

Article

# Petrographic Markers for Archaeometric Identification of Montjuïc Sandstone, the Flagship Stone of Barcelona (NE Spain)

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**Abstract:** The present study deals with a particular clastic rock from the Montjuïc hill exploited since Roman times in Barcino (present-day Barcelona (NE Spain)). Polarized and cathodoluminescence microscopies have been used to describe the main petrographic features of Montjuïc sandstones. Several characteristic provenance markers have been identified; among them the most specifically restricted to Montjuïc sandstone are the K-feldspar clasts with authigenic overgrowths. A petrographic survey oriented to the detection of such markers has been fruitfully applied to sculptures, architectural elements, mosaics, and pottery. The petrographic approach has demonstrated that some Roman heritage materials had been erroneously assigned to Montjuïc sandstone and the revision of all the pieces macroscopically assigned to this provenance is advised. The use of Montjuïc sandstone in Roman tesserae has been reported for the first time with interesting implications on previously unreported evidence of Roman extraction at the bottom part of the Montjuïc cliff. Finally, Montjuïc crushed sandstone used as pottery temper has been also reported in the productions of a medieval (12–13th century) workshop in Barcelona. This encourages the study of the distribution of pottery with this particular temper.

**Keywords:** archaeometric provenance study; petrography; sandstone; authigenic cement; Roman period; sculpture; mosaics; temper

## 1. Introduction

Archaeometry is a multidisciplinary research area where scientific methods and techniques originally developed for other scientific areas are adapted to the analyses of archaeological finds and other heritage materials. One of its main branches encompasses the physical and geochemical analyses applied to provenance studies. These involve characterization and location of the natural sources of the studied materials [1]. Provenance studies are well developed for pottery artefacts with the usual approach being the chemical [2–5] and/or petrographical analyses [6–8]. However, provenance studies are not so well established and generalized for stone objects.

Stone is actually an essential cultural heritage material that can be found in a very large variety of forms and sizes: Cyclopedian, ashlar and other masonries, columns, sculptures, sarcophagi, stone mills, decorative and/or functional objects, opus sectile, tessellated mosaics, gravel- and

sand-sized inclusions in mortars, pottery, etc. To undertake archaeometric provenance studies of stone materials, the chemical approach (often based on trace element analyses) is the common choice for small objects made of gemstones consisting of one or few minerals [9–11] and for objects made of volcanic rocks with vitreous or microcrystalline components [12–14]. Regarding other stones and uses, the usual approach is a standard petrographic characterization [6] often combined with X-ray diffraction and/or chemical analyses [15–17]. For white marbles, a multitechnique approach has been developed including petrography, chemical analyses, cathodoluminescence, and particularly stable isotopes [18–21]. Besides marbles, gemstones, and volcanic materials, the provenance studies for stone materials are relatively scarce in scientific literature [22] due to the difficulty of finding robust provenance markers. A particularly complicated case of provenance determination is that of the clastic rocks because these are inherently formed by fragments of multiple sources. Statistical counting of components and evaluation of grain roundness are commonly applied in the field of sedimentary petrography [23] to undertake sedimentary provenance studies (identification of sediment sources and transport distances) but this represents a provenance marker for the sedimentary rock itself only in very particular cases. A much more effective provenance marker for a clastic rock would be a particular feature of the cement formed during the lithification process such as authigenic overgrowths [24].

In this paper we present a case study of robust identification of provenance markers for a particular clastic rock (Montjuïc sandstone) exploited since Roman times in Barcino, i.e., what is now Barcelona (NE Spain). The paper firstly presents the historical uses of this sandstone and then detailed geological and petrographic data is given stressing the relevant provenance markers that can be used to identify this sandstone in heritage materials. Several examples of this archaeometric identification are presented, including Roman sculptures and figurative reliefs, tesserae from a mosaic, and pottery shards. The archaeological implications of positive and negative identification on the studied heritage materials are also discussed.

## 2. Montjuïc Sandstone

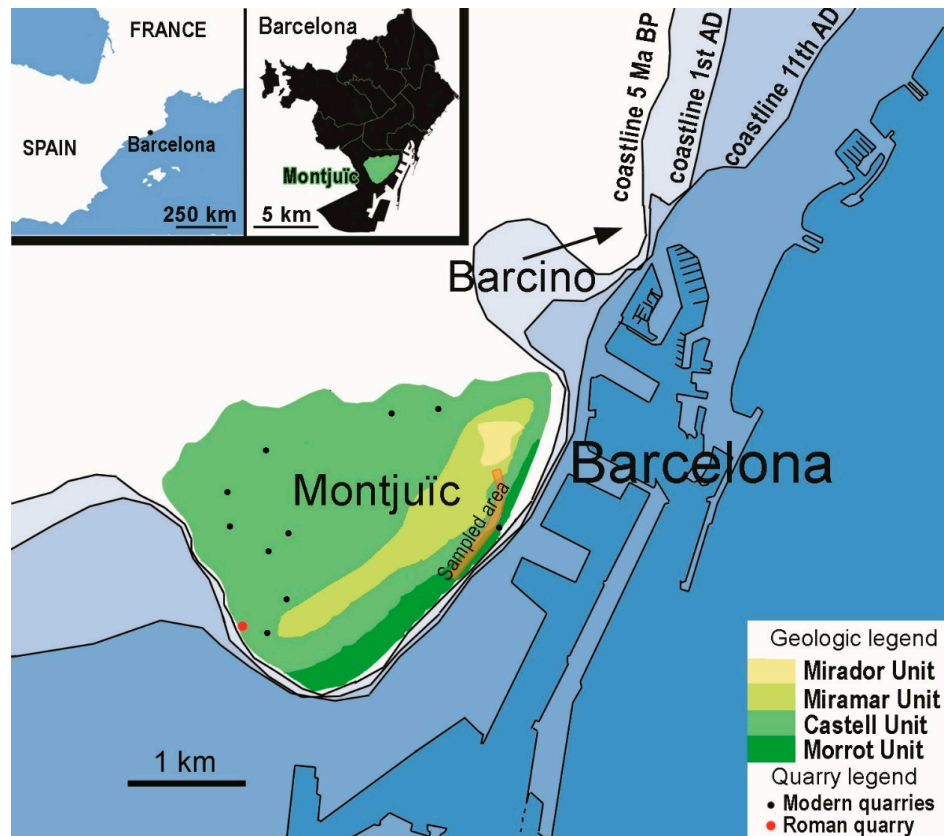
### 2.1. Historical Use

Barcelona (NE Spain) is a city developed in a plain surrounded by hills and sea. Among the hills, Montjuïc is one of the most remarkable because of its position on the seafront, at the south of the city. The sandstones from Montjuïc have been exploited intensively until the mid-20th century. As a result of this, the morphology of the mountain has been modified and many quarry scars are still visible mainly in its western slope where some of them have been turned into facilities like arenas or theaters. During the recent exploitation two varieties were distinguished according their quality: *Blanquet* (Catalan for whitish) which was hard and compact, and *rebuig* (Catalan for reject) which was softer and much more friable. However the history of exploitation goes back to prehistorical times (as attested by an Epipaleolithic workshop for extraction of jasper [25]) and was already intensive during Roman times.

#### 2.1.1. Roman Quarry

The exploitation of Montjuïc stone began in antiquity as suggested by some evidence back to prehistoric settlements of the Neolithic and Bronze Age period [26]. However, the intensive exploitation of the Montjuïc stone dated back to the Roman times in correlation with the foundation of the colony of Iulia Faventia Paterna Barcino by the emperor Augustus around the year 15 BC to 10 BC (see Barcino location in Figure 1). Archaeological remains of the Roman period that appeared in the underground of the city seem to support this theory. In particular, some surveys carried out on the south-southwestern slope of the hill—near the streets *Negrell* and *Ferrocarrils Catalans*—showed an open-air stepped exploitation where prismatic blocks of stone were extracted [27]. This is the only known Roman quarry in Barcelona to date [28], (see red dot in Figure 1). During the years 1989 and 1990, the works carried out in occasion of the Olympic Games (1992) on this area unearthed 220 m<sup>2</sup> of the quarry, revealing a

quarry rock face of ca. 50 m long and 10 m high. Due to some restrictions of the infrastructure project, the final limit of the quarry could not be excavated. However, there is no doubt about it being a large open-air extraction of the local sandstone. The extraction work was carried out on the vertical as well as the horizontal plains and a minimum of three different work surfaces were identified. In the unearthed part of the quarry, it has been possible to differentiate three sectors where different rock extraction has been identified. This could correspond to different varieties of the local sandstone, which could imply different extraction methods [29] although the outcropping formation is always the same (see Castell Unit in Section 2.2). The extracted blocks were possibly transported by ship as the quarry was located at the coastline in the Roman period (Figure 1).



**Figure 1.** Geological map of the Montjuïc hill and its quarry sites. The position of Roman Barcino and the evolution of the coastline are also depicted. Upper insets: Location of Barcelona in Spain and location of Montjuïc within the Barcelona municipality.

The main use throughout history of the stone extracted from the Montjuïc quarries has undoubtedly been construction [30]. It has been the principal material used in the building of the Roman city walls [31]; the use of both *blanquet* and *rebuig* varieties is documented already. The preserved remains (part of the podium, four columns with their corresponding bases, capitals, and the architrave) of the Roman temple that stands in the heart of Barcelona's Gothic Quarter, in the courtyard of a house in *Paradís* street, are also carved in this stone [32]. Sculptures, all worked in local workshops and used for urban, public, and private buildings provide more evidence of the use of the Montjuïc sandstone [30,33,34]. Within this category, a subset worthy of note is represented by funerary monuments and inscriptions [30,35–38]. Such pieces are of exceptional importance to know the provincial Roman art of the region and in general of Hispania, and to establish comparison with the products that came out of the workshops in the north of Italy and south-east of Gaul [39–41]. In fact, it seems that since ancient times, there existed a sculpture school in Barcino that copied motifs and styles that arrived from the capital, and especially from the workshops of Narbonne [29].

### 2.1.2. Medieval and Modern Uses

The extraction of Montjuïc stone continued during the Middle Ages, reaching its maximum exploitation intensity from the second part of the 19th century thanks to major development of the city of Barcelona. The last active quarries were closed during the 1950s, marking the end of 2000 years of dependence on the mountain [42]. The Barcelona Cathedral (late 13–15th centuries), the churches of Santa Maria del Mar, Santa Maria del Pi (14th century), and Sant Pau del Camp (ninth century); the building bearing the Presidency of the Catalan Government (15–17th centuries) and the City Hall (14th century), the historical building of the University of Barcelona (late 19th century) or the Rec Comtal irrigation canal (10th century) are some examples of relevant constructions built using Montjuïc stone. But it is especially during the *Modernisme* period (late 19th to early 20th centuries) that we see a revival in the use of this material for the decorative architecture. Many modernist buildings of Barcelona were constructed using Montjuïc stone such as the Hospital de la Santa Creu i Sant Pau, Palau Güell, Sagrada Família Temple, Casa Batlló, Casa Milà, Casa Vicens, and the Park Güell [42–44]. Besides the function as construction material, another important use of the stone, documented since the 11th century, is that of making millstones, hand grinders, as well as runner stones [45]. They were commercialized not only in the areas around Barcelona and the immediate neighboring areas, but also in many other places such as Majorca and Murcia (Spain), Pays Catalan [46], and Provence (France) or Genoa (Italy) [47].

Finally, besides sandstones, another material extracted from the Montjuïc hill and with a small trade industry was that of *terra d'escudelles* (sand for cleaning purposes), the abrasive silt-sand material from Montjuïc was particularly useful for cleaning kitchen pots and pans. Its use is documented from 1586–1587 and lasted until the mid-20th century [48].

### 2.2. Geology and Petrography

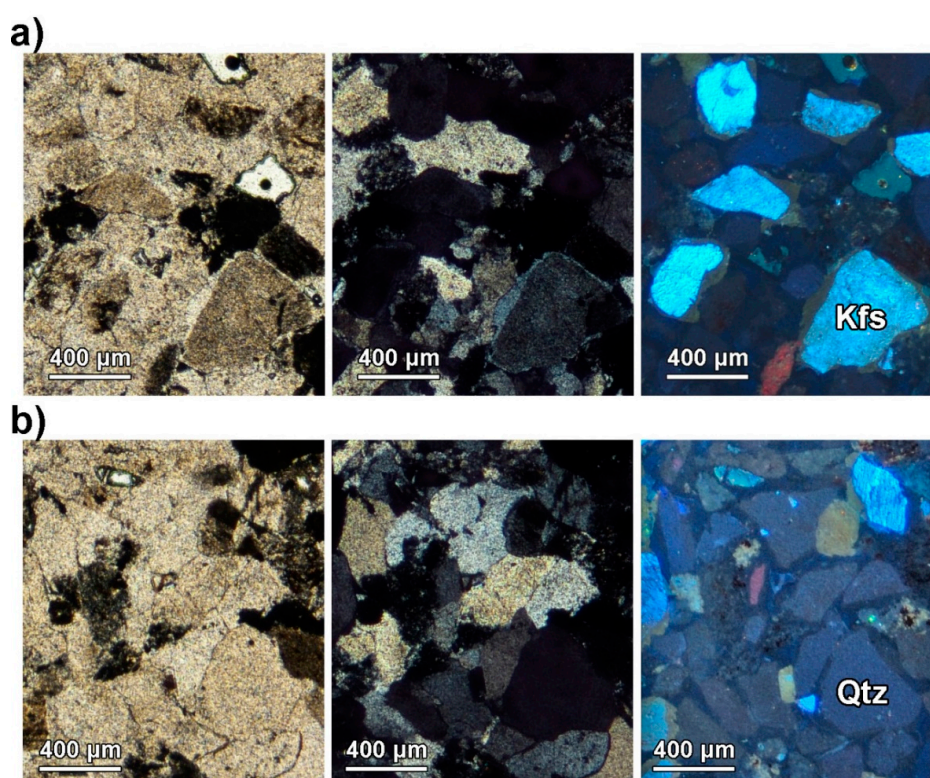
Geologically the Montjuïc hill is a tilted block situated between the Collserola Mountain (made of Palaeozoic rocks that outcrop at the north-west of the city) and the Barcelona half-graben (an offshore Neogene depression roughly parallel to the coastline). The hill towers over the city plain which is formed by marine Pliocene and Quaternary sediments [49,50]. In contrast with the plain, the hill is made of an alternation of older (middle Miocene, Langhian to Serravallian) conglomerates, sandstones, and accessorially mudstones that formed by diagenesis of delta sediments; these were mainly eroded from the Collserola mountain [51].

The geology and petrography of Montjuïc has been a subject extensively investigated previously by many authors and one of us (D.P.) has decisively contributed to it. The materials outcropping at Montjuïc have a thickness that exceeds 200 m and they have been lithostratigraphically divided into four units [50], from base to top: (1) The Morrot conglomerate and sandstone unit; (2) the Castell conglomerate, sandstone, and mudstone unit; (3) the Miramar marlstone unit, and (4) the Mirador conglomerate, sandstone, and mudstone unit. Sandstones appear in units (1), (2), and (4). However, from the areal point of view, unit (2), i.e., the Castell conglomerates, sandstones, and mudstones is the most represented unit (Figure 1). Covering around 70% of the Miocene outcrop at Montjuïc, the sandstones from unit (2) are the materials historically mainly exploited. All the other units outcrop basically in the cliff facing the sea.

The Montjuïc sandstones and conglomerates contain siliciclastic fragments essentially made up of quartz (Q), rock fragments (Rf), and feldspar (F) and they are characteristically cemented by silica and feldspar authigenic cements [52]. They can be classified as sublitharenites/rudites to litharenites/rudites and occasionally lithic wackes (if the matrix amounts >15%) according to Dott classification [53]. Carbonatic components are quite scarce (<5%) and they are restricted to the marliest strata in form of bioclasts (sparitic moldic-pore fillings) and micritic intraclasts. In the rest of the sandstones, carbonatic fragments have been dissolved, replaced by silica, or are simply absent.

The sandstones historically exploited (Castell unit) were deposited in a delta front environment, the clasts (Q 44–65%; Rf 14–45%; F 10–23%) are well sorted, displaying uniform medium or coarse

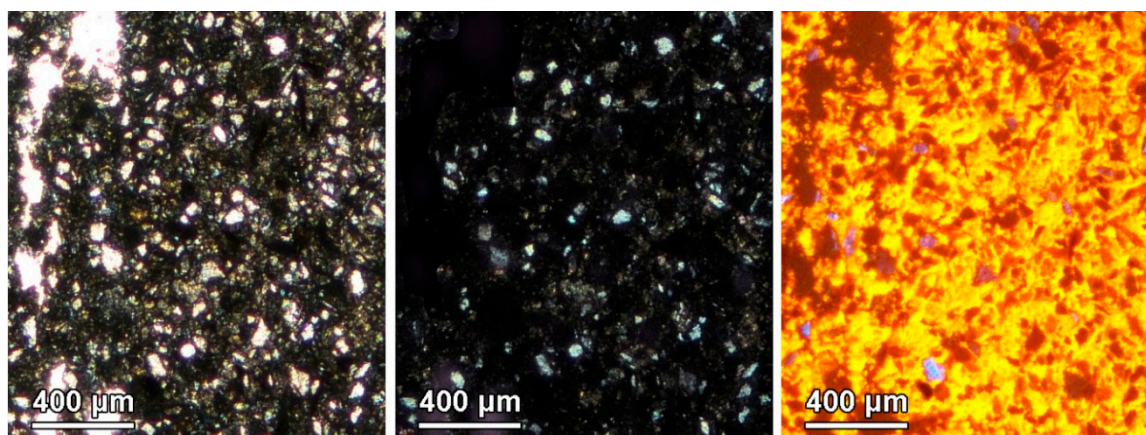
grain size and high roundness [51]. The rock fragments are both metamorphic and plutonic, prevailing the latter; the K-feldspars content (usually >12%) is always higher than plagioclase content (usually <6%). The matrix is almost absent and this has facilitated cementation processes. This cementation consists mainly in authigenic overgrowths on detrital K-feldspar and quartz grains [52]. Authigenic K-feldspar forms euhedral overgrowths (Figure 2a) defined by a slight optical discontinuity due to compositional differences between the grain and the cement and different alteration state. Authigenic quartz overgrowths are also euhedral but instead they show good optical continuity with the quartz grains (Figure 2b). The overgrowths enclose the clasts leaving little or no residual porosity, giving the sandstone a hard consistency and a massive appearance. A particularly useful technique to expose the existence of authigenic overgrowths is cathodoluminescence (Figure 2, images on the right).



**Figure 2.** Petrographic images of two samples of Montjuïc sandstone from the Castell Unit corresponding to macroscopic sample C1 (in Figure 5). Images include plane polarized light, PPL (left), cross polarized light, XPL (middle), and cathodoluminescence, CL (right) micrographs of the same areas. (a) K-feldspar (Kfs) clasts show distinguishable authigenic overgrowths, (b) quartz (Qtz) clasts also have authigenic overgrowths but these are only exposed using CL microscopy, shadows have been adjusted in the CL micrograph to emphasize quartz overgrowths.

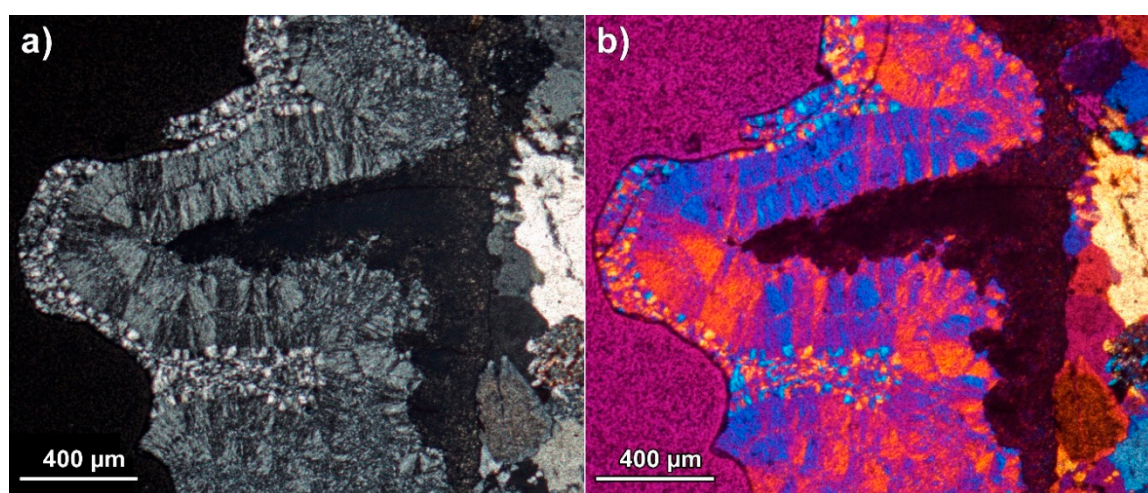
In contrast with the Castell sandstones, the underlying sandstones of the Morrot unit are poorly sorted and with an important proportion of interstitial matrix (up to 50%). The proportion among quartz, rock fragments, and feldspar is similar, but it is worth noting the total absence of plagioclase. Silicification is also important in this unit but authigenic overgrowths are less well developed due to the presence of interstitial matrix. This matrix has been replaced and transformed into opal and microquartz. Iron oxide cement is also common. The upper part of the Morrot unit is worthy of special mention; these are 11 m of marls and siltstones that sometimes have been described as a separate unit (see el Far unit in [51]). In these lower-sized sediments, metamorphic lithic fragments were more abundant, and their alteration contributed to the formation of pseudo-matrix that prevented silicification. Silicification in these layers appears to be irregular, some parts of the same layer are fully silicified and others unsilicified [54]. In unsilicified areas cementation is scarce and generally

forms layers or nodules (up to decimetric size, Figure 3) of calcite spar filling interparticle porosity [52]. Locally, this spar cement can fill porosity in previously silicified sandstones.

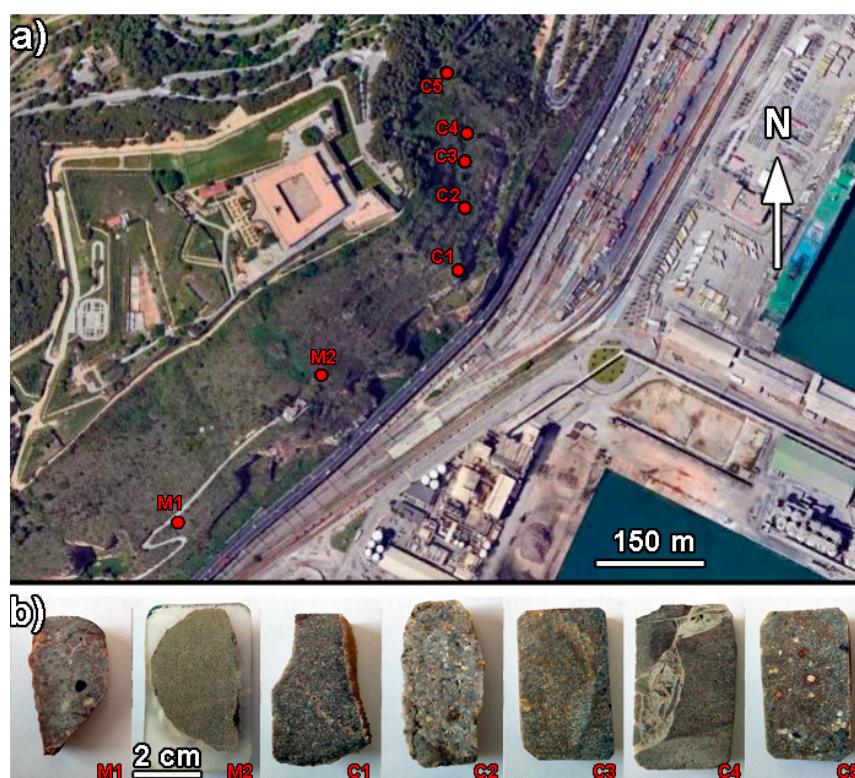


**Figure 3.** Petrographic images of a sample of an unsilicified carbonatic nodule from upper Morrot unit corresponding to macroscopic sample M2 (in Figure 5). Images include PPL (left), XPL (middle), and CL (right) micrographs of the same area. The presence of calcite cement is clearly seen by CL microscopy with its characteristic orange color.

Besides early silicification primarily filling the intergranular volume of the sandstones, another characteristic feature of Montjuïc sandstones is the presence of a late silicification filling rock fractures (see Figure 4 and C4 in Figure 5). During Pliocene times Montjuïc was uplifted and faulted [55] and the sandstones were pervasively affected by joints that were subsequently silicified. In the lower parts of the series (i.e., within the Morrot unit) the joints appear only filled with thin amounts of chalcedony, but at the upper part (i.e., the Castell unit and above) the joints exhibit a complex filling, which can be observed in the widest open joints, usually formed by amorphous opal (sometimes with barite), then chessboard chalcedony and microquartz, and finally fibrous chalcedony towards the center of the joint (Figure 4); for details of such late silicification see [56]. It is worth noting that chalcedony is predominantly of the length-fast (LF) type [57].



**Figure 4.** Petrographic images of postdiagenetical silicification in Montjuïc sandstone from the Castell unit corresponding to macroscopic sample C1 (in Figure 5). (a) XPL micrograph displaying complex joint filling (in this instance the sequence is amorphous opal, fibrous chalcedony, and chessboard chalcedony); (b) same image in XPL with compensator plate to highlight that chalcedony is basically of the length-fast type.



**Figure 5.** (a) Satellite image where some representative sampling sites (red dots) along the Monjuïc cliff have been highlighted; (b) the corresponding samples, belonging to Morrot (M1 and M2) and Castell (C1 to C5) units. To note: M2 corresponds to a calcite nodule from the upper Morrot unit; silicified late fractures are clearly seen in C4 sample and sample C1 (right side).

### 3. Materials and Methods

#### 3.1. Studied Materials

As part of the present study, field sampling was performed following a trail descending from up the hill all the way down to the city port, going across the different lithostratigraphic units (note sampled area in Figures 1 and 5). Some of the obtained geological samples have been used to illustrate the petrographic features of Montjuïc as presented in previous Section 2.2.

Several heritage materials were also sampled, including Roman sculptures and figurative reliefs presumably carved using Montjuïc sandstone. Two tesserae from a Roman mosaic and finally nineteen pottery shards from an excavated medieval workshop were also sampled. Table 1 summarizes all the sampled heritage materials; these will be presented in detail in the next subsections.

**Table 1.** Summary of the retrieved samples from the studied heritage materials.

Heritage Material	Typology	Chronology (Century AD)	Samples	Finding Site	Hosting Museum	ID
Sculptures	Togatus	1st	1	Sant Miquel (Barcelona)	MAC	19008
	Woman with toga	1st	1	Viladecols (Barcelona)	MAC	19018
	Togatus	1st	1	Sant Miquel (Barcelona)	MAC	19075

Table 1. Cont.

Heritage Material	Typology	Chronology (Century AD)	Samples	Finding Site	Hosting Museum	ID
Figurative reliefs	Stele	1st	1	Bellvitge-Z.Franca (Barcelona)	MdC	-
	Frieze	1st	1	Seu d'Ègara (Terrassa)	MdT	25992
	Acroterion	1st	1	Seu d'Ègara (Terrassa)	MdT	26000
Mosaics	-	4th	2	Ataül (Barcelona)	MAC	19004
Pottery	Oxidized ware	12–13th	6	Carders (Barcelona)	MUHBA	-
	Reduced ware	12–13th	8	Carders (Barcelona)	MUHBA	-
	Glazed reduced ware	12–13th	5	Carders (Barcelona)	MUHBA	-

### 3.1.1. Sampled Roman Sculptures and Figurative Reliefs

There are many archaeological pieces carved in the local sandstone of Montjuïc in the museums of Barcelona (mainly in the Archaeological Museum of Catalonia (MAC) and in the Museum of the History of Barcelona (MUHBA)). These are basically architectural elements, some of them decorated, that were often part of the necropolises located in the surroundings of Roman Barcelona and that were later reused as part of the fourth century AD fortified elements of the city [35,37]. Publications dealing with pieces usually assign it to Barcino workshops and it is often assumed that they are made of Montjuïc sandstone without any characterization of the material apart from visual inspection.

Besides Barcino, artworks presumably carved by itinerant workshops and artisans have also been found in nearby areas, such as Baetulo (now Badalona, 10 km from Barcino), Iluro (now Mataró, 30 km from Barcino), Egara (now Terrassa, 25 km from Barcino), and near Lauro (now Llerona, 30 km from Barcino) [41]. These have been identified as produced by Barcino artisans for iconographic and stylistic reasons but especially because, again, it is assumed that these were also carved from Montjuïc sandstone.

To corroborate that artworks found in Roman Barcelona (Barcino) and nearby areas are really carved in Montjuïc sandstone, six archaeological pieces have been selected (Figure 6). All of them were sampled with a small hammer and a sharp chisel to discreetly extract millimetric samples (~0.2 cm<sup>3</sup>) from old fracture surfaces.

Three of them are sculptures that were found reused as part of the city walls and they are presently deposited in the MAC Museum (Figure 6a–c). The other three are pieces (Figure 6d–f) which were found further from Barcino and they are deposited in two smaller museums around Barcelona.

The three sculptures from the MAC Museum feature a togatus (a person wearing a toga) and their catalogue numbers are MAC-19008, MAC-19018, and MAC-19075. All of them would be funerary sculptures, attributed to the first century AD and all are attributed to Montjuïc sandstone through macroscopic inspection [33]. MAC-19008 and MAC-19075 were found together in the south-western sector of the city walls (in the area of the present-day *Baixada de Sant Miquel* alley) in the 19th century. Both are the inferior fragment of a masculine togatus and have similar dimensions. MAC-19018 was found broken in three parts in the south-eastern sector of the city walls (in the area of the present-day *Baixada de Viladecols* alley). The parts have been reassembled showing a sculpture of a woman wearing a toga, it is also assumed that it was carved into Montjuïc sandstone [41].





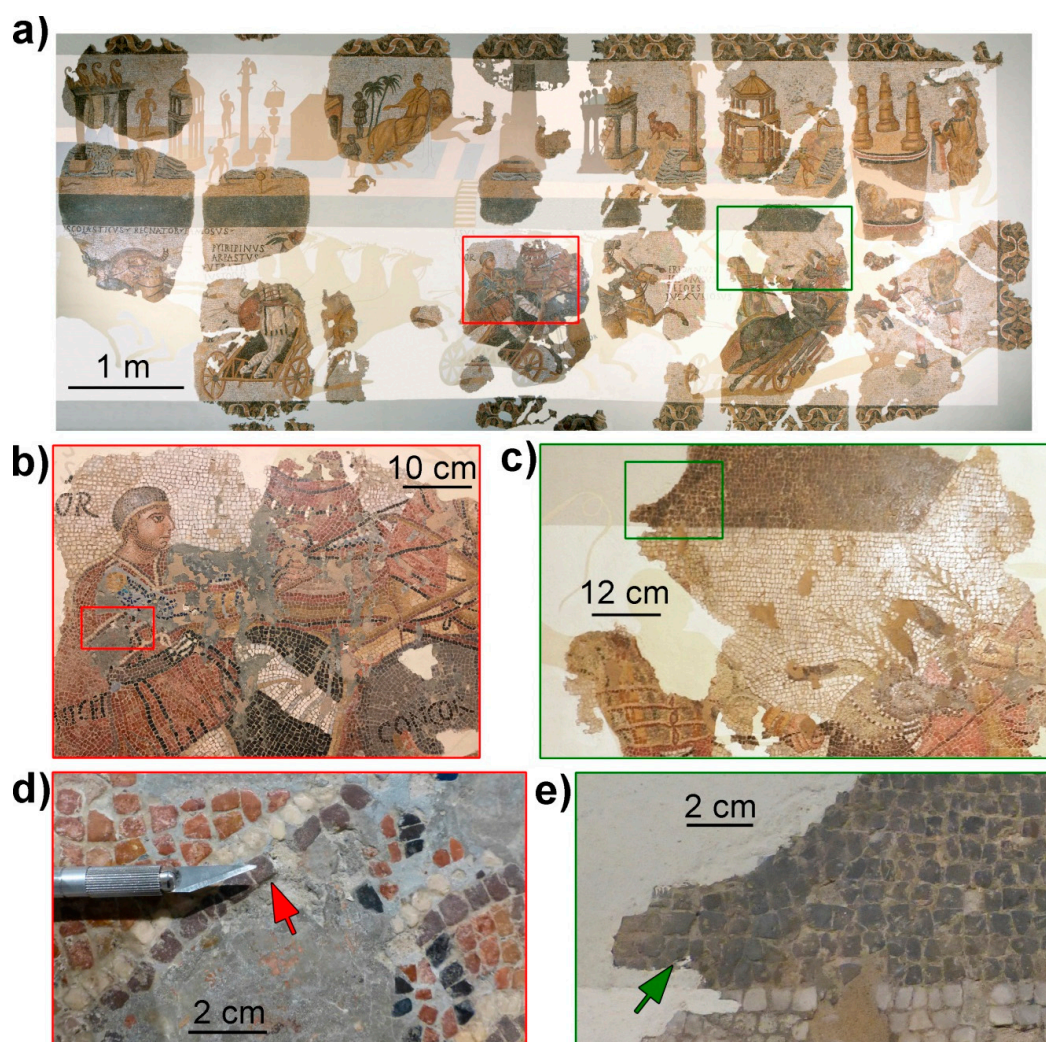
**Figure 6.** The sampled archaeological pieces. (a) Togatus sculpture MAC-19008 found in the south-western sector of the city walls of Barcino. (b) Sculpture of a woman wearing a toga, MAC-19018, found in the south-eastern sector of the city walls of Barcino. (c) Togatus sculpture MAC-19075 found together with MAC-19008. (d) Stele on display at MdC, found between Zona Franca and Bellvitge districts (Barcelona). (e) Frieze MdT-25992 decorated with acanthus scrolls motives, found in the Seu d'Ègara precinct (Terrassa). (f) Acroterion MdT-26000 featuring a satyr head, found together with MdT-25992. Red arrow indicates the location of the retrieved samples.

The pieces found outside Barcino were unearthed in the late 20th century; these are a funerary stele, an acroterion and a frieze. The stele (Figure 6d) was retrieved from an indeterminate place between Zona Franca and Bellvitge districts (6–7 km from Barcino) and it is presently on display at the museum of Cerdanyola (MdC) [35]. This stele has the particularity of depicting the deceased (which is rare in Hispania though common in Roman Gaul). It is attributed to the first century AD and the material is assumed to be Montjuïc sandstone [35]. The acroterion and the frieze (Figure 6e–f) were found together with signs of having been reused as parts of a press, in the medieval infillings of a silo [41,58], during the excavations in the Seu d'Ègara precinct (Terrassa, 25 km from Barcino). Both pieces belong to the Terrassa Museum with catalogue numbers MdT-25992 (frieze) and MdT-26000 (acroterion) and it is often assumed that both were part of a single funerary monument [41]. The monument would have been erected in the first century AD using Montjuïc sandstone [41]. The frieze contains a decoration with acanthus scrolls motifs very similar to several fragments found in the southern sector of the city walls of Barcino. These would correspond to a funerary monument also carved in Montjuïc sandstone and with the same chronology [30].

### 3.1.2. Sampled Roman Mosaic

Tessellated pavements (*opus tessellatum*) is another potential use for Montjuïc sandstones. From the first century BC onwards, these decorated floors experienced a steady expansion. The bichrome (black-and-white) mosaics were usually built using limestones (or occasionally volcanic rocks for black tesserae). However, the appearance of polychrome mosaics (in the second century AD) motivated the introduction of a variety of materials to cover the required wider range of colors [59]. Among them, artificial materials such as colored glass and pottery were used, but also other types of rocks like sandstones. Colorimetry measurements on Montjuïc sandstones from the Castell unit reveal that these sandstones can present a variety of colors with many types of gray, red, and brown, including greenish light olive brown.

The Archaeology Museum of Catalonia (MAC) hosts a large collection of Roman mosaics from Roman cities (mainly Barcino, but also Baetulo or Emporiae) and rustic villas. A large (7.98 m × 3.53 m) polychrome mosaic from Barcino depicting a scene of circus and known as “circus mosaic” was selected to investigate the possible use of Montjuïc sandstone as tesserae (Figure 7).

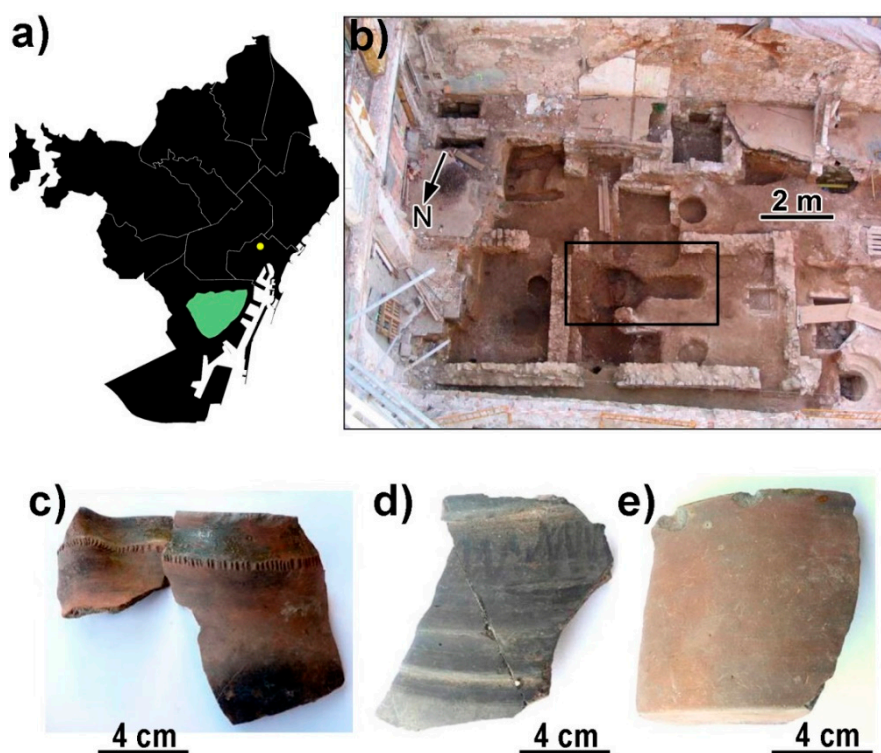


**Figure 7.** Sampled “circus mosaic” MAC-19004. (a) Full view of the mosaic (7.98 m × 3.53 m) including preserved fragments and interpreted parts. Details of (b) the central quadriga and (c) the spina framework. (d) Original location of the sampled dusky red tessella (red arrow). (e) Original location of the sampled greenish gray tessella.

This mosaic is on display at MAC (catalogue number MAC-19004); it was found in 1860 during the demolition works of the Minor Royal Palace (in the area of the present-day *Ataülff* street) and it would have been assembled during the fourth century AD [60]. A total of 13 tesserae of different colors were extracted from its position within the mosaic using a scalpel. They were cut to prepare thin sections to analyze their lithology, and the two that were lithologically identified as sandstones (dusky red and greenish gray colored) were selected to check for Montjuïc features.

### 3.1.3. Sampled Pottery

The particular petrographic markers identified in Montjuïc sandstones provide the opportunity to determine Montjuïc provenance even for very small lithic elements such as aplastic inclusions in pottery pastes. An exhaustive and oriented petrographic search on pottery productions from Barcelona could be done. Here, we investigated 19 pottery shards from a medieval workshop excavated in 2004 at *Carders* street. The workshop remains consist in a building that hosted a single kiln and it is assigned to the 12–13th century [61,62]. The workshop was located next to what had been the *Via Augusta* (a major road build by the Romans) and produced common glazed and unglazed ceramics in both reducing and oxidizing firing conditions (Figure 8). The pottery shards are currently stored in the Museu d’Història de Barcelona (MUHBA).



**Figure 8.** (a) Location of the Montjuïc hill (green) and the excavated workshop (yellow dot) within the Barcelona municipality. (b) Archaeological remains of the pottery production of the kiln. (c–e) Examples of the pottery production of the kiln: (c) Glazed jug, (d) pot, (e) serving pot.

### 3.2. Methods

Preliminary examination of all the samples was undertaken using a stereomicroscope. A total of 15 geological petrographic thin sections were cut from the samples collected during fieldwork. Thin section is the ideal preparation to observe the textural and compositional features of a rock using polarized and cathodoluminescence microscopes. The characteristic authigenic overgrowths found in Montjuïc sandstones (see Section 2.2) could hardly be identified without this sample preparation.

Therefore, small bits of the investigated heritage materials had to be removed to prepare thin sections with them (27 archaeological petrographic thin sections).

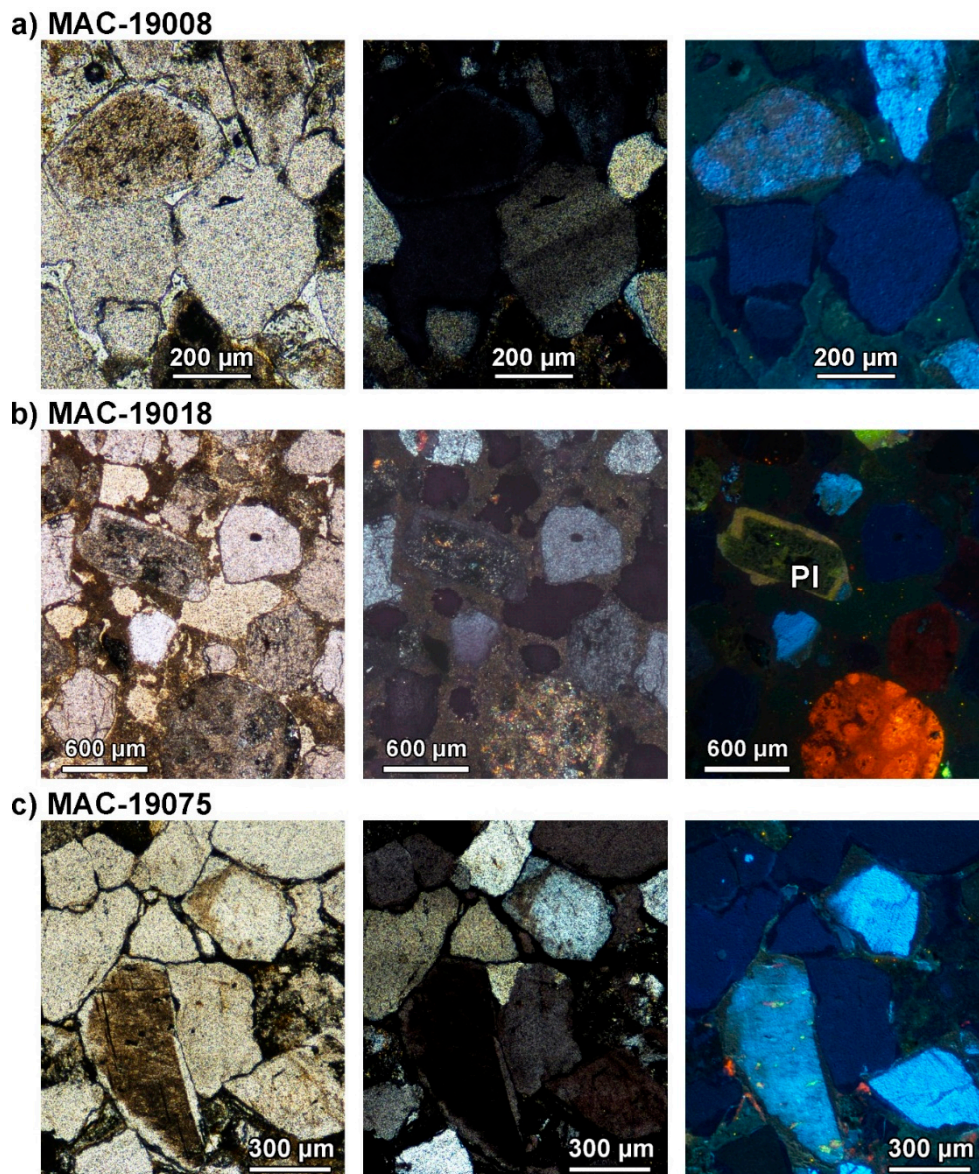
All the thin sections were examined under a polarizing microscope (Nikon Eclipse E600 POL, Tokyo, Japan) under plane polarized light (PPL) and cross polarized light (XPL). Additionally, uncovered thin sections were also analyzed by cathodoluminescence (CL) microscopy using a CL8200 Mk5-1 equipment (Cambridge Image Technology Ltd., Welwyn Garden City, UK), with operating conditions of 15–18 kV and gun current of ~200  $\mu$ A.

#### 4. Results

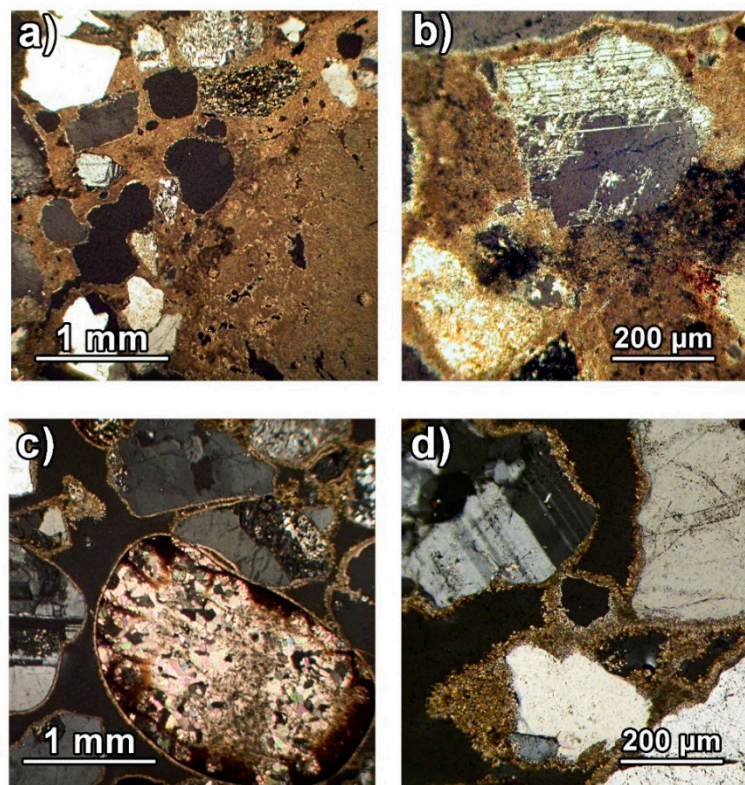
An accurate petrographic inspection of the archaeological samples was undertaken in order to detect the distinct provenance markers of Montjuïc sandstones. The aim was to corroborate previous macroscopic attribution to Montjuïc sandstone of several Roman sculptures and figurative reliefs and to unveil, for the first time, other historical uses of this material.

##### 4.1. Identification in Roman Sculptures

From the three sampled sculptures found reused in the city walls of Barcino, the detailed petrographic inspection revealed that both MAC-19008 and MAC-19075 are indeed carved into Montjuïc sandstone. Authigenic overgrowths are easily found both in K-feldspar and quartz clasts (Figure 9a,c), particularly visible using CT microscopy. The petrographic features are fully compatible with the historically exploited Castell unit. In contrast, for the MAC-19018 sculpture petrography (Figure 9b) does not match with Montjuïc sandstones for several reasons: (i) Authigenic overgrowths are totally absent around K-feldspar clasts and quartz; (ii) there are carbonatic clasts, both intraformational and extraformational, while in Montjuïc the first are rare and the latter completely absent; (iii) micritic-size cement occupies a significant part of the sample (>35%) intergranular space (Figure 10a), while such cement is rare in Montjuïc. Additionally, the composition of the MAC-19018 sculpture deviates from the typical Montjuïc sandstone (lithic fragments are too scarce and plagioclase too abundant compared with it). The petrographic features of the sandstone of sculpture MAC-19018 are consistent with a beachrock [63] (Figure 10). This type of recent coastal formation has been reported in core logging at drilling sites in the nearby Llobregat delta [64] and even at the Barcelona port (just in front of the Montjuïc cliff, see Figure 10c,d).



**Figure 9.** Petrographic images of the sampled sculptures from Barcino including PPL (left), XPL (middle), and CL (right) micrographs of the same areas. (a) sample of MAC-19008 togatus, exhibiting typical features of Montjuïc sandstone (K-feldspar and quartz clasts with authigenic overgrowths). (b) sample of MAC-19018 feminine sculpture, exhibiting micrite-size cement, plagioclase (PI) and a carbonatic intraclast made of pellets (orange in CL mode). (c) Sample of MAC-19008 togatus, exhibiting typical features of Montjuïc sandstone.

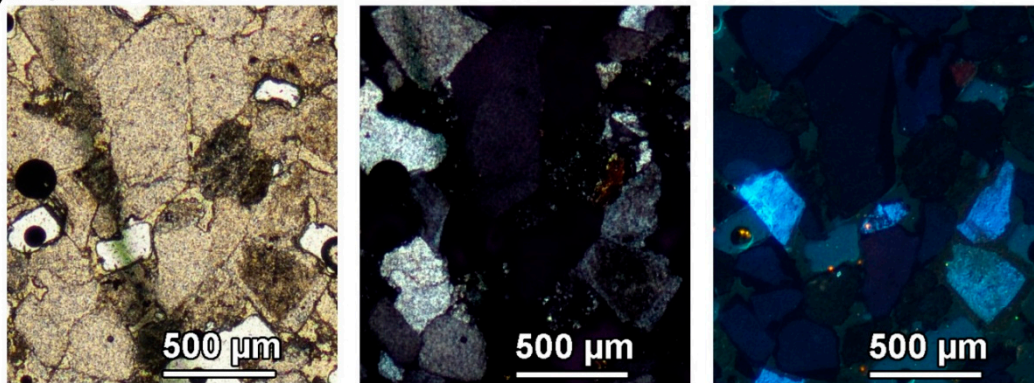


**Figure 10.** Comparison between the petrography of the sampled MAC-19018 feminine sculpture (a,b) and that of a beachrock lithology from a core drilled at the Barcelona port (c,d). Both samples show substantial intergranular space but in the archaeological sample this is more prominently filled with micritic-size cement. Composition for both samples is similar (including quartz, feldspars, and carbonatic components), the angularity of clasts is higher for the archaeological samples.

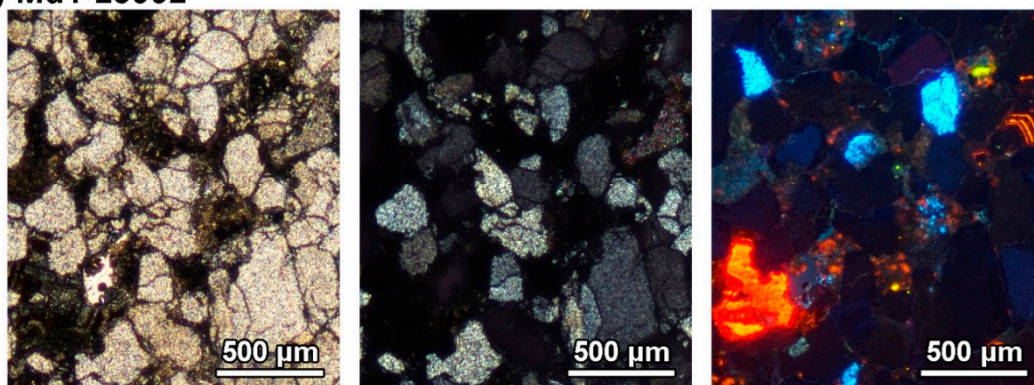
#### 4.2. Identification in Roman Figurative Reliefs

Concerning the figurative reliefs found outside Barcino, detailed petrographic inspection (Figure 11) revealed that both the stele (MdC) and the acroterion (MdT-26000) are certainly carved into Montjuïc sandstone; again the presence of authigenic overgrowths in K-feldspar is very conclusive and quartz clasts also exhibit similar overgrowths (Figure 11a,c), both clearly visible using CT microscopy. Thin section observation of the sampled frieze (MdT-25992) also reveals authigenic quartz overgrowths (Figure 11b, micrograph on the right). However, in this case the quartz overgrowth is preceded by a thin iron oxide coating (pore-lining) that allows the distinction between the clast and the quartz cement under PPL light (Figure 12a). This pore-lining is rarely observed in the Montjuïc sandstones. But again, the complete absence of K-feldspar overgrowths (Figure 11b, micrograph on the right) is particularly conclusive (this time to reject the Montjuïc origin). Besides that, other petrographic features of the MdT-25992 frieze are discordant with that of Montjuïc: On the one hand there are both sparitic and micritic carbonate components (Figure 11b), and on the other hand there is clear evidence of compaction (long contacts between grains instead of the typical point contacts of low compacted Montjuïc as well as fracturing of feldspar clasts (see Figure 12b). Additionally, the composition of the frieze is too rich in quartz grains compared to typical Montjuïc sandstone. The petrography of the frieze matches that of the Triassic quartzarenitic sandstones outcropping in the Catalan Coastal Ranges [65] (Buntsandstein and Middle Muschelkalk facies) not far from Barcelona.

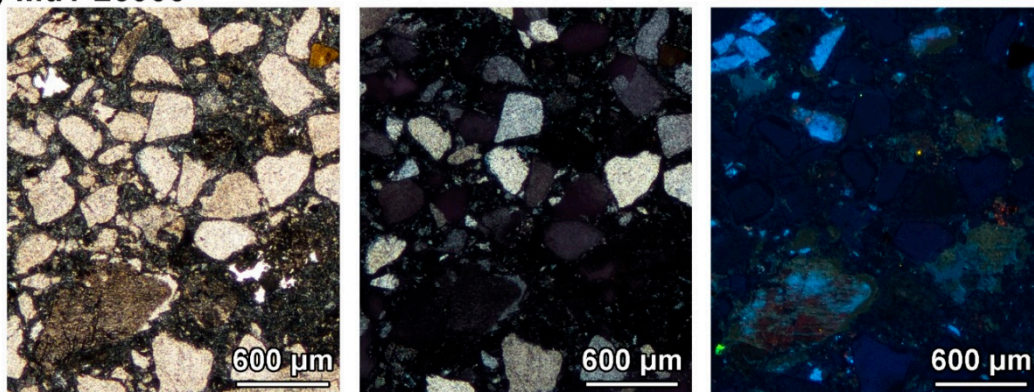
## a) MdC



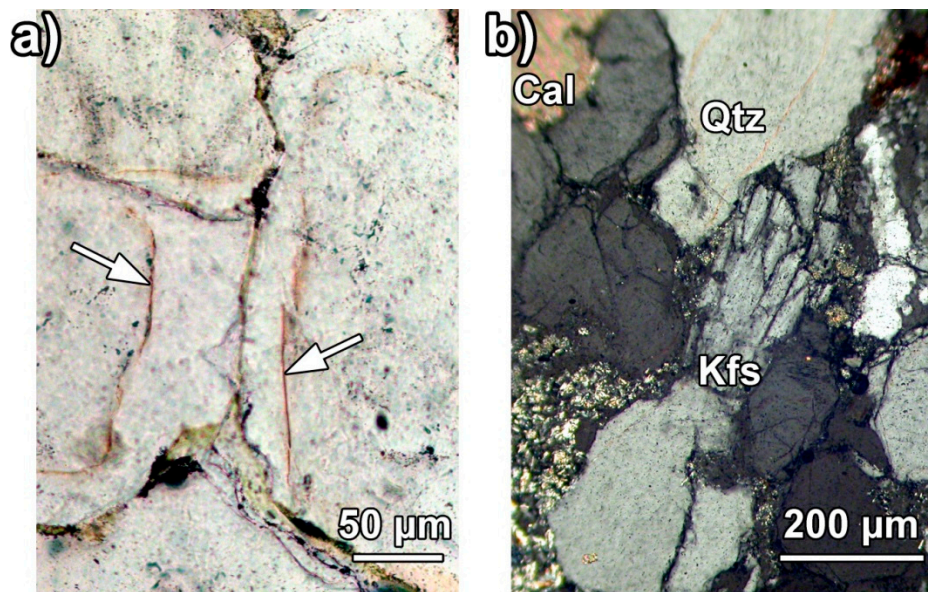
## b) MdT-25992



## c) MdT-26000



**Figure 11.** Petrographic images of the sampled pieces found outside Barcino including PPL (left), XPL (middle), and CL (right) micrographs of the same areas. (a) Sample of MdC stele, exhibiting typical features of Montjuïc sandstone (K-feldspar and quartz clasts with authigenic overgrowths). (b) Sample of MdT-25992 frieze, exhibiting K-feldspar and quartz clasts, only the latter with authigenic overgrowths and carbonate components (orange in CL mode). (c) Sample of MdT 26000 acroterion, exhibiting typical features of Montjuïc sandstone.



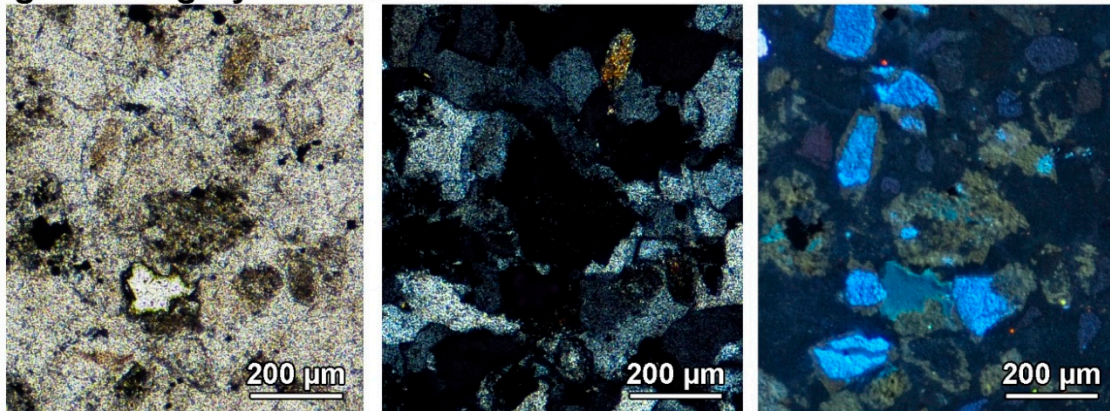
**Figure 12.** Petrographic features of the sampled MdT-25992 frieze. (a) PPL micrograph exhibiting iron oxide pore-lining (arrows) delimiting quartz clasts and quartz cement. (b) XPL micrograph presenting quartz (Qtz), calcite (Cal), and K-feldspar (Kfs) clasts, the latter appear fractured due to mechanical compaction.

#### 4.3. Identification in Roman Mosaic

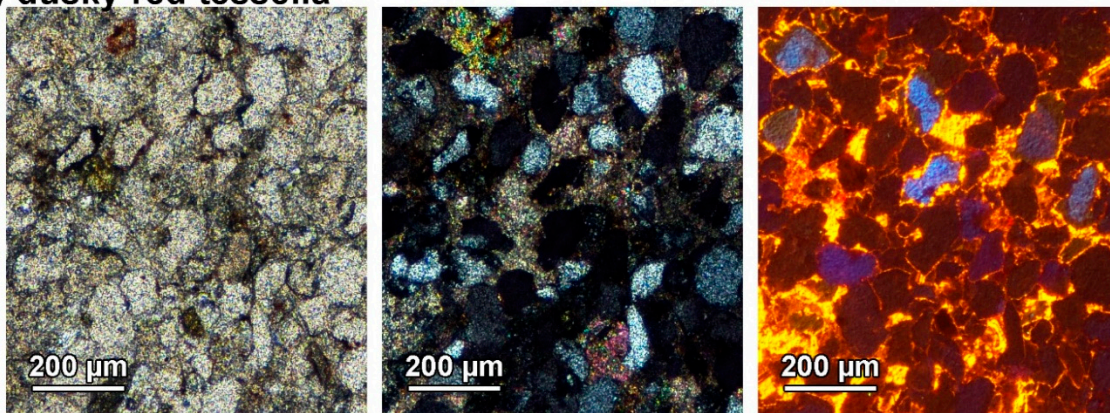
Petrographic inspection of the sandstone tesserae reveals at first sight that the greenish gray tessella (Figure 13a) contains the main provenance markers of Montjuïc sandstone (K-feldspar and quartz clasts with authigenic overgrowths and absence of carbonatic elements). In contrast, the dusky red tessella (Figure 13b) includes abundant carbonatic cement clearly visible using a cathodoluminescence microscope (Figure 13b, image on the right) which is uncommon to Montjuïc sandstones. However, and despite the presence of the carbonatic cement, a detailed examination of the feldspar clasts reveals that they also contain the typical K-feldspar authigenic overgrowths. These overgrowths are the main diagnostic feature of Montjuïc sandstone and therefore all evidence points to locate the provenance of this tessella in the upper layers of the Morrot unit (Figure 3 and M2 in Figure 5b) where carbonatic cement occasionally fills the porosity of previously silicified sandstones.



### a) greenish gray tessella



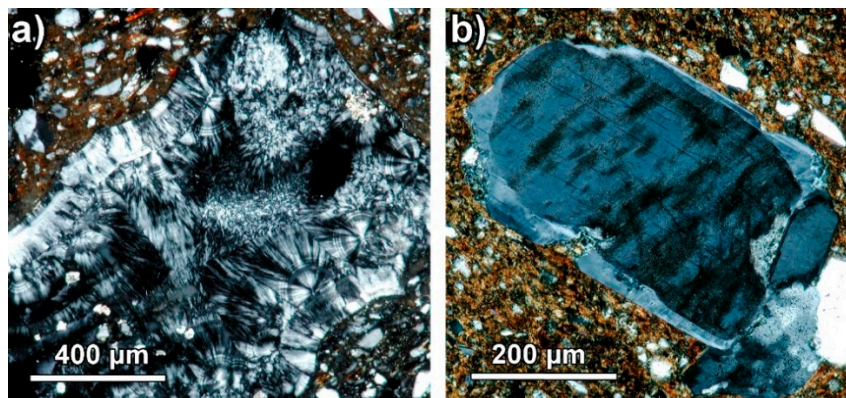
### b) dusky red tessella



**Figure 13.** Petrographic images of the sampled sandstone tesserae from the mosaic MAC-19004 including PPL (left), XPL (middle), and CL (right) micrographs of the same areas (a) sampled greenish gray tessella exhibiting typical features of Montjuïc sandstone (K-feldspar and quartz clasts with authigenic overgrowths). (b) Sampled dusky red tessella exhibiting K-feldspar and quartz clasts with authigenic overgrowths embedded in a carbonatic cement (orange in CL mode).

#### 4.4. Identification in Pottery

Petrographic thin sections of 19 samples of pottery shards obtained from the excavated kiln revealed the predominance of fine fraction aplastic inclusions (<0.1 mm) composed mostly of quartz and feldspar and subordinate micas. The coarse fraction (>1.5 mm) is formed by fragments of granitoids and derived single mineral grains, subordinate micaschists, quartz-micaschists, quartzites, occasional slates and biotite, subordinate fragments of limestone, sandstone, chert, and sparry calcite crystals. It is likely that aplastic coarse inclusions were intentionally added; they are lithologically consistent with the rocks of local Paleozoic substrate (outcropping at the Collserola Mountain). However, in terms of provenance, the presence of chalcedony (Figure 14a) and fragments of K-feldspar minerals with authigenic overgrowths (Figure 14b) is particularly conclusive; both features can be considered diagnostic of Montjuïc provenance [66].



**Figure 14.** (a) XPL micrograph of a chalcedony inclusion found within the paste of shard depicted in Figure 8d. (b) XPL micrograph of a K-feldspar inclusion found within the paste of shard depicted in Figure 8e containing a clear authigenic overgrowth.

## 5. Discussion and Perspectives

The use of Montjuïc sandstone has been identified with a high level of confidence using a number of petrographic provenance markers described in Section 2.2. Specifically, the presence of K-feldspar clasts with authigenic overgrowths is especially convincing. This feature can be easily spotted in petrographic thin sections of the sampled heritage materials using a petrographic microscope and particularly using a cathodoluminescence microscope. Besides these, no additional methods are required; other common techniques such as geochemical analyses or stable isotopes measurements would be of no use for an inherently heterogeneous rock such as this sandstone.

Concerning sculptures and architectural elements, the detailed analyses revealed that macroscopic identification of Montjuïc sandstones can be misleading. In particular, the sculpture of a woman wearing a toga (MAC-19018, Figure 6b) was not really carved in Montjuïc sandstone as it had been previously assumed and this clearly indicates that the Roman workshops in Barcino were not only using Montjuïc sandstones as local stone. Another interesting case is that of the two pieces found together in the Seu d'Ègara precinct (MdT-25992 (frieze) and MdT 26000 (acroterion), Figure 6e,f). These pieces had been previously interpreted as belonging to the same funerary monument. However, the frieze was not carved using Montjuïc sandstone. The fact that both pieces are not carved using the same lithology does not exclude the possibility that they belonged to the same monument, but now this interpretation appears more unlikely. Besides that, the stylistic closeness between this frieze and other frieze fragments found in the southern sector of the city walls of Barcino prompts to verify if these are really carved in Montjuïc sandstone. In contrast, the acroterion, which is iconographically strange in the Barcino area, resulted to be carved in Montjuïc sandstone. Generally, acroteria feature Gorgon heads in Barcino [34], whilst satyrs heads are frequently found in southern Hispania [67]. This could be used to argue that the acroterion found in Ègara was imported from the south. However, as the acroterion is carved in Montjuïc sandstone it was clearly produced locally.

To summarize, it should be highlighted that a given carved iconography or typology is not necessarily bounded to a given lithology. The fact that we have found that some pieces are not carved in Montjuïc sandstone does not exclude that they were actually produced in the Barcino workshops. It is clear that besides the Miocene Montjuïc sandstones, other local lithologies were also exploited in Barcino, such as the older Triassic and the younger Quaternary sandstones also available in the area. In any case, the accurate identification of the lithology of the sampled heritage material does not modify substantially the historical discourse on the development and the activities of the local workshops in Barcino, but encourages the reevaluation of all the heritage materials that had been previously assigned (macroscopically) to Montjuïc provenance. Precise identification of this sandstone opens the door to face meticulous studies of the distribution of objects carved using this material and not only within the

Roman period. In particular, it would be worthy to add the petrographic approach to the studies on the distribution of Montjuïc millstones.

Concerning mosaics, the obtained data are very important since the use of Montjuïc sandstone in tesserae from Roman Barcino mosaics has been verified for the first time. Such attestation could have hardly been established without the presented petrographic approach. However, such local use for this material makes sense, taking into account that Montjuïc sandstone outcrops close to the city are chromatically rich and available as a byproduct from its use in architecture and sculpture. In fact, two different lithotypes from Montjuïc have been identified in the tesserae; not only the commonly exploited sandstones of Castell unit but also carbonate-rich nodules from the underlying Morrot unit. The identification of the latter is particularly interesting because the Morrot unit only outcrops in the bottom part of the cliff facing the sea (Figures 1 and 5). This would constitute the first evidence of other Roman exploitation points apart from the only known Roman quarry [28]. The bottom part of the south-east steep cliff of Montjuïc has been heavily altered due to the construction of the port and the coastal ring road (Figure 5a). It would be no surprise if all traces of ancient exploitation in this area had been completely erased.

It is worth mentioning that tesserae from Montjuïc have been identified in a single big polychrome mosaic bearing a wide range of colors. There are other similar mosaics from Barcino that could be analyzed. These could help to determine if such use of the Montjuïc sandstone was either anecdotal or prevalent. Colorimetric analyses are suggested as a non-destructive approach to perform a first screening on the mosaics.

The identification of Montjuïc sandstones in tesserae appeared already unlikely without an accurate petrographic approach (specifically oriented to the detection of distinct provenance markers). Without such an approach, the identification in ceramics would be almost impossible. However, K-feldspar overgrowths and chalcedony fragments have been spotted as temper in the pottery productions of a specific workshop ascribed to the 12–13th century. The use of crushed Montjuïc sandstone as temper can be considered the result of the use of a byproduct of the quarry activities, or maybe relates to the small trade industry of Montjuïc sand (*terra d'escudelles*), even if this industry is only documented since the 16th century. Similar to mosaics, it would be necessary to supervise the known pottery productions from Barcelona to check if the use of Montjuïc temper is secondary or usual and what is its temporal range. It is worth keeping in mind that the local pottery production in the Barcelona area started in prehistorical times [68] and it is more or less continuous from the Roman period until recently [69].

If an accurate and systematic survey could determine that the use of Montjuïc temper was more or less prevalent, at least for a certain time period, this would pave the way to track accurately the distribution of these ceramic productions. The potential is higher than that of the study of the distribution of stone objects, for pottery objects are much easier to transport. Actually, other 12–13th century pottery productions from Barcelona are known to have travelled to Provence [66,70] though these were identified on account of their typology and paste features. The markers commonly used for provenancing pottery are based on typological or technological features [71–73], particular mineral inclusions or paste textures (actually only rarely truly related to a single possible geographical origin). The particularity of Montjuïc sandstone are the K-feldspar clasts with authigenic overgrowths, those along with other less exclusive features (i.e., chalcedony, quartz overgrowths) provide a potentially powerful tool for tracking Montjuïc temper in pottery.

## 6. Conclusions

The present study illustrates the potential of archaeometric provenance studies dealing with clastic rocks. Despite being inherently formed by fragments of multiple sources, occasionally these rocks can bear specific features homogeneously bounded to the stone under study, such as the cement. Specifically, in the case of Montjuïc sandstones the particular provenance markers are:

- K-feldspar clasts with authigenic overgrowths
- Quartz clasts with authigenic overgrowths

- Fractures filled with chalcedony (mainly of length-fast type)
- Absence of carbonatic clasts
- Absence of early carbonatic cement (except for carbonatic nodules of the upper Morrot unit)

From all the mentioned markers, the most specifically restricted to Montjuïc sandstone are the K-feldspar clasts with authigenic overgrowths and the ideal technique to spot such overgrowths is the observation of petrographic thin sections using a cathodoluminescence (CL) microscope. The CL color of K-feldspar clasts is light blue (almost cyan) and their corresponding overgrowths are dark brownish blue. The CL color of quartz clasts is dark blue (with variations depending on the clast: Mainly navy but occasionally even violaceous) and the CL color of the corresponding overgrowths is very dark blue (almost black).

A petrographic survey oriented to the detection of the relevant Montjuïc provenance markers has been fruitfully applied to sculptures, architectural elements, mosaics, and pottery. The main conclusions from each reported use of Montjuïc sandstone are:

- Some Roman sculptures and architectural elements attributed macroscopically to Montjuïc sandstone do not bear their characteristic provenance markers. We have demonstrated that Montjuïc was not the only source of local sandstones used by the Romans to carve sculptures and architectural elements in Barcino. A reevaluation of all the heritage materials that had been previously assigned to Montjuïc provenance would be recommendable.
- The use of Montjuïc sandstone in Roman tesserae has been reported for the first time. Two different lithotypes from Montjuïc have been identified: The commonly exploited sandstones of Castell unit and carbonate-rich nodules from the underlying Morrot unit. This constitutes the first evidence of Roman extraction points located at the bottom part of the south-east steep cliff of Montjuïc. Colorimetric analyses are suggested as a non-destructive approach to perform a first screening on other Roman mosaics liable to contain Montjuïc tesserae.
- The use of Montjuïc sandstone as pottery temper has been reported in the productions of a medieval (12–13th century) workshop in Barcelona. A systematic petrographic survey oriented to identify the specific provenance markers would be useful to determine the temporal range of such particular use of the Montjuïc sand and the spatial distribution of the corresponding pottery productions.

The adoption of the described provenance markers entails the possibility to perform confident recognition of Montjuïc sandstones in all its uses throughout history.

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