U–Pb zircon ages (SHRIMP) for Cadomian and Early Ordovician magmatism in the Eastern Pyrenees: New insights into the pre-Variscan evolution of the northern Gondwana margin

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1. Introduction

In recent years, the development of isotopic geochemistry and the improvement in geochronological methods have made great strides in the reconstruction of the Cadomian orogen and its geodynamic evolution and in the transition of the Cadomian to the Variscan cycle in the European realm (e.g., Stampfli et al., 2002; Linnemann et al., 2004; Samson et al., 2005; Gerdes and Zeh, 2006). In this regard, the Iberian Massif has played a major role for three main reasons: (i) good preservation of pre-Variscan sediments, unaffected or affected by very low-grade metamorphism, (ii) fairly complete stratigraphic sequences with thick Precambrian series, and (iii) the presence of volcanic and plutonic rocks emplaced in several stratigraphic levels at different times and with diverse geochemical signatures (Nägler et al., 1995; Fernández-Suárez et al., 1998; Valladares et al., 2002; Bandrés et al., 2004; Rodríguez-Alonso et al., 2004; Silva and Pereira, 2004; Díaz García, 2006 and references therein).

In the Pyrenees, the presence of an important Early Ordovician magmatic event (Delapeniere and Respaut, 1995; Deloule et al., 2002; Cocherie et al., 2005) and its relationship with the lower levels of the pre-Upper Ordovician metasedimentary sequence have led some authors to rule out the existence of Cadomian magmatism (e.g., Launonier et al., 2004). Recently, Cocherie et al. (2005) obtained an Ediacaran age for a metatuff from the lower levels of the sequence. Nevertheless, the age of the upper part of the metasedimentary...
sequence and the location of the Ediacaran-Cambrian boundary is still a matter of debate because of its unfossiliferous character.

We present new geochronological data obtained from six samples of three Eastern Pyrenean massifs in an attempt to gain further insight into the Cadomian orogen, its transition to the Variscan orogen, and on the controversy over the importance of the Cadomian signature in the Pyrenees. From west to east, these massifs are (Fig. 1): Canigó (known as Canigou in the French literature), Roc de Frausa (also known as Roc de France) and Cap de Creus. The new data confirm the existence of an Ediacaran-Lower Cambrian metasedimentary sequence with coeval volcanism in the Pyrenean pre-Variscan massifs. We also obtained Cadomian and Ordovician ages for orthogneisses emplaced at different structural levels in these metasediments. Inherited ages and detrital zircons analyzed in the volcanic and volcano-sedimentary samples provide some insights into the sources of these rocks and their correlation with other pre-Variscan complexes. The geological data obtained from several areas enable us to characterize the Cadomian orogenic stages (collision, arc magmatism and breakup and amalgamation of the peri-Gondwanan basins) along the northern margin of Gondwana, and to understand the evolution of the Variscan cycle with the opening and closure of the Rheic Ocean (von Raumer et al., 2002; Stampfli et al., 2002; Murphy et al., 2004).

2. Geological setting

In the Pyrenees, Alpine tectonics has exposed an extensive E-W trending area, the so-called Axial Zone, where a thick pre-Variscan succession crops out in several massifs (Fig. 1).

The lower part of this succession is a thick azoic metasedimentary sequence (Fig. 2), pre-Upper Ordovician in age, locally cut by orthogneiss sheets near the base of the sequence (Cavet, 1957). This author described a heterogeneous sequence at the base made up of metapelites and metagreywackes with interbedded metavolcanic rocks. At the top, the sequence consists of a monotonous succession of shales, sandstones and quartzites. Laumonier (1988) established a more detailed subdivision of the sequence based on lithological criteria.

A well-dated Upper Ordovician succession, with Caradocian conglomerates (Cavet, 1957; Hartveit, 1970) generally at the base, lies unconformably over the former metasediments (Santanach, 1972b; Casas and Fernández, 2007). The absence of a biostratigraphic control in the pre-Upper Ordovician sequence makes the evaluation of the magnitude of this unconformity difficult. Nevertheless, it has been suggested that at least the Lower and Middle Ordovician sediments were removed before deposition of the Upper Ordovician rocks (Muñoz and Casas, 1996). Silurian and Devonian strata, consisting respectively of siliciclastic sediments and limestones, follow in stratigraphic continuity. On top of the series, black shales, cherts and limestones constitute a Carboniferous pre-orogenic sequence.

The whole succession was affected by Variscan deformation (late Visean to Serpukhovian) accompanied by high temperature-low pressure metamorphism (Guitard, 1970; Zwart, 1979). Syn- to late orogenic (Moscovian-Kasimovian) granitoids intruded mainly into the upper levels of the succession, producing local contact metamorphism (Autran et al., 1970).

The pre-Upper Ordovician sequences are well developed in the Eastern Pyrenees, whereas in the Central Pyrenees thick Devonian and Carboniferous series are predominant. In the studied massifs (Canigó, Roc de Frausa and Cap de Creus), the pre-Upper Ordovician sequence is mainly composed of a metapelitic series sporadically interbedded with numerous layers of metabasites, ryholitic tufts, marbles, quartzites and calc-silicates. The orthogneisses are variably thick, from ~2 km (at the Canigó massif) to 100 m (at the Cap de Creus massif).

2.1. Canigó massif

This massif exhibits the most complete pre-Upper Ordovician metasedimentary succession of the Eastern Pyrenees (Fig. 2). The succession can be divided into three series (Cavet, 1957). The lowermost part
Fig. 2. Synthetic stratigraphic columns of the pre-Upper Ordovician rocks of the Canigó, Roc de Frausa and Cap de Creus massifs with the location of the studied samples. Data compiled from Guitard (1970), Santanach (1972a) and Ayora and Casas (1986) for the Canigó massif; Liesa and Carreras (1989) for the Roc de Frausa massif and Carreras et al. (1994), and Losantos et al. (1997) for the Cap de Creus massif.

consists of a thick sequence, the Balaig micaschists, made up of biotite-rich micaschists with interlayered marbles, metabasites and quartzites (Guitard, 1970). The intermediate part is formed by the Canavelles Series (the Canaveilles Series of Cavet, 1957), a heterogeneous sequence mainly composed of alternating metagreywackes and metapelites interbedded with numerous layers of marbles, quartzites, ilmenite-rich micaschists, black phyllites, calc-silicates and a variety of metavolcanic rocks (Cavet, 1957; Guitard, 1970; Casas et al., 1986; Ayora and Casas, 1986). At the top, the Jujols Series consists of a monotonous rhythmic sequence composed of shales, sandstones and quartzites (Cavet, 1957).

In addition, three different granitic orthogneissic bodies are located in the basal levels of the pre-Upper Ordovician sequence (Fig. 1). The Cadi gneiss (Guitard, 1970) constitutes the deepest rock outcropping in this area below the Balaig micaschist. The Casemi gneiss (Guitard, 1970; Delaperrière and Soliva, 1992) is a leucocratic, fine-grained orthogneiss interlayered into the Balaig micaschist. Finally,
2.2. Roe Frausa orthogneisses (Autran and Guitard, 1969), divide the sequence into two series according to the lithologies. The lower series is an 800 m thick monotonous alternance of predominant greywackes, subordinate pelites and discontinuous layers of plagiophyric amphibolites. Bandied quartzites form distinct continuous layers ranging in thickness from a few centimeters to a few meters. This sequence is overlain by a sequence of carbonaceous black slates interbedded with marbles and acidic metamorphites. The upper series is mainly formed by conglomerates, siliciclastic sediments and carbonates with marked lateral changes (Carreras et al., 1994). Granite orthogneisses (known as Port gneisses, Carreras and Ramírez, 1984) and metabasites crop out at the bottom and mid part of the lower sequence, whereas metautufs are interstratified at the top. The protolith of the Port gneiss is a small intrusion derived from subalkaline granites to quartz-monzonites composed of scarse K-feldspar megacrysts in a fine-grained matrix. Metabasites comprise gabbrodioleritic intrusions and metabasaltic lens-shaped bodies. Minor element geochemistry indicates that they are low-K tholeiites (Navidad and Carreras, 1995).

The metautufs contain feldspar and quartz porphyroclasts in a fine-grained matrix. They correspond to Al-rich calc-alkaline rhyolites and rhyodacites (Navidad and Carreras, 1995).

Five samples from the Cap de Creus were selected for U-Pb zircon analysis, including the Port gneiss (sample CC-7), metabasites (samples CC-4 to CC-6) and a metatuff (sample CC-2), in order to ascertain the age of the bimodal magmatism from this massif but again, there was no zircon yield from the metabasites.

2.3. Cap de Creus massif

In this massif, the pre-Upper Ordovician sequence can be divided into two series according to the lithologies. The lower series is an 800 m thick monotonous alternance of predominant greywackes, subordinate pelites and discontinuous layers of plagiophyric amphibolites. Bandied quartzites form distinct continuous layers ranging in thickness from a few centimeters to a few meters. This sequence is overlain by a sequence of carbonaceous black slates interbedded with marbles and acidic metamorphites. The upper series is mainly formed by conglomerates, siliciclastic sediments and carbonates with marked lateral changes (Carreras et al., 1994). Granite orthogneisses (known as Port gneisses, Carreras and Ramírez, 1984) and metabasites crop out at the bottom and mid part of the lower sequence, whereas metautufs are interstratified at the top. The protolith of the Port gneiss is a small intrusion derived from subalkaline granites to quartz-monzonites composed of scarse K-feldspar megacrysts in a fine-grained matrix. Metabasites comprise gabbrodioleritic intrusions and metabasaltic lens-shaped bodies. Minor element geochemistry indicates that they are low-K tholeiites (Navidad and Carreras, 1995).

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3. Geochronological background

The age of the pre-Upper Ordovician metasediments and the granitic orthogneisses in the pre-Variscan massifs of the Pyrenees has been a matter of debate since the work of Fontboté (1949), Cavet (1957) and Guittard (1970). Initial studies (see Table 1) concluded that large granitic orthogneisses were situated at the core of metamorphic masses, representing a Cadomian basement overlain by a lower Paleozoic cover (Autran and Guittard, 1969; Guittard, 1970; Vitrac-Michard and Allegre, 1975). In contrast, pioneering geochronological work in the Central Pyrenees pointed to an Ordovician age for the orthogneisses (see Table 1, Jäger and Zwart, 1968; Majoor, 1988). The advances made in U-Pb geochronology have revealed that some of the granitic orthogneisses are Early Ordovician in age. These granites intruded into a pre-Upper Ordovician metasedimentary sequence, thus invalidating the basement-cover model (Deloule et al., 2002; Cocherie et al., 2005). The implications of the nonexistence of a Cadomian granitic basement and the presence of an Ordovician magmatism in the Eastern Pyrenees (comparable to the one described in other areas of northern Gondwana; Pin and Marini, 1993; Santos Zalduegui et al., 1995; Valverde-Vaquero and Dunning, 2000) have been extensively discussed in subsequent alternative interpretations (e.g., Autran and Guittard, 1996; Barbey et al., 2001; Deloule et al., 2002; Laumonier et al., 2004; Cocherie et al., 2005).

The exact age of the pre-Upper Ordovician metasediments has also been a matter of debate due to their azaic character. According to its stratigraphic position, this sequence has been termed pre-Caradocian (Fontboté, 1949) or Cambro-Ordovician (Cavet, 1957) and it has been correlated with the Cambrian and Ordovician successions of the southern slopes of the Montagne Noire. High-grade paragneisses located in the lowermost part of the pre-Upper Ordovician sequences
Table 1
Available geochronology in the pre-Variscan massif of the Pyrenees

<table>
<thead>
<tr>
<th>Massif</th>
<th>Lithology</th>
<th>Age</th>
<th>Method</th>
<th>Reference</th>
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</thead>
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<tr>
<td>Central Pyrenees</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Aston</td>
<td>Augengneiss</td>
<td>475</td>
<td>Rb/Sr isochron</td>
<td>5</td>
</tr>
<tr>
<td>Hospitalet</td>
<td>Leucogneiss</td>
<td>470</td>
<td>Rb/Sr</td>
<td>2</td>
</tr>
<tr>
<td>Santi</td>
<td>Sil orthogneiss</td>
<td>566±12</td>
<td>Single zircon evaporation</td>
<td>3</td>
</tr>
<tr>
<td>Barthelemont</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Santi</td>
<td>Milictonic</td>
<td>526±7</td>
<td>Single zircon evaporation</td>
<td>3</td>
</tr>
<tr>
<td>Santi</td>
<td>Paragneiss</td>
<td>539±26</td>
<td>Rb/Sr isochron</td>
<td>4</td>
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<tr>
<td>Easten Pyrenees</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Agly Paragneiss</td>
<td></td>
<td>~550</td>
<td>Rb/Sr</td>
<td>5</td>
</tr>
<tr>
<td>Canigó</td>
<td>Augengneiss</td>
<td>580±20</td>
<td>U-Pb TIMS</td>
<td>5</td>
</tr>
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<td>Canigó</td>
<td>Augengneiss</td>
<td>451±14</td>
<td>Single zircon evaporation</td>
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</tr>
<tr>
<td>Canigó</td>
<td>Núria orthogneiss</td>
<td>570±12</td>
<td>Single zircon evaporation</td>
<td>6</td>
</tr>
<tr>
<td>Canigó</td>
<td>Núria orthgneiss</td>
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<td>Single zircon evaporation</td>
<td>6</td>
</tr>
<tr>
<td>Canigó</td>
<td>La Preste</td>
<td>446±20</td>
<td>Single zircon evaporation</td>
<td>7</td>
</tr>
</tbody>
</table>

1: Jäger and Zwart (1986); 2: Majoor (1988); 3: Delapérrière et al. (1994); 4: Marshall (1987); 5: Vitrac-Michard and Allègre (1975); 6: Guitard et al. (1996); 7: Delapérrière and Respaut (1955); 8: Deloule et al. (2002); 9: Delapérrière and Soliva (1982); 10: Cocherie et al. (2005).

In some Pyrenean massifs have been dated, yielding late Neoproterozoic–Early Cambrian ages (Rb–Sr method in the Agly paragneisses, see Table 1). According to Autran and Guittard (1996), these ages represent a Cadomian homogenization of the Rb–Sr system superposed on older Precambrian protoliths (Sm–Nd model age of 1.6 Ga, Othman et al., 1984).

For the upper part of the sequence, a Mi–Late Cambrian age (Abad, 1987; Laumonier, 1988) or Late Cambrian/Early Ordovician (Guittard et al., 1998) has been proposed. Early Cambrian fossils have been found in an isolated outcrop located in the Eastern Pyrenees and bounded by Alpine faults (Abad, 1987; Perejón et al., 1994). However, the lack of continuity of this outcrop with the neighbouring massifs precludes any correlation with the pre-Upper Ordovician successions of the Pyrenees. Recent radiometric dating of an interlayered metatuff has yielded a Neoproterozoic age (581 Ma) for the middle part of the succession (U–Pb SHRIMP in zircon, Cocherie et al., 2005; see Table 1). This age measurement has rekindled the controversy over the existence of a Cambodian basement and raised new interesting questions such as the position of the Neoproterozoic–Cambrian boundary, the age of the upper part of the succession, and the extent and the significance of the Upper Ordovician unconformity (Casas and Fernández, 2007).

Cocherie et al. (2005) also present some SHRIMP results obtained from inherited zircon in the igneous rocks, which have a variety of Pan-African (600–800 Ma), Mesoproterozoic (~1.0 Ga), Palaeoproterozoic (~2.0 Ga) and Archean ages (2.5, 2.8 and 3.5 Ga).

4. SHRIMP U–Pb geochronology

4.1. Analytical techniques

U–Th–Pb analyses of zircon were conducted on the Sensitive High Resolution Ion Microprobe-Reverse Geometry (SHRIMP-RG) operated by the SUMAC facility (USGS-Stanford University) during one analytical session in February 2006.

Mineral separation was performed at the Universidad Complutense (Madrid) and the U.S. Geological Survey (Denver). The samples were crushed using a jawcrusher and pulverized with a disc mill. The zircons were separated using heavy fraction enrichment on a Wilfley table, magnetic separation with a Frantz isodynamic separator and density separation with methylene iodide. The zircons were handpicked under a binocular microscope and representative grains were chosen in accordance with size, length-to-breadth ratio, roundness, colour, and other salient morphological features. They were then mounted on a double-sided adhesive on glass slides in 1 x 6 mm parallel rows together with some chips of zircon standard R33 (Black et al., 2004). After being set in epoxy resin, the zircons were ground down to expose their central portions. Internal structure, inclusions, fractures and physical defects were identified with transmitted and reflected light on a petrographic microscope, and with cathodoluminescence on a JEOl 5800LV electron microscope (housed at USGS-Denver).

The mounted grains were washed with 1 N HCl and distilled water, dried in a vacuum oven, and coated with Au. Mounts typically sit in a loading chamber at high pressure (~10^-7 Torr) for several hours before being moved into the source chamber of the SHRIMP-RG.

Secondary ions generated from the target spot with an O2¹ primary ion beam varying from 4–6 nA. The primary ion beam produced a spot with a diameter of ~25 μm and a depth of 1–2 μm for an analysis time of 8–10 min. Twelve peaks were measured sequentially in a single collector: 204Pb/206Pb, background (0.050 mass units above 204Pb), 206Pb, 207Pb, 208Pb, 238U160, 235U160, 166Er160, 172Yb160, 188Hf160. Five scans were collected, and the counting time for 206Pb was increased according to the Paleozoic age of the samples to improve counting statistics and precision of the 206Pb/238U age. Measurements were made at mass resolutions of 6000–8000 (10% peak height) which eliminates all interfering atomic species. The SHRIMP-RG employs magnetic analysis of the secondary beam before electrostatic analysis to provide higher mass resolution than the forward geometry of the SHRIMP I and II (Clement and Compston, 1994). The reverse geometry of the USGS-Stanford SHRIMP provides very clean backgrounds. This geometry combined with the high mass resolution and the acid washing of the mount ensures that surface contamination is removed and that counts found at mass 204Pb are in fact Pb from the zircon. Moreover, before collecting the data the primary beam was rastered for 90–120 s over the area to be analyzed. Concentration data for zircons are standardized against zircon standard C23 (550 ppm U, Pidgeon et al., 1995), and isotope ratios were calibrated against R33 (206Pb/238U = 0.06716, equivalent to an age of 419 Ma, Black et al., 2004) which were analyzed repeatedly throughout the duration of the analytical session.

Data reduction follows the methods described by Williams (1998) and Ireland and Williams (2003), and SQUID (version 1.08) and ISOPLOT (version 3.00) software (Ludwig, 2001, 2003) were used. The Pb composition used for initial Pb corrections was 204Pb = 0.0554, 207Pb = 0.864 and 206Pb = 0.719, calculated by SQUID using the age of the standard R33 and Stacey and Kramers (1975) model.

4.2. Sample descriptions

The metamuff collected in the Canigó massif (NU-3) has a medium-grained schistose fabric defined by lepidoblastic biotite-rich layers and granoblastic quartz-feldspathic domains, enclosing porphyroclasts of plagioclase, quartz and K-feldspar. Other minerals of this metamorphic paragenesis include muscovite and opaque ore. Chlorite and sericite replace biotite and feldspar, respectively.

In this sample, zircon morphologies vary from rounded grains to idiomorphic prisms, suggesting multiple zircon sources, consistent with a calc-silicate origin of the host rock. Under cathodoluminescence (CL), most zircons exhibit oscillatory zoning and...
xenocrystic cores (Fig. 3), although other textures, such as soccer-ball (grain 19) and homogeneous (grain 22) zoning can also be found.

The metatuff collected in the Roc de Frausa massif (RF-3) consists of variably recrystallized plagioclase porphyroclasts in a lepidoblastic groundmass of biotite, defining a planar fabric. Quartz, muscovite and opaque ore can also be found. Retrograde chlorite and sericite replace biotite and plagioclase. Zircons from sample RF-3 are moderately rounded stubby prisms (aspect ratio 1:2) and elongated prisms (aspect ratio 1:4). Under CL, these zircons display a faint moderately luminescent oscillatory zoning with scarce xenocrystic weakly luminescent cores (Fig. 4).

The Mas Blanc orthogneiss (RF-4) contains centimetre-scale K-feldspar porphyroclasts in a heterogranular matrix composed of quartz, K-feldspar, plagioclase, biotite and garnet. Quartz has undulose extinction and subgrains, evidencing dynamic recrystallization. Secondary chlorite, clinozoisite, epidote and sericite are found. The Roc de Frausa orthogneiss (RF-5) is characterized by the presence of K-feldspar in a groundmass composed of quartz, plagioclase, biotite and garnet.

Zircons from the Mas Blanc and Roc de Frausa orthogneisses (RF-4 and RF-5, respectively) are similar: moderately rounded prisms with variable aspect ratios (1:2 to 1:4). CL images of both samples reveal an oscillatory zoning with some inherited xenocrystic cores (Fig. 4).

The Cap de Creus metatuff (CC-2) was sampled from a metre-scale lense of strongly foliated rocks with K-feldspar and albite porphyroclasts enclosed in a fine-grained matrix formed by abundant quartz, plagioclase, biotite and opaque minerals. Clinozoisite, sericite and chlorite replace the metamorphic mineral assemblage.

Zircons from this sample are stubby grains (aspect ratio lower than 1:2) with scarce inclusions and moderately rounded tips. Under CL a variety of textures become evident (Fig. 5), which is consistent with a volcano-sedimentary origin of this rock. These textures include oscillatory, sector (grain 15) and soccer-ball zoning (grain 17). Some grain cores are surrounded by a moderately luminescent homogeneous rim (grain 9).

The Port orthogneiss (CC-7) is a highly homogeneous, massive and rather leucocratic sill. It displays a recognizable relict porphyritic texture with sporadic K-feldspar phenocrysts. The rock has a blastoporphyrritic texture, with tectonically induced foliation. The orthogneiss is composed of perthitic microcline, quartz, plagioclase and biotite. Accessory minerals are zircon, apatite, allanite and abundant titanite. Porphyroclasts composed of microcline containing amphibole, chlorite and albite inclusions and others consisting of polycrystalline albite are frequent.

In the Port gneiss, zircon grains are usually idiomorphic prisms (aspect ratios between 1:2 and 1:3) with small inclusions, although rounded grains and broken prisms can also be found. Under transmitted light microscopy, core-rim features are evident in some grains. Cathodoluminescence images reveal that most of the grains exhibit complex textures, commonly rounded xenocrystic cores with variable CL response, from low to high luminescence, and weakly luminescent overgrowths (Fig. 5). Other grains show oscillatory zoning with a varied luminescence response from core (moderate) to rim (weak).
4.3. U-Pb results

One hundred and nine analyses were performed on 101 zircon grains. The whole set of analytical data is provided as supplementary material. Uncorrected radiogenic compositions are plotted in Tera-Wasserburg concordia diagrams (Figs. 6, 7 and 8). In order to visualize their complexity, results from some samples are also plotted in probability density diagrams (Fig. 9). Ages younger than 1000 Ma are reported as 207Pb-corrected 206Pb. Otherwise, the reported age is 206Pb-corrected 207Pb/206Pb. The correction method is described in Ludwig (2001).

In the metatuff from the Canigo massif (NU-3), the variety of zircon textures results in an assortment of ages (Fig. 6) that can be clearly visualized in a probability density diagram (Fig. 9), where data with less than 10% of discordance were plotted. In this plot, the age profile presents significant peaks at ~640, ~680, ~800 and ~970 Ma. In addition, some older ages can also be found, with peaks at ~1.0, ~2.3 (with a discordance greater than 10%, and therefore not included in Fig. 9) and 3.1 Ga.

Because of its volcanoclastic nature, it is not possible to obtain an exact age for this sample. Instead, a maximum age could be inferred for its formation, considering the youngest concordant age obtained from a magmatic zircon. However, the youngest age in this sample corresponds to a single analysis (~540 Ma, Fig. 3, grain 4), which makes it unsuitable for inferring the maximum deposition age of sample NU-3 owing the possibility of lead loss. The next youngest age group clusters around an age of 640 Ma, which is too old to be considered close to the deposition age given earlier geochronological work (see above).

In the Roc de Frausa massif, the results in themetatuff(sample RF-3) and the Mas Blanc orthogneiss (sample RF-4) are dominated by Neoproterozoic to Early Cambrian ages, whereas, data from the Roc de Frausa orthogneiss (sample RF-5) yield an Early Ordovician age and a string of Neoproterozoic inheritances (Figs. 7, 9). In sample RF-3, only weakly luminescent areas with oscillatory zoning were analyzed to obtain the protolith crystallization age. The analyzed areas have low U contents (90–140 ppm) and a tight range of Th/U ratios (0.3–0.4). Even though their measured isotopic compositions are concordant within analytical uncertainty (Fig. 7a) the best age is obtained from a set of seven analyses, yielding a concordia age of 548.4±8.4 Ma (95% confidence). The outlier analyses are omitted owing to their high U content (points 9.1 and 11.1), the low Th/U ratio (point 10.1) and the possibility of lead loss (point 1.1).

In the Mas Blanc orthogneiss (RF-4), the analyzed spots correspond to weakly luminescent magmatic oscillatory zones with moderate U content (150–800 ppm) and Th/U ratios (0.09–0.35). The measured radiogenic compositions are equally distributed between two populations, ~510 and ~560 Ma. There are two possible explanations for this arrangement: 1) the younger age could represent the crystallization age with the result that the older one would be interpreted as inheritance; 2) the older age could be regarded as the crystallization age and the younger age would reflect lead loss. There appears to be no relation between age difference and CL features (Fig. 4), common lead, uranium, thorium contents or Th/U ratio, which is to be expected in zircons grown at different times (see Supplementary material). Thus, we prefer the latter interpretation and consider that ~560 Ma represents the crystallization age of the igneous protolith. By pooling the oldest ages, we obtain a concordia age of 560.1±7.2 Ma (95% confidence) for the crystallization of this rock.

In the Roc de Frausa orthogneiss (RF-5), twelve analyses were made in moderately luminescent areas with oscillatory zoning to obtain the crystallization age of the protolith. The U content and the Th/U ratio vary widely in range (100–1200 ppm and 0.07–2.30, respectively). After rejecting analyses 5.1, 10.1, 12.1 (probable
lead loss) and 1.2 (probable hit in a core), a mean age of $475.9 \pm 4.7\text{ Ma}$ (95% confidence) was obtained from eight analyses (Fig. 7c).

Additionally, four xenocrystic cores were analyzed to trace the inheritance component, yielding single ages in the interval from $\sim 530$ to $\sim 870\text{ Ma}$ (Fig. 7).

In the Cap de Creus metatuff (CC-2), the range of ages obtained is attributed to the diverse CL textures found in zircon grains. The spots were aimed to magmatic areas with oscillatory or homogeneous zoning, avoiding weakly luminescent rims. The U and Th contents of the analyzed grains are low to moderate, with Th/U ratios typical of melt-precipitated zircon. The youngest set of analyses yields a concordia age (sensu Ludwig, 1998) of $560.1 \pm 10.7\text{ Ma}$ (Fig. 8a). The remaining results are inherited ages in the $600-900\text{ Ma}$ and $2.5\text{ Ga}$ ranges (Fig. 9). In the Port orthogneiss (CC-7), 21 grains were analyzed in the central and rim areas with oscillatory zones, hitting some xenocrystic cores to trace inheritance. The U and Th contents of the analyzed spots are variable, ranging $100-7000\text{ ppm}$ and $50-1900\text{ ppm}$, respectively (see Supplementary material). The measured radiogenic $^{206}\text{Pb}/^{238}\text{U}$ values are equivalent to ages from $540$ to $2500\text{ Ma}$. Bearing these parameters in mind, three groups can be differentiated (Fig. 10).

The first group is composed of analyses with the lowest $^{206}\text{Pb}/^{238}\text{U}$ values and moderate U and Th contents obtained from spots in the...
centra areas of magmatic zircons (points 4.1, 8.1, 9.1, 14.2 and 20.1). Isotopic ratios form a cluster giving a mean age of 576±8 Ma (95% confidence). The second group has also moderate U and Th contents, and ages range from 620 to 2500 Ma (interpreted as inheritance). The analyses were made in xenocrystic cores (points 13.2, 16.2, 19.2 and 21.2) and zircon magmatic grains without weakly luminescent (meta­morphic) rims (points 2.1, 3.1 and 12.1). The third group has high U contents (over 4000 ppm) obtained from spots in weakly luminescent magmatic rims. The reverse discordance of these analyses (Fig. 8) and the strong correlation between U and radiogenic 206Pb/238U (Fig. 10), suggest that these are more probably due to U-dependent changes in sputtering and secondary ionization efficiency than to accumlated radiation damage (Butera et al., 2001). This situation resembles that described in McLaren et al. (1994) and Williams and Herdt (2000). This analytical bias can be compensated, applying a correction of 2% per 1000 ppm of U over 2500 ppm (see Supplementary material), which reduces the dispersion in radiogenic 206Pb/238U (Fig. 8). However, to obtain a weighted mean 206Pb/238U age we must reject some of the analyses with the highest U and Th (points 10.1, 15.1, 16.1, 18.1 and 19.1). The youngest age was ruled out (point 1.1) because it deviated from the
5.2. West African craton provenance, which is similar to other peri-Gondwanan groups can be visualized. The Variscan belt has been used to lend support to the existence of South America derived zones. This is not consistent with the absence of Grenvillian ages in detrital zircons and by the higher ENd values in the South American derived zones.

Given that 207Pb/206Pb ratios are not affected by the U content, as noted by McLaren et al. (1994), a weighted mean can be calculated for all the high U analyses (except 16.1), yielding an age of 546±7.3 Ma (95% confidence). By pooling both ages, we can obtain a best estimate of 553±4.4 Ma (95% confidence) for the protolith age of the Port orthogneiss. Finally, older ages ranging from 1.8 to 2.5 Ga are regarded as inheritance.

5. Interpretation and discussion

5.1. Zircon inheritance

Despite of the scarcity of inherited ages obtained in the Canigó and Cap de Creus metatuffs (samples NU-3, and CC-2) and the Roc de Frausa orthogneiss (sample RF-5), some conclusions on the characterization of the sedimentary sources can be drawn. The presence of Pan-African (600-800 Ma) and Mesoproterozoic (~3.1 Ga) detrital zircon ages point to a West African craton provenance, which is similar to other peri-Gondwanan areas (e.g., Fernández-Suárez et al., 2002b; Linnemann et al., 2004).

However, the Grenvillian (0.9-1.1 Ga) signature in the detrital zircon age patterns in the Neoproterozoic and Paleozoic sediments of the Variscan belt has been used to lend support to the existence of South American derived crust in Central Europe (Friedl et al., 2000). Fernández-Suárez et al. (2002b) and Gutiérrez-Alonso et al. (2003) have proposed two contrasting source areas during the Neoproterozoic in the pre-Variscan exposures of the Iberian massif: the West African craton for the Ossa-Morena zone and the South American craton for the Cantabrian, West Asturian Leonese and Central Iberian zones. This paleogeographic model is supported by the presence of Grenvillian ages in detrital zircons and by the higher ENd values in the South American derived zones. This is not consistent with the absence of Grenvillian ages and the lower ENd values in the West African derived zone.

However, this is not the only explanation to account for a Grenvillian signature in the detrital zircon spectra of samples from the peri-Gondwanan realm. Recently, some authors have documented Grenville zircon ages in sediments from northern Gondwana, at a considerable distance from Amazonian sources, favouring a North African source (Keay and Lister, 2002; Avigad et al., 2003; Drost et al., 2007).

Finally, Neoarchean ages (~2.5 Ga) can be found in both Amazonian and North African sources (Abdesalam et al., 2002; Fernández-Suárez et al., 2002a).

5.2. Age of the pre-Upper Ordovician sequences and magmatism in the Pyrenees

Based on our geochronological data, we recognize two pre-Variscan magmatic episodes related to the pre-Upper Ordovician sequence. The oldest magmatic episode is represented by intrusive and extrusive rocks. The results from the extrusive rocks yield information on the age of the sequences. Thus, from the Cap de Creus metatuff (CC-2) we estimate the age of the top of the Lower Series to be at least 560 Ma. In the Upper Series of the Roc de Frausa massif, the results from an interbedded metatuff (sample RF-3) indicate that it was deposited approximately at ~548 Ma, close to the Ediacaran–Cambrian boundary. To the SW of the Canigó massif, the analyzed metatuff (NU-3) occupies an equivalent stratigraphic position of sample RF-3. However, owing to the abundance of inherited zircon grains, only one analysis yielded an age close to ~540 Ma, making it difficult for us to obtain a maximum deposition age. Cocherie et al. (2005) analyzed zircons that also provided ~545 Ma in an equivalent sample from the southern slope of the Canigó massif (GRA-1). These authors discarded this result in favour of an older age of 580 Ma, arguing that it was difficult to regard all the zircons dated between 570 and 600 Ma as inherited zircons. This is a surprising statement given the continued Cadomian igneous activity from 600 to 550 Ma along the northern Gondwana margin.

In the light of these findings, it is reasonable to assume that the Neoproterozoic–Cambrian boundary is situated fairly close to the studied samples in the three massifs. In the Canigó massif it would be located inside the Canavelles Series, probably in the lithologically varied group (black shales, carbonates and metavolcanic rocks) located in the lower part of the Canavelles Series, where sample NU-3 was collected (Fig. 2). In the Roc de Frausa, the presence of a syn- orogenic Variscan granitoid masks the upper part of the metasedimentary series. However, this boundary would be situated in the Upper series close to sample RF-3 (Fig. 2). In Cap de Creus, the presence of the Roses and Rodes granodiorites and the reduced thickness of the series make it more difficult to place the boundary. However, it could be located in the conglomerates and carbonates of the Upper series (Fig. 2). Should this interpretation be correct, the Jujols Series of Cavet (1957), or Jujols Group of Laumonier et al. (2004) could represent a sequence deposited in a quiet environment, probably ranging in age from Mid Cambrian to Early Ordovician (Fig. 2). Nevertheless, further research and geochronological work is needed to assess the age of the intermediate levels of the Canavelles and Jujols Series and the magnitude of the Upper Ordovician unconformity.

Moreover, the metaplutonic rocks located in the lower part of the series in the Cap de Creus and Roc de Frausa massifs (Port and Mas Blanc orthogneisses, respectively) yield Late Neoproterozoic ages for their intrusion (~553 and 560 Ma, respectively). These ages correspond to the emplacement of the plutonic rocks, and are roughly equivalent to the age of the metatuffs interbedded in the upper part of the metasedimentary sequence. However, the age of the lowermost series in the three massifs remains unresolved, even though an age slightly older than the plutonic rocks could be proposed (older than 600 Ma?). Further geochronological studies are warranted to gain further insight into the age of these series that represent the deepest rocks cropping out in the Eastern Pyrenees.

Field relationships and geochemistry of metatuffs, metabasites and metaplutonic rocks suggest that this syn-sedimentary Late Neoproterozoic (Ediacaran)-Early Cambrian magmatism is bimodal. No tectonic or metamorphic Late Neoproterozoic–Early Cambrian activity related to this igneous event has been described in the study area, to date.

The second magmatic episode is represented by sample RF-5 (Roc de Frausa gneiss) which yields 476 Ma, confirming the presence of an important Early Ordovician magmatic event. This event is recognized in the Pyrenees (Delaperrière and Respaut, 1995; Deloule et al., 2002; Cocherie et al., 2005) and in other sectors of the European Variscides, where the intrusion of Early Ordovician granites is widely documented (Delaperrière and Lancelot, 1989; Pin and Marini, 1993; Valverde-Vaquero and Dunning, 2000; Roger et al., 2004; Helbing and Tiepolo, 2006).
These ages are in agreement with the models proposed for the pre-Variscan evolution in the Central and Southeastern European Alpine mountain belts. In these models, an Andean-type continental margin triggered by the southward subduction of the Iapetus or Proto-Thethys Ocean (570–520 Ma) is followed by a back-arc rifting episode (520–500 Ma) prior to the separation of a terrane from northern Gondwana (490–485 Ma) [Neubauer, 2002; Stampfl et al., 2002; von Raumer et al., 2002]. Cambrian igneous activity related to a rifting episode is widespread in the Iberian massif (Simancas et al., 2004), but it has not been recognized in the Pyrenees to date.

In addition, the subsidence pattern from Lower Ordovician to Carboniferous calculated by von Raumer and Stampfl (2008–this issue) for the Pyrenees documents the continuation of the extensional episode and the opening of the Paleotethys.

6. Concluding remarks

U–Pb SHRIMP dating of metagneous rocks from the pre-Variscan basement allowed us to differentiate two magmatic episodes. The older episode is constituted by Late Cadomian intrusive and extrusive rocks (560–580 Ma), whereas the younger episode corresponds to Early Ordovician magmatism (476 Ma).

The ages obtained in extrusive rocks from the pre-Upper Ordovician metasedimentary sequence for the series of the Roc de Frausa and Cap de Creus massifs allow us to estimate the deposition age as Ediacaran–Early Cambrian (~540 Ma). In the Canigó massif, this age would correspond to the lower part of the Canavelles series. We suggest that the lowermost series of the three massifs (Balaig series, in the Canigó and the Lower and Intermediate series in the Roc de Frausa and Cap de Creus massifs) are Neoproterozoic. It should be noted that neither of the two magmatic episodes described can be related to deformational structures. All this confirms the absence of a Cadomian basement in the Pyrenees.

Moreover, despite the scarcity of inherited zircon ages, these ages are sufficiently distinctive to identify a source for the metavolcanosedimentary rocks from the Eastern Pyrenean massifs. This source may be comparable with the Cantabrian, West Asturian Leonese and Central Iberian zones in the Iberian Massif, and would probably be located in North Africa.

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Appendix A. Supplementary data


Navidad, M., Carreras, J. 1995. Pre-Hercynian magmatism in the eastern Pyrenees (Cap de Creus and Albera Massifs) and its geographical setting. Geol. Mblg. 74, 65-77.

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