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Diagenesis, provenance and tectonic setting of siliciclastic rocks. A case study from Upper Devonian of the Iberian Chain (Tabuenca, Spain)

Diagénesis, análisis de procedencia y ambiente tectónico de rocas siliciclásticas. Un caso de estudio en el Devónico Superior de la Cordillera Ibérica (Tabuenca, España)

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ABSTRACT

This paper describes the petrography and infers the provenance of siliciclastic rocks from the Upper Devonian of the Iberian Chains (Tabuenca, NE Spain), and outlines the tectonic setting associated with the Ebro Massif. These Devonian deposits are constituted by four different siliciclastic units: the Rodanas, Bolloncillos, Hoya and Huechaseca Formations. The provenance and diagenesis of over 400 sedimentary rocks samples are studied with a combination of petrographic polarizing microscope, scanning electron microscopy, atomic absorption spectroscopy, X-ray fluorescence and X-ray diffraction. In this sense, AAS and XRF analysis were used to determine the content of Ca, Mg, Fe, Mn, Na, K and Sr, among others; and XRD analysis was used to determine the clay´s crystalline phases. These rocks experienced intense compaction and quartz cementation processes after deposition. No primary porosity remains nowadays and secondary porosity is rare. The formation of these siliciclastic rocks occurred mainly under subtropical climatic conditions, given the paleogeographical position of the current Iberian landmass during the Devonian.

Keywords: Devonian; Diagenesis; Paleoclimate; Siliciclastic rocks; Provenance; Iberian Chains.

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RESUMEN

Este trabajo describe la petrografía e infiere la procedencia de los materiales que constituyen las rocas siliciclásticas del Devónico Superior de la Cordillera Ibérica (Tabuenca, España), y esboza el ambiente tectónico asociado con el Macizo del Ebro. El Devónico superior de Tabuenca está dividido en cuatro formaciones: Rodanas, Bolloncillos, Hoya y Huechaseca. Se ha llevado a cabo el análisis de la procedencia y el estudio de los procesos diagenéticos de mas de 400 rocas sedimentarias combinando diversas técnicas: microscopía petrográfica clásica, microscopía electrónica, espectrometría de absorción atómica (AAS) , fluorescencia mediante rayos X (XRF) y difracción de rayos X (XRD). En este sentido, los análisis mediante AAS y XRF se usaron para determinar el contenido de Ca, Mg, Fe, Mn, Na, K y Sr, entre otros elementos mayores; y el análisis mediante XRD fue utilizado, fundamentalmente, para caracterizar las distintas fases cristalinas de los minerales arcillosos. Estas rocas han experimentado unos intensos procesos de compactación y cementación silícea después de su depósito. No se ha detectado la existencia de porosidad primaria y la secundaria es escasa. La formación de estas rocas siliciclásticas ha tenido lugar de manera generalizada bajo condiciones climáticas subtropicales, en consonancia con su posición paleogeográfica durante el Devónco.

Palabras clave: Devónico; Diagénesis; Paleoclima; Rocas Siliciclásticas; Análisis de procedencia; Cordillera Ibérica.

Introduction

The rocks of the Rodanas, Bolloncillos, Hoya and Huechaseca Formations were deposited during the upper Devonian in the Iberian Peninsula, in a foreland basin setting adjacent to the Ebro Massif (Carls, 1983; Oliveira *et al.*, 1986).

Provenance studies of siliciclastic rocks have been studied in sedimentary basins, such as foreland basins (i.e., DeCelles & Hertel, 1989; Critelli & Le Pera, 1994; White *et al.*, 2002; Critelli *et al.*, 2003; Garzanti *et al.*, 2003) or rift basins (i.e., Garzanti *et al.*, 2001, 2003; Arribas *et al.*, 2003, 2007, 2014; González-Acebrón *et al.*, 2007, 2010), or in areas of relative tectonic quiescence (i.e., Marenssi *et al.*, 2002; Amorosi & Zuffa, 2011; Amorosi *et al.*, 2012; González-Acebrón *et al.*, 2017).

For that, in this work, a detailed petrographic study of Upper Devonian rocks from the Iberian Chain and the diagenetic history of these quartz-rich sedimentary rocks is presented. So, it makes inferences on the provenance, and outlines the tectonic setting associated with the Ebro Massif. The provenance and diagenesis of the Upper Devonian rocks in the Iberian Chains are studied by means of a combination of optical microscopy (Basu *et al.*, 1975; Young, 1976; Tortosa *et al.*, 1991), scanning electron microscopy (Kwon & Boggs, 2002), atomic absorption spectroscopy, X-ray fluorescence and X-ray diffraction (OM/SEM/AAS/XRF/XRD). In this sense, AAS and XRF analysis were used to determine the content of Ca, Mg, Fe, Mn, Na, K and Sr (Torrijo, 2003); and XRD analysis was used to determine the clay´s crystalline phases.

Geological setting

The Devonian materials of the Iberian Chains have been included into the Herrera Unit (Carls, 1983; Gozalo & Liñán, 1988; Fig. 1.A).The outcrops of Upper Devonian rocks are located in two areas: The Montalbán Anticline from the NE Iberian Chains (Carls & Lages, 1983; Carls *et al.*, 2004) and the Tabuenca and Rodanas Area from the NW Iberian Chains (Gozalo, 1986, 1994; Bauluz, 1997; Bauluz *et al.*, 2000; Torrijo *et al.*, 2000, 2001; Gozalo *et al.*, 2001, 2017; García-Alcalde *et al.*, 2002; Torrijo, 2003; Carls *et al.*, 2004). The Upper Devonian rocks of the Tabuenca and Rodanas Area (Fig. 1.B) are composed of 1300 m of mostly siliciclastic materials with up to 30 m interbedded carbonatic rocks (Gozalo, 1986, 1994; Fig. 2). The age of these rocks span from the Early Frasnian to the Late Fammenian (Gozalo, 1986, 1994; Montesinos & Gozalo, 1987; Montesinos *et al.*, 1990; Carls & Valenzuela, 2002; Valenzuela *et al.*, 2002; Dojen *et al.*, 2004), and they are subdivided into four lithologic units by Gozalo (1994; Fig. 2) named, in stratigraphical order, as Rodanas Formation, Bolloncillos Formation, Hoya Formation and Huechaseca Formation.

The Rodanas Fm. comprises thin-to medium-bedded, light brown, quartzose sandstones with hardgrounds, limolites as well as lutites with carbonate concretions and narrow episodes of carbonates. The

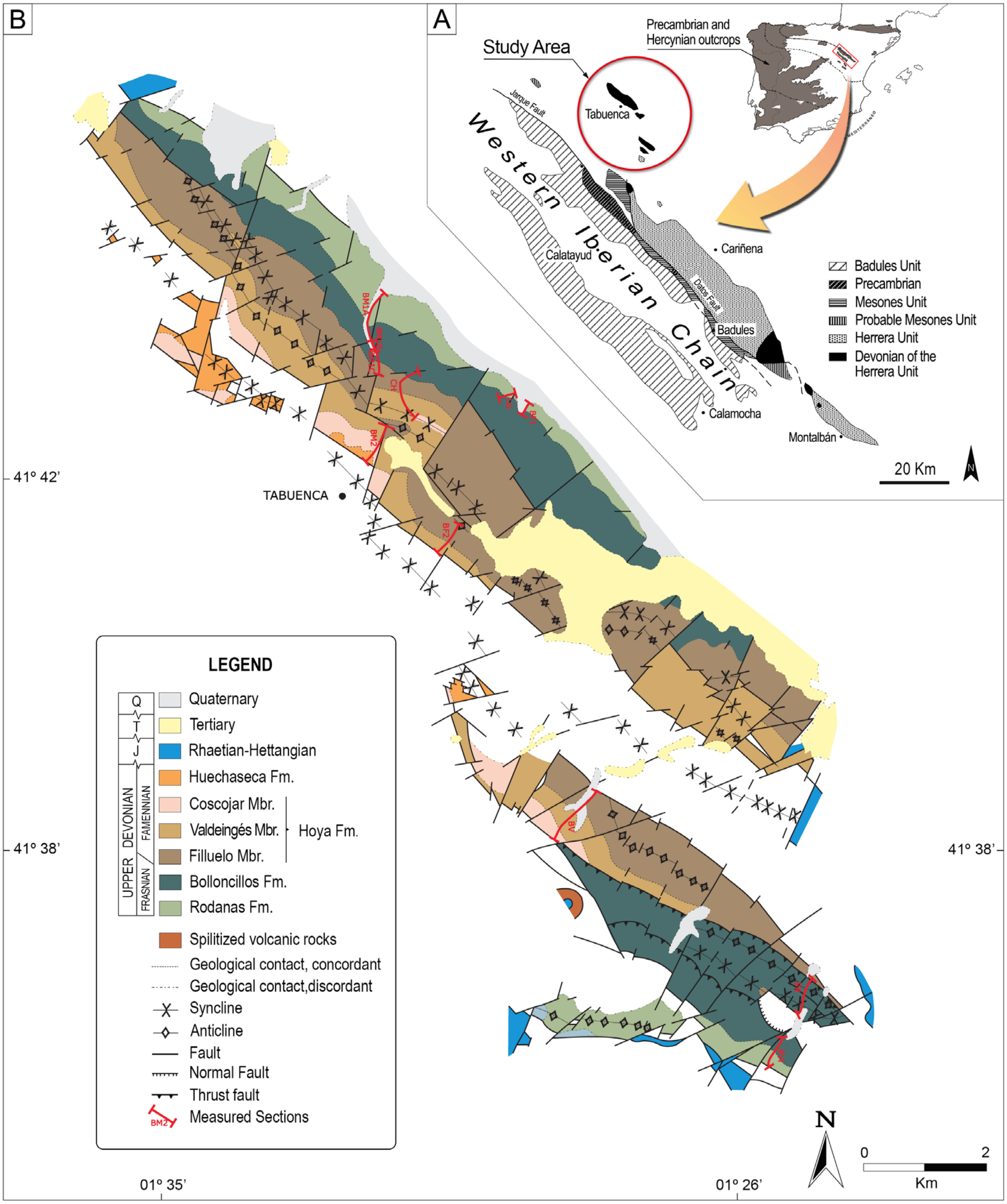

Figure 1.— A) Sketch map of the Iberian Peninsula showing the location of the Iberian Chain (based Gozalo and Liñán, 1988) with indication of Devonian outcrops (based on Dojen *et al.,* 2004) and location of the studied area. B) Simplified geologic map of the study area (Gozalo, 1990, unpublished) showing the local stratigraphy and the location of the studied sections (based on Gozalo, 1994 and Torrijo, 2003).

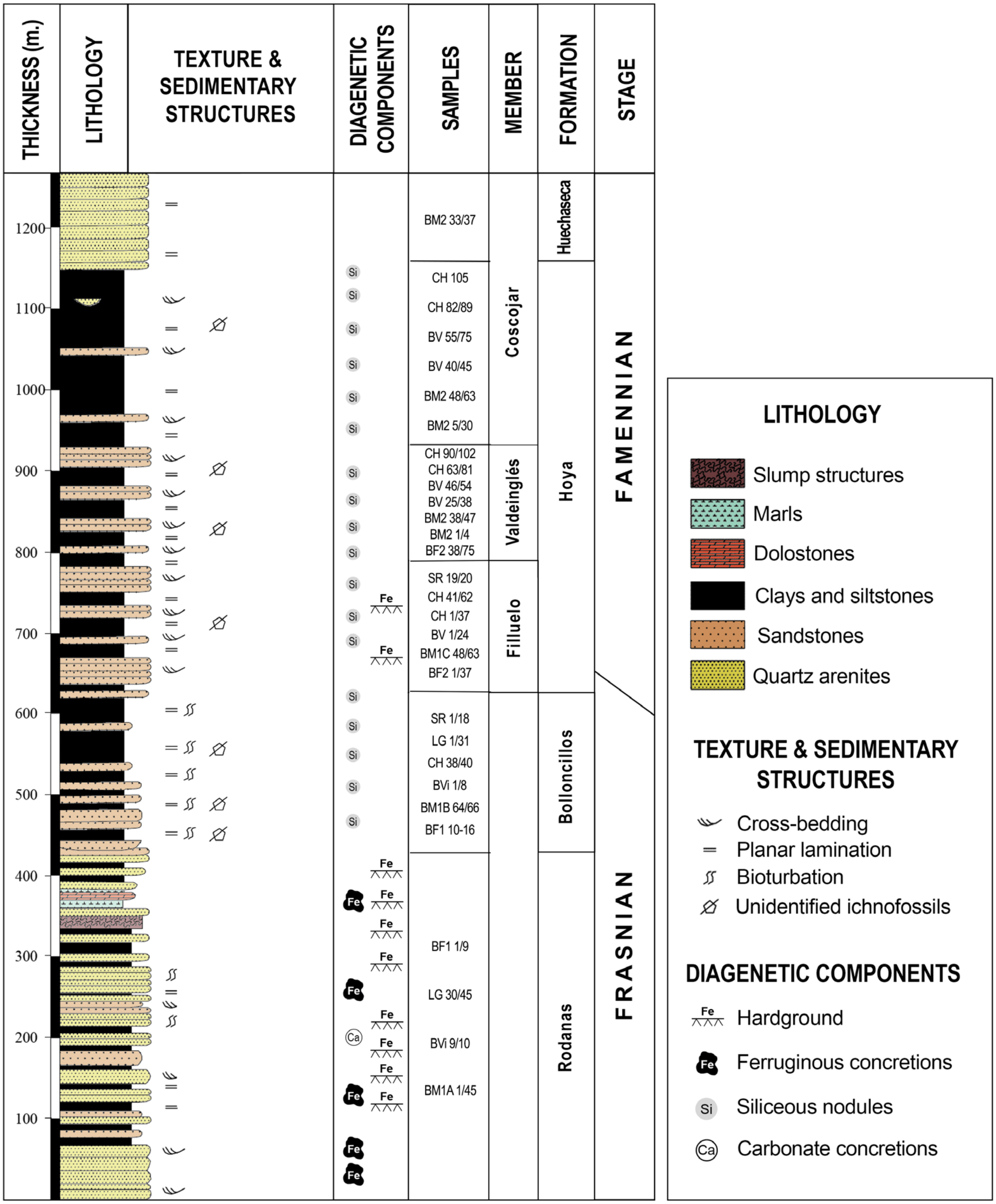

Figure 2.— Simplified stratigraphic section for the upper Devonian of the study area (Western Iberian Chain, NE Spain).

fossil content hints at a deep sublittoral, or event circalittoral environment; above it, very important siliciclastic sedimentation, indicative of shallower environments, was developed (Gozalo, 1994; Gozalo *et al.*, 2017). The Bolloncillos Fm. is an alternance of coarse to fine sandstone and lutites, the sandstone are predominant in the lower part and the lutites increasing in the upper part of the formation; the beginning of this formation represents a shallowing, that was followed by a deepening, but environments keep always within the sublittoral; the fossil and ichnofossil content of the upper part and the presence of organic matter would indicate a poorly oxygenated benthos, or even below the minimum oxygenation zone relate with the upper Kellwasser Event (Gozalo *et al.*, 2017). A transitional contact exists between this formation and the Bolloncillos and the Hoya formations, which consist of interbedded sandstones and lutites with siliceous nodules. Channel sand bodies-fills, ripple marks and through cross-bedding, typical sedimentary structures for shallow braided rivers (Miall, 1996) can be identified in the sandstone layers of the Hoya Fm. in the study area, possibly representing a fluvial/coastal environment. The transition to the overlying Huechaseca Fm. is again gradational. This last formation consists almost exclusively of quartz arenites. The sandstones of these four units are clastic sedimentary rocks with >95% of quartz as the main constituent of the framework grains (Torrijo, 2003).

The gradational transitions existing between these formations seem to reflect sea-level changes (Gozalo, 1994: Fig. 78) in a shallow epicontinental sea, which existed in this area at least from the Late Devonian (Torrijo, 2003), to the deposition of the overlying Carboniferous materials.

Materials and methods

General Concepts

Eleven representative stratigraphic sections were logged in the upper Devonian sedimentary sequence (Fig. 1) and over 620 samples were collected for analysis (Fig. 2). The position of samples in stratigrafic sections are provided in the electronic supplementary material accompanying this report (named Stratigraphical Profies). These samples were studied using combined OM/SEM/AAS/XRD analysis.

Quantitative Petrographic Analysis

Over 424 samples were analyzed to study their provenance and diagenesis, including those of the Rodanas Fm. (132 samples), the Bolloncillos Fm. (69 samples), the Hoya Fm. (218 samples) and the Huechaseca Fm. (5 samples). A Zeiss Jena 30-G0060 Jenapol polarizing microscope with 25x, 50x and 100x magnification was used for petrographic analysis. Standard point counting techniques were applied, using the integrated Gazzi-Zuffa point counting method (Zuffa, 1985), as recommended by Ingersool *et al.* (1984) and González-Acebrón (2017). 300 points were counted per thin section, distinguishing thirteen different components. This data was applied for sandstone classification and for estimating the amount of porosity lost by compaction, cementation and matrix/pseudomatrix, assuming 45% original porosity (Torrijo, 2003; Bernet *et al.*, 2007).

SEM Analysis

SEM analyses were carried out using ZEISS DSM 940A and JSM 6400 microscopes provided with with a Thechnosym 8200 MKII. Mineral identifications were evaluated by chemical spot analyses using an eXL-10 Link Analytical X-ray EDS analysis. Digital SEM images were taken at 150 to 200 μA beam current and 10-13 kV acceleration voltage at an average working distance of 20 mm. Images of 30x, 60x and 100x magnification were taken at 8000-pixel average. The samples studied are rock fragments included in resin.

Atomic Absorption Spectroscopy and X-Ray Fluorescence Analysis

The content of various chemical elements were determined with atomic absorption spectroscopy and X-ray fluorescence. AAS analysis was done with a Perkin-Elmer 3030 and the elements analysed and detection limits were the following: Ca (1000 ppm), Mg (20 ppm), Fe (100 ppm), Mn (6,5 ppm), Na (10 ppm) and K (50 ppm).

On the other hand, XRF was done in X-Assay Laboratory (XRAL) of Toronto and the elements analysed and detection limits were the following: SiO_2 (0,01%), Al_2O_3 (0,01%), CaO (0,01%), MgO (0,01%), Na_2O (0,01%), K_2O (0,01%), Fe_2O_3 (0,01%), MnO (0,01%), TiO_2 (0,01%), P_2O_5 (0,01%), Rb (2 ppm), Sr (2 ppm), Y (2 ppm), Zr (2 ppm), Nb (2 ppm) and Ba (20 ppm).

XRD Analysis

A Pananalytical Xpert with Detector XCeleretor and a Bruker D8 Advance with SOL-X were used for X-ray diffraction analysis. The determination of the clay's crystalline phases was made as recommended by Galán & Martín Vivaldi (1973).

Results

Petrological Characteristics

The arenites from the Rodanas Fm. are quartz arenites (Pettijohn *et al.*, 1987) composed of detrital quartz grains, mainly monocrystalline quartz, although some polycrystalline quartz grains with undulating extinction are observed. Silicate minerals (muscovite, tourmaline, zircon, potassium feldspar), apatite grains and lithic fragments are scarce, as well as no plagioclase grains were observed. Grain contacts in arenites are concavo-convex to sutured. The phyllosilicate matrix is very rare and is associated with the diagenetic alteration of the labile components (epimatrix) and the deformation of the ductile grains (mica) during the first phase of the compaction processes. The scarce interparticle porosity is sealed by siliceous cement of a syntaxial type and ferruginous cements. Limolites are a quartz wackes and lithic wackes (Pettijohn *et al.*, 1987), formed by phyllosilicate matrix of quartz grains, lithic fragments and micas (muscovite), in variable proportion.

In the Bolloncillos Fm., the arenites, similar to those of the Rodanas Fm., are also quartz wackes to lithic wackes (Pettijohn *et al.*, 1987), or quartz arenites to sublitharenites (Pettijohn *et al.*, 1987), although the latter in smaller proportion. The wackes are constituted, mostly, by quartz clasts and by lithic fragments and micas (muscovite) in minor proportion, and also, phyllosilicate and quartz matrix (15-20 %). The quartz arenites and sublitharenites are mainly by quartz clasts basically monocrystalline, although some polycrystalline quartz grains with undulating extinction are observed, and in minor proportion, sedimentary rock fragments, zircon, mica and apatite grains.

The Hoya Fm. has been subdivided into three members (Gozalo, 1994) that from base to up are Filluelo Mb., Valdeinglés Mb. and Coscojar Mb. The Filluelo Mb. is a detrital alternance constituted by arenites and lutites-limolites. The Valdeinglés Mb. consists of an arenite-lutite alternance. Its limit with the underlain Filluelo Mb. is based on the augment of lutitic material and in a colour change (white to green and brown colours). The Coscojar Mb. consists of lutites with narrow intecalations of arenites. Generally, the arenites are sublitharenites to lithic wackes (Pettijohn *et al.*, 1987) formed by quartz clasts, basically monocrystalline, and in minor proportion, lithic fragments, very altered potassium feldspar, tourmaline (partially silicified) and mica grains. Grain contacts in arenites are concave-convex to sutured, and the phyllosilicate and quartz matrix (2-16 %) is associated with the diagenetic alteration of the labile components (epimatrix) and the deformation of the ductile grains (mica) in the first phase of the compaction processes.

In Huechaseca Fm., the arenites are quartz arenites (Pettijohn *et al.*, 1987) formed by quartz clasts, mainly monocrystalline quartz, although some polycrystalline quartz grains with undulating extinction are observed. Silicate mineral (muscovite, tourmaline, zircon, apatite), and lithic fragments grains are scarce, as well as no plagioclase grains were observed; and the phyllosilicate matrix is very rare (<1 %). Grain contacts are concave-convex to sutured (Fig. 3).

Analyses done using combined SEM and OM on individual quartz grains showed that the majority of the sand grains in the studied samples were identified as of plutonic origin, thanks to the presence of spider-web microcracks within the grains (Sprunt & Nur, 1979; Berner *et al.*, 2007). Grains of potential volcanic affinity are almost exclusively restricted to the finest sand-sized fraction of the samples (Fig. 4).

Geochemical Characteristics

The most pronounced compositional differences between the analysed samples are in the in SiO_2 and

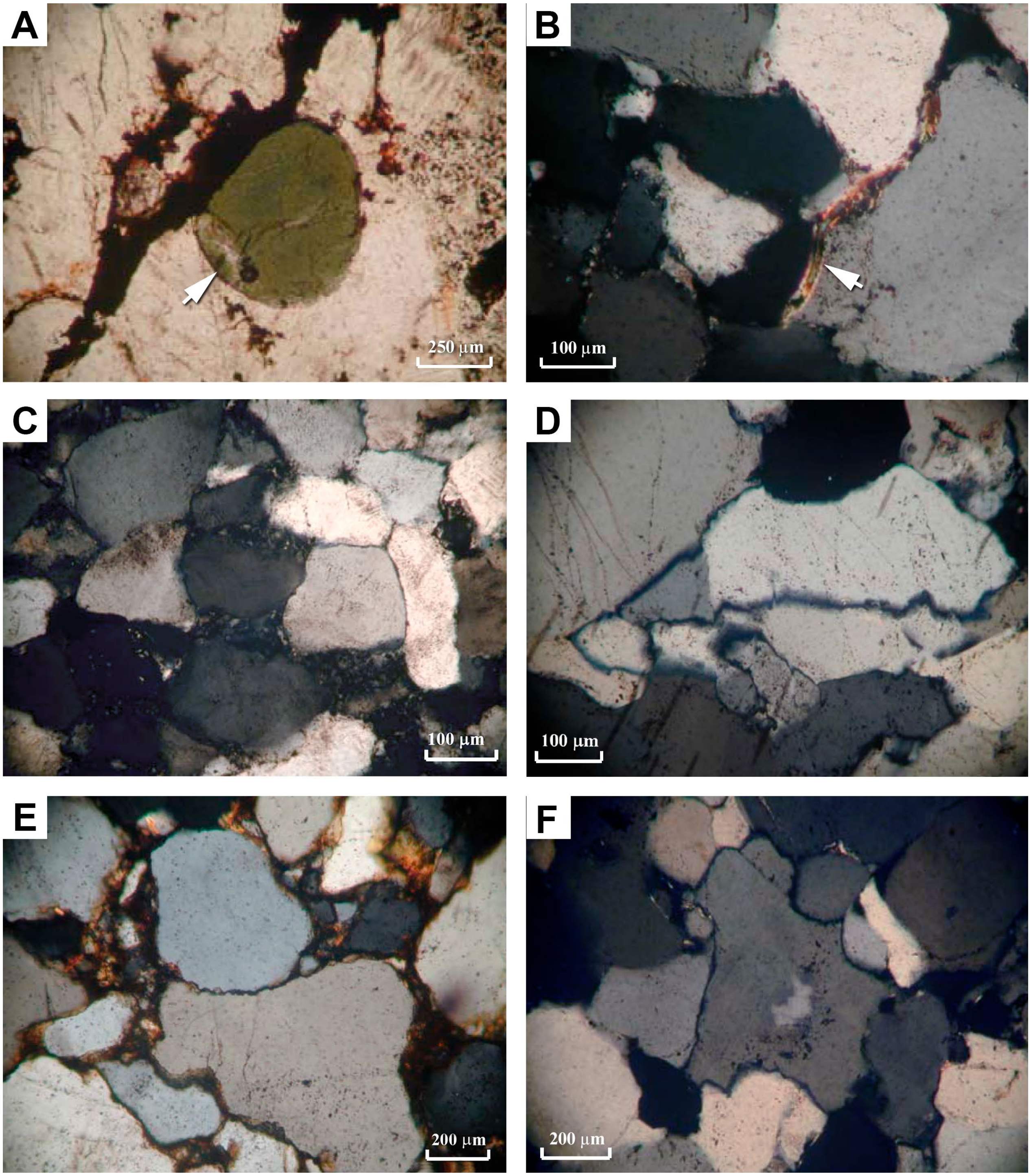

Figure 3.— Thin section photomicrographs of the upper Devonian sandstones from the Western Iberian Chain (Tabuenca, Spain). a) Cross-polarized OM image showing a tourmaline grain (white arrow) included into a quartz-grain (sample LG-022, Bolloncillos Formation). b) Cross-polarized OM image showing a detail of the sample CH-042 from the Hoya Formation. The mica (muscovite) grain (white arrow) has been mechanically deformed by the differential stress caused by the overburden. The surrounding quartz grains have been partly dissolved at the mica-quartz interfaces, explaining the highly conformable quartz/mica interfaces (Bjørkum, 1996). c-f) Cross-polarized OM images showing overgrowth quartz cement around detrital grains and concave-convex to sutured contacts. c) Sample BM1-013, Rodanas Formation. d) Sample BVi-004, Bolloncillos Formation. e) Sample CH-017, Hoya Formation. f) Sample BM2-035, Huechaseca Formation.

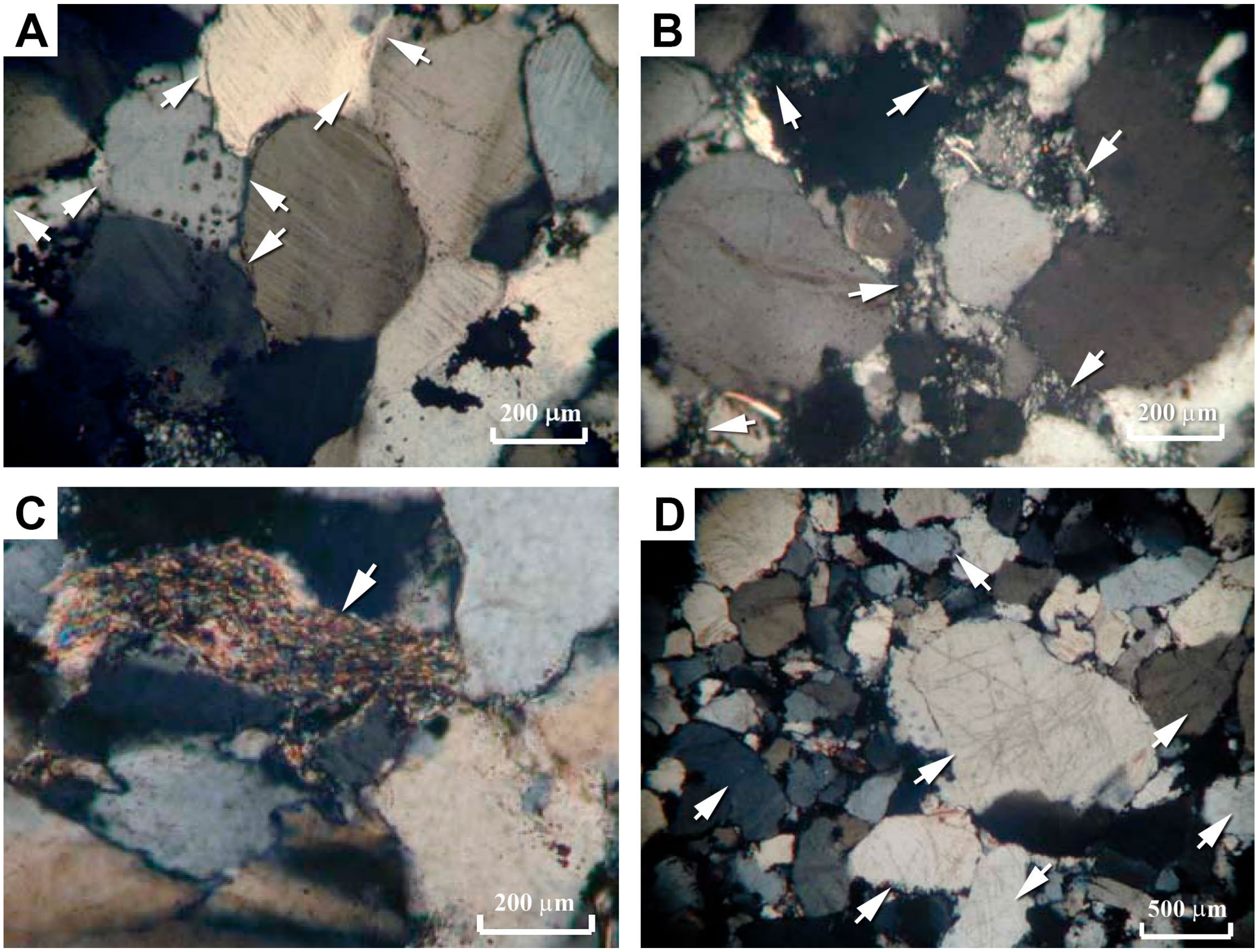

Figure 4.— Thin section photomicrographs of the Upper Devonian sandstones from the Western Iberian Chain (Tabuenca, Spain). a) Cross-polarized OM image showing overgrowth quartz cements (white arrows) around detrital grains and concave-convex to sutured contacts in sample CH-038 of the Bolloncillos Formation. b) Cross-polarized OM image of sample BVi-004, Bolloncillos Formation, showing the presence of clay matrix (white arrows) between the quartz grains. c) Epimatrix (white arrow) associated to diagenetic alteration of a lithoclast appear in this cross-polarized OM image of sample CH-010, Hoya Formation. d) Cross-polarized OM image showing spider-web microcracks within the quartz grains (white arrows) in sample BM-002 of the Hoya Formation.

Al_2O_3 content, which would reflect the different proportions of phyllosilicates and quartz present in each type of rock. In addition, there are noticeable differences in the average content in Fe_2O_3 due to synsedimentary and diagenetic processes. The proportion of TiO_2 is also higher in the lutites than the other lithologies. The high correlation presented by Al_2O_3 with K_2O and TiO_2 would indicate that they are being provided primarily by phyllosilicates, while those with lower Fe_2O_3, would indicate they are also found in other mineral phases such as Fe-oxyhydroxides. In the case of CaO and MgO, no significant correlations are observed with Al_2O_3, suggesting they should be present in other mineral phases, possibly carbonates, especially in the Rodanas Fm. The samples analyzed by X-ray fluorescence, except the carbonated samples of the Rodanas Fm., were geochemically classified (Fig. 5) according to Herron (1988), as suggested by Rollinson (1998).

In addition, it has been observed that the Rb, Cs, Ba, Sr, Th and U contents significant correlations with the amount of Al_2O_3. This would suggest that, in different proportions, the distribution of all of them is controlled by the presence of phyllosilicates (Dumitru *et al.*, 2015). However, Sr presents a behavior lightly erratic, probably due to its presence in several mineral phases (i.e., carbonates, phyllosilicates...). Besides, the good linear interrelations that

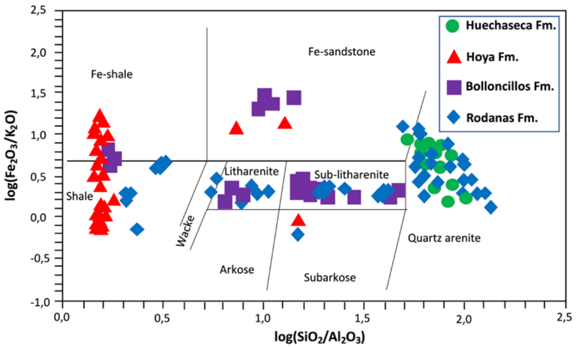

Figure 5.— Classification of the analyzed samples following the rules of Herron (1988).

the Rb and the Cs present with the K_2O, might indicate that, possibly, they are being controlled by the micaceous phases (Fig. 6). Zr exhibits a geochemical behavior similar to Hf (Bauluz, 1997), reflected in the high correlation between both (Zr/Hf = 30-40). This would indicate that they are being contributed mainly by zircons (Murali *et al.*, 1983).

Finally, with regard to rare-earth elements (REE), they are provided with the information contributed by Bauluz (1997). It show that both light and heavy rare earth elements (LREE and HREE) have a similar geochemical behavior. That is, they tend to concentrate on the same type of mineral phases. However, LREE and HREE tend to have slightly different geochemical behaviors.

Detailed cuantitative tables containing the results derived from the complete geochemical analyses are provided in the electronic supplementary material accompanying this paper (named Geochemical Data).

Sand-Sized Grains

The sand-sized grains from the Hoya Fm. samples are mostly angular to sub-rounded in shape. Most of the sand-sized grains in the Rodanas, Bolloncillos and Huechaseca formations' samples have a higher textural maturity, being mainly sub-rounded to rounded. Grain contacts in all these formations range from concave-convex to sutured. Additional petrographic information of arenites in upper Devonian Formations are provided in Table 2.

Porosity and Cementation

Only 63 samples (14.8 % of total samples) presented secondary porosity, ranging between 0.1 and

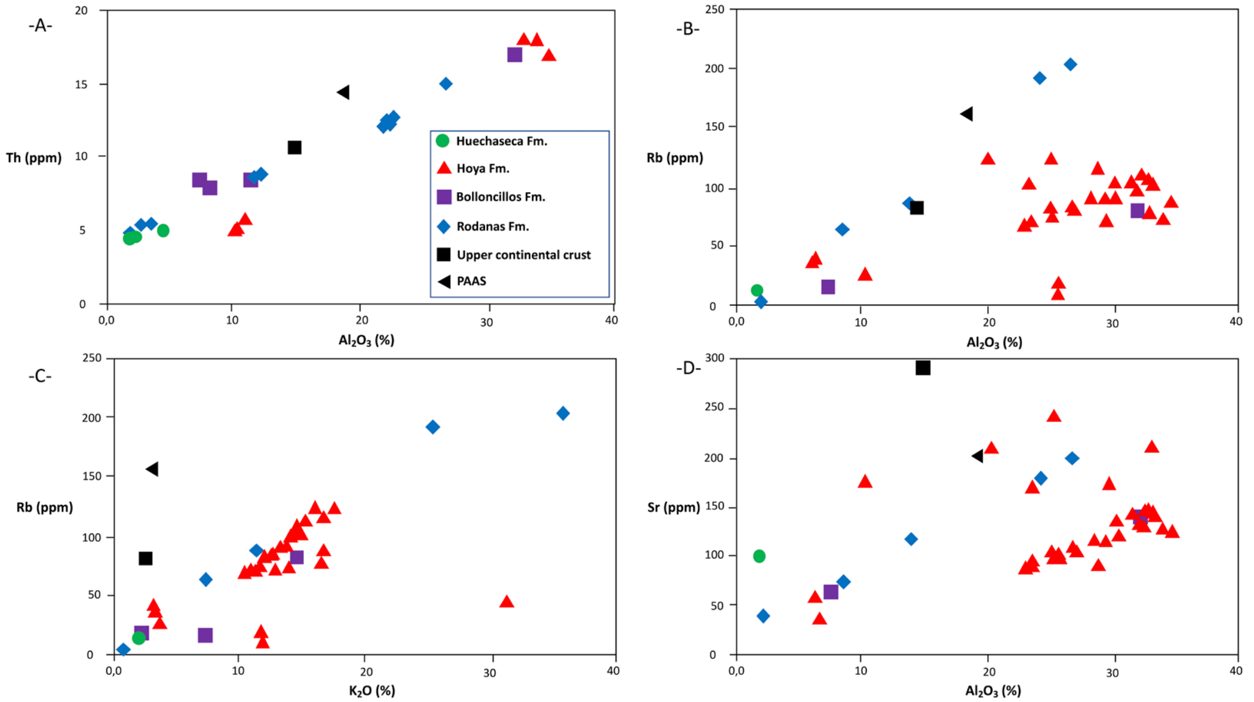

Figure 6.— Graphical representation of the global samples analyzed, differentiated by formations. In addition, the compositions of PAAS and the upper continental crust (Taylor & McLennan, 1985) have been also projected: A) Th-Al_2O_3; B) Rb-Al_2O_3; C) Rb-K_2O; D) Sr-Al_2O_3.

1.2 %, and no primary porosity was found in any of the samples because it was occluded by compaction, post depositional cementation and by the formation of matrix/epimatrix. Compaction is, in general, the most important factor of loss of primary porosity and resulted in an increase in contact strength, with more concave-convex and sutured contacts than long or point contacts, brittle deformation and deformation of ductile grains (e.g. muscovite). Quartz cementation is the second important factor for porosity reduction. Quartz cement appears in most samples with various grey shades in panchromatic SEM images. Samples with a relatively high matrix content were less affected by compaction.

Discussion

Diagenetic History

Compaction is the most important diagenetic process along the first 2 km of burial, whereas cementation increases at higher depths, especially when temperatures reach 85ºC or more (Worden & Morad, 2000; Torrijo, 2003; Bernet *et al.*, 2007; Surpless, 2015). In the Iberian Chains at present approximately 2 km thickness of the stratigraphically overlying Carboniferous, Permian and Triassic deposits exist (Torrijo, 2003). Therefore, upper Devonian quartz arenite samples from the Iberian Chains are characterized by post-depositional compaction and quartz cementation.

Several diagenetic processes have been distinguished in the samples studied. The development of the diagenetic structures (nodules and concretions) that appear in the Rodanas and Hoya formations, took place within the early diagenetic processes (Torrijo *et al.*, 1998, 2000, 2001, 2004a, 2004b, 2005; Torrijo, 2003). As final processes, the presence of concave-convex and sutured contacts between the skeleton constituents can be noted. The presence of iron oxides in these materials is both synsedimentary (indicated by the presence of banding) and properly diagenetic or hydrothermal (presence of hematites associated with crack fillers and epidiagenetic (genesis of Fe and Mn oxides and oxyhydroxides).

The post-depositional quartz cementation produced syntaxial overgrowths. These cements have diverse chemical compositions, indicating that these cements are not genetically linked. Silica sources for cementation in these formations were controlled by the lack of lithic fragments in the rocks and feldspars (e.g. Worden & Morad, 2000; Caja *et al.*, 2010). Therefore, the dissolution of the quartz grains by pressure-dissolution during compaction, or the infiltration of silica-bearing pore fluids from adjacent areas, may be the most likely silica source (Torrijo, 2003; Bernet *et al.*, 2007). Evidences for this process were found in the studied samples.

Aparicio *et al.* (1991) indicate, for the Paleozoic of the Iberian Chains, a progressive decrease in the diagenetic degree reached by the materials from the Precambrian (with values of illite crystallinity index of 3.25) and Cambrian materials (with an crystallinity average value of 3.6) to the Ordovician (with increasing values ranging from 3.8 at the base to 5.2 at the top of the sequence). These cristallinity indexes place the Precambrian rocks in conditions of the epizone; the Cambrian rocks in the Epizone-Anchizone boundary, and the Ordovician rocks in the anchizone conditions.

Similar conclusions are obtained for by Bauluz *et al.* (1998) in Cambrian-Ordovician rocks of the Iberian Chains. They indicate a gradual evolution from the Epizone-Anchizone limit for Cambrian rocks or to the Anchizone – Diagenesis-Anchizone for the Ordovician rocks. These data would be consistent with a simple model of progrant evolution of the phyllosilicates, being the progressive increase of temperature during the burial the main controlling factor: the oldest materials reach higher depths of burial (i.e. higher temperatures) and are those achieve a more advanced stage in the mineralogical transformation sequence of the phyllosilicates.

The results presented by Aparicio *et al.* (1991) and Gimeno (1999) for the Silurian of the Iberian Chains would also be consistent with this model: the recorded illite crystallinity indexes and the observed diagenetic processes are typical for an advanced diagenetic environment (Fig. 7). Bescós (1988) for Silurian-Devonian deposits from the Iberian Chain also indicates an evolutionary stage proper to the diagenetic environment and far away from the conditions of the anchizone.

Therefore, only to be explained the cause of the abnormally high values of the crystallinity of the il-

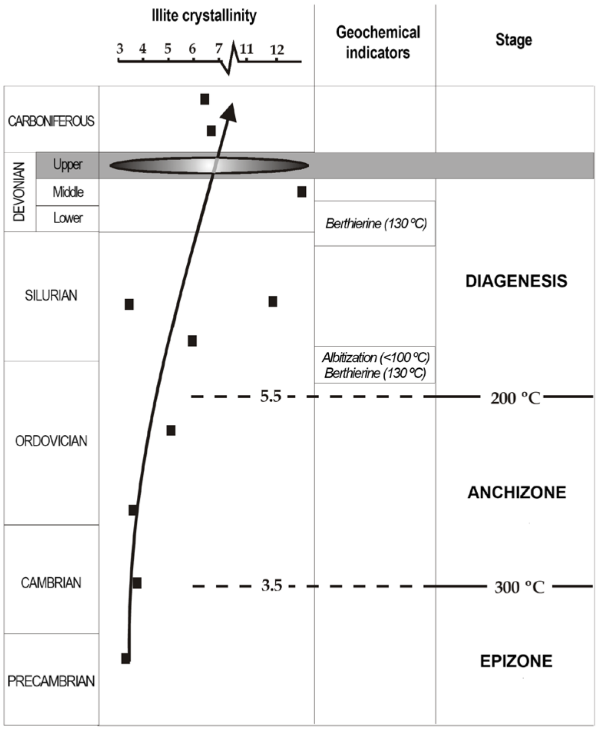

Figure 7.— Schematic Diagram showing the diagenetic evolution of the Paleozoic materials (Precambrian-Carboniferous) of the Iberian Chain. Illite crystallinity data were retrieved from Aparicio *et al.* (1991). Only geothermometric processes are considered. Note that the history of the crystallinity of the illite is consistent with the usual diagenetic evolution of the materials that gradually reach higher values of Temperature and Pressure with burial deep. The illite crystallinity values of Devonian are abnormally high according with Aparicio *et al.* (1991). Nevertheless, the presence of berthierine in the Silirian-Devonian materials led to refer simple burial diagenesis model. Modified from Gimeno (1999).

lite for Lower-Medium Devonian rocks (Aparicio *et al.*, 1991). However, according to their mineralogy (Fig. 7), they would fall within the general pattern of simple burial models. It is therefore reasonable to think that the crystallinity values recorded in the Upper Devonian from Tabuenca area would fall within the ranges defined by the materials placed immediately under and by the overlying materials, always within the diagenetic environment. According to Aparicio *et al.* (1991), the appearance of prograde or retrograde effects on the degree of crystallinity of the illite from the Devonian rocks is due to the effect of the hydrothermal activity of igneous intrusions (Gimeno, 1999).

As a summary, it can be inferred that the materials constituting the Upper Devonian of the Iberian Chains in the area of Tabuenca did not reached an advanced diagenetic stage. They would be placed in the proper diagenetic environment, reaching maximum temperatures of 125-150ºC (Torrijo, 2003).

Source Area Composition

The abundance of trace elements in detritic sedimentary rocks has been commonly used to trace the source area composition (e.g. McLennan *et al.*, 1983; Bhatia, 1985; Taylor & McLennan, 1989; Wronkiewicz & Condie, 1990; Shukla *et al.*, 2020; George & Ray, 2021). In this sense, for the case of the rocks studied, the triangular diagrams La-Th-Sc and Th-Hf-Co have been chosen. And besides, to calculate the approximate percentage of felsic and maphic volcanic rocks in the source area, a Co/Th vs. La/Sc diagram has been used (McLennan *et al.*, 1983; McLennan, 1989).

The results suggest that the lutites are similar to the PAAS-type slates, which would indicate that their composition would be similar to that of the upper continental crust. Therefore, the primitive source area would be predominantly of a felsic type. Besides, the lutites of the Fm. Rodanas could come from source areas where the maphic material represented about the 15% of the constituents, while in the more recent formations (Bolloncillos Fm. and Hoya Fm.) the proportion increases up to the 25% (Fig. 8).

Detritic Rocks Provenance and Paleoclimate

The arenites of the Rodanas and the Huechaseca formations are composed of mono- and rare polycrystalline quartz (total quartz > 90%). Silicate minerals (muscovite, tourmaline, zircon, potassium feldspar), apatite grains and rock fragments grains are scarce, as well as no plagioclase grains were observed. This composition corresponds that of sediments derived from recycled orogenic or craton interior sources according to Dickinson *et al.* (1983). On the other hand, the arenites of Bolloncillos and the Hoya Formations have a sand-sized framework composed of mono- and polycrystalline quartz (total > 75%). Feldspar grains are scarce, and no plagioclase grains were observed. However, muscovite, tourmaline, zircon and apatite are more abundant than in the Rodanas and in the Huechaseca formations. These

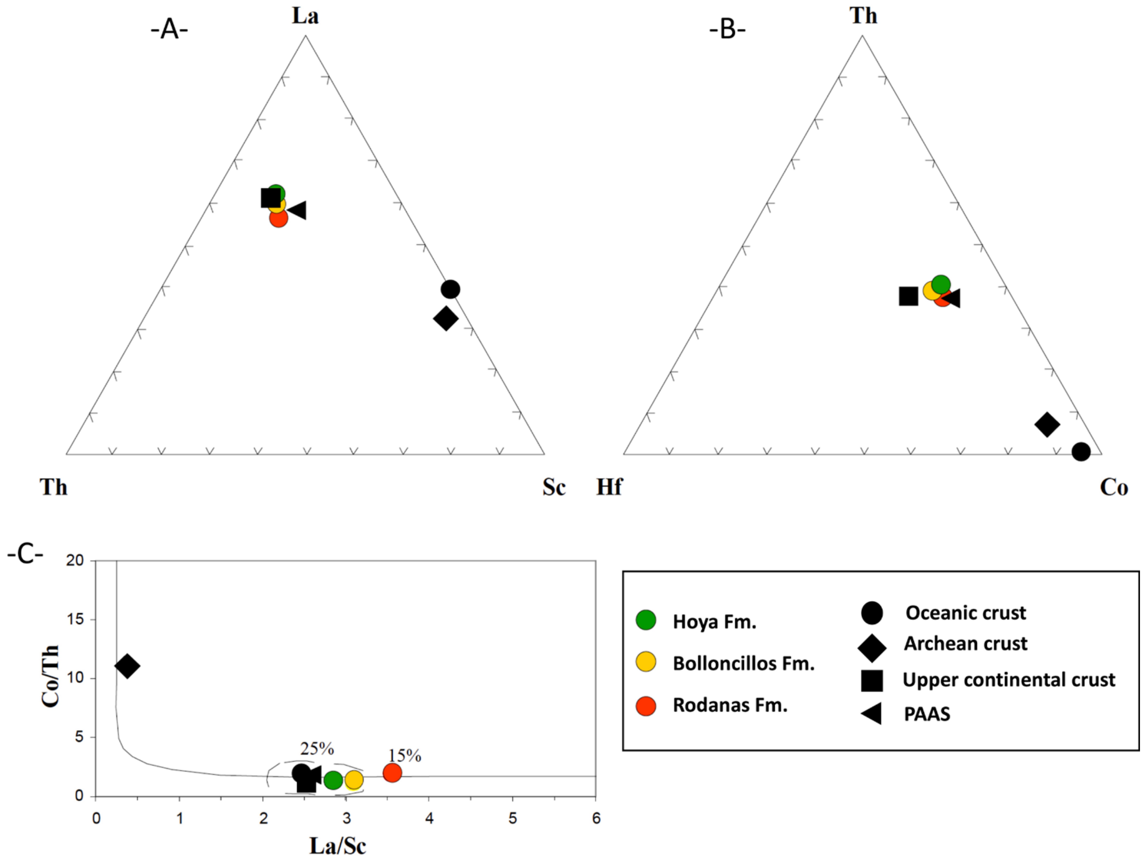

Figure 8.— Graphical representation of the average compositions of the lutites of the upper Devonian of Tabuenca. In addition, the compositions of PAAS, and the oceanic, upper continental and archean crusts (Taylor & McLennan, 1985) have been also projected: A) Th-La-Sc; B) Th-Hf-Co; C) Co/Th vs. La/Sc.

compositions (Fig. 9) are typical from craton interior or recycled orogenic sources (Dickinson *et al.*, 1983).

In this way, primary source areas of the siliciclastic materials from the upper Devonian formations are located to the Northeast (Oliveira *et al.*, 1986) and are part of a crystalline basement and quartz-rich sedimentary cover units (the Ebro Massif, see Carls, 1983, 1999). Given that no plutonic rocks were exposed in this area during the Devonian (Gozalo & Liñan, 1988; Carls, 1999), it seems probable that the quartz grains were second-cycle grains derived from sedimentary rocks which could have lost their inherited quartz overgrowths by abrasion during transport. The few available lithic fragments found in these sediments also reflect a combination of crystalline basement and sedimentary sources (Arribas *et al.*, 2014; Konstantinou *et al.*, 2014). Sediment recycling in upper Devonian materials of the Iberian Chains was also suggested by Bauluz (1997) and Torrijo (2003).

All the arenites of these above-mentioned formations are coastal sediments (Fig. 10) derived from quartz-rich sources. The most likely candidate to explain the high maturity of the studied arenites is a climatic influence (Torrijo, 2003). Currently, it is considered that, from Cambrian to Devonian ages, with the exception of the South Portuguese Zone that probably belonged to Avalonia, the whole Iberian Peninsula was part of the North Gondwana Province that extended along the northern margin of the African part of the Gondwana landmass (Gutiérrez-Marco *et al.*, 2001, and references therein). Thereby, the latitudinal position of the present Iberian can thus be estimated only roughly as intermediate between the rather warm or cold temperature of high lati-

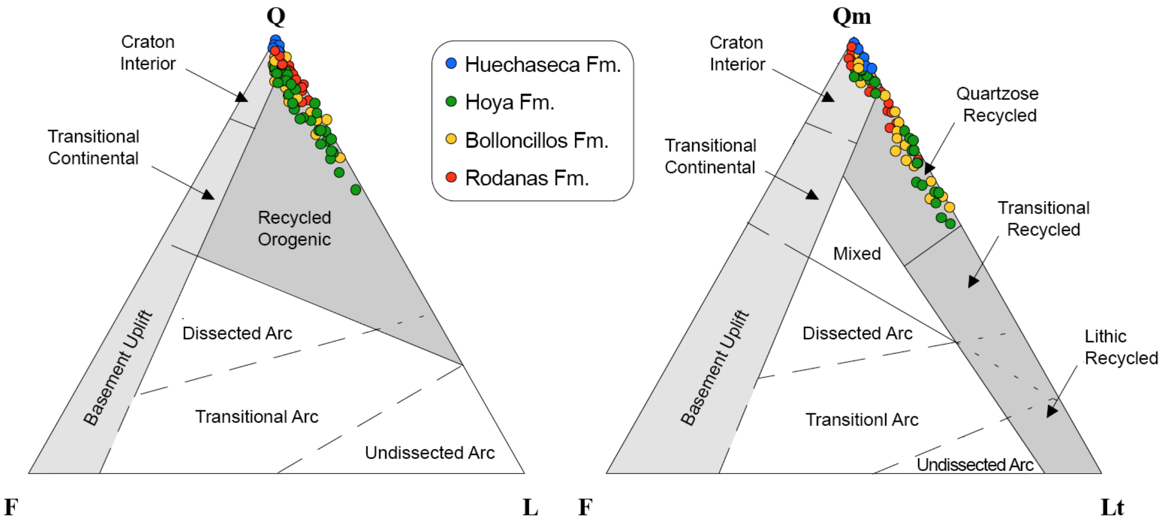

Figure 9.— Sandstone provenance diagrams after Dickinson *et al.* (1983). Abbreviations: Q – total quartz (includes mono- and polycrystalline quartz); Qm – monocrystalline quartz; F - Feldespars; L – Lithoclasts (including polycrystalline quartz).

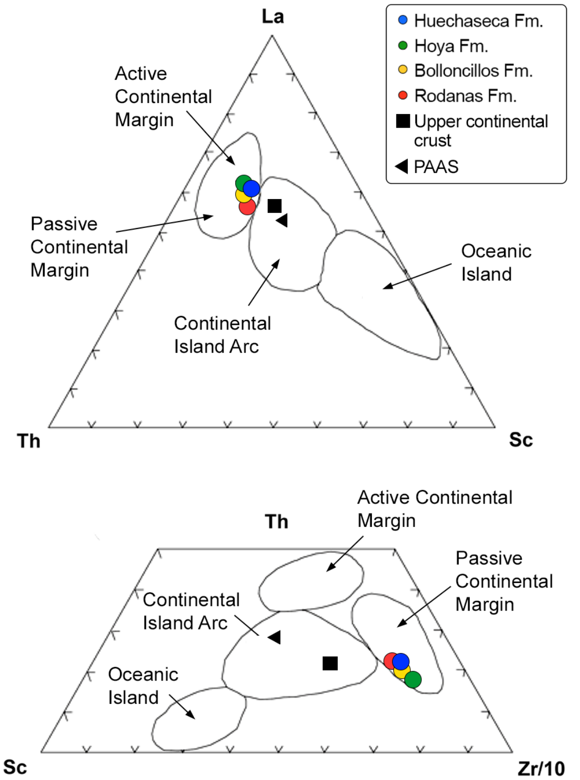

Figure 10.— Average composition of the arenites from the western Iberian Chain (Tabuenca, Spain). Data obtained from Bauluz (1997) are represented in ternary trace elements diagrams proposed by Bhatia & Crook (1986). In addition, the compositions of PAAS and the upper continental crust (Taylor & McLennan, 1985) have been also projected: “Oceanic Island Arc”; “Continental Island Arc”; “Active Continental Margin” and “Passive Continental Margin”.

tudes (ca. 50ºS) of the latest Ordovician times, and the warmer temperatures of the subtropical latitudes (ca. 35ºS) of the Devonian succession (Robardet & Gutiérrez-Marco, 2002), with a high rainfall regime and high mean-annual temperatures. So, these climatic conditions may have led to a rapid weathering and breakdown of unstable rock fragments during transport, deposition and along the early diagenesis, enriching the sediments in quartz grains (Basu, 1985a, b).

Conclusions

A geological characterization of the Upper Devonian siliciclastic formations of Tabuenca area (Iberian Chains, Spain) was conducted, studying their provenance and diagenesis. Results show that these materials are mainly composed of quartz-rich sediments that have experienced intense weathering before and during their deposition and posteriorly, underwent alteration, compaction, and quartz cementation during diagenesis. No primary porosity was recorded in the quartz arenites of any unit, and the development of secondary porosity was rare. Sand-sized grains of the Hoya Formation samples were mostly angular to sub-rounded, while most of the sand-sized grains of the Rodanas, Bolloncillos and the Huechaseca formations samples exhibited higher textural maturity, were mainly sub-rounded to rounded in shape, and showed concave-convex to sutured grain contacts. The majority of sand grains in the studied samples were regarded to be of plutonic origin. Quartz grains of potential volcanic affinity are almost exclusively restricted to the finest sand-sized fraction of the samples. All samples show evidence of quartz cementation after deposition.

Several diagenetic processes have been distinguished in the samples studied. Thus, the presence of concave-convex and sutured contacts between the skeleton constituents can be noted. Besides, the presence of iron oxides in these materials is both synsedimentary and properly diagenetic or hydrothermal:

1. Upper Devonian quartz arenite samples from the Iberian Chains are characterized by post-depositional compaction and quartz cementation. The dissolution of the quartz grains by pressure-dissolution during compaction, or the infiltration of silica-bearing pore fluids from adjacent areas, may be the most likely silica source. The post-depositional quartz cementation produced syntaxial overgrowths.
2. It can be inferred that the materials constituting the Upper Devonian of the Iberian Chains in the area of Tabuenca did not reached an advanced diagenetic stage. They would be placed in the proper diagenetic environment, reaching maximum temperatures of 125-150ºC.
3. The results suggest that the lutites are similar to the PAAS-type slates, which would indicate that their composition would be similar to that of the upper continental crust. Therefore, the primitive source area would be predominantly of a felsic type. Besides, the lutites of the Fm. Rodanas could come from source areas where the maphic material represented about the 15% of the constituents, while in the more recent formations (Bolloncillos Fm. and Hoya Fm.) the proportion increases up to the 25%.
4. Primary source areas of the siliciclastic materials from the upper Devonian formations are located to the Northeast and are part of a crystalline basement and quartz-rich sedimentary cover units (the Ebro Massif). It seems probable that the quartz grains were second-cycle grains derived from sedimentary rocks which could have lost their inherited quartz overgrowths by abrasion during transport.
5. In this way, all the arenites studied are coastal sediments derived from quartz-rich sources. The most likely candidate to explain the high maturity of the studied arenites is a climatic influence. Thereby, the latitudinal position of the present Iberian can thus be estimated only roughly as intermediate between the rather warm or cold temperature of high latitudes (ca. 50ºS) of the latest Ordovician times, and the warmer temperatures of the subtropical latitudes (ca. 35ºS) of the Devonian, with a high precipitation regime and high mean-annual temperatures.

Supplementary materials

Supplementary information is available in the online version of the paper.

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