Supplementary File: Zircon geochronology and Multidimensional Scaling statistics

Annex to the Research paper:

A tectonic carpet of Variscan flysch at the base of an unrooted accretion prism in NW Iberia: U-Pb zircon age constrains from sediments and volcanic olistoliths

González Clavijo, E.[a]; Dias da Silva, I.[b,c]; Martínez Catalán, J.R.[d]; Gómez Barreiro, J.[d]; Gutíerrez Alonso, G.[d]; Díez Montes, A.[a]; Hofmann, M.[e]; Gärtner, A.[e]; Linnemann, U.[e]

a) Instituto Geológico y Minero de España. Plaza de la Constitución 1, 3º, 37001 Salamanca, Spain
b) Instituto Dom Luiz, Faculdade de Ciências, Universidade de Lisboa, Campo Grande, 1749-016 Lisboa, Portugal
c) Departamento de Geologia, Faculdade de Ciências, Universidade de Lisboa, Campo Grande, Lisboa, 1749-016, Portugal;
d) Departamento de Geología, Universidad de Salamanca. Plaza de la Merced, s/n, 37008 Salamanca, Spain
e) Senckenberg Naturhistorische Sammlungen Dresden, Königsbrücker Landstr. 159, D-01109 Dresden, Germany

Content

1. Zircon Geochronology

1.1. Samples preparation procedures and U-Th-Pb isotope analyses

Zircon concentrates were separated from 2-4 kg sample material. Zircon grains of the selected samples were separated using traditional techniques. Samples were crushed with a jaw mill reaching 1-2 cm2 size rock fragments. Further grinding to sand grain-size was made with a WIDIA TEMA grinder or a RETSCH cross beater pulverizer (sample preparation laboratories in the Departamento de Geología de la Universidad de Salamanca, Spain and in the Facultad de Geociencias de la Universidad Complutense de Madrid, Spain). After granulometric sieving under 500 micra, all samples were processed in a wet Wilfley mineral separation table (hosted at the Universidad Complutense de Madrid), passing several times until achieving a zircon super-concentrate. No magnetic separation was used to avoid artificial biases. We have avoided environmentally harmful and toxic heavy liquids in this process. The ultimate zircon concentrate was obtained using hand picking with a binocular lens (Universidad Computense de Madrid, Universidade de Évora, Portugal and Senckenberg Naturhistorische Sammlungen Dresden, Germany). The selected zircon grains were mounted by hand with a binocular lens on a double-sided tape where a plexiglass ring was attached and filled with crystal high-grade epoxy resin, following the specifications of the LA-ICP-MS mount port. After epoxy-resin stabilization, zircon cores were exposed by sanding and polishing.

The study of zircon internal structures was made with cathodoluminescence (CL) or backscattering (BS) electron imaging. CL images of the samples CR-ZR-01, MEI-ZR-01, GIM-ZR-01, PICON-2, P381 and P385 were obtained at the REM Labor of the Technische Universität Bergakademie Freiberg (Germany) with a JEOL JSM 6400 electron microscope. CL-images of samples EC-PO-293, AD-PO-66, AD-PO-48B, AD-PO-49, AD-PO-55, AD-PO-57, EC-PO-337 and EC-PO-419 were realized using a Zeis EVO50 scanning electron microscope at the Senckenberg Naturhistorische Sammlungen Dresden (Germany). CL imaging of sample MIR-41 was made in the USGS Denver Microbeam Laboratory (United States) using the JEOL 5800 LV electron microscope. PET-01 and RAB-01 zircons were documented by backscattered electron and cathodoluminescence (CL) images using a scanning electron microscope (JEOL JSM-820 at the Research Assistance Centre of Geological Techniques, Complutense University, Madrid, Spain) to study their internal structure to select the best areas for laser ablation.

U-Pb geochronology was made by different methods. U, Th and Pb isotope measurements of samples CR-ZR-01, MEI-ZR-01, GIM-ZR-01, PICON-2, P381, P385, EC-PO-293, AD-PO-66, AD-PO-48B, AD-PO-49, AD-PO-55, AD-PO-57, EC-PO-337, EC-PO-419, PET-1 and RAB-

1 were made at the Geochronology laboratory of the Senckenberg Naturhistorische Sammlungen Dresden (Museum für Mineralogie und Geologie, Germany) using the Thermo-Scientific Element 2 XR sector field mass spectrometer coupled to a RESOlution 193 nm Excimer laser system. Sequential sampling of heterogeneous grains (e.g., growth zones) during time resolved data acquisition was possible. Each analysis consisted of 15 s background acquisition followed by 30 s data acquisition, using a laser spot-size of 20, 25 and 35 µm, respectively. The fluence on the grain surface during laser ablation was monitored and kept below 2.0 J/cm^2. A common-Pb correction based on the interference- and background-corrected ^{204}Pb signal and a model Pb composition (Stacey and Kramers, 1975) was carried out if necessary. The necessity of this correction was judged on whether the corrected ^{207}Pb/^{206}Pb lies outside of the internal errors of the measured ratios (Frei and Gerdes, 2009). Discordant analyses were always interpreted with care. Raw data were corrected for background signal, common Pb, laser induced elemental fractionation, instrumental mass discrimination, and time-dependant elemental fractionation of Pb/Th and Pb/U using the Excel®-based AGeRAp program version 2.07 developed by Dr. Axel Gerdes, Dr. Richard Albert Roper, and Linda Marko (Radiogenic Isotope Lab FIERCE, Goethe University, Frankfurt/Main, Germany). Data acquisition and age calculation including all necessary corrections follow the protocol described in Gerdes and Zeh (2006, 2009) and Zeh and Gerdes (2012).

The GJ-1 (c.a. 608 Ma) (Jackson et al., 2004) external standard zircon was used in the measurements of samples CR-ZR-01, MEI-ZR-01, GIM-ZR-01, PICON-2, P381, P385, PET-1 and RAB-1. Additionally, to the test the accuracy of the measurements and data reduction in the isotopic measurements of samples EC-PO-293, AD-PO-66, AD-PO-48B, AD-PO-49, AD-PO-55, AD-PO-57, EC-PO-337 and EC-PO-419 we also used the Plesovice zircon as a secondary standard in our analyses. Repetitive measurements of the Plesovice zircon resulted in the age of c. 337 Ma, which fits the results of Sláma et al. (2008).

Isotope measures in the zircon grains of sample MIR-41 was made using a NU Instruments double focusing Single-Collector Sector-Field High-Resolution Inductively-Coupled Plasma Mass-Spectrometer (SF-HR-ICPMS) AttoM™ in combination with a Photon Machines Excite™ 193 nm ArF excimer laser with burst count of 200 shots, repetition rate of 5 Hz, laser energy of 5 mJ at 70%, and fluence of 5.9 J/cm2. ICP-MS data were collected in peak jumping mode, using 1 point per mass peak with a 40 s gas blank and a 40 s signal. Measured masses were: 202Hg, 204(Hg+Pb), 205Tl, 206Pb, 207Pb, 208Pb, 232Th, 235U, and 238U. Raw data were reduced off-line using Iolite™ program (http://www.iolite.org.au/Iolite.html) to subtract baseline signals, normalize for down-hole fractionation and instrumental bias using external mineral standards R-33 zircon (419

Ma) (http://www.plasmage.org/reference-materials), used as a primary standard and WRP 03-68 zircon (1707 Ma) used as an in-house secondary standard. Data were collected in 3 analytical sessions.40 unknowns were analyzed in each session. Each session started and ended with 3 analyses of R-33 and 3 analyses of WRP 03-68. Each of the standards was also analyzed twice after 5 unknowns.

Concordia diagrams (2σ error ellipses) and concordia ages (95% confidence level) were produced using Isoplot/Ex 4.15 (Ludwig, 2001) and frequency and relative probability plots using AgeDisplay (Sircombe, 2004) and ISOPLOT-R (https://www.ucl.ac.uk/~ucfbpve/isoplotr/home/index.html; Vermeesch, 2018). The $^{207}Pb/^{206}Pb$ age was taken for interpretation for all zircons >1.0 Ga, and the $^{206}Pb/^{238}U$ ages for younger grains.

All zircons showing a degree of concordance in the range of 90-110 % in this paper are classified as concordant because of the overlap of the error ellipse with the Concordia. Th/U ratios are obtained from the LA-ICP-MS measurements of investigated zircon grains. U and Pb content and Th/U ratio were calculated relative to the GJ-1 zircon standard and are accurate to approximately 10%.

The complete isotopic analyses are given in Supplementary Tables 1-17.

2. Sample description and zircon age populations

2.1. Siliciclastic rocks

CR-ZR-01

This sample was collected in the southern rim of the Morais Complex, at the base of the Meirinhos Formation (Pereira et al., 2003, 2009), belonging to the Lower Parautochthon of the GTMZ (i.e. above the BLPD). It is a well-sorted fine-grained greywacke, presenting abundant greenish-grey pelitic mainly consisting of chlorite. Angular quartz and feldspar are dominant, pyrite, organic matter and oxide minerals are secondary. Deformation is weak and was developed under low- to very low-grade metamorphism.

Yellowish, light-pink and colourless zircon grains vary in size from 60 to 200 µm. Few grains are rounded showing a core surrounded by a rim or are simple with oscillatory zoning. Many grains have angular shapes with bipyramidal prismatic and acicular habits and simple oscillatory zoning, suggesting a proximal magmatic source. Other zircon grains are sub-rounded to sub-angular (SI-1A).

From a total of 149 analyses performed on 136 zircon grains, 93 yielded concordant ages (90-110% concordance) (SI-1A). Precambrian ages represent 36% of the zircon age population distributed by: Paleoproterozoic (1% from ca. 2.5 to 1.8 Ga), Mesoproterozoic (3% ca. 1.8-1.0 Ga) and 32% Neoproterozoic (32%: 18% Ediacaran from 635 to 540 Ma, 8% Tonian ca. 975 Ma, 6% Cryogenian from 850 to 635 Ma). Paleozoic zircon ages represent the dominant group (64%) including Cambrian (40.9%: 14% Furongian, 10% Terraneuvian, 9% Series 2 and 9% Miaolingian), Ordovician (21.5%: 15% Lower Ordovician, 5.4% Middle Ordovician and 1.1% Upper Ordovician) and Silurian (1.1%).

The youngest age is 439±8 Ma and the estimated maximum depositional age based on the youngest population in the age range from 462 Ma to 469 Ma, is 467±4Ma (Middle Ordovician, 4 ages, 95% concordance interval). A second, most abundant youngest age population is 484±3 Ma with 12 zircon ages from 480 to 486 Ma (SI-1A).

MEI-ZR-01

This sample also belongs to the Meirinhos Formation, in a higher structural/stratigraphic position than CR-ZR-01. It is a medium-grained greyish laminated lithic sandstone. Angular to sub-angular quartz, K-feldspar, plagioclase and lithic fragments are dominant. Normal and reverse sediment gradation occurs in the same bed.

Zircon grains are colourless, pale-yellow and light pink and with size ranging from 80 to 240 µm. Most zircon grains are angular to sub-angular while other are sub-rounded. Many crystals are bipyramidal, prismatic to acicular. Zircon grains are mostly simple presenting banded or oscillatory zoning, and few grains include a core (SI-1B).

We have obtained a total of 76 concordant ages (90% interval) out of 149 analysis preformed on 139 zircon grains (SI-1B). No Archean or Mesoproterozoic ages were found. Palaeoproterozoic represents 4% of the ages, 32% are Neoproterozoic of which 5% are Tonian, 6% are Cryogenian and 21% are Ediacaran. Palaeozoic zircons are the most abundant population in this sample including 57% Cambrian ages (14% Terraneuvian, 9% Series 2, 17% Miaolingian and 17% Furongian), 5 % Ordovician ages (4% Lower Ordovician, 1% Middle Ordovician) and 1% Silurian.

The youngest age is 443±6 Ma and the maximum depositional age of 486±3Ma (upper Furongian) was estimated using the youngest population including grains with age range from 485 Ma to 488 Ma SI-1B.

GIM-ZR-01

This sample was collected from the Gimonde Formation (Meireles, 2013; Meireles et al., 1999a, 1999b), in the northern rim of the Bragança Complex, as part of the Lower Parautochthon of the GTMZ. It represents a coarse-grained light-brown lithic sandstone to microconglomerate showing thin-laminations of brownish-grey pelite. Angular to sub-angular shaped quartz, K-feldspar, plagioclase and lithic fragments are dominant.

Zircon grains size ranges from 80 to 180 µm. Most grains are rounded to sub-rounded but there are also several subangular crystals. Many crystals show bipyramidal prismatic habit and some are stubby. They frequently present simple banded or oscillatory zoning, and few include a core. Zircon colours range from colourless, to pale-yellow and light pink (SI-2A).

A total of 74 concordant ages (90% interval) out of 121 analysis on 122 zircon grains were obtained (SI-2A). 14% of the ages were Archean, 27% are Palaeoproterozoic, 4% are Mesoproterozoic and 29% are Neoproterozoic of which 1.3% is Tonian, 1.3% is Cryogenian and 26.4% are Ediacaran. Palaeozoic zircons represent a total of 26% including 14% Cambrian ages (14% Terraneuvian, 9% Series 2, 17% Miaolingian and 17% Furongian), only 1 % Middle Ordovician, 5% Devonian (Givetian-Fammenian) and 5% are early Carboniferous (Tournaisian-Serpukovian).

The youngest zircon grain has 327±9 Ma and the maximum depositional age of 354±3 Ma was obtained based on the youngest detrital zircon population with ages in the range of 351 Ma and 359 Ma (2 ages, 95% concordant). The second youngest detrital zircon population is 382±4 Ma (Frasnian) with 3 ages in the range from 375 to 386 Ma (3 ages) (SI-2A).

EC-PO-293

Although this sample was collected in a higher tectonic sheet of the LPa than GIM-ZR-01, few meters below the Lower Allochthon in the Bragança Complex in the Gimonde Fm. It is texturally and compositionally identical, even though it presents black quartzite clasts (<10 cm) in the coarser-grained beds.

Zircon grains size ranges from 80 to 150 µm. Most have subangular to sub-rounded shapes. Many crystals have euhedral bipyramidal prismatic habits and some have stubby shapes. The bipyramidal frequently present simple banded or oscillatory zoning, and many include a core (SI-2B). Some show thin rim overgrowths. Zircon colours range from colourless, to pale-yellow and light pink.

From 118 analysis preformed on 101 zircon grains, a total of 97 concordant ages (90% interval) were obtained (SI-2B). 6% of the ages were Archean, 34% are Palaeoproterozoic, 11% are Mesoproterozoic and 32% are Neoproterozoic of which 3% is Tonian, 7% is Cryogenian and 22% are Ediacaran. Palaeozoic zircons represent a total of 17% including 7% Cambrian (6% Terraneuvian, 1% are Series 2, 2% Miaolingian and 3% Furongian) and 1 % Silurian.

The youngest zircon grain has 426±44 Ma and the maximum depositional age (youngest population) has 435±29 Ma (2 ages, 95% concordant). The second youngest population is 495±24 Ma (4 ages).

AD-PO-66

This sample belongs to the Gimonde Fm. in a similar tectono-stratigraphic position as EC-PO-293, laying some meters below the basal thrust of the Lower Allochthon in the Bragança Complex. This sample is composed by an alternance of middle-coarse grained greywackes and greyish slates. The coarser grained rocks can have a strong volcanoclastic component with broken quartz and feldspar crystals.

Zircon grain size ranges from 80 to 180 µm, with rounded to sub-rounded shapes. Few are subangular. Many crystals have bipyramidal prismatic habits and other are stubby. The most elongated zircon prisms have simple banded growth (SI-3A). The bipyramidal show oscillatory zoning, frequently exhibiting a core. Many grains have thin rim overgrowths. Zircon colours range from colourless, to pale-yellow and light pink.

We made 159 analysis on 144 zircon grains, obtaining 101 concordant ages (90% interval) (SI-3A). 12% of the ages were Archean, 23% are Palaeoproterozoic, 7% are Mesoproterozoic and 34% are Neoproterozoic (6% Tonian, 6% Cryogenian and 22% Ediacaran). Palaeozoic zircons represent a total of 24%, namely 19% Cambrian (2,8% Terraneuvian, 5,5% are Series 2, 5,5% Miaolingian and 5,5% Furongian), 4% Ordovician (3% is Lower and 1% is Middle Ordovician) and 1% Silurian.

The youngest zircon age has 431±31 Ma and the maximum depositional age (youngest population) of this sample is 483±18 Ma (3 ages, 95% CONC). The second youngest population is 507±14 Ma (5 ages) (SI-3A).

AD-PO-48B

This sample belongs to the Curros Fm. (Lower Parautochthon, W Morais Complex) (Rodrigues, 2008; Rodrigues et al., 2010). It is composed of fine-grained arkosic sandstones with 5-10 mm lithoclasts, presenting low-grade metamorphism and folds with spaced axial planar cleavage.

Zircon grain sizes range from 70 to 220 µm with most grains showing rounded to sub-rounded shapes and few subangular. There are abundant bipyramidal prismatic grains, but some have stubby shapes. The longer prismatic grains frequently show simple banded growth (SI-3B). The bipyramidal show oscillatory zoning, often exhibiting a core. Zircon colours range from colourless, to pale-yellow and light pink

This sample yielded a total of 61 concordant ages (90% interval) out of 98 analysis preformed on 58 zircon grains (SI-3B). 11% of the ages were Archean, 13% Palaeoproterozoic, 7% Mesoproterozoic and 66% Neoproterozoic (26% Tonian, 25% Cryogenian and 15% Ediacaran). Palaeozoic zircons represent only 3% of the ages, of which 1 age is Fammenian and 1 age is Tournaisian.

The youngest zircon grain has 355±34 Ma and the maximum depositional age (youngest population) has 364±22 Ma (2 ages, 90% conc.). The second youngest population is 561±23 Ma (4 ages) (SI-3B).

MIR-41

This sample was collected in a Lower Parautochthon tectonic window, a few meters below the MTMT. It belongs to the Upper Schist Fm. (Pereira et al., 2006; Rodrigues et al., 2010) previously attributed to the Silurian, and it is formed by fine-grained greywackes and laminated slates. Zircon grains have sizes ranging from 120 to 220 µm. Most grains have rounded to sub-rounded shapes and are colourless, to pale-yellow and light pink. Most have bipyramidal prismatic habits but there are abundant stubby crystals. The most elongated prismatic zircons have simple banded growths while the bipyramidal show oscillatory zoning and xenocrystic cores (SI-4A).

We have performed a total of 119 analysis in 119 zircon grains (SI-4A). We have obtained a total of 83 concordant ages (90% interval). 5% of the ages are Archean, 34% Palaeoproterozoic, 4% Mesoproterozoic and 50% Neoproterozoic (6% Tonian, 6% Cryogenian and 22% are Ediacaran). Palaeozoic zircons represent a total of 7% being 5% Cambrian (2,4% Series 2, 1,2% Miaolingian and 1,2% Furongian), 1% is Lower Ordovician and 1% is Famennian.

The youngest zircon age is 369±7 Ma. It presents one single 479±6 Ma age and a youngest population of 497±5 Ma (3 ages, 90% concordant). The second youngest population is 552±8Ma (3 AGES, 95% concordant) (SI-4A).

AD-PO-49

This sample was collected in the same site as MIR-41, 30-40 cm stratigraphically above. It is made of fine-grained greywackes.

Zircon grains have sizes ranging from 70 to 200 µm. Most grains have rounded to sub-rounded shapes. Many crystals have bipyramidal prismatic habits but there are also stubby crystals. The long prismatic grains show simple banded growth and the bipyramidal show oscillatory zoning and, commonly a core (SI-4B). Zircon colours range from colourless, to pale-yellow and light pink.

We have made 170 analysis in 105 zircon grains, including rims and cores (SI-4B). They yielded a total of 112 concordant ages (90% interval) in which 5% are Archean, 30% Palaeoproterozoic, 11% Mesoproterozoic and 52% Neoproterozoic (16% Tonian, 17% Cryogenian and 19% Ediacaran). Palaeozoic zircons only represent 2% (1% Cambrian Stage 2 and 1 % Middle Ordovician).

The youngest zircon age has 468±39 Ma and a youngest population with 494±27 Ma (2 ages, 90% concordant). The second youngest population is 574±12 Ma (5 ages, 95% concordant) (SI-4B).

AD-PO-55

This sample was also collected in the Upper Schist Fm., in a lower structural/stratigraphic position. It is made of medium grained sandstones with positive granular selection, showing intercalations of dark-grey slates.

Zircon grains have sizes ranging from 70 to 180 µm. Most grains are rounded to sub-rounded. Many crystals are bipyramidal prismatic and some have stubbier shapes. The prismatic grains frequently show simple parallel growths (SI-5A). The bipyramidal frequently show oscillatory zoning with a core and thin rim growths. Zircon colours range from colourless, to pale-yellow and light pink

We have made 150 analysis preformed on 142 zircon grains, including rims and cores (SI-5A). A total of 92 concordant ages (90% interval) were obtained in which 7% are Archean, 17%

are Palaeoproterozoic, 3% are Mesoproterozoic and 63% are Neoproterozoic, including 16% Tonian, 15% Cryogenian and 32% Ediacaran. Palaeozoic zircons represent a total of 10% of the concordant ages of which 4% is Cambrian (1% Terraneuvian and 3% Furongian), 4% is Ordovician (3% is Lower and 1% is Middle Ordovician) and 1% is Silurian.

The youngest zircon age is 444±26 Ma and the maximum depositional age (youngest population, 3 ages, 95% concordant) of this sample is 488±16 Ma. The second youngest population is 565±18 Ma (3 ages, 95% concordant) (SI-5A).

AD-PO-57

This sample belongs to the Culminating Slates and Greywackes Formation (Pereira et al., 2006; Rodrigues et al., 2010). It is made of middle grained sandstones with fine layers of dark-grey slates.

Zircon grain sizes range from 70 to 180 µm, showing colourless, to pale-yellow and light pink coloration. Most grains are rounded to sub-rounded with bipyramidal prismatic habits and some with stubby shapes. The prismatic grains frequently show simple banded growths and the bipyramidal show oscillatory zoning. Many zircon grains have cores and/or thin rim overgrowths (SI-5B).

We have made 177 analysis on 142 zircon grains, including rims and cores (SI-5B). A total of 116 concordant ages (90% interval) were obtained, in which 9% of the ages are Archean, 14% are Palaeoproterozoic, 4% are Mesoproterozoic and 64% are Neoproterozoic (9% Tonian, 22% Cryogenian and 33% Ediacaran). Palaeozoic zircons represent a total of 9% of the ages, including 6% Cambrian (2% Terraneuvian, 1% Series 2, 1% Miaolingian and 2% Furongian), 1 % Middle Ordovician and 3% Upper Devonian (Frasnian-Famennian).

The youngest zircon age has 368±14 Ma and the maximum depositional age (youngest population) of this sample is 372±6 Ma (3 ages, 90% concordant). The second youngest population is 488±16 Ma (3 ages, 90% concordant) (SI-5B).

PICON-2

This sample was collected at the base of the Rio Baio unit (Lower Parautochthon in this work) (Marcos et al., 2002) in the eastern Rim of the Cabo Ortegal Complex, immediately over the BLPD in the Picón beach (Spain). This sample is a rock aliquot made of fine- to middle-grained black quartzites with pyrite and black pelithic laminations. Quartz grains are clast supported, with organic matter in the voids. The pelithic lamellae are mostly composed by chlorite.

Zircon grains have sizes ranging from 50 to 180 µm. Most grains are rounded to sub-rounded and there are some subangular with colourless, to pale-yellow and light pink colours. Many crystals are bipyramidal prismatic and some have stubbier shapes. They frequently present simple banded growth or oscillatory zoning many times including a core (SI-6).

We have made 149 analysis preformed on 145 zircon grains, of which 73 are concordant ages within the 90% confidence interval (SI-6). 9% of these ages are Archean, 23% are Palaeoproterozoic, 10% are Mesoproterozoic and 29% are Neoproterozoic (4% Tonian, 6% Cryogenian and 19% Ediacaran). Palaeozoic zircons represent a total of 29% of the ages, including 7% Cambrian (3,9% Terraneuvian, 1,3% Miaolingian and 1,3% Furongian), 4 % Ordovician (1% is Early and 3% Middle Ordovician), 11% Devonian (Eifelian-Fammenian) and 6% early Carboniferous (Tournaisian)

The youngest zircon grain with 350±7 Ma and maximum depositional age (youngest population) of 357±4 Ma (4 ages, 95% CONC). The second and third youngest age groups are respectively 375±5 Ma (3 ages) and 384±4 Ma (3 ages) (SI-6).

2.2. Igneous rocks

P-381

This sample was collected in NE Morais Massif, in the Peso Fm. (Dias da Silva et al., 2016), in the UPa (former Vulcanic-Siliceous Complex of the Lower Allochthonous Complex of Pereira et al., 1998, 2006), few meters below the basal Lower Allochthon thrust zone, represented here by the Macedo de Cavaleiros Fm. (Pereira et al., 1998). It is an intensely foliated (mylonitic) white rhyolite with quartz phenocrysts and white micas.

We have made 180 analysis preformed on 177 zircon grains, including rims and cores. Zircons are often prismatic acicular, with banded growth, showing sharp contours (angular to sub-angular grains). Others are bypiramidal with oscillatory zoning and sometimes exhibiting a core (SI-7A). Zircon colours range from colorless to pale pink.

We have obtained a total of 85 concordant ages (90% interval) (SI-7A). There are no Archean nor Mesoproterozoic ages. 3% of the ages are Palaeoproterozoic, 14% are Neoproterozoic (1,1% Tonian, 1,1% Cryogenian and 11,4% Ediacaran). Palaeozoic zircons represent a total of 83% of ages, of which 68% are Cambrian (14% Terraneuvian, 16% are Series

2, 24% Miaolingian and 15% Furongian) and 15% are Ordovician (8% are Lower, 6% are Middle and 2% are Upper Ordovician).

The youngest zircon age has 456±9 Ma and the magmatic age (youngest population) of this sample is 461±4 Ma (6 ages, 95% interval), with the second and third youngest populations respectively with 481±6 Ma (2 ages) and 498±3 Ma (6 ages) (SI-7A).

P-385

This sample is made of foliated dacite with quartz, feldspar and plagioclase phenocrysts, collected in a similar stratigraphic and structural position as P-381, in the Peso Fm. of the UPa, NW of the Morais Complex.

Zircon grains have morphologies and colors similar to those described in the previous sample (SI-7B).

We have made 180 analysis preformed on 172 zircon grains, in which a total of 99 are concordant ages in the 90% confidence interval (SI-7B). As in the previous case, there are no Mesoproterozoic ages, but just 1% of the ages are Archean and 2% ages are Palaeoproterozoic. 14% are Neoproterozoic (1,1% Tonian, 1,1% Cryogenian and 11,4% Ediacaran) with the Palaeozoic zircons represent a total of 83% of the obtained ages. In these, 73% are Cambrian (17% Terraneuvian, 11% Series 2, 16% Miaolingian and 29% Furongian) and 11% are Ordovician (10% are Lower and 1% is Middle Ordovician).

The youngest zircon age has 468±13 Ma and the magmatic age (youngest population) of this sample is 475±5 Ma (5 ages, 95% concordant). The second and third youngest age groups are 487 3 Ma (7 ages) and 495 2 Ma (19 ages) (SI-7B).

EC-PO-337

This sample belongs to an hectometric block of epiclastic/volcanic acidic tuff within the Upper Member of the Infraquartzite Fm. (equilavent to Rabano Fm. in Alcañices) (Meireles, et al., 1999b).

Zircon grain habits range from prismatic tabular with banded zoning, bipyramidal prismatic and stubby with oscillatory zoning and commonly cores (SI-8A). Zircon colours spam from colorless to pink.

We have made 148 analysis on 119 zircon grains, including rims and cores. A total of 72 concordant ages (90% interval) were obtained (SI-8A). No Archean nor Paleoproterozoic ages were found. 4% of the ages are Mesoproterozoic, 17% are Neoproterozoic (1% Tonian, 3% Cryogenian and 13% Ediacaran) and 79% are Palaeozoic, including 54% Cambrian (7% Terraneuvian, 17% are Series 2, 10% Miaolingian and 21% Furongian), 22% Ordovician (18% are Lower, 3% are Middle and 1% is Upper Ordovician) and 3% Silurian.

The youngest zircon age has 435±40 Ma and the magmatic age (youngest population) of this sample is 442±22 Ma (3 ages, 95% concordant). The second, third and fourth youngest ages are 471±14 Ma (8 ages), 484±16 Ma (6 ages) and 494±13 Ma (9 ages) respectively (SI-8A).

EC-PO-419

This sample was collected in the Supraquartzite Fm. (Meireles et al., 1999b) in the northern limb of the Verín-Alcañices Synform, to NW of the Bragança Complex. It is a medium-grained quartz-eyed acid tuff, with pervasive foliation, in a compromising structural and stratigraphic position within the LPa. New field data points for an olistolithic nature of this rock, laying within a synorogenic polygenic mélange.

grains have shapes spamming from long prismatic with banded growth, bypiramidal prismatic to stubby with oscillatory zoning and frequently including a core (SI-8B). Colors range from colorless to pink.

We have made 70 analysis preformed on 75 zircon grains including rims and cores (SI-8B). Zircon We have obtained 45 concordant ages (90% interval). The Archean, the Meso and the Paleoproterozoic are not represented in this sample. 9% of the concordant ages are Neoproterozoic (4,4% are Cryogenian and 4,4% are Ediacaran), 79% are Palaeozoic including 24% Cambrian (4% Series 2, 11% Miaolingian and 9% Furongian), 62% Ordovician (29% Lower, 20% Middle and 11% Upper Ordovician) and 4% Silurian.

The youngest zircon age has 439±8 Ma and the magmatic age (youngest population) of this sample is 442±12 Ma (5 ages, 95% concordant). The second and third youngest populations are respectively 465±11 Ma (6 ages) and 474±13 Ma (5 ages) (SI-8B).

PET-1

This sample is a reddish rhyolite olistolith within the Supraquartzitic/Rábano Fm. (Meireles et al., 1999c; González Clavijo, 2006) flysch sequence.

Zircon grains have shapes spamming from long prismatic with banded growth, bypiramidal prismatic to stubby with oscillatory zoning and frequently including a core. Colors range from colorless to pink. Only magmatic zircons and rims were analyzed to obtain a magmatic age of this sample (SI-9A).

We have obtained 50 concordant ages (90% interval) out of 60 analysis preformed on 60 magmatic zircon grains (SI-9A). The youngest zircon age is 483±8 Ma and the magmatic age (youngest population) of this sample is 493±1 Ma Ma (28 ages, 95% concordant). The second youngest population is 552±4 Ma (4 ages) (SI-9A).

RAB-1

This sample is a porphyritic whitish rhyolitic tuff, belonging to a one-meter-thick olistolith, within fine grained grey sandstones and dark-grey slates of the synorogenic Manzanal del Barco Fm (González Clavijo, 2006; Martínez Catalán et al., 2016).

Zircon grains have shapes spamming from long prismatic with banded growth, bypiramidal prismatic with oscillatory zoning, sometimes including a core (SI-9B). Colors range from colorless to pink. Only magmatic zircons and rims were analyzed to get a magmatic age of this sample, although some inherited ages were found.

We have obtained 39 concordant ages (90% interval) preformed on 39 magmatic zircon grains. The youngest zircon age is 472±5 Ma and the magmatic age (youngest population) of this sample is 473.8.7±3.2 Ma (5 ages, 95% concordant). The second and third youngest populations are 479.1±2.8 Ma (5 ages) and 496.1±2.8 Ma (6 ages), respectively. Other scattered older ages spam from 550 Ma and 670 Ma (SI-9B).

3. MDS analysis of the NW Iberia Synorogenic basins

We used Multidimentional Scaling (MDS) (ISOPLOT-R by Vermeech, 2018) to compare the new and the already published data on the zircon age populations of the NW Iberia synorogenic basins, with potential sources within the Iberian Massif terranes. This approach has proven successful in the improvement of paleogeographic reconstruction models in the southwestern branch of the Iberian Variscan belt (Pereira et al., 2020a; 2020b) where the zircon age fingerprinting of the possible sedimentary sources and the Devono-Carboniferous synorogenic marine basins have shown these basins were fed by sediments from both continental margins, Laurussia and Gondwana.

In this study, the zircon age data used to represent the possible source areas of the synorogenic marine basins of NW Iberia is a data selection of published U-Pb ages of detrital zircons of pre-Upper Devonian siliciclastic rocks of the NW Iberian Autochthon and Allochthonous Complexes. The geochronological data gathered by Puetz et al. (2018) and Stephan et al. (2018) were used in combination to have the best quantity and quality of data. This dataset is used to fingerprint the source areas using MDS and works as a tool to plot the our team age data collection of the synorogenic siliciclastic rock samples, which was expanded in this work from 13 to a total of 24 samples. We have also included new zircon age data on volcanic rocks from the UPa (2 samples) and from large olistoliths in the LPa (4 samples) to compare their age spectra with the detrital zircon samples, thus tracing the source areas for some of the large olistoliths and the flysch sequence.

The detrital zircon age records from the Ediacaran to Devonian siliciclastic strata in the possible source areas were selected and organized in an Excel spreadsheet (Supplementary Table 18), according to their age, structural and stratigraphic characteristics. Because the databases provided by Puetz et al. (2018) and Stephan et al. (2018) engage isotopic analysis from the late 90s to 2018, we have performed a quality test to the U-Pb isotopic data in each sample, recalculating all the zircon ages following the procedure used in our samples. In this way, it was possible to select those samples that show the biggest amount of concordant ages (90% concordance interval) that can be used for statistical correlation purposes. We have found that much of the "old" data cannot be used either by the large amount of unconcordant ages and/or by the reduced number of ages in a sample. The fingerprinting of the zircon age populations in NW Iberia source areas using MDS, used only data falling in the 90% concordance interval and samples with over 25 concordant ages.

The age data of the possible source areas was selected according a conceptual paleogeographic model for the Upper Devonian-lower Carboniferous (as provided in Dias da Silva et al., 2015 and Martínez Catalán et al., 2016). In this way, Source A represents the peripheral bulge developed in the Autochton (CIZ, WALZ-CZ and OMZ) and the UPa slice as the most continental section of North-central Gondwana margin; Source B reflects the NW Iberian Allochthon (GTMZ), a rootless accretionary complex emplaced onto the autochthon in the Devonian-Carboniferous using the Parautochthon as the lower tectonic sheet. We highlight that although the UPa is in Source A, it could have been at either side of the synorogenic basin margins, as belonging to the peripheral bulge in early Variscan times (Late Devonian), or to the GTMZ basal tectonic sheet in the early Carboniferous, as the tectonic front moved towards inland Gondwana.

3.1. Source A – Autochton (CIZ, WALZ-CZ and OMZ) and Upper Parautochthon

In this source, samples were separated by stratigraphic age, considering that the general stratigraphy of the Autochthon and UPa was not substantially shuffled by major Variscan thrust zones as in the case of the GTMZ allochthons. We assumed a simplistic model where the stratigraphy of the autochthon is unevenly exposed in the lower Carboniferous due to the action of the first Variscan folding event in the Autochthon and UPa. This model is used as a filter to define the possible sedimentary sources of the synorogenic basins in NW Iberia. However, it is important to emphasize that there are still many stratigraphic units without geochronological information in both the Autochthon and UPa.

Detrital zircon age populations of all selected samples from Source A were plotted individually, zone by zone, in MDS diagrams (Figs. SF1.1 to SF1.4). The result of this analysis shows a large dispersion, meaning that the sediment sources along the north-central margin of Gondwana in Iberia changed during time and space throughout the Ediacaran-Paleozoic period.

To evaluate the variability of the sources over time in each autochthonous zone, the age populations of detrital zircon from samples of Source A were combined according to their stratigraphic ages, leading to the definition of 8 categories: A.1.- Ediacaran (CIZ, WALZ-CZ and OMZ); A.2.- Early Cambrian (UP, CIZ, WALZ-CZ and OMZ); A.3.- Upper Cambrian (CIZ); A.4.- Lower Ordovician (UP, CIZ and WALZ-CZ); A.5.- Upper Ordovician (CIZ); A.6.- Silurian (CIZ and WALZ-CZ); A.7.- Lower Devonian (CIZ and WALZ-CZ); A.8.- Upper Devonian (CIZ and WALZ-CZ).

The MDS diagrams (Figs. SF1.1 to SF1.4) show the population age variations within the different Source A categories. Although we have produced the Lower Devonian, Silurian and Upper Ordovician age clusters of the CIZ, they will not be used in the final MDS diagram because they have a reduced number of samples and/or have few concordant zircon ages and so they do not comply with the above requests.

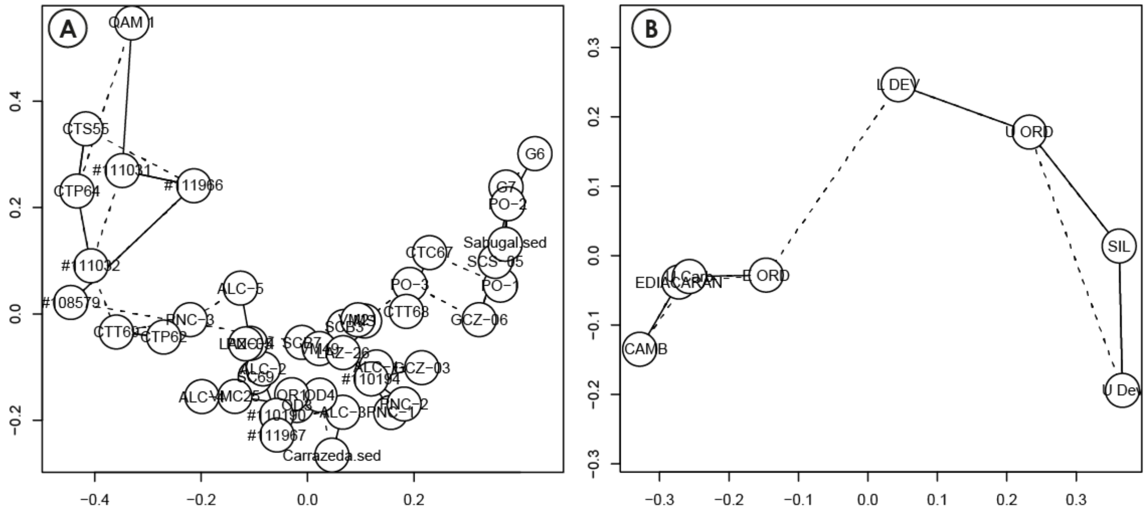

Figure SF1.1 – MDS diagrams for the CIZ. Sample codes (A) and reference populations (B) are given in Supplementary Table 18.

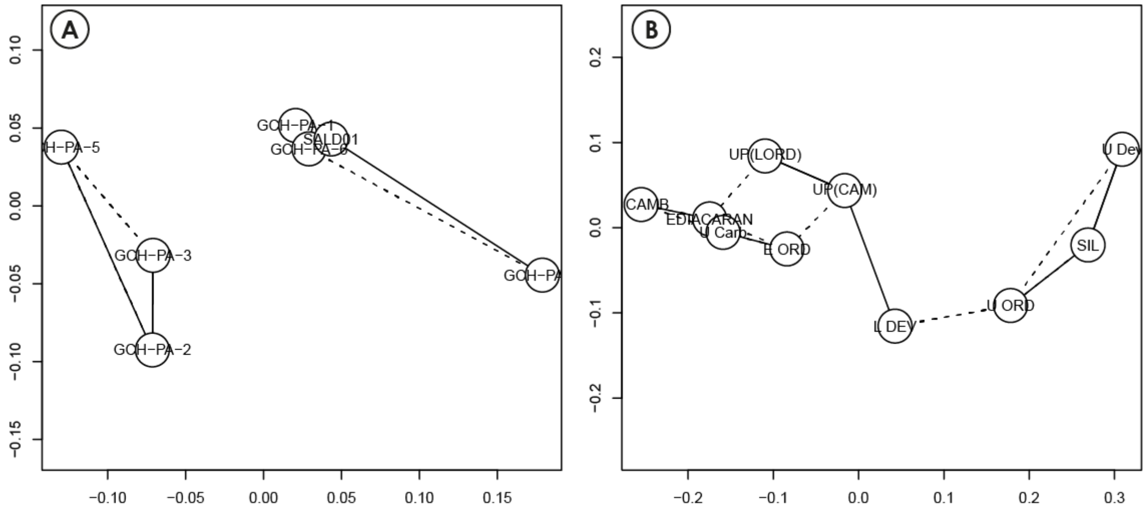

Figure SF1.2 – MDS diagrams for the UPa (A) and CIZ, including the UPa reference groups (B)

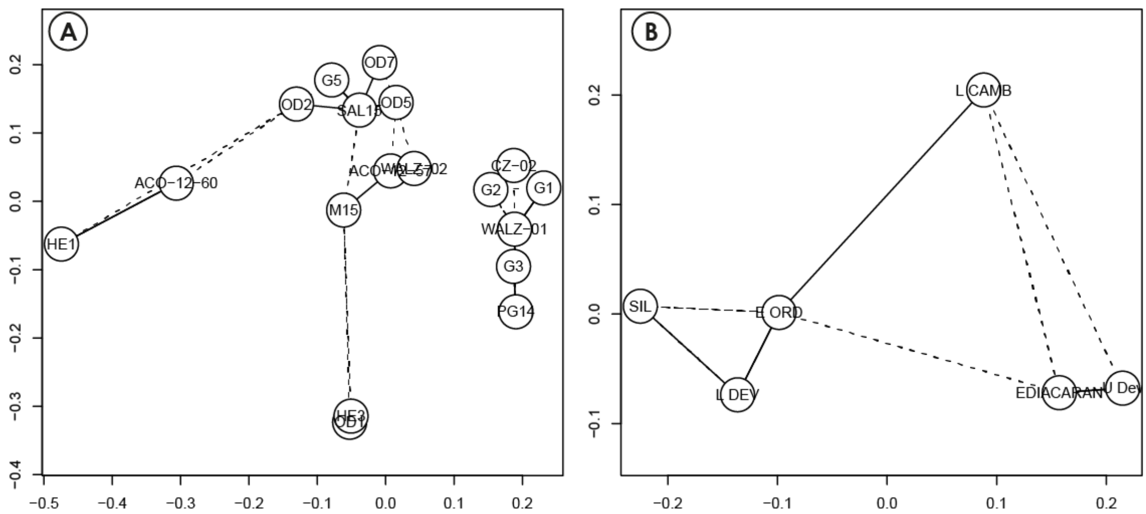

Figure SF1.3 – MDS diagrams for the WALZ-CZ. Samples (A) used to make the reference populations plotted on the MDS diagram in B.

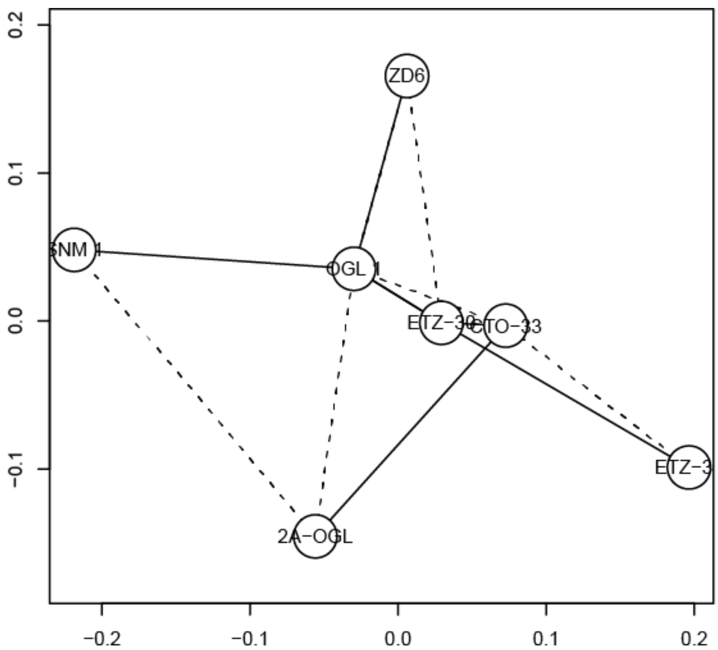

Figure SF1.4 – MDS diagram for the Edicarian-early Cambrian siliciclastic rocks of the OMZ. These points collapse in one population, representative of the N-Gondwana margin, acording Pereira et al. (2020a, 2020b)

3.2. Source B – NW Iberian Allochthon (GTMZ)

In Source B, the GTMZ Allochthonous complexes were separated according to their tectono-metamorphic unit/domain with all samples ranging from Ediacaran to Lower Ordovician stratigraphic ages and belonging to the Galician allochthonous units (Malpica-Tuy, Ordenes and

Cabo Ortegal complexes). The main characteristics of the GTMZ allochthonous complexes is the tectonic shuffling of differently metamorphosed and deformed rocks due to Eo-Variscan (Devonian) subduction, obduction, out-of-sequence thrusting and extensional detachments. Together they define a rootless accretionary prism set tectonically onto the UPa. This tectonic framework favors a model where the GTMZ units were exposed to erosion during a long time period and it is expected that it was the main contributor of detrital zircons of the Devono-Carboniferous foreland basins preserved today in NW Iberia. In this way, Source B samples (Figs. SF1.5) were separated into four categories, according to their structural position in the nappe pile and/or their metamorphic grade (as in the case of the Upper Allochthon). The four categories are: B1 - Upper Allochthon (M-HT/HP); B2 - Upper Allochthon (LT/MP); B3 - Middle Allochthon; B4 - Lower Allochthon.

In MSD diagram the age clusters of allochthonous units of the GTMZ show a more refined differentiation in two LT/MP Upper Allochthon (B2a and B2b) and two Lower Allochthon (B4a and B4b) age populations (Fig. SF1.5; UA(LT)1 and UA(LT)2; LA1 and LA2, respectively). Although we have plotted the detrital zircon age populations of the Middle Allochthon (MA), we will not use this data for the provenance study due to the reduced number of samples and concordant ages. The relationship between Source A and Source B categories is presented in the MDS diagram Fig. SF1.6.

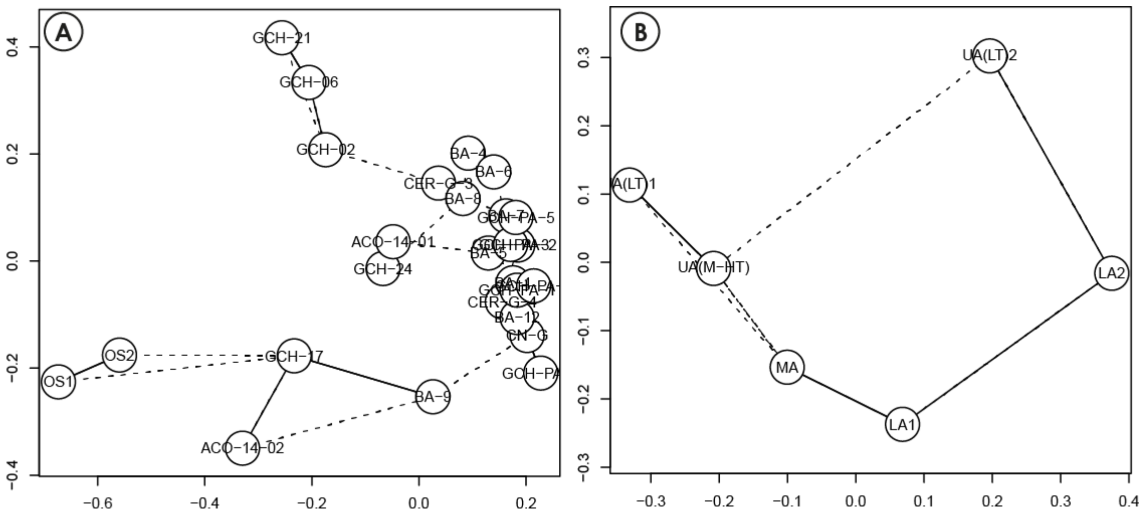

Figure SF1.5 – MDS diagrams for the GTMZ. On A the samples used to make the reference categories of Source B (B).

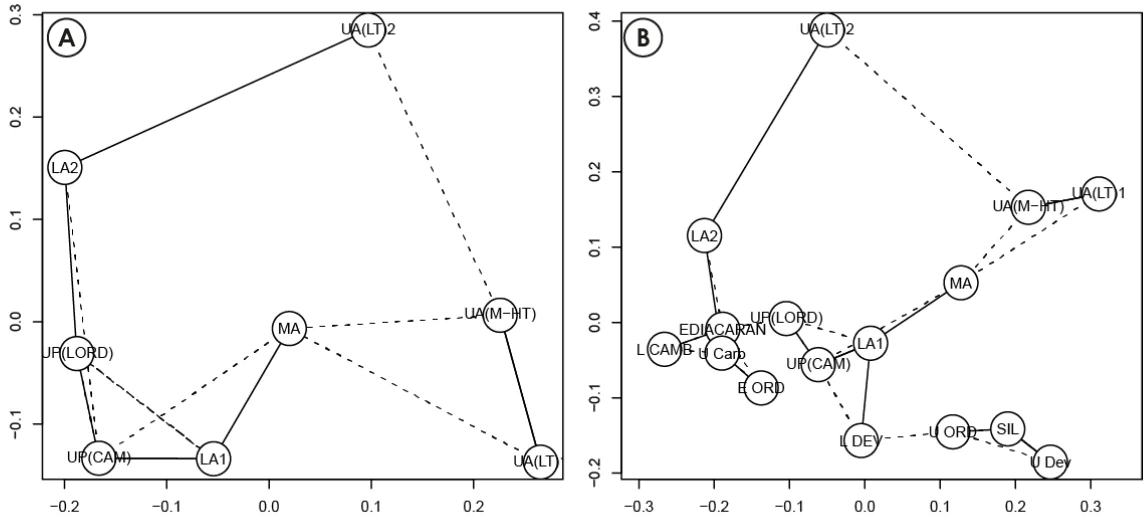

Figure SF1.6 – MDS diagrams for the GTMZ including the UPa (A) and both UPa and CIZ (B), showing the relationships between Source A and Source B categories in NW Iberia. Note the proximity of the LA1, LA2, UP (LORD) and UP (CAM) age clusters with the more Gondwanan sources within the CIZ and their distancing with the UA and MA clusters.

3.3. Synorogenic sediments VS potential sources

The diagram of Fig. SF1.6 is the base plot where we input the new synorogenic siliciclastic samples of the LPa and the new inherited and magmatic ages obtained in the Middle-Upper Ordovician/lower Silurian volcanic rocks embedded in the synorogenic sediments as large olistoliths or as igneous rocks in the upper stratigraphic units of the UPa. The field and microtextural aspects of these igneous rocks indicate that they belong to the same Middle-Upper Ordovician magmatic event, as reported in previous works in the UPa in the eastern rim of the Morais Complex (Dias da Silva et al., 2016).

Besides the new detrital zircon data presented in this work, we have added previously published ages given by Martínez Catalán et al. (2004) (samples SO-1 and SO-2), Martínez Catalán et al. (2008) (SO-4 to SO-7), Dias da Silva et al. (2015) (VC21, VC45 and VC57) and Martínez Catalán et al. (2016) (SO-8 to SO-14) (Supplementary Table 19).

Observing the MDS diagram with the reference populations and the synorogenic sediments of NW Iberia we have obtained 2 major age clusters (Fig. SF1.7):

Cluster 1 (Fig. 11, manuscript) – “Upper Parautochthon Middle Ordovician-Silurian volcanism” type characterized by synorogenic sediments with high abundance of Cambrian and Ordovician zircons, namely a high concentration of Middle to Upper Ordovician ages and minor amounts of Silurian and Devonian ages: samples VC-21ZIR (youngest zircon: 323 Ma; maximum depositional age: 393 Ma), SO-6 (YZ: 378 Ma), CR-ZR-01 (YZ: 439 Ma) and MEI-ZR-01 (YZ: 443 Ma). Another highlighting aspect is the lack of “old” zircon ages (Archean to Ediacaran). These samples show direct relation with the analyzed volcanic rocks of Middle Ordovician-lower Silurian magmatic and inherited ages from the UPa (P-381 and P-385) and form LPa olistoliths (EC-PO-337 and EC-PO-419), as they show the same age distribution as the related synorogenic sediments. This means a direct relationship between the source rocks (UPa volcanics of the Peso Fm.; Dias da Silva et al., 2016) and some segments of the synorogenic basin (Gimonde, Travanca and Meirinhos Fms.);

Cluster 2 (Fig. 12, manuscript) – Samples with a wide variety of sub-clusters (groups 1 to 7), including the Autochthon, the UPa (Source A) and the Allochthonous complexes (Source B). The defined groups show direct relations with the most probable sources (A and/or B), but they also represent different grades of sediment mixing and recycling. These can be defined as follows.

Group 1 – Upper Allochthon (M-HT+LT1): This cluster includes the Upper Allochthon’s reference groups UA (M-HT) and UA (LT)1 (source categories B1 and B2a) and the San Vitero Fm. sample SO-13 (YZ: 350 Ma). They are characterized by age distributions with a large Cambro-Ordovician age population and few Cadomian ages, scattered Mesoproterozoic ages and Paleoproterozoic (1.8-2.25 Ga) and Neoarchean (2.5-2.8 Ga) age populations. The larger proportion of Mesoproterozoic ages in the sample SO-13 can evidence mixing with other “more Gondwanan” type sources (e.g. Group 5, Source A).

Group 2 – undefined sources: This sample cluster include the already published samples from the Vila Chã Fm. (VC-57ZIR, youngest zircon 532 Ma) and two siliciclastic olistoliths in the Gimonde Fm. (SO-7 and SO-8, youngest zircon with 552 and 553 Ma, respectively), showing no apparent connection with any reference samples. The age distribution that characterize this group present a large Neoproterozoic (Cadomian) population with 2 or more age spikes, a Mesoproterozoic gap and 2 age populations, one in the Paleoproterozoic (1.7 to 2.3 Ga) and the other in the Neoarchean (2.5 to 2.8 Ga), with scattered older ages down to the Paleoarchean (<3.5 Ga). The presence of Mesoproterozoic ages in VC-57ZIR also shows some proximity to sources belonging to groups 4 and 6. There are no zircons younger than the early Cambrian in this sample group.

Group 3 – undefined sources: It includes the reference samples for the Silurian strata of the WALZ+CZ and the Upper Devonian record of the CIZ. They appear to have no relationship with the synorogenic sediments of NW Iberia. This group is characterized by an age distribution with few Cambro-Ordovician ages, abundant Neoproterozoic and Mesoproterozoic populations, and in lesser extent Paleoproterozoic and Neoarchean age populations.

Group 4 – Cambrian Upper Parautochthon and Lower Allochthon: This group includes the reference samples of the early Cambrian rocks of the UPa, the Source B4a (LA1) of the Lower Allochthon, the Early Ordovician and Lower Devonian clusters of the WALZ+CZ and the samples from the Upper Slates (AD-PO-49, YZ: 467 Ma), Vila Chã (VC-45ZIR, deformed early Cambrian olistolith in the LPa, YZ: 527 Ma) and Curros Fms. (AD-PO-48B, YZ: 355 Ma). Although the reference samples for Early Ordovician and Lower Devonian of the WALZ+CZ are within this group, their high amount of Mesoproterozoic ages led us to consider the most probable sources to be the Lower Allochthon (LA1) and the Cambrian strata from the Upper Parautochthon.

Group 5 – "Gondwana type": It includes most of the CIZ reference populations (Ediacaran, lower Cambrian, and Early Ordovician), the Ediacaran and Upper Devonian of the WALZ+CZ, the Early Ordovician of the UP, the OMZ and the Source B4b (LA2) of the Lower Allochthon. Note the presence of Cambro-Ordovician and Variscan (390-330 Ma) ages in the reference Ediacaran population of the CIZ (Fig. 12, in the manuscript), which is reflecting the presence of HT-LP events suffered by these siliciclastic rocks (Orejana et al., 2015). The fingerprint of this group is the typical Gondwana zircon age population with some small peaks of Archean and Paleoproterozoic ages (1.7 to 2.25 Ga with scattered older ages), few or no Mesoproterozoic, locally few Tonian (700-1000 Ma), abundant Ediacaran (Cadomian) and Lower Cambrian (520 to 700 Ma) and late Cambrian- Early Ordovician ages (500-470 Ma). The synorogenic samples associated to this group usually also show zircon ages in the range of 470-440 Ma, evidencing the contribution of Middle/Late Ordovician-lower Silurian magmatic sources, excluding sample SO-11 ("Nuez quartzite", YZ: 551 Ma), which is an Armorican Quartzite olistolith, bearing Lower Ordovician *skolithus*. Because the reduced number of ages in this sample, we have decided not to use it in this correlation. All the other synorogenic samples in this group have similar age distributions, namely the samples collected in Rio de Onor (probable UPa; SO-10, YZ: 458 Ma), Upper Schists (MIR-41, YZ:369 Ma; and AD-PO-55, YZ: 444 Ma) and Culminating Slates and Greywackes Fms. (AD-PO-57, YZ: 368 Ma).

Group 6 – Cambrian WALZ+CZ: This group includes the samples that are somewhat related to the reference population for the Lower Cambrian of the WALZ+CZ. Maybe there is a relation with the Middle Allochthon (MA), but because of the lack of representativity of the detrital zircon ages in the MA we decided not to include it in our study. However, we believe that the MA must be also a contributor for detrital zircons in the synorogenic basin of NW Iberia, but this has to be confirmed using inherited ages of the magmatic rocks and with further geochronology studies in the detrital and magmatic rocks of this possible source. The fingerprint of this age group is an age population with Lower Ordovician-Neoproterozoic ages, with maxima around 540 Ma and 480 Ma, a Mesoproterozoic gap with scattered ages, a Paleoproterozoic (1.8/1.9-2.3 Ga) population with maxima around 2.0 Ga and scattered ages between 2.3 and 3.1 Ga. The samples that fit in this group belong to the Rábano (SO-9, youngest zircon=362±3 Ma), San Vitero (SO-5, youngest zircon age =355±4 Ma) and Gimonde Fms. (EC-PO-293, youngest zircon age=426±44 Ma).

Group 7 – Upper Allochthon LT2/Mix with "Gondwana type": It includes all the samples that are directly or indirectly (recycling?) related to the UA(LT)2 reference group of the GTMZ. The age population distribution of this reference group is characterized by the presence of 480-750 Ma ages with maxima at 520-540 Ma, a Mesoproterozoic gap or scattered ages, Paleoproterozoic ages (1.8 to 2.35 Ga) with maxima at 2.1 Ga and several small peaks between 2.4 and 3.0 Ga. The synorogenic samples that are most directly related to this reference population are those from the CIZ synorogenic San Clodio Fm. (SO-1 and SO-2, YZ: 325 Ma). The remaining synorogenic samples appear to trend into Group 5, exposing the increasing contribution of Gondwana type sources, mixed with the UA (LT)2 source. For instance, all the Gimonde Fm. samples (AD-PO-66, GIM-ZR-01 and SO-12, YZ: 431 Ma, YZ: 327 Ma and YZ: 340 Ma) are very similar and are also close to the sample SO-9 of Group 6 and evidence about the same amount of source mixing or recycling of different zircon populations within the basin. Samples SO-4 (YZ: 380±4 Ma) from the San Vitero Fm. and PICON-2 (YZ: 350 Ma) from the Rio Baio thrust sheet in Cabo Ortegal Complex are also very similar to the previous. The sample from the Almendra Fm. (SO-14, YZ: 333 Ma) shows intermediate characteristics between the San Clodio Fm. and the other samples and may be reflecting recycling of sediments at both margins of the synorogenic basin.

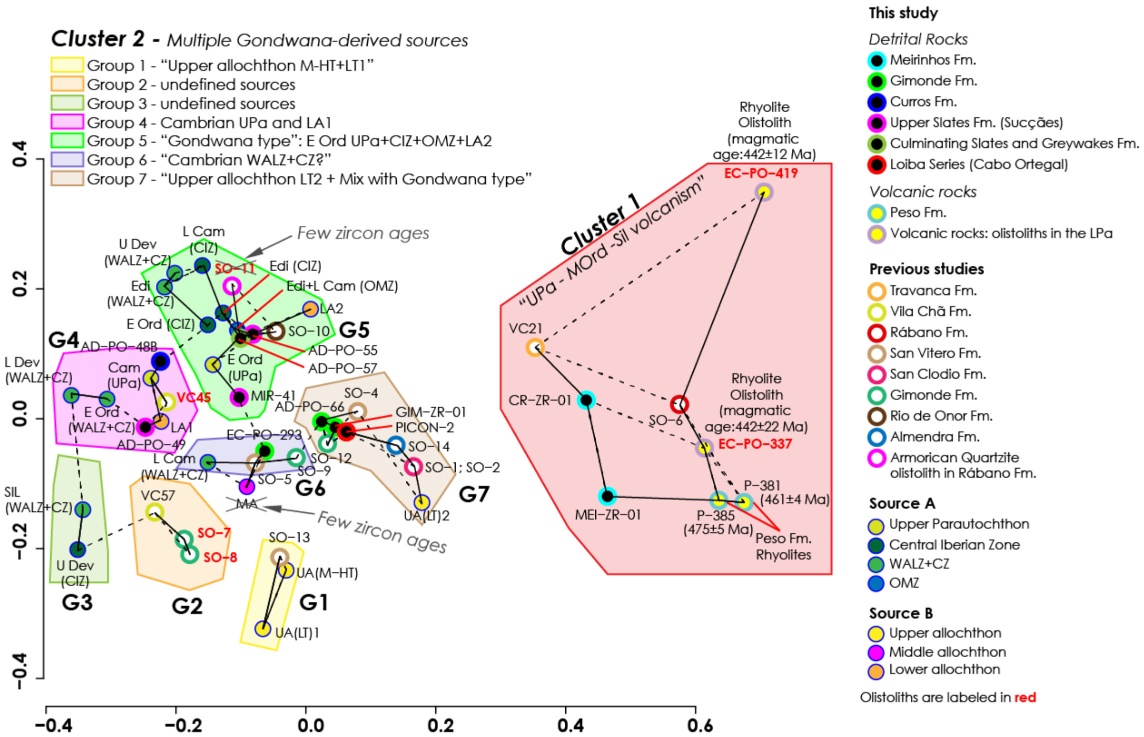

Figure SF1.7 – Final MDS diagram for the studied samples and the reference populations in the autochthon and allochthon. Data to construct this plot is provided in the Supplementary tables 18 and 19.

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