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Significance of detrital zircons in Siluro-Devonian rocks from Iberia

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Abstract: Seven samples of Siluro-Devonian sedimentary rocks from the Cantabrian and Central Iberian zones of the Iberian Variscan belt have been investigated for provenance and contain four main age populations in variable relative proportion: Ediacaran–Cryogenian (*c*. 0.55–0.8 Ga), Tonian–Stenian (0.85–1.2 Ga), Palaeoproterozoic (*c*. 1.8–2.2 Ga) and Archaean (*c*. 2.5–3.3 Ga). Five samples contain very minor Palaeozoic (Cambrian) zircons and six samples contain minor but significant zircons of Middle and Early Mesoproterozoic (Ectasian–Calymmian, 1.6–1.8) age. These data highlight the transition from an arc environment to a stable platform following the opening of the Rheic Ocean. Variations in detrital zircon populations in Middle–Late Devonian times reflect the onset of Variscan convergence between Laurussia and Gondwana. The presence of a high proportion of zircons of Tonian–Stenian age in Devonian sedimentary rocks may be interpreted as (1) the existence of a large Tonian–Stenian arc terrane exposed in the NE African realm (in or around the Arabian–Nubian Shield), (2) the participation, from the Ordovician time, of a more easterly alongshore provenance of Tonian–Stenian zircons, and (3) an increase in the relative proportion of Tonian–Stenian zircons with respect to the Ediacaran–Cryogenian population owing to the drift of the Avalonian–Cadomian ribbon continent, or the progressive burial of Ediacaran–Cryogenian rocks coeval with the denudation of older source rocks from the craton interior.

Supplementary material: Tables with the analytical data and the geochronological results are available at http://www.geolsoc.org.uk/SUP18812.

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The Palaeozoic ocean that bordered the northern coast of Gondwana from the Ordovician until its closure in the Late Devonian–early Carboniferous is known as the Rheic Ocean (e.g. Nance *et al*. 2010, and references therein). It separated Gondwana from Laurussia (Laurentia–Baltica–Avalonia) following the closure of the Iapetus Ocean (e.g. Nance & Linnemann 2008). The closure of the Rheic Ocean produced a suture that extends over 10000 km from Mexico to Turkey, gave rise to the vast Variscan–Alleghanian–Ouachita orogen (e.g. Nance *et al*. 2010, and references therein) and assembled the supercontinent Pangaea.

One of the most complete sections of the Rheic passive margin sequence of Gondwana is exposed in the Iberian Massif (western Iberia, Fig. 1) where Palaeozoic rocks lie within a complex arcuate segment of the Variscan orogenic belt (Weil *et al*. 2001, 2013; Catalan 2011, 2012; Shaw *et al*. 2012). The Rheic Ocean was bordered on its southern side by an extensive passive margin (Pastor-Galan *et al*. 2013*a*,*b*) that originated after the rift–drift of Avalonia from northern Gondwana at the Cambrian–Ordovician boundary (Murphy *et al*. 2006, 2008). The passive margin, now preserved in Iberia, was more than 200 km wide (Murphy *et al*. 2008) and previous interpretations are consistent with palaeogeographical reconstructions that place NW Iberia adjacent to North Africa along the southern flank of the Rheic Ocean during the Palaeozoic (e.g. Robardet 2002, 2003; Catalan *et al*. 2007; Fernández-Suárez *et al*. 2014; Shaw *et al*. 2014).

The provenance of the clastic sediments that constitute much of the Rheic passive margin sequence is not well constrained. To date, the main constraint is provided by abundant Tonian–Stenian detrital zircons whose source in the African basement is not known. Neoproterozoic rocks of NW Iberia are characterized by detrital zircon U–Pb ages between *c*. 0.9 and 1.2 Ga (Fernandez-Suarez *et al*. 2000, 2002; Gutierrez-Alonso *et al*. 2003; Pereira *et al*. 2012*b*,d; Pastor-Galan *et al*. 2013*b*; Fernández-Suárez *et al*. 2014; Shaw *et al*. 2014) and detrital micas with ^{40}Ar–^{39}Ar ages between *c*. 0.85 and 1.1 Ga and between *c*. 1.55 and 1.8 Ga (Gutiérrez-Alonso *et al*. 2005), the latter mica ages being missing in the detrital zircon record. The *c*. 0.85–1.2 Ga zircons and micas were initially interpreted as having been derived from a terrane of Amazonian affinity and deposited on the unknown Iberian basement, which was subsequently transported along the northern margin of Gondwana in late Ediacaran times (Fernandez-Suarez *et al*. 2000). However, the existence of detrital zircons with similar age in Cambrian and Ordovician rocks of North and NE Africa, which it is now known as the Arabian–Nubian Shield (Wilde & Youssef 2002; Avigad *et al*. 2003, 2007, 2012; Cox *et al*. 2004, 2012; Kolodner *et al*. 2006; Horton *et al*. 2008; Ramos *et al*. 2008; Be'eri-Shlevin *et al*. 2009; Meinhold *et al*. 2011; Ustaomer *et al*. 2012; Williams *et al*. 2012; Altumi *et al*. 2013; Kydonakis *et al*. 2014), and the large proportion of *c*. 0.85–1.2 Ga detrital zircons in the Phanerozoic successions of Central Africa (de Wit *et al*. 2008; Linol *et al*. 2014) suggest construction of the Iberian platform along the Central to NE African margin of northern Gondwana, and open new possibilities for the source region of these detrital

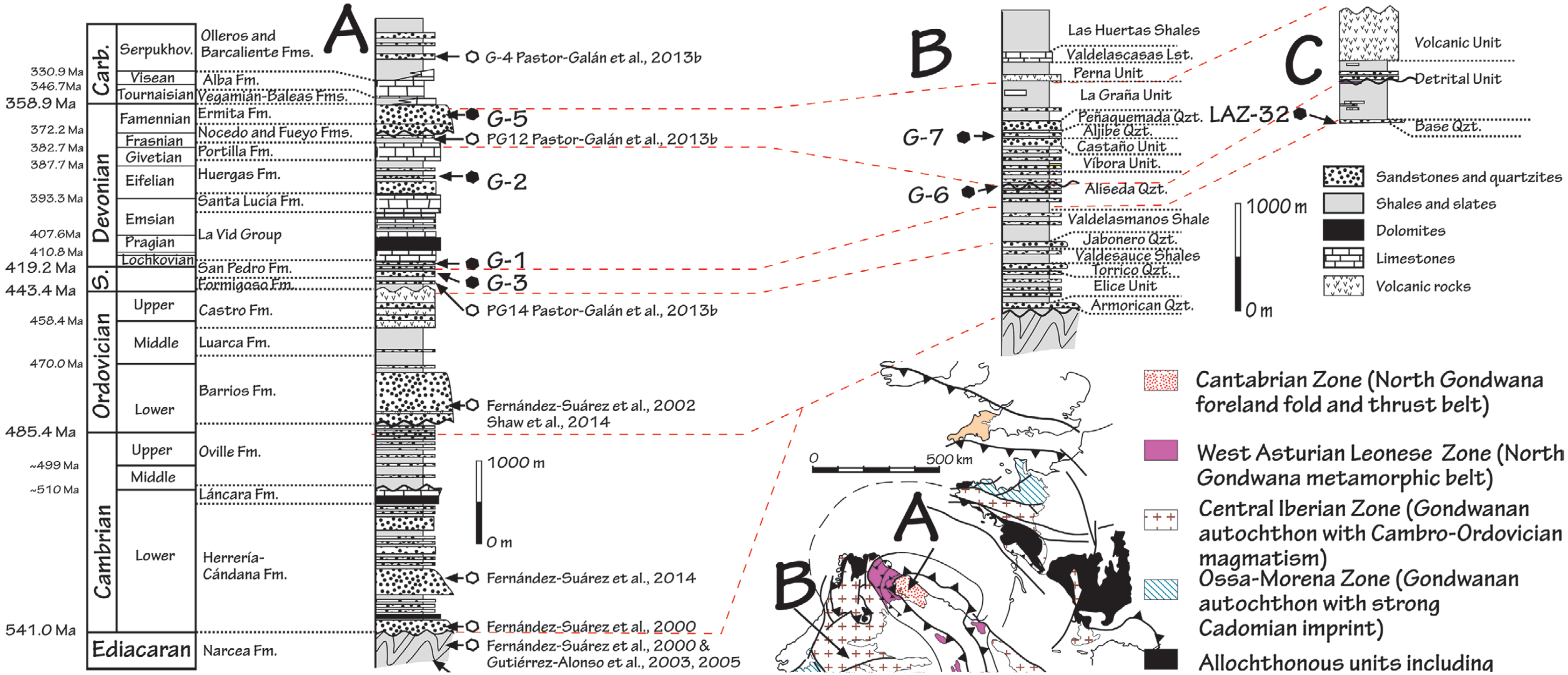

Fig. 1. Geological sketch of the Western European Variscan Belt with the location of the studied samples in the Cantabrian Zone and the Central Iberian Zone. Synthetic stratigraphic columns for the sampling areas: column A, Cantabrian Zone (based on Aramburu *et al*. 2004; Fernández *et al*. 2004); column B, Sierra de San Pedro, Central Iberian Zone (based on Soldevila Bartolí 1992); column C, Almadén (based on Pardo & García-Alcalde 1984).

zircons (e.g. Gómez Barreiro *et al*. 2007; Fernandez *et al*. 2010; Díez Fernández *et al*. 2012; Pastor-Galan *et al*. 2013*b*; Fernández-Suárez *et al*. 2014; Shaw *et al*. 2014). A NE African (close to the Nubian Shield) Cambro-Ordovician position for Iberia has also been proposed on the basis on Sm–Nd isotope studies of rocks from Iberia and North Africa (Bea *et al*. 2010).

Geochronological studies of detrital zircon from Ordovician sedimentary rocks in NW Iberia (Fernandez-Suarez *et al*. 2002; Catalan *et al*. 2004; Shaw *et al*. 2014) show a predominance of mixed Tonian–Stenian and Ediacaran–Cryogenian sources. There are few detrital zircon data for the overlying Silurian sedimentary rocks (Catalan *et al*. 2004; Pastor-Galan *et al*. 2013*b*) but available U–Pb ages show age populations to be similar to those of the Ordovician rocks.

Devonian clastic sedimentary rocks in NW Iberia were deposited during the closure of the Rheic Ocean. Samples from the basal Devonian (Catalan *et al*. 2004) and Givetian–Frasnian, and two more of uncertain Late Devonian age (Catalan *et al*. 2008; Pereira *et al*. 2012*a*), are interpreted to show a close relationship between migration of Variscan synorogenic depocentres towards the foreland with the advance of a wedge of exotic terranes (Catalan *et al*. 2008). Continuing foreland-directed migration of synorogenic depocentres related to the final emplacement of the upper plate above the subducting Gondwanan margin is recorded in U–Pb detrital zircon ages from Late Devonian–Carboniferous samples in NW Iberia (Catalan *et al*. 2004; Pastor-Galan *et al*. 2013*b*).

In this paper we present detrital zircon U–Pb age data for a set of seven samples of clastic rocks (interpreted in conjunction with three previously published samples from Pastor-Galan *et al*. (2013*b*)) the depositional age of which ranges from Late Silurian to Early Carboniferous, including all Devonian stages.

The main objective of this geochronological study is to further constrain the provenance of the Siluro-Devonian clastic sequences of Iberia by (1) establishing the nature and age of the source rocks shedding detritus onto the foreland, (2) testing for secular variations in the sediment sources and determining their tectonic significance for the evolution of the Gondwanan margin, and (3) constraining the possible palaeogeographical scenarios that could explain the age patterns of detrital zircons in the sedimentary rocks.

Geological setting

The Iberian Massif is divided into zones that are based on their Lower Palaeozoic stratigraphy and Variscan structural style, metamorphism and magmatic activity, which broadly reflect decreasing proximity to the Gondwanan margin (Lotze 1945; Julivert *et al*. 1972*a*; Farias *et al*. 1987; Fig. 1). Several zones, broadly encompassing the Rheic coastline to outer platform, can be identified. The nearshore Cantabrian Zone, which forms the foreland fold and thrust belt of the Variscan orogen, is where samples G-1, G-2, G-3 and G-5 were collected (Fig. 1). The West Asturian–Leonese, Central Iberian (where samples LAZ-32, G-6 and G-7 were collected, Fig. 1), Galicia–Tras-os-Montes (Schistose Domain) and Ossa–Morena zones are interpreted to represent progressively more distal, offshore facies (Julivert *et al*. 1972*b*; Pérez Estaún *et al*. 1990; Quesada 1990; Ribeiro *et al*. 1990; Quesada *et al*. 1991; Catalan *et al*. 1997; Gutiérrez-Marco *et al*. 1999; Marcos & Farias 1999; Aramburu *et al*. 2002; Robardet 2002, 2003; Robardet & Gutiérrez-Marco 2004; Murphy *et al*. 2008). In addition, the southernmost portion of the Iberian Massif, the South Portuguese Zone, is interpreted to have been part of the northern margin of the Rheic Ocean (the Meguma terrane; Catalan *et al*. 1997). Boundaries between these zones are major faults and shear zones (Julivert *et al*. 1972*a*; Catalan *et al*. 1997, 2003). The most outboard sedimentary rocks deposited on the alleged basement of the northern Gondwana margin are the so-called Basal Units, the lowermost units within the allochthonous complexes of the Galicia–Tras-Os-Montes zone (Fig. 1). The Basal Units have a continental affinity and are considered to represent the most external part of the Gondwanan margin (Catalan *et al*. 1996; Fernandez *et al*. 2010). In Devonian times, the Gondwana passive margin is interpreted to have comprised several troughs, parallel to the putative coastline and opening to the ocean towards the south and west (relative to the Cantabrian Zone) in present-day coordinates (Oliveira *et al*. 1986).

The Rheic Ocean is recorded in NW Iberia as oceanic remnants preserved in rocks with ophiolitic affinity in the Galicia–Tras-Os-Montes zone (Fig. 1). The geometry of these ophiolites is complex, leading to a variety of interpretations (e.g. Catalan *et al*. 2007; Arenas *et al*. 2014). The first record of ocean closure is indicated by evidence of subduction beneath the Laurussian (northern) margin of the Rheic Ocean prior to *c*. 400 Ma (Ibarguchi *et al*. 1990; Mendía Aranguren 2000; Fernandez-Suarez *et al*. 2007; Gómez Barreiro *et al*. 2007; Catalan *et al*. 2009). By *c*. 395 Ma, an increase in the inferred rate of convergence, and indications of coupling of the Gondwana margin to a subducting slab, is thought to reflect northward subduction of the Rheic mid-oceanic ridge (Woodcock *et al*. 2007; Gutierrez-Alonso *et al*. 2008).

Continental collision is interpreted to have started at *c*. 365–370 Ma (Dallmeyer *et al*. 1997; Rodriguez *et al*. 2003; López-Carmona *et al*. 2014) with initiation of the subduction of the Gondwanan margin below Laurussia, giving rise to an eastward migration of deformation and related synorogenic sedimentation.

Immediately after construction of the Variscan orogen during the Carboniferous, and driven by the closure of the Rheic Ocean, dramatic buckling of the mountain chain about vertical axes of rotation during the Late Pennsylvanian gave rise to the Cantabrian (Weil *et al*. 2001, 2013; Gutiérrez-Alonso *et al*. 2004, 2011; Johnston *et al*. 2013) and Central Iberian (Aerden 2004; Catalan 2012; Shaw *et al*. 2012) oroclines (Fig. 1).

The Siluro-Early Carboniferous succession in the Cantabrian Zone

The nearly complete Palaeozoic succession in the Cantabrian Zone has been divided classically into pre-, syn- and post-orogenic units (Julivert 1978; Marcos & Pulgar 1982), although a new early synorogenic unit should be included. The pre-orogenic unit formed by the Palaeozoic pre-Fammenian sequence is wedge-shaped, thins and shallows toward the foreland, and is preserved in the core of the Cantabrian Orocline. Samples G-1, G-2 and G-3 were collected in this unit. On top of it, the early synorogenic sequence comprises siliciclastic Fammenian rocks (where sample G-5 was collected) and three Mississippian condensed carbonate units. This sequence records the inversion from a passive margin to an orogenic marginal basin influenced by the continuing Variscan collision taking place further west. The synorogenic sequence *sensu stricto* (*s.s.*) consists of a thick succession of dominantly siliciclastic continental rocks of Late Mississippian and Early to Middle Pennsylvanian age deposited within a foreland fold and thrust belt. Finally, the post-orogenic sequence consists of Late Pennsylvanian and Early Permian continental intramontane coal basins (Colmenero *et al*. 2008). The detrital zircon geochronology of the early syn-, syn- *s.s.* and post-orogenic rocks has been described in detail by Pastor-Galan *et al*. (2013*b*).

In the Cantabrian Zone, the Lower Cambrian series lies unconformably on Ediacaran turbiditic sediments and subduction-related volcanic rocks (Gutiérrez-Alonso & Fernández-Suárez 1996; Fernandez-Suarez *et al*. 1998; Rubio-Ordoñez *et al*. 2014). The Lower Palaeozoic Cambro-Ordovician successions may reach 4000 m in thickness and comprise mostly siliciclastic sediments with minor carbonates and volcanic rocks accumulated in braid deltas to marine shelf environments in a graben-type basin located in the northern border on the Gondwana continent (Aramburu & García-Ramos 1993). Silurian rocks in the Cantabrian Zone are represented by the Formigoso Fm (Fig. 1, column A), a 70–200 m thick succession of dark siltstones and lutites of Late Llandovery to Early Wenlock age that lie para- or disconformably on Ordovician rocks; sample PG-14 from this unit (Pastor-Galan *et al*. 2013*b*) has been included in this study for comparison. Above the Formigoso Fm, the Furada–San Pedro Fms (Fig. 1) consist of intensely bioturbated ferruginous sandstones with rare ooid sandstones and local volcaniclastic beds (van den Bosch 1969; Suárez de Centi 1988; Gallastegui *et al*. 1992). These formations were deposited in a shallow epeiric sea characterized by frequent storms (Suárez de Centi 1988; García-Ramos *et al*. 1989). Their facies distribution implies that the sediments were derived from an emergent land area located to the east, in what is now the core of the Cantabrian Orocline (Suárez de Centi 1988; Aramburu *et al*. 2002). The high iron content of the sediments is interpreted as being derived from subaerial weathering of basic volcanic rocks (García-Ramos *et al*. 1987) or as being related to an intra-Rheic event that changed the redox conditions of the ocean (Pastor-Galan *et al*. 2013*a*). The lower part of this formation, where sample G-3 was collected (Fig. 1, column A), has yielded Ludlow fauna (Comte 1934, 1959; Poll 1970) whereas the upper part is Lochkovian (Poll 1970). Sample G-1 was collected in the uppermost beds of this unit (Fig. 1, column A).

The Devonian succession of the Cantabrian Zone is up to 2000 m thick and consists of nearshore deposits, with alternating clastic rocks and bioclastic and reefal limestones (Fig. 1, column A). The Furada–San Pedro Fms are succeeded by *c*. 500 m of limestones, dolomites, marls and shales of the Rañeces–La Vid Groups, interpreted as representing deposition on a storm-dominated homoclinal carbonate ramp with scarce biostromal reef episodes (Vera de la Puente 1989). Its age is bracketed between Middle Lochkhovian and Late Emsian (García-López 2002). The overlying Moniello–Santa Lucía Fms (Fig. 1, column A) comprise a 250 m thick massive bioclastic and reefal limestone with shallower lagoon facies towards the core of the Cantabrian Orocline (Méndez-Bedia 1976). Its age is Late Emsian to Early Eifelian (Arbizu *et al*. 1979).

The 200–400 m thick Naranco–Huergas Fm (Fig. 1, column A) varies from mostly iron-rich sandstones, deposited on a high-energy siliciclastic shelf with sand bars in the northern realm of the Cantabrian Zone, to sediments mostly comprising shales, deposited on an outer shelf below storm wave base (García-Ramos 1978) in the southern realm. Sample G-2 was collected in the middle part of this unit. The age of this formation is early Eifelian–Early Givetian (García-Alcalde *et al*. 2002).

The 60–200 m thick Candás–Portilla Fm (Fig. 1, column A) comprises a limestone unit of biostromal origin and is Givetian–earlier Frasnian in age (García-Alcalde *et al*. 2002).

A second laterally variable siliciclastic complex, the Nocedo–Piñeres Fms (Frasnian–middle Famenian; García-Alcalde *et al*. 2002), lies conformably above the Candás–Portilla Fms. In the northern realm, the Piñeres Fm is up to 500 m thick and comprises ferruginous and siliceous sandstones. In the south, the Nocedo Fm consists mainly of calcareous sandstones with thin limestone and shale beds. The formations are Frasnian in age (García-Alcalde *et al*. 2002) and are interpreted as prograding shallow marine wedges (Colmenero 1976; García-Ramos & Colmenero 1981; Keller *et al*. 2008).

The Nocedo Fm is conformably overlain by the Fueyo Fm. This formation consists of up to 120 m of shale with pelagic faunas and sandstones and some conglomerate intercalations, deposited in an outer shelf setting (Colmenero 1976; García-Ramos & Colmenero 1981). Its age is Late Frasnian–Early Famennian and it is thought to have originated in an outer shelf–slope environment (Colmenero 1976; García-Ramos & Colmenero 1981; van Loevezijin 1986*a*,*b*).

Detrital zircon ages from this formation have been described by Pastor-Galan *et al*. (2013*b*) and have been included this study (sample PG-12).

Palaeocurrent directions for Cambrian to Devonian successions change as a function of structural strike yielding a radial pattern that fans out around the Cantabrian Orocline. Palinspastic reconstruction of the orocline restores the palaeocurrent directions to a common orientation indicating that sediment transport was from east to west (Colmenero 1976; García-Ramos 1978; Aramburu & García-Ramos 1993; Shaw *et al*. 2012).

The Upper Famennian is represented in the Cantabrian Zone by the Ermita Fm. This formation, from which sample G-4 was collected, consists of 10 to *c*. 100 m of quartzitic sandstones deposited across a low-angle regional unconformity. This unconformity, which lies either below or within the lower Ermita Fm, is interpreted to represent the effects of the initial stages of the Variscan Orogeny (García-Ramos & Colmenero 1981; Rodríguez-Fernández *et al*. 1985; Keller *et al*. 2008). The upper part of the Ermita Fm is considered to record the onset of the Variscan early synorogenic and synorogenic sedimentary record in NW Iberia, which has a complex distribution in time and space and has been separated into six sedimentary sequences (Colmenero *et al*. 2002).

The first sequence is restricted to the Tournaisian and Visean and is represented by the Baleas, Vegamian and Alba condensed formations. This sequence records the inversion from a passive continental margin to an orogenic peripheral basin owing to the Variscan deformation in the inner parts of the orogen (Colmenero *et al*. 2002). The Baleas Fm, which is of Late Famenian to Early Tournaisian age, occurs in the western part of the Cantabrian Zone and is a 5–10 m thick condensed succession of bioclastic sandy limestones deposited in high-energy shallow-water marine shelf environments. In the eastern part of the Cantabrian Zone, the Baleas Fm is replaced, at least partially, by the Vegamián Fm, comprising a 10–60 m thick succession of black shales and mudstones with chert lenses and phosphate nodules deposited in relatively deep, locally stagnated intra-shelf and shelf edge environments (Colmenero *et al*. 2002). The overlying Visean Alba Fm consists of *c*. 30 m condensed nodular red limestones and red radiolarian shales. Its age spans the Visean and extends to the earliest Serpukhovian (Sánchez de Posada *et al*. 2002). This unit is ubiquitous in the Cantabrian Zone and is a good stratigraphic marker. It is interpreted as a transgressive unit deposited on a relatively well-oxygenated pelagic platform (Colmenero *et al*. 2002).

On top of this synorogenic sequence lies a second, Serpukhovian to early Bashkirian sequence comprising the Barcaliente and Olleros Fms (Colmenero *et al*. 2002).

This second sequence records the first arrival of terrigenous debris from the developing orogenic belt to the west and south of the Cantabrian Zone in present-day coordinates (Colmenero *et al*. 2002). The Barcaliente Fm is a 300–350 m thick, mostly azoic dark and grey laminated limestone. It was deposited on a restricted shallow water platform passing to the west and south into a slope environment where a deeper turbidite trough was filled by the coeval siliciclastic equivalent, the Olleros Fm, from which sample G-4 (Pastor-Galan *et al*. 2013*b*) was collected. The Olleros Fm consists mostly of *c*. 400–500 m of shales and sandstones of turbiditic origin, and conglomerates, sandstones, shales and limestones, and its age extends to at least the earliest Bashkirian (Wagner *et al*. 1971).

Above the Barcaliente Fm the synorogenic deposits change profoundly from one thrust unit to the next within the Cantabrian Zone imbricate system (Marcos & Pulgar 1982; Colmenero *et al*. 2002).

The Devonian in the Central Iberian Zone

In the northern subdivision of the Central Iberian Zone (referred to as the Ollo de Sapo Domain; Catalán *et al*. 2004), Devonian clastic sedimentary and volcanic rocks are mainly exposed within the core of the Alcañices and Truchas synforms, from which detrital zircon is available (Catalan *et al*. 2004, 2008). The studied Devonian sedimentary rocks in the Alcañices synform crop out near the Rheic suture and their detrital zircon populations are interpreted to reflect the emplacement of the Laurussian margin on top of the Gondwana continental shelf in the Late Devonian.

In the central and southern Central Iberian Zone, Devonian rocks crop out in the core of synclines and consist mainly of siliciclastic sequences with some volcanic rocks (Gutierrez-Alonso *et al*. 2008; Higueras *et al*. 2013). Distinguishable lithostratigraphic units are referred to in the literature with a wealth of local names (see Pardo Alonso & García-Alcalde 1996; García-Alcalde *et al*. 2002). In the studied sections of the Central Iberian Zone, there is a widespread hiatus or paraconformity encompassing the Eifelian and most of the Givetian (Puschmann 1967). For this study, the northern limb of the Sierra de San Pedro syncline and one locality in the southern limb of the Almadén syncline (Fig. 1, columns B and C, respectively) were chosen for sampling and two of the collected samples lie below and one above the above-mentioned paraconformity (hiatus).

The Devonian sequence in the San Pedro Syncline (Fig. 1, column B) has been described by López Díaz (1991) and Soldevila Bartolí (1992) and consists of a siliciclastic succession (Fig. 1, column B) that starts with the 300 m thick Aliseda Quartzite, from which sample G-6 was collected. Its fossil-based age is Early Devonian (Emsian) (Arbizu *et al*. 1989). Above this formation lies the 250 m thick Víbora Unit, consisting of shales and quartzites, within which the presence of a mid-Devonian unconformity is surmised on the basis of palaeontological evidence (Puschmann 1970; Vergés 1980; Pardo & García-Alcalde 1984). On top of the Víbora Unit is a distinct *c*. 40 m thick formation known as the Aljibe Quartzite (Soldevila Bartolí 1992), from which sample G-7 was collected, which has yielded fossils of Frasnian age. Above the quartzites, the *c*. 100 m thick Castaño Fm is composed of black shales and quartzites with fossils of late Devonian age (Soldevila Bartolí 1992), it is overlain by the Peñaquemada quartzites (*c*. 50 m thick) and greywackes and shales of the Graña Unit, the uppermost Devonian rocks of the region.

The 700–1800 m thick Devonian sequence in the Almadén Syncline (Fig. 1, column C) is composed mainly of siliciclastic rocks with volcanic deposits and minor limestones toward the top. The base of the Devonian is represented by the Base Quartzite Formation, from which sample LAZ-32 was collected (Fig. 1, column C). It has been dated palaeontologically as Lochkovian (Pardo & García-Alcalde 1984). The Base Quartzite Fm is interpreted to have formed as a littoral bar prograding towards the west, in present-day coordinates (García-Sansegundo *et al*. 1987; Vergés 1980). Above the Base Quartzite, there is a sequence of predominantly detrital rocks, known as the 'Detrital Unit', the upper part of which contains an unconformity marking a mid-Devonian hiatus (e.g. Pardo & García-Alcalde 1984). Above the Detrital Unit, a thick sequence of predominantly volcanic and volcaniclastic rocks of Frasnian age is widely recognized in the area ('Volcanic Unit', Pardo & García-Alcalde 1984).

Analytical methods

U–Pb LA-ICP-MS analysis

Initial preparation of samples was conducted at the Salamanca and Complutense (Madrid) universities. Samples were crushed with a jaw crusher and pulverized with a disc mill. Zircons were separated by heavy fraction enrichment on a Wilfley table followed by density separation using di-iodomethane (CH_2I_2) and magnetic separation in a Frantz isodynamic separator. Zircons were selected from the least magnetic fraction and handpicked in alcohol under a binocular microscope. Zircon grains were set in synthetic resin mounts, polished to approximately half their thickness and cleaned in a warm HNO_3 ultrasonic bath.

Zircons were analysed for U and Pb isotopes by laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at the Museum für Mineralogie und Geologie (Senckenberg Naturhistorische Sammlungen Dresden). For U/Pb analyses the laser spots were placed in zones with monophase growth patterns that show no metamorphic overprint. Zircons were analysed for U, Th, and Pb isotopes by LA-sector field (SF)-ICP-MS techniques at the Senckenberg Naturhistorische Sammlungen Dresden (Museum für Mineralogie und Geologie, GeoPlasma Lab), using a Thermo-Scientific Element 2 XR SF-ICP-MS system coupled to a New Wave UP-193 excimer laser system. A teardrop-shaped, low-volume laser cell (modified version of the NERC Isotope Geosciences Laboratory in UK; see Bleiner & Gunther 2001; Gerdes & Zeh 2006) constructed by Ben Jähne (Dresden) and Axel Gerdes (Frankfurt/M.) was used to allow sequential sampling of heterogeneous grains (e.g. growth zones) during time-resolved data acquisition. Raw data were corrected for background signal, common Pb, laser-induced elemental fractionation, instrumental mass discrimination, and time-dependent elemental fractionation of Pb/Th and Pb/U using an Excel® spreadsheet program developed by Axel Gerdes (Gerdes & Zeh 2006); (Frei & Gerdes 2009).

Reported uncertainties were propagated by quadratic addition of the external reproducibility obtained from the standard zircon GJ1 (*c*. 0.6% and 0.5–1% for $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{238}U$, respectively) during single analytical sessions and the within-run precision of each analysis. Repeated analyses of the standard GJ1 during analytical sessions yielded a mean $^{207}Pb/^{206}Pb$ age of 607.8 ± 2.2 Ma (certified isotope dilution thermal ionization mass spectrometry age: 608.5 ± 0.4 Ma; Jackson *et al*. 2004). Further details on analytical protocol and data processing have been given by Frei & Gerdes (2009).

No common lead correction was applied as all analyses used are concordant. In LA-ICP-MS analyses the carrier gas contains mercury and the ^{204}Pb correction based on the 202/204 ratios may result in overcorrection for common lead. Furthermore, the effect of small amounts of common Pb will not significantly affect the results within the analytical uncertainty associated with single analyses (e.g. Jackson *et al*. 2004). The effect of possible common lead was further tested with the method of Andersen (2002), confirming that the effect of common Pb is negligible in all the analyses listed in the tables and used in the interpretation.

Age assignment for each analysis was as follows. For analyses whose 2σ error ellipse overlaps the concordia curve, the chosen age and 2σ uncertainty are the concordia age and error (Ludwig 1998) as calculated by Isoplot 3.75 (Ludwig 2012). For analyses that are less than 10% discordant but whose corresponding 2σ ellipses do not intercept the concordia curve we have chosen either the $^{207}Pb/^{206}Pb$ age or the $^{206}Pb/^{238}U$ age depending on which corresponding isotope ratio was measured with more precision in that particular analysis.

U–Pb data treatment and presentation

After data reduction and age calculation, analyses with discordance higher than 10% (i.e. concordance <90% or >110%) were rejected. The number of concordant (non-rejected) analyses ranged between 70 and 150 per sample, and was around 100 for most. The U–Pb data are visually represented for comparative purposes in concordia diagrams (Fig. 2), kernel density estimation (KDE, Fig. 3; Sircombe & Hazelton 2004), and probability density distribution (PDD) plots (Fig. 3). Concordia, KDE and PDD plots were drawn using the programs Isoplot 3.75 (Ludwig 2012; Vermeesch

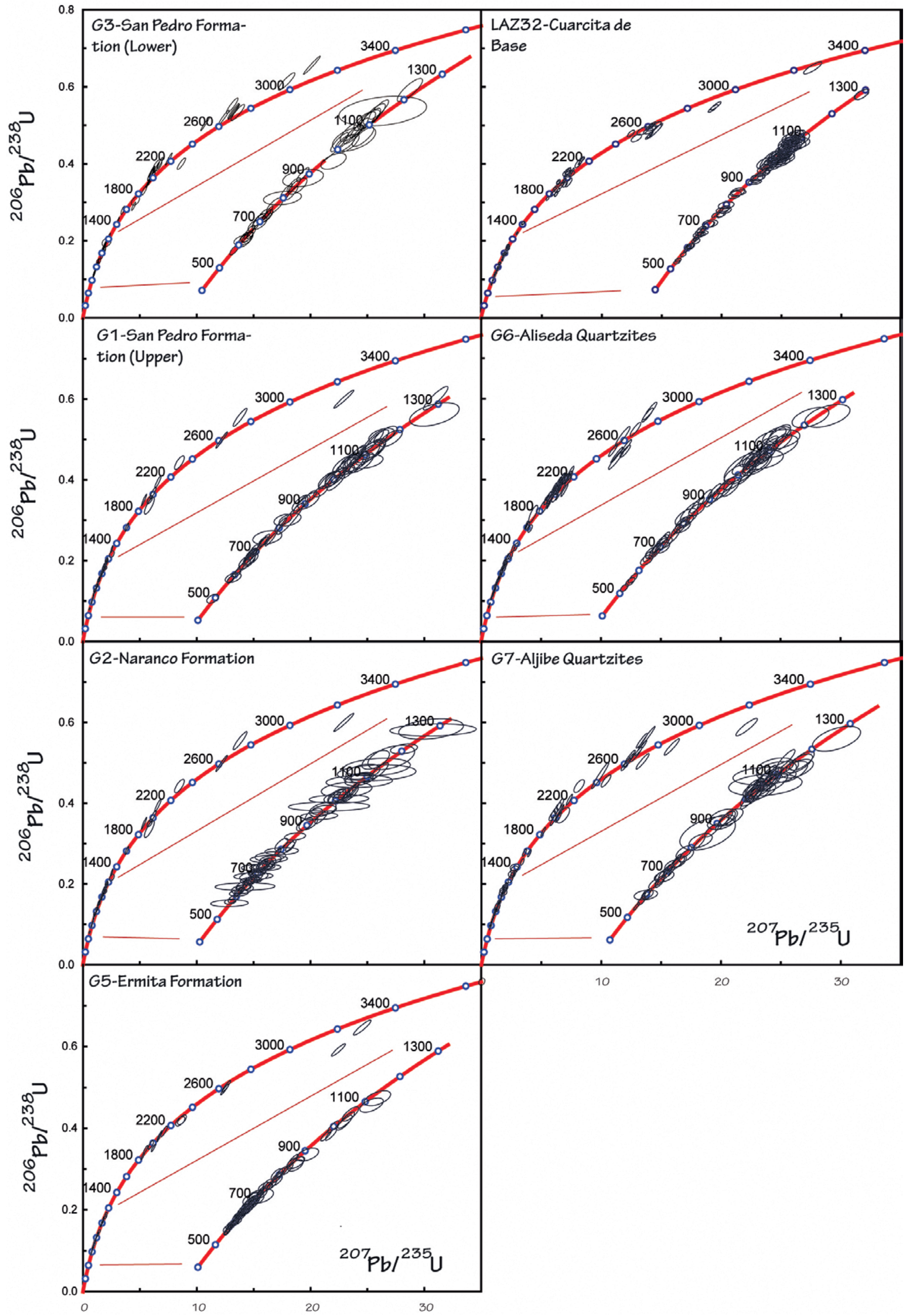

Fig. 2. Concordia diagrams for samples G-1, G-2, G-3, G-5, G-6, G-7 and LAZ-32 (location shown in Fig. 1). Ellipses represent 2σ uncertainties.

2012), Density Plotter v. 3.1 (Vermeesch 2012) and AgeDisplay (Sircombe 2004), respectively. For discussion purposes (see below) we have also added a plot (short-dashed lines in Fig. 3) representing the difference in probability for a given age between the U–Pb age distribution for each sample and a 'baseline' represented by the U–Pb zircon age distribution in the Ediacaran samples from the same area taken from Fernández-Suárez *et al.* (2014). These plots, constructed by substracting the PDD values obtained using AgeDisplay (Sircombe 2004) for each age value (every 1 Ma), provide an illustrative test for determining whether or not a certain zircon age distribution can be generated solely by recycling of the underlying sedimentary rocks. Positive values can be interpreted as representative of the input of zircons from a new source not present in the reference (older) population, whereas negative values are indicative of a lower input from the same sources as in the older rocks. Zero or very small PPD values indicate that, for a given age, the proportion of zircons is similar in both populations and, hence, they were derived from the same source.

Results

A total of 764 analyses with concordance between 90 and 110% are reported in Figures 2 and 3. For further comparison we have drawn a synoptic chart (Fig. 4) illustrating the proportions of the main age populations in the samples analysed in this study, and two samples taken from Pastor-Galan *et al.* (2013*b*), as well as the age groups in zircons from Ordovician (Shaw *et al.* 2014) and Neoproterozoic (Fernández-Suárez *et al.* 2014) rocks of Iberia to facilitate visualization of the changes in the relative proportions of the zircon age populations up the stratigraphic section.

Inspection of Figures 2, 3 and 4 shows that all samples contain four main age populations displaying variable relative proportions: Ediacaran–Cryogenian (*c.* 0.55–0.8 Ga), Tonian–Stenian (*c.* 0.85–1.2 Ga), Palaeoproterozoic (*c.* 1.8–2.2 Ga) and Archaean (*c.* 2.5–3.3 Ga). The two first groups constitute *c.* 60–80% of the total population in all samples. In addition, samples G-1, G-4, G-6, LAZ-32 contain very minor Palaeozoic (Cambrian) zircons and

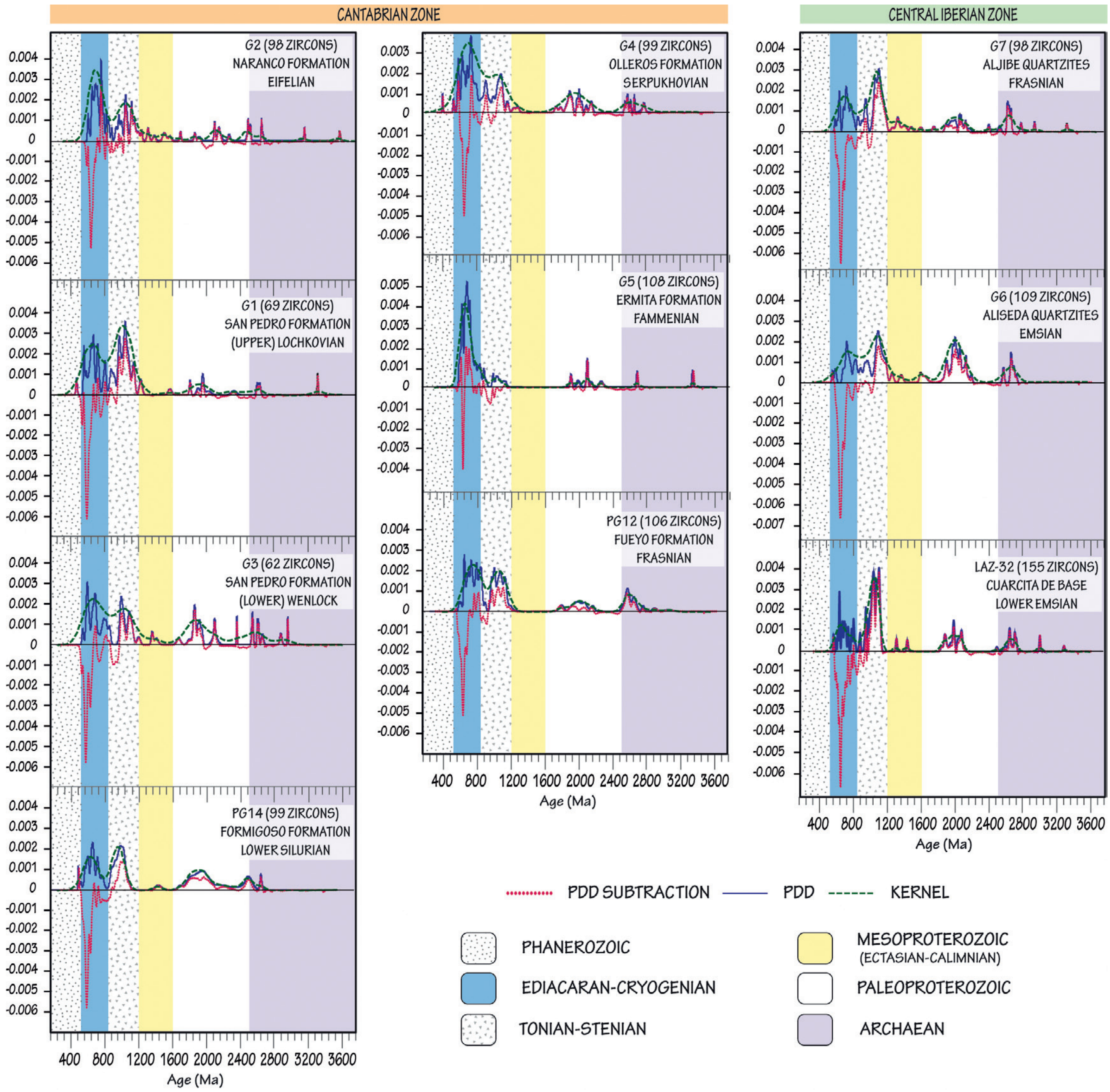

Fig. 3. Kernel density estimation (KDE) and probability density plots for the studied samples. Graphics were drawn using the reported age data. Also shown is the curve representing subtraction of each population from a baseline defined by detrital zircon ages in the Ediacaran rocks.

samples G-1, G-2, G-3, G-6, G-7 and LAZ-32 contain minor but significant zircons of Middle and Early Mesoproterozoic (Ectasian–Calymmian, 1.6–1.8 Ga) age.

The data also show an increase in the peak age of the Tonian–Stenian population (Fig. 5) from *c.* 940 Ma in the Ediacaran rocks (Fernández-Suárez *et al.* 2014) through *c.* 1000 Ma in the Ordovician rocks (Shaw *et al.* 2014) to *c.* 1050 Ma in the Siluro-Devonian rocks (this study). The peak age then shifts back to *c.* 980–1010 Ma in the Carboniferous rocks (sample G-4 and other Carboniferous samples from this region reported by Pastor-Galan *et al.* (2013*b*)). Interpretation of this trend (see discussion below) depends on the limitations imposed by the uncertainty of single U–Pb analyses determined by LA-ICP-MS (see discussion by Kosler *et al.* 2013). In other words, although all samples were analysed in the same laboratory under the same conditions, it is not possible to ascertain with certainty whether these peak-age shifts reflect a geological phenomenon or are an analytical or statistical artefact.

Discussion

We consider our results from Siluro-Devonian strata in conjunction with data from the available datasets from Ordovician (Shaw *et al.* 2014) and Ediacaran (Fernández-Suárez *et al.* 2014) strata of NW Iberia and more limited data from Devonian strata (Catalan *et al.* 2004, 2008).

Late Ediacaran to Ordovician sedimentary rocks in the Cantabrian Zone–West Asturian–Leonese Zone and Central Iberian Zone show a decrease in the Ediacaran–Cryogenian population (from *c.* 70% to *c.* 50%) and a concomitant increase in the Tonian–Stenian population (from *c.* 15–20% to *c.* 30–35%) together with a less significant increase in the Palaeoproterozoic and Archaean populations in the Ordovician strata (Fig. 4). The Silurian to Devonian samples (G-3 to G-1, Fig. 4) are marked by an increase in the proportion of Tonian–Stenian zircons (up to *c.* 50%). This increase occurs at the expense of the

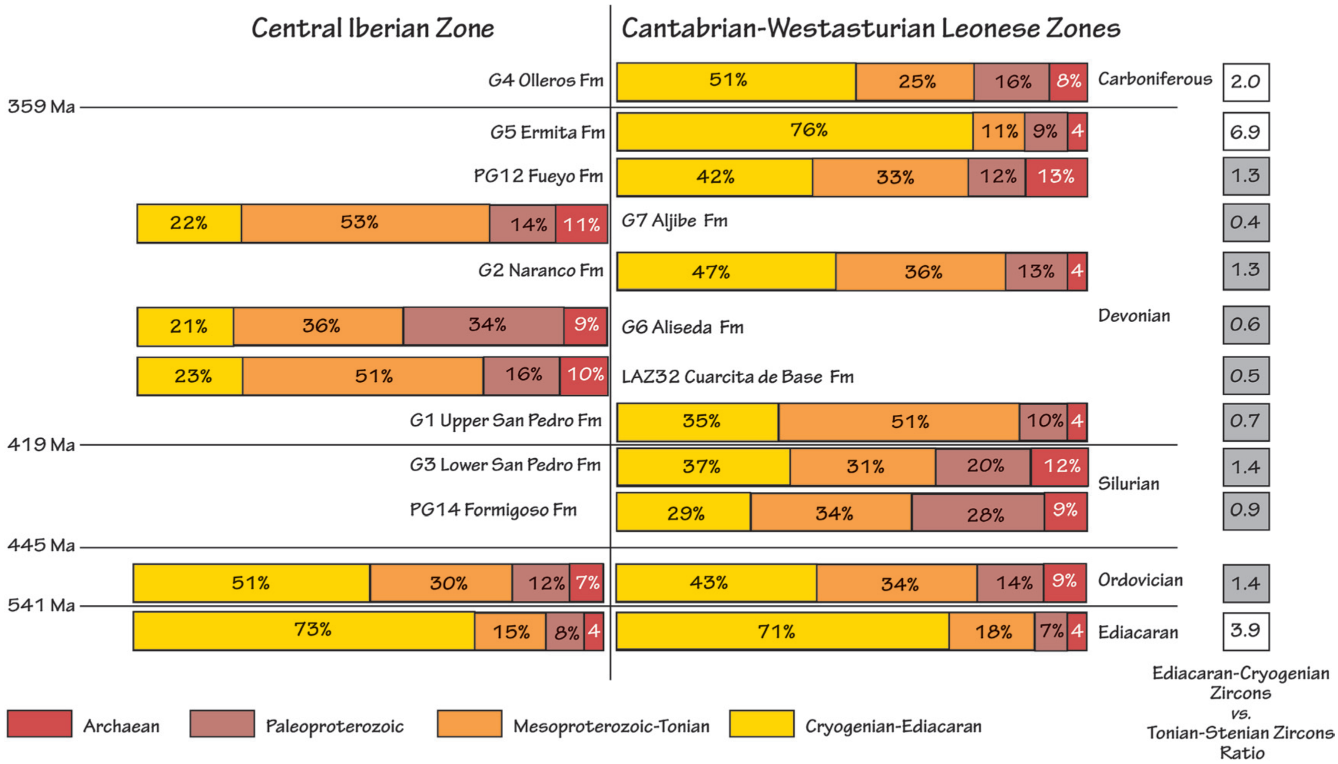

Fig. 4. Comparative table showing the relative abundance of the main detrital zircon age groups in the studied samples and in the Ordovician Armorican Quartzite (Shaw *et al*. 2014) and the Ediacaran rocks (Fernández-Suárez *et al*. 2014) of NW Iberia.

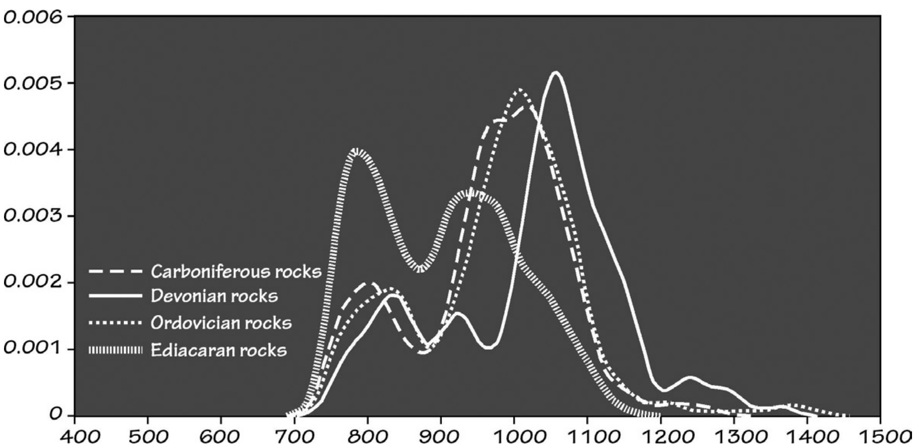

Fig. 5. KDE plot of the age distribution within the Tonian–Stenian population for the Ediacaran, Ordovician, Devonian and Carboniferous rocks of NW Iberia.

Ediacaran–Cryogenian and Palaeoproterozoic and Archaean populations. The Palaeoproterozoic and Archaean populations reach their maximum abundances in Silurian strata (combined *c*. 30–35% of total population) and decrease to *c*. 15–25% in Devonian and Carboniferous strata (Pastor-Galan *et al*. 2013*b*).

In all the Siluro-Devonian samples, the youngest zircon age is older than their fossil constrained depositional age (Figs 2 and 3), a feature that is characteristic of sedimentary rocks deposited in stable platforms (passive margins) (e.g. Cawood *et al*. 2012). Only the Carboniferous (Serpukhovian) sample G-4 contains a Devonian zircon (Pastor-Galan *et al*. 2013*b*). Silurian and Devonian samples (except G-5; see below) plot in or near the passive margin field on a crystallization age–depositional age (CA–DA) diagram (Fig. 6). In contrast, Ediacaran samples of the same realm include a significant population of syndepositional zircons and hence plot in the subduction-related arc settingfield (see Fernández-Suárez *et al*. 2014). Plotting zircon crystallization versus depositional age (Fig. 6) illustrates the transition from an Ediacaran–Early Cambrian convergent margin (e.g. Fernandez-Suarez *et al*. 1998; Gutierrez-Alonso *et al*. 2003; Pereira *et al*. 2012*c*; Fernández-Suárez *et al*. 2014; Rubio-Ordoñez *et al*. 2014) to a passive margin following the opening of the Rheic Ocean in the latest Cambrian–earliest Ordovician (see Murphy *et al*. 2006; Nance *et al*. 2010). The Ordovician rocks (*c*. 480 Ma Armorican Quartzite) (Shaw *et al*. 2014) plot between the Siluro-Devonian passive margin samples (this study) and the Ediacaran subduction-related arc convergent margin samples (Fernández-Suárez *et al*. 2014), consistent with their deposition recording the transition from arc to passive margin setting as the Rheic Ocean opened and widened.

In contrast, the youngest Devonian sample in the Cantabrian Zone (G-5, Fammenian Ermita Fm) does not fit this pattern (Fig. 6). With respect to sample PG-12 of the underlying unit, there is a three-fold decrease in the Tonian–Stenian population, a smaller decrease in the Palaeoproterozoic–Archaean populations, and a concomitant increase in the Ediacaran–Cryogenian population. These changes occurred between *c*. 375 and 370 Ma, the depositional ages of samples G-5 and PG-12, and are interpreted to reflect the effect of the onset of the Variscan Orogeny; the Variscan

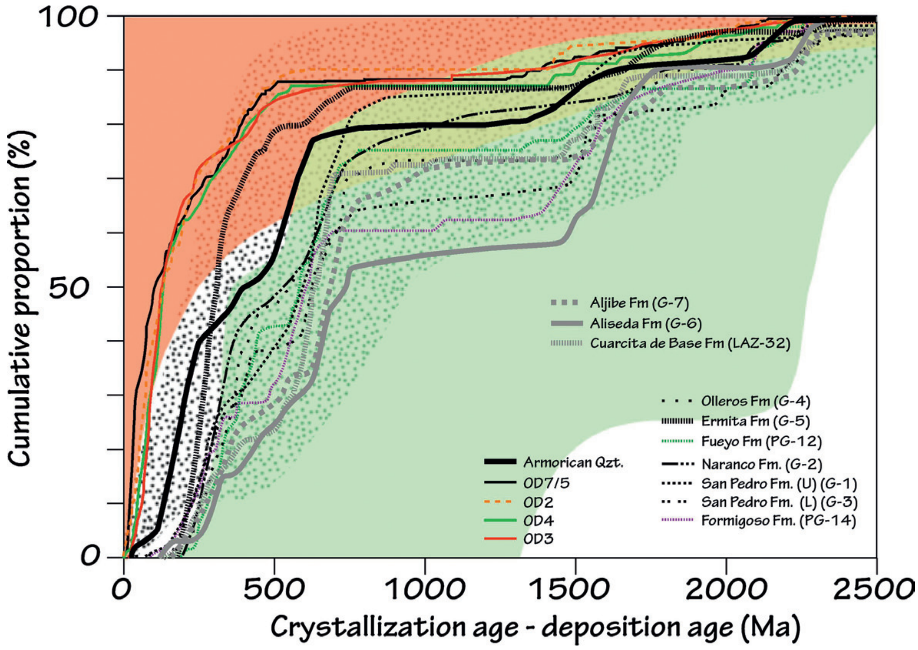

Fig. 6. CA–DA diagram of Cawood *et al.* (2012) showing the cumulative curve for each sample (data are obtained by subtracting the sedimentation age of the sample from the crystallization age of each zircon).

collision is considered to have started at *c.* 365–370 Ma (Dallmeyer *et al.* 1997; Rodriguez *et al.* 2003; López-Carmona *et al.* 2014).

The change in provenance marking the onset of the Variscan Orogeny has been detected in older rocks within the Central Iberian Zone (Catalan *et al.* 2004; Catalan *et al.* 2008). Givetian–Frasnian (*c.* 385–380 Ma) siliciclastic deposits from the Alcañices syncline are interpreted to have been derived from upper plate terranes that sit structurally above the ophiolite-like units. Variations in the age of onset of the Variscan Orogeny may indicate along-strike changes in the timing of Rheic Ocean closure. The Carboniferous (Serpukhovian) Olleros Formation (sample G-4) reverts to a pattern similar to the (pre-G-5) Devonian samples. Because the Olleros Formation is a synorogenic deposit (Colmenero *et al.* 2002; Pastor-Galan *et al.* 2013*a*,*b*), its detrital zircon age admixture can be interpreted to reflect, at least in part, recycling of the underlying passive margin sequence that is being deformed to the west, without direct input from the upper plate rocks. This is consistent with the trend reversal in the age peak of the Tonian–Stenian population of the Carboniferous samples, which is younger than that of the Devonian samples (Fig. 5).

In the Central Iberian Zone, the Emsian sample G-6 (with no age equivalent in the Cantabrian Zone in the samples used for this study) has a distinctive (high) proportion of Palaeoproterozoic zircons (*c.* 34%). The Frasnian sample G-7 shows a higher proportion of Tonian–Stenian zircons than its equivalents in the Cantabrian Zone and points to along-strike variations in the northern margin of Gondwana.

Based on the data summarized above and taking into consideration previous work (Murphy *et al.* 2006; Nance *et al.* 2008; Fernández-Suárez *et al.* 2014; Shaw *et al.* 2014) the following first-order interpretations can be made.

(1) The Ediacaran peri-Gondwanan basins were flooded by arc sediments with a small but significant contribution of detritus derived from rocks bearing Tonian–Stenian (*c.* 15–20%) and Palaeoproterozoic and Archaean (*c.* 10–15%) zircons (Fig. 5) (see Fernández-Suárez *et al.* 2014, and references therein).

(2) By the early Ordovician (Fig. 7b), the proportion of Tonian–Stenian and Palaeoproterozoic–Archaean zircons had significantly increased (Fig. 5) (Shaw *et al.* 2014). Silurian (PG-14 and G-3) and Ordovician samples have equal proportions of Tonian–Stenian zircons, but the proportion of Palaeoproterozoic–Archaean zircons is greater in the Silurian samples, forming up to 30–40% of the total population.

(3) The Tonian–Stenian zircon population reaches its maximum (*c.* 50% of total zircon population) in the Lower Devonian samples LAZ-32 (Central Iberian Zone) and G-1 (Cantabrian Zone).

(4) The Middle and Upper Devonian sedimentary rocks in the Central Iberian Zone and the Cantabrian Zone–West Asturian–Leonese Zone display contrasting detrital zircon records. In the Cantabrian Zone, the Middle–Upper Devonian rocks (except G-5; see above) show a relative decrease in the proportion of Tonian–Stenian zircons to percentages similar to those of Silurian strata albeit with a smaller proportion of Palaeoproterozoic and Archaean zircon. In the Central Iberian Zone the Frasnian Aljibe Quartzite (sample G-7) shows the highest proportion of Tonian–Stenian zircons of all the samples analysed for this study (53% of total population), a 20% increase with respect to the underlying Emsian Aliseda Quartzite.

(5) Neither Silurian nor Devonian sedimentary rocks contain a significant population of *c.* 470–490 Ma zircons. Because 470 and 490 Ma is a time of intensive magmatic activity on the northern margin of Gondwana (e.g. Valverde-Vaquero & Dunning 2000; Valverde-Vaquero *et al.* 2005; Gutierrez-Alonso *et al.* 2007; Murphy *et al.* 2008; Montes *et al.* 2010; Talavera *et al.* 2013) the Siluro-Devonian strata cannot have been recycled from the underlying rocks and were probably derived from a source terrane in the Gondwanan mainland.

(6) The abundance of Tonian–Stenian zircons (*c.* 30–50%) in Siluro-Devonian sedimentary rocks of NW Iberia is 2–4 times higher than their abundance in underlying Ediacaran rocks. Hence, the studied samples could not have been generated by the recycling of the Ediacaran sediments (Figs 3 and 4), nor could the Siluro-Devonian rocks have been derived from recycling of the underlying Ordovician rocks because the Ordovician strata, although they contain significant proportions of Tonian–Stenian zircons (Fig. 4), are characterized by the presence of a *c.* 470–490 Ma population (Shaw *et al.* 2014) that is absent in the Siluro-Devonian rocks.

Here we consider three possible scenarios that might explain the origin of the Tonian–Stenian zircons.

A large late Mesoproterozoic–early Neoproterozoic terrane in NE Africa and/or Central Africa

An areally significant Stenian–Tonian source terrane (capable of generating up to *c.* 50% of the total zircon population in Lower Devonian rocks) may have furnished sediment to the

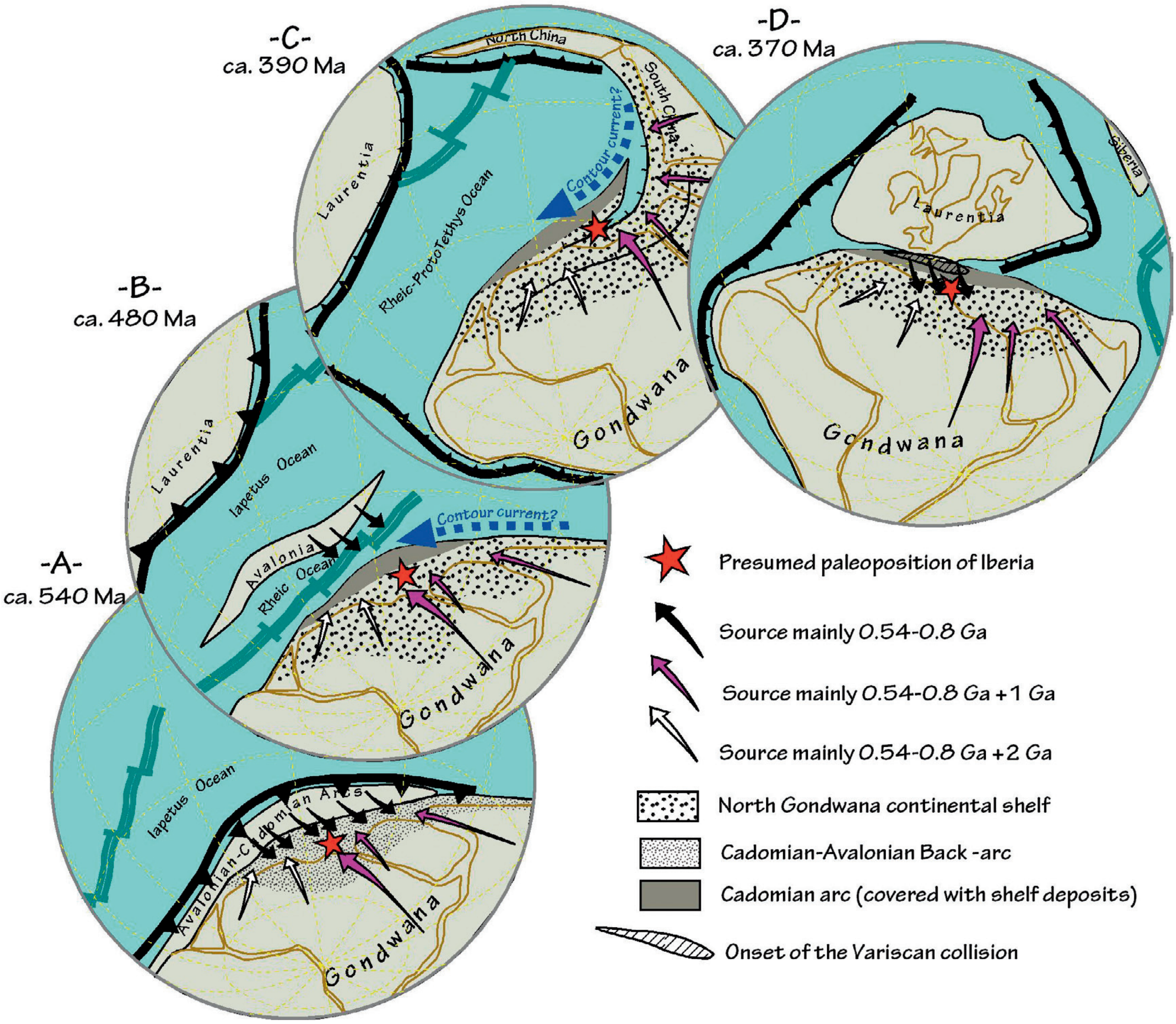

Fig. 7. (**a**–**d**) Tentative palaeogeographical reconstructions of the northern margin of Gondwana from the Ediacaran–Cambrian boundary (*c*. 540 Ma) to the Late Devonian (*c*. 370 Ma) based on, and modified from, the various reconstructions proposed by Stampfli *et al*. (2002, 2013), Stampfli & Borel (2002), Cocks & Torsvik (2006), Murphy *et al*. (2006), Gutierrez-Alonso *et al*. (2008), Gehrels *et al*. (2011), von Raumer *et al*. (2012), Dong *et al*. (2013) and Torsvik & Cocks (2013).

northern Gondwanan realm. NE Africa is characterized by arc terranes that contain zircons covering the *c*. 1.2–0.6 Ga time slice (e.g. Eyal *et al*. 2014). Erosion of these terranes could have produced the continuous spectra of zircon ages seen in the Ediacaran and Palaeozoic sedimentary sequences of NW Iberia (Fig. 7a).

The increase of the Tonian–Stenian population at the expense of the Ediacaran–Cryogenian population could be explained by denudation of the Cadomian–Avalonian arc(s) system (Fernández-Suárez *et al*. 2014; Shaw *et al*. 2014) and progressive exposure of the older basement terranes (Fig. 7b and c). The detrital zircon populations in Lower Devonian strata point to this time slice as the period of maximum erosion of the pre-Ediacaran terranes exposed in the Gondwanan mainland (Frizon de Lamotte *et al*. 2013), Also, recent studies indicate the abundance of large amounts of zircons with Tonian–Stenian ages in Central African sediments, interpreted to have been shed from the Sahara 'metacraton', which are likely to be the main source for the zircons of this age in the studied rocks (Iizuka *et al*. 2013; Be'eri-Shlevin *et al*. 2014; Linol *et al*. 2014).

Alternative or additional sources in northern Gondwana

A source within the South China Block located along-strike to the east may have provided the Stenian–Tonian zircons (Fig. 7) according to some reconstructions (Zhao *et al*. 1996; Cawood *et al*. 2013; Dong *et al*. 2013). The South China Block contains abundant zircons in the age range 1.1–0.6 Ga (e.g. DeCelles *et al*. 2004; Wang *et al*. 2010; Xiang & Shu 2010; Duan *et al*. 2011, 2012; Cawood *et al*. 2013; Xu *et al*. 2013; Li *et al*. 2014). Alongshore currents or margin-parallel river systems (Fig. 7b) may have transported Tonian–Stenian zircons to the Iberian segment of the Gondwana–Rheic margin.

The main problem with this scenario is that it is palaeogeographical model-dependent as there is no consensus on the location of the South China Block during the Palaeozoic (e.g. Domeier & Torsvik 2014, and references therein). Other eastern Gondwana terranes capable of supplying the Tonian–Stenian zircons include the Madurai Block of India (e.g. Plavsa *et al*. 2014), the Annamia–Indochina terranes (e.g. Usuki *et al*. 2013; Burrett *et al*. 2014) and the Rayner Complex–Eastern Ghats regions of Antarctica and India (Rösel *et al*. 2014).

First-order changes in the relative proportion of sources

The Rheic Ocean formed by the partial or complete rift and drift of the Avalonian–Cadomian arc terranes from the periphery of northern Gondwana. The removal of the Avalonian–Cadomian arc would have significantly decreased the availability of Ediacaran–Cryogenian zircons to the Gondwanan nascent platform. Hence the increase in the relative proportion of Tonian–Stenian zircons derived from mainland Gondwanan sources may simply reflect the decrease in Ediacaran zircons. Devonian samples record an approximately five-fold decrease in the Ediacaran–Cryogenian/Tonian–Stenian zircon ratio with respect to the Ediacaran samples. The decrease in this ratio starts in the Early Ordovician, consistent with the postulated timing for the opening of the Rheic Ocean in the latest Cambrian–early Ordovician (Nance *et al*. 2010) and falls throughout the Palaeozoic. This 'removal' of the Ediacaran (Cadomian)–Cryogenian sources can also be explained by progressive burial of these stratigraphic units and related magmatic arc rocks and the coeval gradual denudation of older source rocks located within the mainland.

However, although the proportion of Ediacaran–Cryogenian zircons fell, there remained a strong population of these zircons in the passive margin sequence throughout the Palaeozoic even long after the opening of the Rheic Ocean and the cessation of the Cadomian arc construction. Ediacaran–Cryogenian zircons in Devonian strata must, therefore, have been derived from Gondwana; hence, it remains unclear if the departure of Avalonian–Cadomian terranes is a sufficient explanation for the relative rise in the significance of Stenian–Tonian zircons.

Conclusion

Detrital zircons from Palaeozoic strata of the Iberian margin of Gondwana, including our samples from Silurian and Devonian strata, document two fundamental changes in the character of Gondwana margin sedimentation. The first major change occurred in the late Cambrian and was characterized by a decrease in the availability of Ediacaran–Cryogenian zircons, and a proportional increase in the significance of Stenian–Tonian zircons. This change is inferred to reflect the opening of the Rheic Ocean in response to the partial or complete rift and drift of the Avalonian–Cadomian arc terranes from the Gondwana margin. There are various ways of explaining the increase in Stenian–Tonian zircons, none of which are mutually exclusive; further detailed detrital zircon studies coupled with detailed sedimentological investigations are required to test these competing models. The second major change occurred between 370 and 375 Ma, and was characterized by the demise of passive margin sedimentation, the onset of recycling of the passive margin sequence, and the introduction of exotic source terranes. This change is interpreted to reflect the onset of the Variscan Orogeny in response to continental collision upon closure of the Rheic Ocean.

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