

Article

Provenance Studies of Natural Stones Used in Historical Buildings of the Peninsula de Barbanza, Galicia, Spain (North-Western Iberia)

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Abstract: Traditionally, the building stones used in the architectural heritage of Galicia (Spain) during the past were mainly extracted from quarries located in the surrounding areas of the historical buildings. Thus, a great variety of monuments were built with the same type of granite but with different degrees of weathering depending on local conditions, geological context (facies), and period of construction. The main purpose of this work is to evaluate the probable origin of the construction materials of six historical buildings on the Barbanza Peninsula, Galicia (Spain), based on the degree of weathering and petrographic-mineralogical characteristics. The evaluation was performed on six different samples of granite according to the geological context in outcrops of places where there are old quarries. We used X-ray fluorescence spectrometry (XRF), X-ray diffraction (XRD), and petrographic microscopy (PM) to attempt to address whether the origin is really local to the Barbanza Peninsula or whether the stone was brought from more distant places, based on our knowledge of the geological context of the study area. Based on the chemical, textural, and mineralogical analyses obtained, the material used for construction has a local origin and comes from small quarries spread over a wide area within the Peninsula itself. Barbanza-type granite is the most abundant within the geological context of the area and also the most used as a construction material. Other types of granites identified are the Confurco Granite and other granitoids of the Noya complex. The Chemical Alteration Index (CIA) shows low to moderate weathering in the granites, fitting petrographic observations. In monuments, samples show higher concentrations of Na and K due to salt crystallization. They show microcracks due to extraction, cutting, and finishing processes and common alteration processes of the main minerals, such as sericitization of plagioclase and chloritization of biotite. This work consists of a multidisciplinary study focused on the geological perspective for the identification and preservation of historic quarries. Knowledge of the original material also presents a unique opportunity for the restoration and/or reconstruction of monuments, which allows for the maintenance of their conceptual and constructive homogeneity.

Keywords: geological resource; historical quarries; granite; weathering



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1. Introduction

The granite massifs cover most of the territory of Galicia (NW Spain) and Northern Portugal. As granite is the most frequent stone type in these areas, it played a very important role in the construction of historic buildings from prehistoric times. This type of stone is usually highlighted as a building material due to its hardness, low porosity, and, in general, great durability [1–4]. The Barbanza Peninsula is located on the western coast of Galicia (Figure 1). It has a vast architectural heritage of all historical periods [5,6],

from megalithic constructions such as Dolmens to villages of the Iron Age referred to as ‘Castros’. There are also remains of heritage buildings from Roman times, like those of Santa Mariña de Xuño. Religious buildings include Romanesque and Gothic styles, but also Baroque style (like the Church of San Xoán de Macenda), and finally, Neoclassical style buildings (the majority, especially due to the reforms carried out) can be found on the peninsula. Some of the most abundant buildings, mostly constructed between the 17th and 19th centuries, are the so-called ‘Pazos’ (large manor houses). Unfortunately, many of the most prominent cultural heritage walls in the study area were destroyed over time and replaced with new construction materials, but, on the other hand, there are also remains that have survived the urban reforms carried out. Despite that, granite has been the main rock construction material used in both the original buildings and these reforms over the centuries. From this perspective, natural stones can be understood not only as geological resources but also as cultural resources [7].

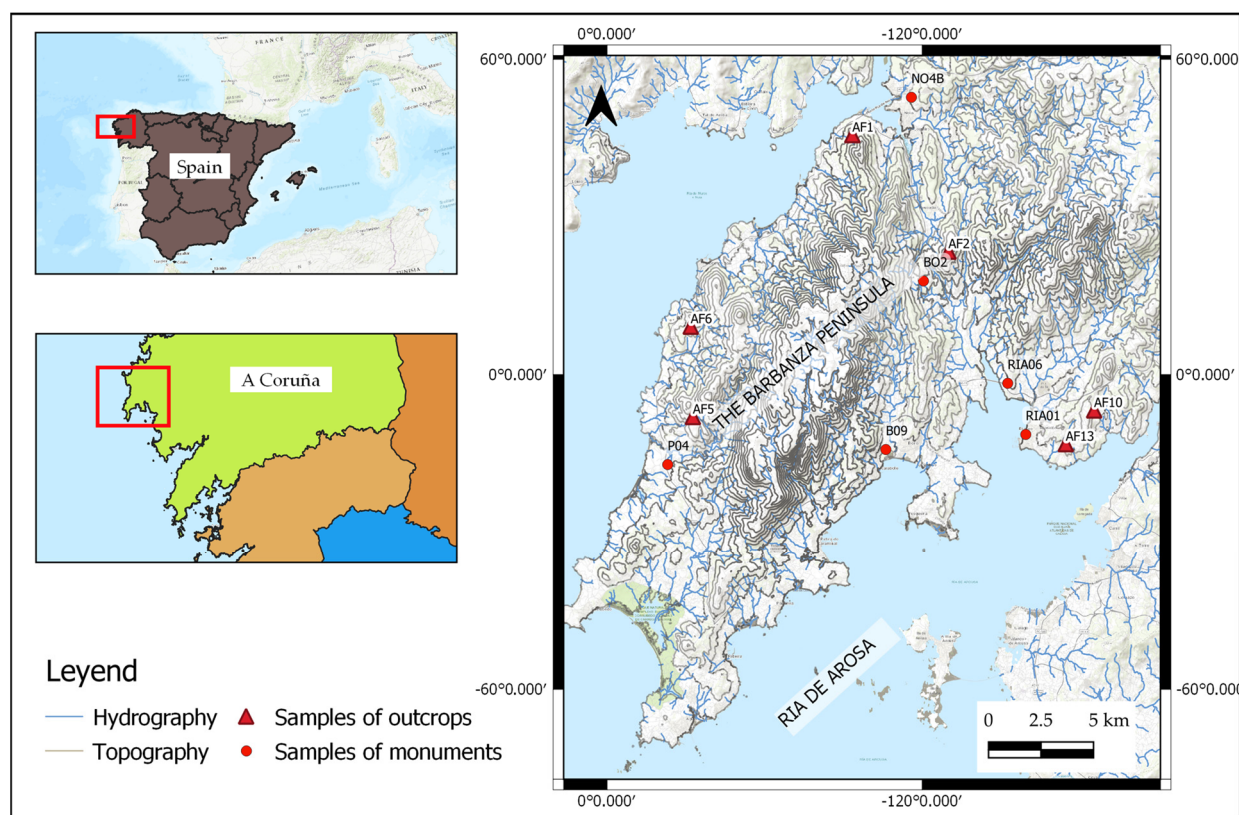


Figure 1. Location of the Barbanza Peninsula.

Traditionally, on the Barbanza Peninsula, the building stones used in the past were mostly extracted from quarries located in the surroundings of buildings [8,9]. Thus, a great variety of monuments have been built with the granite types that can be found on the peninsula, according to the geological context (Figure 2). The same granite type can show different degrees of weathering according to the local outcrop conditions (facies). During the historical periods of construction, there was no current technology for carrying out deep excavations. Thus, the old quarries were shallower than the present ones, and the granite used for construction weathered to some degree. According to this aspect, the granites used in historical monuments tend to have a more yellowish color than the granites extracted today for construction. At the same time, such granites should have greater porosity and lower density than the granites used in modern buildings [10,11].

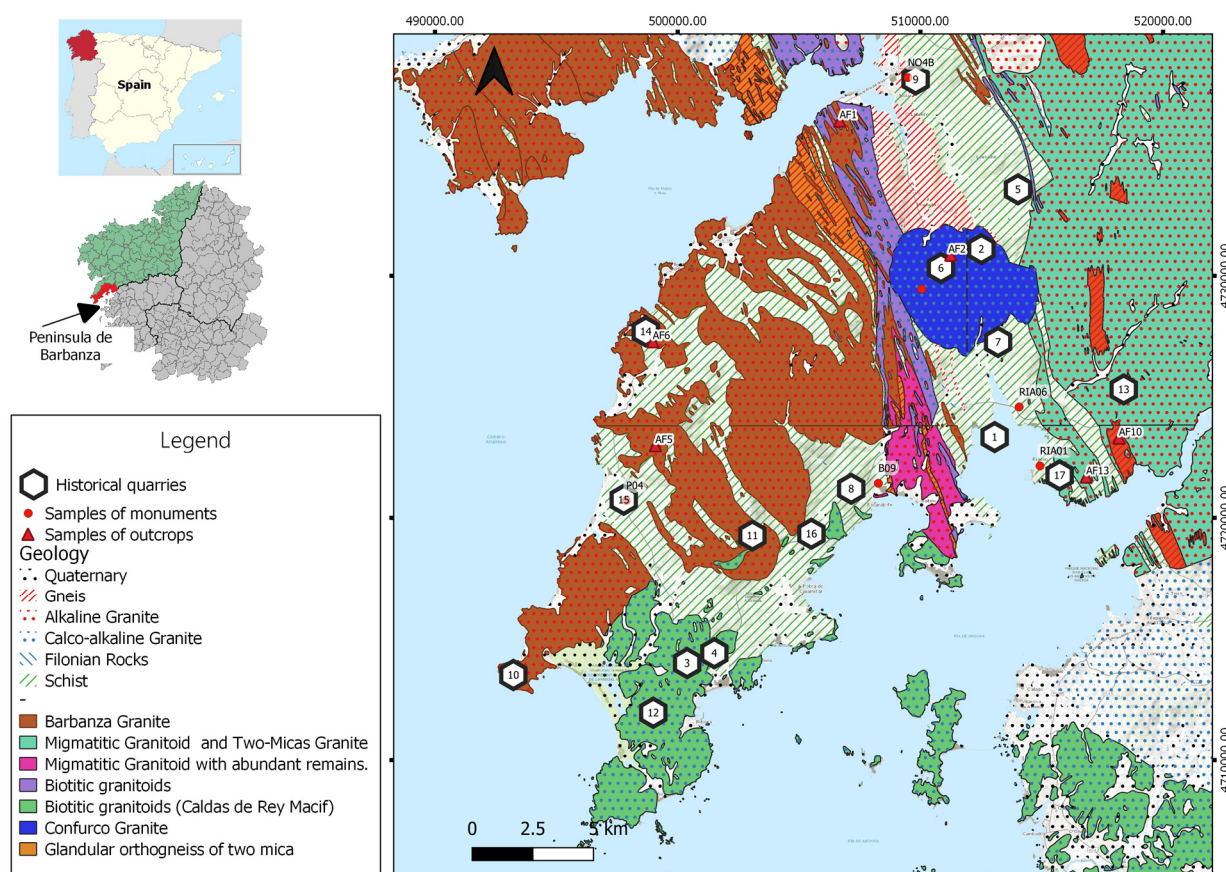


Figure 2. Location of the outcrop and monuments samples in geological map on Barbanza Peninsula. Historical quarries: 1. Neixón; 2. Confurco; 3. San Alberte; 4. Carballa; 5. Minas de San Finx; 6. Moimenta; 7. Bealo; 8. Lampón; 9. Santa Cristina de Barro; 10. Corrubedo; 11. Monte de la Curota; 12. Ribeira (mountain zone); 13. Asados; 14. Baroña; 15. Santa Maria de Parish (Xuño); 16. San Isidro de Postmarcos.

In addition, natural stones can decay over time at higher rates outside of their natural environment [12]. For this reason, there are a large number of studies that indicate that granite can undergo deterioration processes when exposed to weathering, due to their intrinsic properties (mineralogy, porosity, permeability) together with extrinsic (fluid circulation, climatic conditions, environment) and anthropogenic factors [13–18]. If decay is significant in historical periods, it can lead to serious problems in historical buildings with loss of value, and eventually the loss of some physical properties can lead to ruins. Hence, evaluating its characteristics and knowing the origin of the material are essential in conservation and restoration work. Recently, different studies have shown interest in identifying the outcrops of origin of building stone. More specifically, antique quarries provide geographic and geological information on the bedrock and allow study of the stone behavior under several environmental conditions [11,19–26], as well as finding the most suitable material for future restorations and replacements of the construction elements of buildings [27–30].

The main purpose of this work is to evaluate the probable origin of the construction materials of historical buildings on the Barbanza Peninsula based on the petrographic-mineralogical characteristics and the degree of weathering. The evaluation is performed on the granite used in historical buildings in the area and also on the geological context in outcrops of places where there is evidence of old quarrying. Knowledge of these materials is very important for the scientific community, both to understand the characteristics of historic buildings and their possible damage, and to propose solutions for the best possible conservation and preservation of these remains of architectural value. Recently, an effort

has been made to study the degree of deterioration and geochemical properties of granite as a stone material in the heritage of the Barbanza Peninsula [6], but the lack of provenance data for these materials inspires this work as a first approach to the current situation and raises awareness about the importance of heritage stones and the need to save resources for the continuing lack of historic buildings and structures.

2. Geological Setting

2.1. Geology

The geologic bodies in the Barbanza Peninsula are mostly Hercynian or pre-Hercynian intrusive bodies, with the exception of some Quaternary filonian rocks and detritic deposits (the latter located mainly in coastal areas). The classification of Galician granites is based essentially on the works of Capdevila (1969) and Capdevila and Floor (1970) [31,32], which established two general series: one alkaline and the other calc-alkaline (Figure 2).

The granites of the calc-alkaline series are characterized by their porphyritic character and the predominance of biotite over muscovite (sometimes only biotite). They are frequently deformed by the third phase of Hercynian deformation, and microgranular enclaves of highly variable dimensions abound [33]. They include megacrystal-bearing granodiorites and coarse-grained two-mica granite (Confurco Granite) [34]. The Confurco Granite (Figure 2), also called Moimenta Granite, formed after the third phase of Hercynian deformation, being younger than all the others described. Two facies have been described [35]: porphyritic and fine-grained. The porphyritic facies represents more than 80% of this granite area.

The alkaline series occupies the largest area of rocks, comprising all the two-mica granites that are pre- or synkinematic with the third phase of Hercynian deformation, both. It also includes a heterogeneous granitoid, also called migmatitic or first granitoid products of migmatization (Migmatite Domain and of the Granitic Rocks-Laxe Group).

The Barbanza Granite (Figure 2) occupies most of the Peninsula [34], distributed mainly in the central zone. It consists of two micas of coarse-grained alkaline granite. In the north of Barbanza, this granite contacts rocks of the Noya metamorphic zone (the Malpica-Tuy Unit, called here the Noia Complex), formed by schist, paragneiss, and amphibolites. On the Corrubedo zone (in the SW portion of Barbanza), this granite is transformed into a fine-grained variant (Corrubedo Granite) with many xenoliths and also migmatites, oriented [33].

On the other hand, the Migmatite Domain of the Granitic Rocks-Laxe Group is a set of rocks formed mainly by anatectic granites, granitic gneisses, and intensely migmatized schists [33]. It is distributed mainly along the entire SE coast, as well as along the road between Noya and Boiro and in the SW part, near Ribeira.

2.2. Historical Quarries and Mining

Since the dominance of granite on the Peninsula of Barbanza, the main quarries have been dedicated to the extraction of this material, creating important sources of employment and wealth in the area. Several building stones came from a large number of small quarries spread over a wide area (Figure 2). It is beyond the scope of this document to provide a comprehensive review of all extraction points. Most of the information about the location of these extraction sites is obtained from the inhabitants of the village, and the existing documentation is scarce, mainly registered in national texts [33,36].

According to this, the construction material used in the traditional houses of Castros de Neixón came from Neixón [37], but the granite used came mainly from more distant quarries, such as the one in Confurco (Boiro). Furthermore, in San Alberte (Ribeira), large stone blocks, also locally called “penedos”, are located with granite stone extraction marks. Another place of stone and mud extraction is in Carballa (Palmeira, Ribeira), according to information from a woman who worked in this mine [36].

On the other hand, granitic pegmatites emerge in the surroundings of San Finx Tin Mines, which contributed to the first tungsten exploitation started in Spain and one of

the first in Europe [38]. In this period, new mining techniques appear and spread with a rational approach to management, contributing to the evolution of culture and technology in Galicia. Also reported are the mining operations in Moimenta and Bealo (Boiro) [36] and other smaller mines in the Muros-Noia Ria.

In Noia, there is granite exploitation in the village of Iglesia, near Santa Cristina de Barro. On the other hand, on the back cover of the magazine “La Voz de Galicia” (27 October 2007), a dozen disused mining drillings are reported. Numerous mines consisted of family-run exploitations and therefore are difficult to report throughout history, probably underestimating the number of explorations and quarries. An example of this is the LLovo mine in Cabo Ladrón, Corrubedo. Mining activities were also reported in the 20th century in Mondelo (Palmeira) and in the vicinity of Monte de la Curota (Ribeira, on the border with Puebla), related to the Arosa de Lampón tin factory in Boiro. On the other hand, vestiges of mining exploitation are also reported in the surroundings of the mountain surrounding the City of Ribeira, in Baroña (Porto de Son), in Santa Maria de Xuño, in San Isidro de Postmarcos (A Pobra do Caramiñal), and in many places in Rianxo [8,9].

Currently, there are active quarries, mainly Barbanza-type granite (Figure 3) and granodiorite, noted for its potential interest as an ornamental rock due to its pink color. In the case of gneiss and schists, they have less practical interest as industrial rock; however, locally, they present good characteristics and are used.

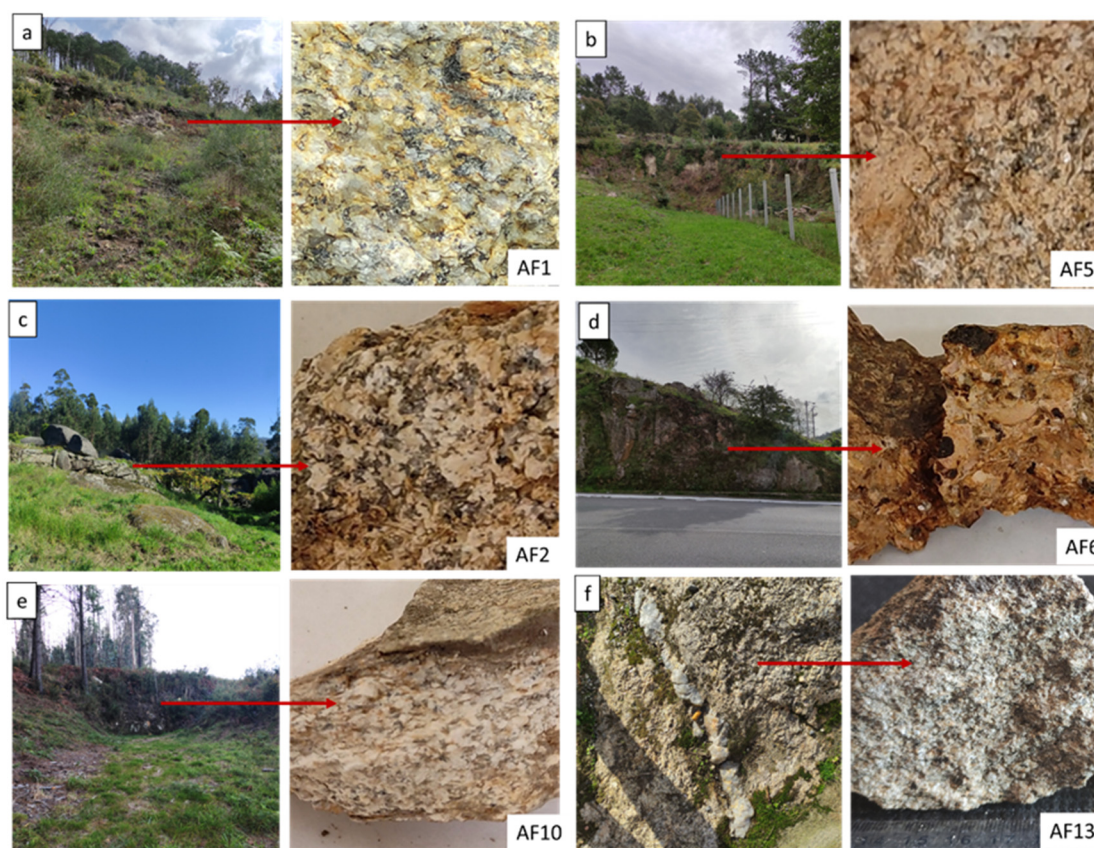


Figure 3. Outcrops and samples. (a) Granite (NW of Noia); (b) Granite found in the areas adjacent to the Petroglyphs of A Gurita; (c) San Xoa de Macenda granite; (d) Granite found near San Vicente de Noal; (e) Granite found near Leiro (Rianxo); (f) Granite found near Desamparados Chape (Rianxo).

As previously mentioned, the information on the oldest quarries used for the extraction of rocks as construction material is limited and does not appear on the maps (Figure 2). Therefore, the only current source is population knowledge. In this work, we have proceeded to obtain samples of granite of different types according to the geological context in bedrock outcrops of places where an old quarry possibly existed (AF1, AF2, AF5,

AF6, AF10, AF13) (Figures 2 and 3) and to assess whether they were used as construction material in the monuments of Barbanza.

3. Historical Buildings Included in This Study

This section describes some monuments of interest that were built with stone material and are shown in Figure 4. Despite the large number of churches and pazos that are part of the cultural heritage, only a few of them will be mentioned. The description of the main type of rock used in each monument is established in Table 1.

- The Pazo de Goiáns (Figure 4a) is an old medieval fortress (symbol of lordship between the 15th and 19th centuries) measuring 1,972 m², located in a unique environment, at the foot of the Corroño river estuary, and surrounded by large gardens full of unique species, a jetty, and some old fisheries. The complex is extensive, with an abundance of buildings, some complementary (e.g., warehouses, winepresses) and others decorative (mainly in the garden area), as well as porticos, pumps, stables, cellars, a chapel, and a garden that descends to the riverbank. Most of these buildings are made of medium- to coarse-grained granite, although many façades, including the wall that delimits the pazo, are built with a mixture of granites and metamorphic rocks (schist and gneiss). In 2019, the complex was declared in danger of collapse, and rehabilitation works have been carried out on the Main Tower, which is currently being restored, with the vision of continuing with the annexed constructions, whose state of deterioration is alarming. An example is the collapse of the chapel roof and the extensive biological colonization of the complementary buildings.
- The Church of Santa Mariña de Xuño (Figure 4b) is located in Xuño (Porto Son), about 70 km from Santiago de Compostela towards the western coast. It is very close to the sea and the Sieira River. The church was built in the 12th century but has undergone numerous modifications and extensions, preserving the original Romanesque fabric of the main chapel (externally in the eaves corbels). Most of these buildings are made of medium- to coarse-grained granite and the weathering is moderate.
- The Church of San Salvador de Taragoña (Figure 4c) was rebuilt in the 19th century, and although its original age is unknown, it is one of the most recent historical buildings. It has a rectangular floor plan with a single nave and two side chapels with a square floor plan. It is located in Taragoña (Rianxo). It is made up of a light tone and medium grain. Weathering is low.
- The Church of San Xoán de Macenda (Figure 4d) is a valuable example of a rural Baroque church, located in Macenda (Boiro), very close to where the Río Grande crosses. It was built in the 18th century. Most of the decorative elements of the temple are found in the bell tower since the rest of the walls of the temple are symmetrical and only altered by the placement of a square tower that consists of a body and a tower that was built in 1778. It is made up of a granite of medium-grain, medium-coarse-grained granite with potassium feldspar phenocrysts.
- The Church of Santa Maria de Barro (Figure 4e) is located in Noia, 38 km from Santiago de Compostela and 1.5 km north of the main town. This church is in the baroque style and was built in the 16th century. It has a rectangular floor plan with a barrel vault, which starts with simple molding on each of the side walls.
- Guadalupe Chapel (Figure 4f) is located in the center of the municipality of Rianxo. It is one of the monuments found on the Camino de Santiago. It is a religious temple built in the 17th century on a previous chapel. It is a very simple chapel made up of a single rectangular nave in which the buttresses stand out, accessed by a staircase.

Table 1. Samples, sample location and macroscopic description.

	Sample Code	Place of Sampling	Macroscopic Description
Outcrops	AF1	In the NW of Noia, on the slopes of the San Lois peak	Outcrop with slight orientation. Granite with two micas of medium grain. Light gray-yellow tone and moderate weathering. Composed mainly of quartz, plagioclase, feldspar, biotite, and muscovite.
	AF2	San Xoán de Macenda (Boiro)	Outcrop of coarse-grained granite with potassium feldspar phenocrysts and light pink tone. Moderate weathering. Phaneritic texture and composed mainly of quartz, plagioclase, feldspar, biotite, and muscovite.
	AF5	Outcrop near the Baroña church, areas adjacent to the Petroglyphs of A Gurita.	Granite with two micas of medium-coarse grain with potassium feldspar phenocrysts. Composed mainly of quartz, plagioclase, feldspar, biotite, muscovite, and dispersed mafic minerals (amphibole). Grey tone and moderate weathering.
	AF6	Outcrop near the church of Porto Son (San Vicente Noal)	Granite with two micas of medium-coarse grain, composed mainly of quartz, feldspar, muscovite, and plagioclase.
	AF10	Outcrop around Leira (Rianxo)	Outcrop with marked orientation. Medium-grained granite with whitish tone and equigranular texture. Composed mainly of quartz, plagioclase, feldspar, and muscovite. High weathering (tends to disintegrate).
	AF13	Outcrop near the chapel “de los desamparados” (Rianxo)	Outcrop of medium- to coarse-grained granite with a light gray tone. Low-medium weathering. Phaneritic, equigranular, and composed mainly of quartz, plagioclase, feldspar, and muscovite.
Monuments	BO9	The Pazo de Goiáns (Boiro)	Coarse-grained granite with potassium feldspar phenocrysts and light gray tone. Phaneritic texture and composed mainly of quartz, plagioclase, feldspar, biotite, and muscovite. There is no orientation.
	BO2	Church of San Xoán de Macenda (Boiro)	Coarse-grained granite with potassium feldspar phenocrysts and light pink tone; moderate weathering. Phaneritic texture and composed mainly of quartz, plagioclase, feldspar, biotite, and muscovite. There is no orientation.
	RIA01	Guadalupe Chapel (Rianxo)	Granite with fine-medium grain. Whitish tone and equigranular texture. Composed mainly of quartz, plagioclase, feldspar, and muscovite. Slight orientation marked by micas.
	RIA06	Church of San Salvador de Taragoña (Rianxo)	Medium-grained Granite with whitish and pink tones. Equigranular texture. Composed mainly of quartz, plagioclase, feldspar, and muscovite. Slight orientation marked by micas.
	PO4	Church of Santa Martiña de Xuño (Porto Son)	Medium- to coarse-grained granite with a light gray tone. Low-medium weathering. Phaneritic, equigranular, and composed mainly of quartz, plagioclase, feldspar, biotite, and muscovite.
	NO4B	Church Santa Maria de Barro (Noia)	Medium- to coarse-grained granite with potassium feldspar phenocrysts and light pink tone, moderate weathering. Phaneritic texture and composed mainly of quartz, plagioclase, feldspar, biotite (sometimes phenocrysts), muscovite, and dispersed mafic minerals (amphibole).



Figure 4. (a) The Pazo de Goiáns, (b) Church of Santa Mariña de Xuño, (c) Church of Taragoña, (d) Church of Macenda, (e) Church of Santa Maria de Barro, (f) Guadalupe Chapel.

4. Methodology

4.1. Sampling Procedure and Experimental Process

First, a macroscopic characterization of outcrops and monuments was carried out to discriminate morphology, grain size, texture, color, mineral composition, alterations, and foliation of the material.

A total of 12 representative granite outcrops (6) and monument (6) samples were collected. For instrumental analyses, a portion of the dried samples was separated and homogenized to be analyzed by X-ray diffraction and X-ray fluorescence spectroscopy.

4.2. X-ray Diffraction (XRD)

Mineralogical composition was also determined by means of X-ray diffraction (XRD) of bulk samples. We prepared fine powders ($<150\ \mu\text{m}$) from representative samples by pulverizing in an alumina mortar until the powdered material easily passed through a 150- μm sieve. Samples were analyzed using a D5000 Siemens (Munich, Germany) powder diffractometer with Ni-filtered $\text{Cu K}\alpha$ radiation. Measurements were performed at 40 kV, 30 mA, with a step size of 0.050° and step time of 2.5 s, a 2θ range of $2\text{--}80^\circ$. The patterns of XRD in all samples have been recorded with DIFFRAC Plus XRD software.

4.3. X-ray Fluorescence (XRF)

The elemental analyses of the samples were performed using a Fluorescence Spectrometer S4 Pioneer of wavelength dispersion Bruker-Nonius under helium purge. The bulk geochemical composition was analyzed on pressed powder pellets by X-ray fluorescence (XRF). All XRF analyses were measured in triplicate. The analyzer was calibrated using USGS standard reference materials. Statistical and geochemical analyzes were performed with the GCDKIT 6.0 program.

4.4. Petrographic Microscopy (PM)

Representative thin sections of outcrop samples were prepared for the petrographic study. The thin sections ($30\ \mu\text{m}$) were observed under a polarizing microscope (Leica DM4500 P, Wetzlar, Germany, University of A Coruña), equipped with a digital FireWire

Camera (Leica DFC 290 HD, Wetzlar, Germany) that worked with the Leica application suite software LAS 4.

5. Results

5.1. Macroscopic Characterization of the Samples

The description of samples includes criteria of morphology, grain size, texture, color, mineral composition, alterations, and foliation. Distinct types of granite have been identified, corresponding to the geological framework of the area (Figure 5).

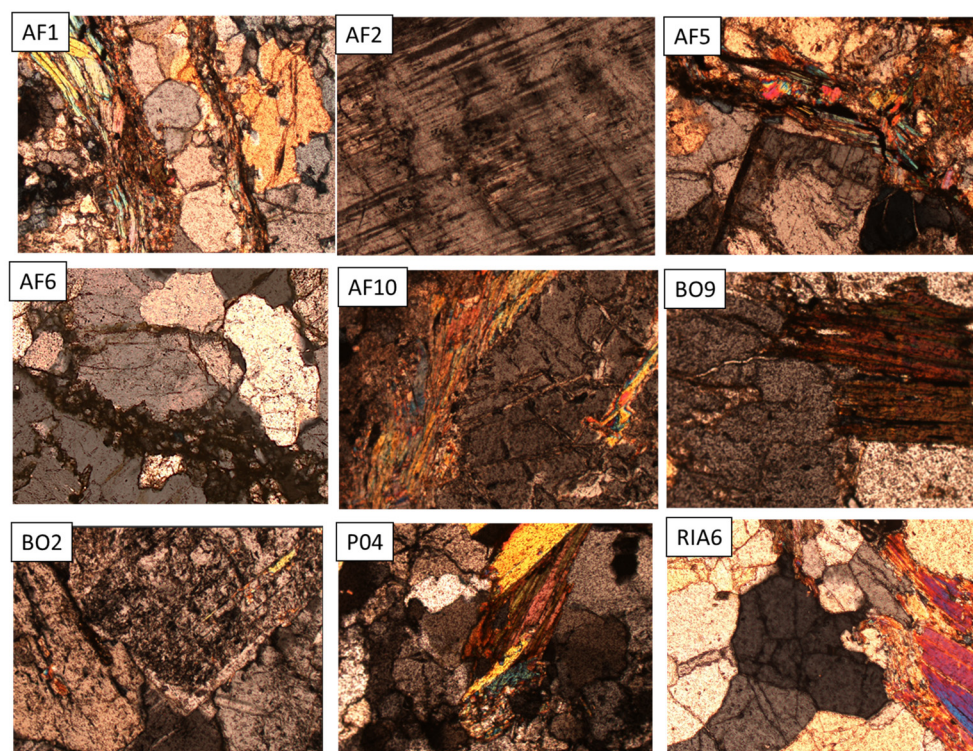


Figure 5. Photomicrographs of granite samples from the study area. All of them were taken with crossed nicols at 40× magnification.

The representative samples of the monuments show macroscopic evidence comparable to some granite outcrops. Samples BO2, BO9, and PO4 are medium- to coarse-grained two-mica granites like AF6, AF5, and AF2, while RIA6 and RIA1 samples present oriented texture and light tonality like the outcrops AF13 and AF10. Finally, sample NO4 is characterized by being medium-grained granite but with biotite megacrystals.

5.2. Petrographic Characteristics

The petrographic characteristics (textures and minerals) of samples taken from both outcrops and monuments are described in a thin section and shown in Figure 5. Moreover, details of the general microscopy and petrography of samples BO9A and BO2 have been previously studied to evaluate the weathering processes in the rocks [6]. Thus, similar petrographic characteristics are evident in the samples: hypidiomorphic and porphyritic texture, composed mainly of quartz, potassium feldspar, plagioclase, biotite, and muscovite.

The AF1 sample exhibits a light orientation generally parallel to sub-parallel alignment (Figure 5) attributed to the crystals of muscovite. Quartz crystals are anhedral, very strained, and commonly show undulatory extinction, generally in mosaics. Plagioclase shows polysynthetic twinning, perititic texture, and the presence of microfractures. It is common for the edges to have simplectitic textures. Biotite occurs in tabular and subhedral crystals

with a very marked pleochroism, dark to light brown, and partially includes anhedral to subhedral apatites. Clinozoisite and opaques are also present.

AF2 granite shows large crystals of quartz (with undulatory extinction), plagioclase, biotite, and feldspar that include small crystals of muscovite. The megacrystals are twinned perthitic microclines in Carlsbad (Figure 5) and include plagioclase and micas. Plagioclase is polysynthetically twinned, and there is sericitization. As for micas, muscovite is strongly pleochroic, and biotite appears altered to chlorite. Also, biotite occurs in tabular crystals with very marked pleochroism, dark to light brown, frequently with zircon and opaque inclusions.

AF5 granite shows quite an altered, hypidiomorphic texture with a porphyry tendency (Figure 5). Quartz with wave extinction is frequently fractured. Plagioclase shows polysynthetic twinning and irregular edges. Potassium feldspar with quartz and plagioclase inclusions is also present. Perthitic texture. Furthermore, perthitic texture, titanite, opaques, tourmaline, muscovite flakes with a certain orientation, and chloritized biotite are also observed.

AF6 Granite shows mainly feldspar with a poikilitic texture and quartz and mica inclusions. Sericitization in plagioclase and wedge finishes due to the effect of deformation and partially recrystallized zones is observed. Muscovite crystals have a platy habit with corroded edges, are either interstitial or occur as inclusions in other crystals, and generally show a preferred orientation. Intergrowth-textured quartz and chloritized biotite are also present. (Figure 5).

The AF10 granite sample exhibits a granular, lepidoblastic texture. Muscovite is a very altered form of lepidoblasts and subhedral crystals (Figure 5). Quartz crystals are anhedral, very strained, and commonly show undulatory extinction. It shows a perthitic texture (Figure 5). Presence of clinozoisite and opaques. K-feldspar crystals are stained and have a perthitic texture.

Samples BO2, BO9, and PO4 (Figure 5) were compared in texture and mineralogy with AF6 and AF2 samples due to their similarities, while the RIA6 sample shows texture and mineralogy similar to AF10, and this coincides so far with the macroscopic characteristics.

5.3. Chemical Composition

Table 2 shows the chemical composition of the rocks extracted from the different outcrops, comparing them with the samples from the monuments. The samples are granites with a very similar elemental composition, but some aspects can be differentiated: The SiO_2 and Al_2O_3 compositions of the monument samples are 68.3%–70.4% and 15.6%–17.5%, respectively, which is consistent with the composition of outcrop samples (65.5%–71.8% and 14.6% vs. 18.4%). Samples AF1, AF13, and RIA06 have the highest values of SiO_2 , while AF2, AF6, AF10, RIA1, PO4, and NO4B, BO2, and BO9AP show middle values between 67% and 69%. However, AF5 is different since it shows the lowest values of Si. Fe_2O_3 was observed below 1% in samples RIA6, RIA1, BO9A, and AF13, in which no (or very limited) presence of biotite or amphibole was observed. Na and Ca are earth elements that present greater migration in weathering processes [3]. In fact, the content of Na_2O is variable between 2.8% and 4.6%, and that of CaO is between 0.36% and 0.78%. K has moderate migration, and in these samples, the content of K_2O is 4.1%–5.6%. Potassium is found mainly in micas and potassium feldspars.

Table 2. Chemical composition of the samples in % by XRF.

		SiO ₂	Al ₂ O ₃	K ₂ O	Na ₂ O	Fe ₂ O ₃	CaO	MgO	P ₂ O ₅	TiO ₂	Rb ₂ O	ZnO	ZrO ₂	SrO	CuO	MnO	BaO	LOI	CIA	WIP
	Samples	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%
Quarries	AF1	71.8	14.6	5.0	2.8	2.0	0.44	0.53	0.40	0.33	0.027	0.015	0.018	0.005	0.006	<0.005	<0.008	2.0	63.9	57.2
	AF2	69.9	15.9	4.9	4.6	1.8	0.36	0.44	0	0.23	0.047	0.012	0.020	0.004	<0.005	<0.005	<0.008	1.6	61.7	66.5
	AF5	65.5	18.4	4.1	4.2	2.4	0.87	0.83	0.11	0.43	0.021	0.012	0.020	0.045	<0.005	0.028	0.083	2.8	66.7	59.0
	AF6	69.5	15.6	6.6	4.3	1.2	0.39	0.18	0.41	0.13	0.035	0.009	0.007	0.008	<0.005	<0.005	<0.008	1.6	58.0	74.1
	AF10	69.5	17.6	4.6	2.8	1.2	0.64	0.43	0.20	0.15	0.017	0.008	0.009	0.012	<0.005	<0.005	0.053	2.7	68.6	54.2
	AF13	71.3	15.7	5.2	4.4	0.92	0.65	0.18	0.27	0.10	0.041	0.011	0.006	0.008	<0.005	0.017	<0.008	1.1	60.5	67.9
Monuments	N04B	68.3	16.2	6.1	3.5	1.4	0.50	0.43	0.37	0.24	0.042	0.016	0.016	0.009	<0.005	<0.005	<0.008	1.8	61.59	70.0
	Ria01	69.5	17.5	5.3	3.6	1.0	0.60	0.26	0.34	0.13	0.036	0.011	0.007	0.007	<0.005	0.012	0.000	1.5	54.42	64.1
	PO4	69.5	17.1	4.8	4.4	1.3	0.26	0.24	0.37	0.11	0.039	0.011	0.006	0.005	<0.005	0.018	<0.008	1.7	64.38	64.2
	Ria06	70.4	15.6	4.8	5.3	0.98	0.62	0.23	0.42	0.10	0.034	0.009	0.006	0.007	<0.005	<0.005	<0.008	1.4	59.27	69.8
	BO9AP	67.9	17.4	6.2	5.1	0.77	0.41	0.26	0.40	0.11	0.036	0.010	0.007	0.008	0.008	<0.005	<0.008	1.3	59.77	79.6
	BO2	68.5	16.4	5.8	5.3	1.3	0.78	0.36	0.14	0.16	0.047	0.010	0.017	0.006	<0.005	0.046	<0.008	1.1	57.99	78.2

The degree and weathering intensity of the samples can be evaluated from the relationship between the alkaline elements and the alkaline earth elements (molar ratio), making use of the Chemical Alteration Index (CIA) [38–41] shown in Table 2 and calculated from the following formula:

$$\text{CIA} = [\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O})] \times 100$$

The fresh rocks have CIA values that oscillate between 40% and 50%. These values begin to increase according to the degree of alteration of the rock. In the samples studied, a range of CIA between 58% and 68.6% is observed; therefore, most of them present moderate weathering.

On the other hand, the Weathering Index of Parker (WIP), derived by Parker (1970) [42], was calculated from the following formula:

$$\text{WIP} = [(2\text{Na}_2\text{O}/0.35) + (\text{MgO}/0.90) + (2\text{K}_2\text{O}/0.25) + (\text{CaO}/0.70)]$$

The Weathering Index of Parker (WIP) is based upon the proportions of the major alkaline metals and their bond strength with oxygen, which is used as a weighting factor. Inversely, as the values decrease, the degree of weathering also decreases (Table 2). This trend is reflected in Figure 6, where both weathering indices are correlated. Furthermore, samples AF1, AF10, and Ria06 form another correlation.

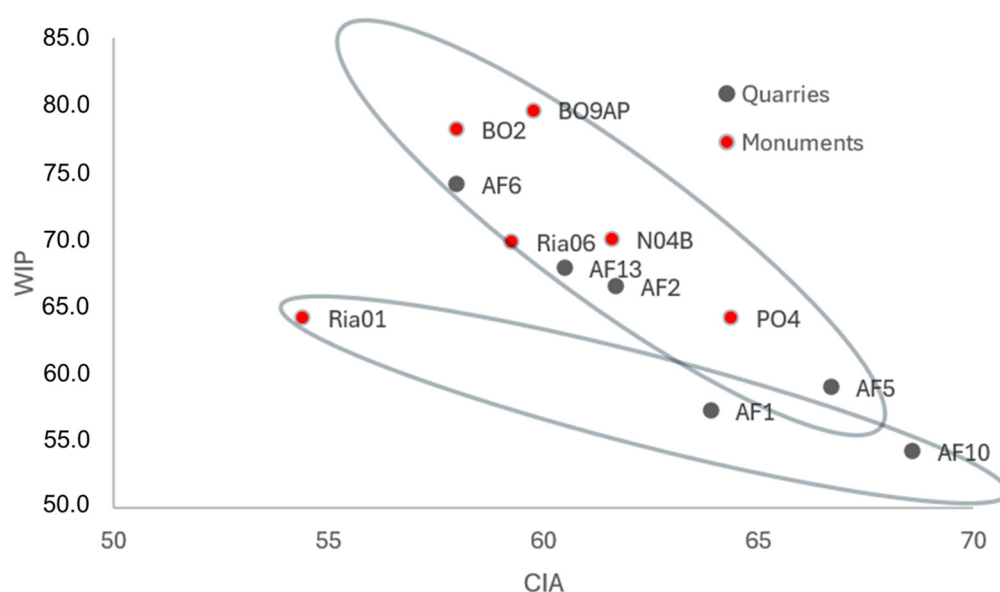


Figure 6. Relationship between the Chemical Alteration Index (CIA) and the Parker Index (WIP).

5.4. Mineralogical Composition

Figure 7 shows the mineralogical composition of the analyzed samples. In general, the granites show a similar XRD pattern, and the mineralogical phases identified are quartz, orthoclase, albite-type plagioclase, microcline, and micas, commonly muscovite. Sample AF13 contains these phases, with the exception of orthoclase. In particular, samples AF2, AF5, and AF6 record vermiculite (mica rich in iron or magnesium) and chlorite (chamosite). In the monument samples, chamosite was present in the BO2 samples.

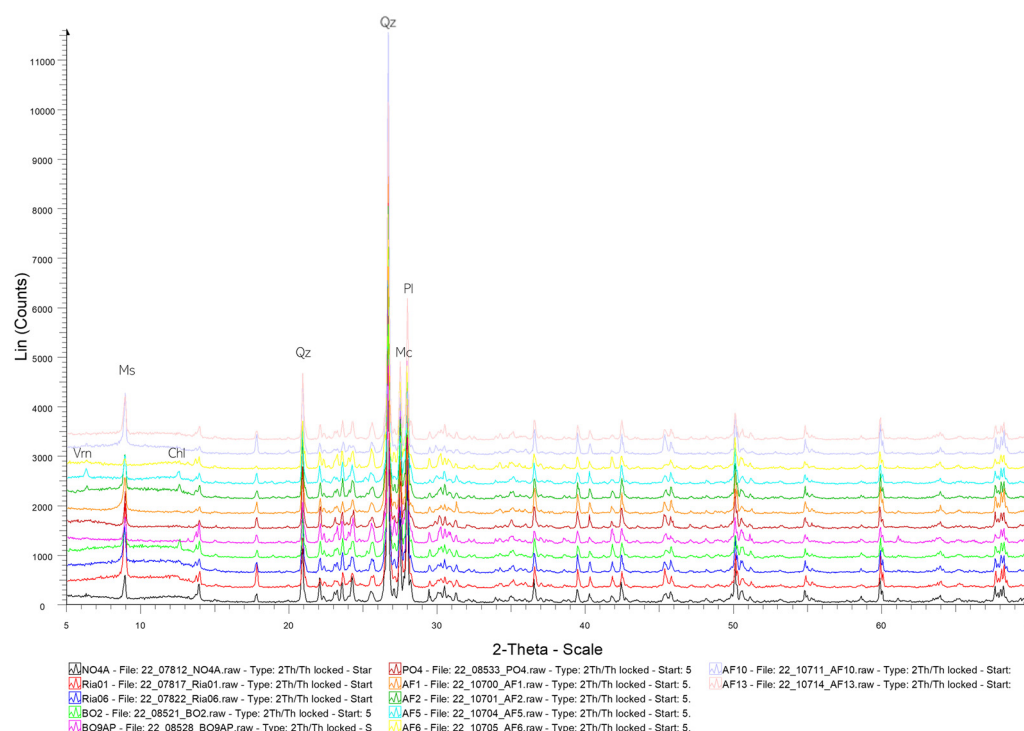


Figure 7. Identification of mineralogical phases by means of XRD (AF1, AF2, 5AF6, AF10, AF13; BO2, NO4B, RIA01, RIA06 and BO9AP). Mineral abbreviations: Quartz (Qtz), Muscovite (Ms), Microcline (Mc), plagioclase (Pl), Vermiculite (Vm), and Chlorite (Chl).

6. Discussion

Given that many quarries are local and managed in a family environment, it is a complex task to establish the exact location of extraction areas in outcrops and spaces that were continuously reused in historical times. The features of historical quarries may have undergone significant changes or even disappeared over time, making their interpretation difficult. In Spain, the efforts made to locate these quarries have allowed them to be classified according to the degree of knowledge, activity, quarry protection, dissemination, and disclosure activities [43]. In this case, the majority of the old granite extraction quarries on the Barbanza Peninsula are considered to be lost, unknown, and abandoned quarries with no administrative protection measures.

There is a correlation between the mineralogical and chemical analyses of the stone material of the historical monuments on the Barbanza Peninsula and the rocks of the selected outcrops. For the outcrop samples, three fundamental aspects were considered:

1. The documentary information on the historical mining sites (which involve small local quarries spread throughout the peninsula with very generic location data [Figure 2]).
2. Exploitation criteria such as geometric holes with curved edges (Figure 3a,b,e) [44] and small-cut stone blocks could raise suspicions of mining activity (Case of AF5) in the areas adjacent to the Petroglyphs of A Gurita. Furthermore, proximity to communication routes is also considered, taking into account transportation to the locations.
3. Geological criteria take into account the types of granite in the areas surrounding the monuments (whether or not the lithology matches the stone of the buildings).

The Barbanza-type granite is the most abundant within the geological context of the area and also probably the most used as a construction material according to the characteristics of the type of granite used in several monuments and previously reported [5,6]. It is a medium- to coarse-grained two-mica granite like sample AF6, and texturally and mineralogically, they are very similar to samples BO9 and PO4. In addition, very close to the AF6 outcrop, small local quarries are reported (more specifically in the Baroña area, Figure 2). Thus, the local differences between some monuments on the Barbanza Peninsula

are due to this variation in the facies of this same granite. On the other hand, other local differences could be related to the existence of several quarries for the same type of granite. For example, mining activity is reported in Xuño and Baroña, and both could be the same type of granite.

On the other hand, outcrop AF2 corresponds to the Confurco granite and is texturally and mineralogically comparable to sample BO2. In both samples, chlorite, feldspar phenocrysts, and the distinctive plaid (cross-hatched) twinning (also known as tartan twinning) were identified, giving a zebra stripe appearance in the microcline. At the same time, historical quarries are reported in this area (Figure 2), and the church of San Xoán de Macenda (Sample BO2) is very close, so it would make sense that the granite used is from this area. Finally, the similarity of samples AF1 and AF10 with RIA01 and RIA06 (medium-grained two-mica granite with light tones and marked orientation) corresponds with the lithological descriptions of the geological strip corresponding to the Noya Complex. Quarries are reported in the region of the outcrop of sample AF13, which also appears to be a rock extraction site (Figure 3). Although AF13 is chemically similar to these samples (geologically located within the Noya Complex), the textural characteristics are different, as it does not have a marked orientation and the grain size is medium to coarse. AF5 is differentiable since it shows the lowest values of Si, and not much correspondence was found with the selected churches.

The extraction, cutting, and finishing processes often develop microcracks, altering the physical and mechanical properties of the rock. The CIA and WIP values show low to moderate weathering and agree with the petrographic images of the samples. In cases of greater alteration, the plagioclase is slightly transformed into sericite, and in some cases, the biotite presents chloritization. Some crystals undergo a fracturing and precipitation process along their internal microfractures. The CIA has been studied at other monuments on the Barbanza Peninsula, and they are similar to these values [6]. Although the CIA values were very similar between samples, the lowest value also corresponds to the oldest churches, in this case the Church of Santa Martiña de Xuño (Porto Son). Therefore, construction time plays a fundamental role in stone wealth. On the other hand, when performing the correlation between both indices, two groups are shown; one of them corresponds to samples AF1, AF10, and RIA06, whose similarities have been previously mentioned in mineralogical and petrographic terms.

In addition, being a coastal city, the stone monuments are constantly subjected to sea foam, which damages their structure due to the crystallization of salts [6,45–47]. It is probable that the Na and K values in the material of the monuments are slightly higher for this reason [5]. The content of these elements could be affected by the direct attack of rainwater (directly on the outcrops), the action of the wind and sea salts mainly on the monuments, and biological colonization [5]. Granites like NO4B, BO9, and BO2 show the highest values of K_2O , and they are similar to AF6. It also coincides that these samples contain large amounts of feldspar phenocrysts. Moreover, the variations in weathering are mainly due to the different sizes, shapes, and orientations of feldspar grains.

The oxidation of minerals rich in iron, such as biotite, causes the stone to turn yellow (like AF1). Moreover, this yellowing can also result from reactions with sulfur dioxide (SO_2) present in the atmosphere and also with ozone (O_3), which acts as an oxidant and can interact with nitrogen oxides, generating nitrogen dioxide (NO_2) [48]. On the other hand, regarding the degree of alteration, superficially, the granitic rocks of the monuments exposed on the facades present greater coatings than in the natural outcrops, due to greater exposure to anthropogenic factors such as air pollution, construction materials, and the structure of the monument [17,49].

Vermiculite present in AF2, AF5, and AF6 is a typical product of the transformation of biotites in an acid environment and in the presence of organic matter [50,51]. The presence of this mineral is possible, taking into account that the samples have been taken at a shallow depth. Considering that in other studies, alteration minerals such as kaolinite and vermiculite are more abundant at the interface and are attributed to the decrease in pH

caused by the acidic substances secreted by the lichens [52], it favors the replacement of K⁺ ions in the interlayer with hydroxylated forms of aluminum. On the other hand, the samples of the monuments are collected in deep quarries, and the alteration products are more typical of primary minerals. In this case, the most common types of hydrothermal alteration were found: sericitization of plagioclase and chloritization of biotite [53]. Additionally, the rock deformation processes in some samples are evident in the orientation trend of the muscovite, mainly in the majority of the samples.

The overall analysis allows us to establish similarities between the outcrops and samples. So, it is considered that the material used for construction has a local origin and is attributed to small quarries spread over a wide area within the peninsula.

7. Conclusions

There are very few research studies that have focused on the characterization of the origin of stone monuments in Galicia. In most cases, the results are only disseminated at the national level, despite the cultural richness that surrounds this region. In this manuscript, six samples extracted from outcrops on the Barbanza Peninsula were compared with six granite samples on the facades of monuments of cultural heritage on the Barbanza Peninsula. This study represents an attempt to address whether the origin is really local to the Barbanza Peninsula or whether the stone was brought from more distant places, based on knowledge of the geological context of the study area. The chemical-mineralogical studies revealed that they have been extracted from local and small quarries, spread over a wide area within the Peninsula itself.

It is beyond the scope of this document to provide a comprehensive review of extraction points. However, the article, focused on the geological perspective, is a call for a multidisciplinary approach to the detailed promotion of these historic quarries, which can offer new perspectives for the promotion of cultural and nature tourism. Knowledge of the original material presents a unique opportunity for restoration and/or reconstruction of monuments, which allows for maintaining their conceptual and constructive homogeneity.

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