ca. 375 Ma and 360 Ma. The paucity of volcanic arc rocks preserved either as bedrock or as clasts in the Horton Group suggests that felsic magmas did not reach the surface in significant amounts, an explanation consistent with the lack of available Devonian zircon grains until the Early Carboniferous.

The significant increase in the number of Mid–Late Devonian detrital zircon higher in the stratigraphy is interpreted as the progressive erosion of the Rheic Ocean magmatic arc(s). Proximity of the Laurussia to Gondwana in Early Carboniferous time is inferred from the presence of Early Carboniferous detrital zircon ages (ca. 359 Ma to 342 Ma) in the Tournaisian Santa Iria greywackes of the Pulo do Lobo Zone (suture; Braid et al., 2011; Pérez-Cáceres et al., 2017), and Visean greywackes of Cabrela and Mértola greywackes (both sides of the suture; Pereira et al., 2012b). Detrital zircon age spectra of the Santa Iria Formation of the Pulo do Lobo zone and Elnhausen Formation of the Rhenohercynian Zone (Hörre nappe in the Lahn-Dill area; Eckelmann et al., 2014) are virtually identical with the only difference being the presence of ~4% of Mesoproterozoic grains in the siliciclastic rocks of the Pulo do Lobo Zone.

Early Carboniferous magmatism is voluminous in the Ossa-Morena Zone (Cambeses et al., 2015; Jesus et al., 2007; Lima et al., 2012; Moita et al., 2015; Pereira et al., 2009, 2015) and in the South Portuguese Zone (Pyrite Belt volcanism and plutonic rocks from the Sierra Norte Batholith; de la Rosa et al., 2002; Dunning et al., 2002; Gladney et al., 2014; Oliveira et al., 2013; Rosa et al., 2008a, 2008b). Similarly, in the Lahn-Dill area of the Rhenohercynian Zone widespread Early Carboniferous volcanism was recognized (Königshof and Linnemann, 2008 and references therein). Early Carboniferous metamafic rocks are also found in the Pulo do Lobo Zone (Dahn et al., 2014; Pérez-Cáceres et al., 2015) and in the Beja-Acebuches ophiolitic complex (Azor et al., 2008).

6.2. Birth and demise of the Rheic Ocean magmatic arc(s)

The Devonian–Carboniferous detrital zircon of SW Iberia siliciclastic rocks from which we obtained the Hf isotope compositions suggest an interval of magma generation and crystallization coinciding with the Rheic Ocean subduction. However, different models for the timing of final closure of the oceanic basin and polarity of the subduction zone or zones have been proposed (Fig. 7): i) a type-1 subduction model assuming subduction of Rheic oceanic lithosphere beneath the Gondwanan margin (Braid et al., 2010; Ribeiro et al., 2010); ii) a type-2 subduction model involving subduction of Rheic oceanic lithosphere beneath the Laurussian margin (Gutiérrez-Alonso et al., 2008; Martínez Catalán et al., 2009; Simancas et al., 2009; Woodcock et al., 2007) and iii) a type-3 subduction model which invokes a doubly divergent system with the opening of the Rhenohercynian oceanic basin (Eckelmann et al., 2014; Franke, 2000; von Raumer et al., 2016 and references therein).

The lack of exposed Devonian magmatic rocks in SW Iberia that could provide a source for the Devonian detrital zircon grains reflect a broader problem, i.e. the widespread paucity of subduction-related igneous rocks that record convergence and closure of the Rheic Ocean (Eckelmann et al., 2014; Pereira et al., 2012b). In the Ossa-Morena Zone (Gondwanan side), there are only few outcrops of Lower Devonian calc-alkaline mafic rocks interbedded in the Odivelas limestones (Fig. 1), which are considered a vestige of a Rheic Ocean magmatic arc (Silva et al., 2011). The Early Devonian arc magmatism, synorogenic deposition (Giese et al., 1994) and crustal shortening with development of southwest-verging recumbent folds (Expósito et al., 2002) suggest convergent tectonics at that stage with north-dipping (present-day coordinates) subduction of the Rheic oceanic lithosphere beneath Gondwanan margin (type-1 subduction model; Ribeiro et al., 2010) (Fig. 7). Nevertheless, based on the striking similarities with the coeval volcanic activity reported in the Lahn-Dill area of the Rhenohercynian Zone (Königshof and Linnemann, 2008 and references therein), we cannot exclude the possibility that the Odivelas volcanism might represent

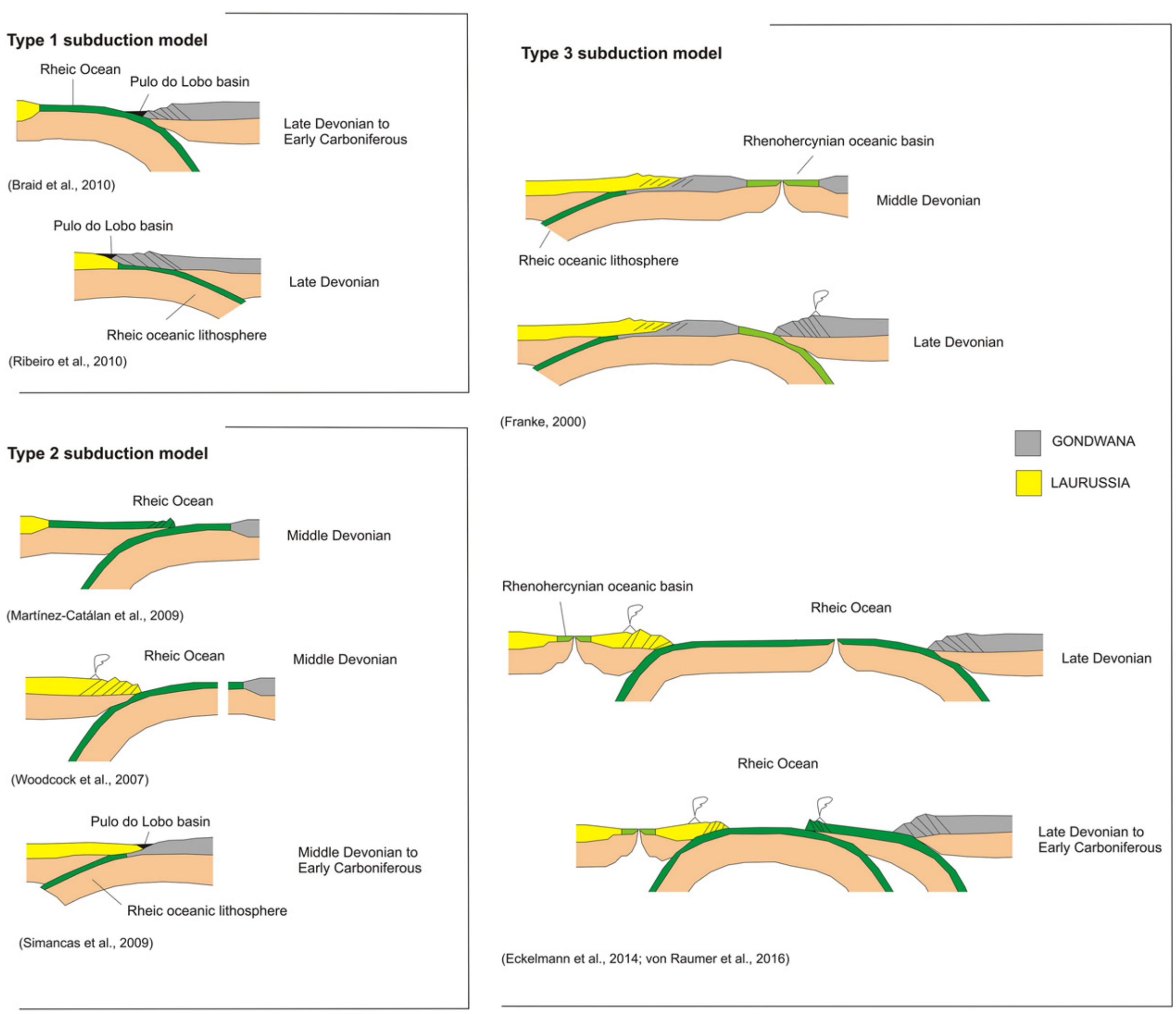

Fig. 7. Types of subduction models proposed for the Rheic suture in Western-Central Europe.
(Adapted from Braid et al., 2010; Eckelmann et al., 2014; Franke, 2000; Martínez Catalán et al., 2009; Ribeiro et al., 2010; Simancas et al., 2009; von Raumer et al., 2016; Woodcock et al., 2007).

a fragment derived from the Laurussian margin, and therefore may not necessarily reflect type-1 subduction model.

Recent studies show that the Rheic Ocean closure and Laurussia–Gondwana collisional record is more readily interpreted from the nature and content of the strata deposited in Late Devonian–Early Carboniferous basins adjacent to the Pulo do Lobo Zone in SW Iberia (Braid et al., 2011; Pereira et al., 2012b, 2013; Pérez-Cáceres et al., 2017; Rodrigues et al., 2014) and in the Rhenohercynian Zone in Central Europe (Eckelmann et al., 2014; Franke and Dulce, 2016; von Raumer et al., 2016). The proportion of Frasnian–Famennian detrital zircon ages is considerably higher in Tournaisian–Visean turbidites of SW Iberia (the Cabrela and Mértola formations; Pereira et al., 2012b; Rodrigues et al., 2014) compared to their proportion in the Devonian formations implying a significant increase in the surface exposure of Middle–Late Devonian magmatic source rocks between Upper Devonian and the Upper Carboniferous times (Fig. 5). In the Rhenohercynian Zone, detrital zircon populations in the Frasnian to Moscovian clastic rocks, that probably overlie a Meguma/Avalonia-type basement, are thought to document the evolution of a Late Paleozoic oceanic basin ("Rhenohercynian Ocean"), but there is no consensus if the ocean was formed during or after the Rheic Ocean closure. Some authors suggest that the Rhenohercynian rift basin along the Laurussian margin following the Early Devonian closure of the Rheic Ocean (type-2 subduction model; Franke, 2000; Fig. 7). Others argue, however, that the opening of the Rhenohercynian basin might have started before closure of the Rheic Ocean as a back-arc basin related to the Late Devonian–Early Carboniferous subduction beneath the Laurussian margin (type-3 subduction model; Eckelmann et al., 2014; von Raumer et al., 2016; Fig. 7).

Thus, the identification and characterization of Devonian detrital zircon is crucial as it could represent witnesses of a magmatic arc that was largely removed by one or a combination of the following processes: (i) erosion, (ii) strike-slip displacement and/or (iii) subduction erosion. Another important feature to understand is whether Devonian zircon crystallized from juvenile magmas, similar to those formed in intra-oceanic magmatic arcs (Pereira et al., 2012b), or from crustally influenced magmas that likely occur in magmatic arcs that form on continental crust. Although our siliciclastic samples contain abundant zircon with positive εHf_T values, particularly in Devonian zircon grains of the Visean Mértola Formation (37 of 40 grains: εHf values of $+8.2$ to $+0.7$; South Portuguese Zone, Laurussian side), no zircon Hf isotope composition overlaps within error with that of contemporary juvenile crust (Fig. 6). Therefore, an entirely intra-oceanic formation of the source rocks is unlikely. Model ages of the majority of the Devonian zircon grains of the Mértola Formation range between 1.13 and 0.67 Ga pointing either to melting of pre-existing crust (Avalonian/Cadomian crust with sedimentary cover) or to mantle derived magmas that were contaminated by older crustal components. The presence of a few grains (3 of 40) with older model ages (1.40 to 1.22 Ga) may be in favor of the latter scenario.

Devonian zircon grains of the Tournaisian-Viséan Cabrela Formation ($\varepsilon Hf_T = +2.53$ to -2.35; Ossa-Morena Zone, Gondwanan side), the Frasnian Gafo ($\varepsilon Hf_T = +1.2$ and $+1.8$) and Atalaia ($\varepsilon Hf_T = +1.4$ to -0.5) formations as well as of the Famennian Represa Formation ($\varepsilon Hf_T = +2.6$ to -2.3) fall in a similar range but show only little overlap with those zircon grains of the Mértola Formation. Compared to the Mértola Formation of the South Portuguese Zone the Devonian Pulo do Lobo and Ossa Morena zones zircon grains have slightly older Hf model ages between 1.34 and 1.01 Ga suggesting to zircon crystallization from a magma derived from an on average slightly older source. This signature can, again, be derived from remelting of pre-existing crust with respective average ages or from contamination of a juvenile magma with older crustal material. There was only one grain (Represa Fm.) from which we obtained ages and Hf isotope data for core (529 ± 12 Ma, $T_{NC} = 0.88$ Ga) and rim (367 ± 9 Ma, $T_{NC} = 1.12$Ga), whereas the data for the core provide evidence for the involvement of Cambrian igneous rocks or sediments derived therefrom. The oldest model ages ($T_{NC} = 1.70$ to 1.45 Ga) and most negative εHf_T values (-4.2 to -8.0) are preserved in Devonian zircon grains of the Famennian to Tournaisian Santa Iria Formation of the Pulo do Lobo Zone. These data show virtually no overlap with Hf data of the remaining stratigraphic units of this study (Fig. 6) and suggest zircon formation from a heavily contaminated juvenile magma or more likely from continental crust.

The Hf model ages of the Santa Iria Formation overlap Nd model ages of ~1.9 to 1.5 Ga reported from the Ossa-Morena Zone Ediacaran and Ordovician sedimentary rocks and crustally derived Cambrian-Ordovician magmatism (López-Guijarro et al., 2008; Cambeses et al., 2017). Those results indicate recycling of Paleoproterozoic (and Archean) material either from underlying crust or more likely from a common and abundant detrital component in Neoproterozoic and Paleozoic siliciclastic rocks.

The younger Hf model ages obtained on Devonian zircon (this study) overlap Nd model ages of ~1.3 to 0.8 Ga of crustally derived Early Silurian volcanic rocks and Late Devonian plutons of the Meguma terrane (Keppie et al., 1997; Moran et al., 2007) as well as Ediacaran–Early Cambrian igneous rocks from West Avalonia (Nance et al., 2008 and references therein). This suggests that the basement upon which the Devonian arc was built is isotopically more juvenile than the basement of the Ossa Morena Zone, and has most likely Laurussian affinity.

As mentioned above, the wide range in εHf_T values indicates that the Devonian zircon probably crystallized in magmas formed in a convergent setting that contained both mantle input and crustal components. An alternative hypothesis is that the island arcs may have nucleated above thinned, ancient continental basement that was rifted and transported from a continental margin flanking the Rheic Ocean, in a manner analogous to the Early Miocene Vanuatu island arc of the western Pacific Ocean which preserves evidence of a continental basement that was rifted from the eastern Australian margin (Buys et al., 2014). The wide range in εHf values can also be explained by mantle recycling, as the volume of sediment input into the mantle by subduction erosion and sediment subduction will be higher in a convergent setting (Kemp et al., 2009; Roberts and Spencer, 2014). The Ronquillo Formation of the Pyrite Belt (also named La Minilla Formation) has been recently interpreted to correspond to material derived from a Devonian Rheic Ocean arc later subducted beneath the Laurussian margin (Rubio Pascual et al., 2013). The Hf model ages of Devonian zircon from the Pulo do Lobo, South Portuguese and Ossa-Morena zones siliciclastic rocks indicate that the Rheic Ocean arcs were built on a Meguma/Avalonia type basement (Fig. 8). The provenance of the Roquillho, Pulo do Lobo, Ribeira de Limas, Gafo and Atalaia formations suggest that the opening of a back-arc basin along the Laurussian active margin favored deposition of voluminous clastic strata in Frasnian times (Fig. 8A). Crustal shortening probably driven by a renewed push convergence with Gondwana at ca. 370 Ma (Arenas et al., 2014; Díez Fernández et al., 2016), probably triggered inversion of the Pulo do Lobo back-arc basin (Fig. 8B), causing deformation of the Frasnian rocksand significant crustal uplift that explain the Late Devonian sedimentary gap of ca. 14 My recognized between the Frasnian and Famennian formations (Pereira et al., 2007).

Few Carboniferous zircon grains from the Early Carboniferous Mértola and Cabrela formations show negative εHf_T values of -5 to -2.7 and $T_{NC} = \sim 1.49$ to 1.34 Ga. The majority of Carboniferous zircon grains has, however, Hf model ages ($T_{NC} = \sim 0.85$ to 1.1 Ga) that largely overlap Nd whole-rock model ages of ~0.9–1.2 Ga obtained from crustally-derived Early Carboniferous volcanic (Pyrite Belt; Rosa et al., 2008a) and plutonic (Sierra Norte Batholith of the South Portuguese Zone; de la Rosa et al., 2002) rocks. The basement of the South Portuguese Zone is not exposed but its identity has been inferred to be Meguma/Avalonian type from Paleozoic reconstructions

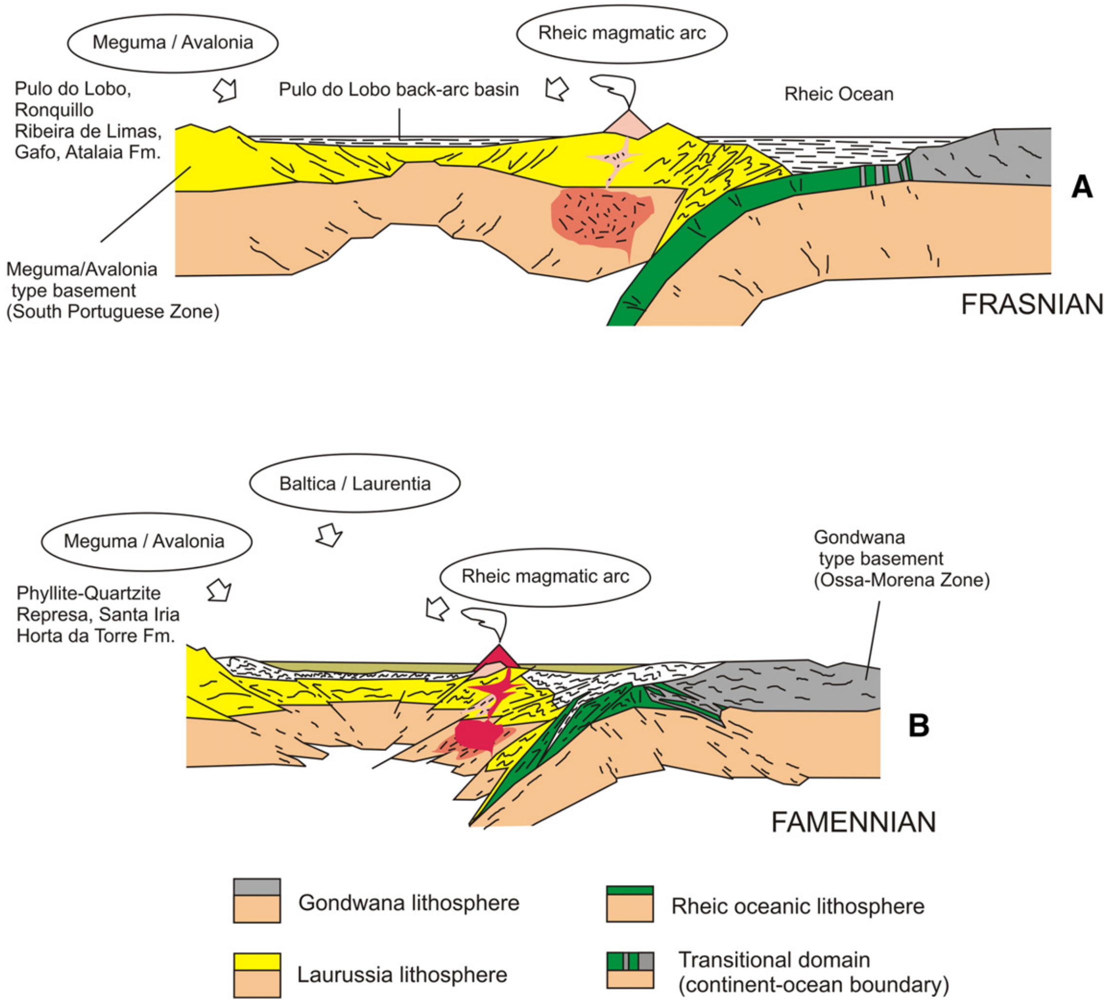

Fig. 8. Proposed evolutionary model of the Pulo do Lobo back-arc basin in Late Devonian with indication of provenance. (Adapted from Eckelmann et al., 2014; Rubio Pascual et al., 2013).

combined with isotopic data from crustally derived igneous rocks (Murphy et al., 2015 and references therein).

7. Conclusions

Detrital zircon age spectra from the Late Devonian–Early Carboniferous Pulo do Lobo Zone formations (excluding the Horta da Torre Formation and Alajar Mélange) are dominated by Neoproterozoic and Proterozoic grains and resemble in many aspects the age distribution patterns common to peri-Gondwanan terranes, such as the Meguma terrane, Avalonia and the Ossa-Morena Zone. The presence of Mesoproterozoic and Early Silurian zircons, however, rules out a provenance from or a source in common with the Ossa-Morena Zone (Fig. 8), and instead points to a linkage with Avalonia and/or the Meguma terrane. In contrast, recycling of Baltica/Laurentia-derived zircon grains is recorded in the Horta da Torre Formation and the Alajar Mélange. Based on the distribution of zircon age populations, the Pulo do Lobo Zone siliciclastic rocks exhibits a typical Laurussian provenance (Fig. 8).

Arc systems which record the Rheic Ocean subduction are scarcely preserved in the Ouachita–Appalachian–Variscan orogenic belt. The preservation of Devonian detrital zircon in Late Devonian–Early Carboniferous siliciclastic rocks of SW Iberia, rather than in arc-related igneous rocks indicates that the arc system may have been largely and progressively destroyed by erosion. In the Pulo do Lobo Zone, that represents a reworked Late Paleozoic suture zone located between Laurussia and Gondwana, the Famennian-Tournaisian formations contain more abundant Mid–Late Devonian zircon than the Frasnian formations. Evaluation of Devonian detrital zircon Hf isotope compositions from Late Devonian to Early Carboniferous formations of the Pulo do Lobo (suture zone), South Portuguese (Laurussian side) and Ossa-Morena (Gondwanan side) zones suggests that the basement upon which the Devonian arc was built is younger (Meguma/Avalonian type basement) than the more evolved basement of the Ossa-Morena Zone (Gondwana side).

Altogether, our data support an evolutionary model of Devonian back-arc basin deposition of the Pulo do Lobo Zone formations (Rubio Pascual et al., 2013) along the Laurussian active margin during Rheic Ocean subduction. A similar tectonic framing is proposed for the Rhenohercynian Zone in Central Europe (Eckelmann et al., 2014; von Raumer et al., 2016) (Fig. 8).

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