

Article

Mineralogical Insights to Identify Göktepe Marble in the Sculptural Program of Quinta Das Longas Villa (Lusitania)

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Abstract: This archaeometric study is focused on the marble used in a group of fragmented sculptures found at the Roman villa of Quinta das Longas (Elvas, Portugal). Dating from the 4th century AD, the pieces are of remarkable quality and correspond to ideal and mythological figures from several iconographic cycles. The numerous fragments, all of very fine-grained white marble, are associated with the ornamentation of an impressive nymphaeum of the villa. Their high level of sculpture technique and style, the models followed and their similar typology to other well-known parallels raise the hypothesis of being linked with Aphrodisian workshops. Using a well-established multi-method approach, with Optical microscopy, X-ray Powder Diffraction (XRPD), qualitative and quantitative cathodoluminescence (CL) by CL-Optical and CL-SEM, and stable C and O isotopic and trace element analytical techniques (IRMS and ICP-AES), together with complementary parameters obtained from electron paramagnetic resonance (EPR) and ⁸⁷Sr/⁸⁶Sr isotopes, the marble provenance can be identified with certainty. The results all point to the best quality of white Göktepe marble, confirming the stylistic connection to the ancient Carian sculptors.

Keywords: archaeometry; marble; Göktepe; Aphrodisian workshops; Roman Lusitania; cultural heritage



Citation: Lapuente Mercadal, M.P.; Nogales-Basarrate, T.; Carvalho, A. Mineralogical Insights to Identify Göktepe Marble in the Sculptural Program of Quinta Das Longas Villa (Lusitania). *Minerals* **2021**, *11*, 1194. <https://doi.org/10.3390/min11111194>

Academic Editors: João Pedro Veiga, Mathilda Larsson Coutinho and Elin Figueiredo

Received: 30 September 2021

Accepted: 21 October 2021

Published: 27 October 2021

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1. Introduction

Studies on marbles used to decorate complex architectural projects are aimed at obtaining detailed historical and archaeological data of the building history of the monuments. In the case of the white marble statuary used to embellish private or public spaces, the identification of the source of marble provides not only valuable information on trading patterns and economic history, but also on the workshops. This contribution reports the results of the archaeometric study of the very fine-grained marble used in a group of sculptured pieces found in the course of an archaeological campaign carried out in 2000 at the Roman villa of Quinta das Longas (Elvas, Portugal) in the Lusitanian Emeritensis territorium (Figure 1). The fragmented pieces were found in a unitary nucleus of destruction and stacking. They are of remarkable quality, corresponding to ideal and mythological figures, forming different iconographic cycles of smaller size than the natural, made for decorating domestic environments, very likely for a *nymphaeum* [1]. They would have originally been distributed as representative sets of scenes from Greco-Latin mythology and literature [2].

Quinta das Longas is located in a rural region of the middle Guadiana valley, characterized by a singular concentration of uillae with great potential for agricultural exploitation due to abundant water resources and very fertile soil. Additionally, the richness in mineral resources favored the development of this territory, which was traversed by a good

network of communication routes that connect the provincial capital, Augusta Emerita, to its sea port, Olisipo. The local routes ensured not only rapid access for soldiers, but also a regular supply of goods, including marble from the Estremoz Anticline, an important area of marble quarries during the Roman imperial age widely used in all Lusitania [3–5].



Figure 1. Quinta das Longas location (38.9479158, -7.6507533) in the Lusitanian territory of Hispania. EA is the Estremoz Anticline marble district.

The villa of Quinta das Longas was integrated into this rural territory's deeply Romanized ways of life and cultural ideals of the Empire for a long period, from the 1st century AD until late Roman times [6]. A residential complex with a peristyle and a nymphaeum was erected in the villa between the 3rd and 4th centuries AD. Placed in an alluvial plain associated with a medium-sized watercourse, the remaining structures have been identified with the *pars urbana* of the villa, richly decorated with *opus sectile*, parietal marble plates, decorative painted walls and sculptures. This decoration gives it truly exceptional character in the rural Lusitanian territory. One of the first signs of this exceptionality concerns the presence of water in the building complex, which seems to be of particular relevance in the decoration of the villa [7]. A wide surface covered with a continuous sheet of water, several water tanks insightfully well distributed to allow a continuous stream of water and a waterfall-type element have been recognized among the remains located in the nymphaeum. Following the style of that time, the courtyard would have presumably been adorned with this singular group of marble sculptures [8].

It is striking that the sculptures were not found in their original placement, but neatly intentionally grouped in one room, separately from the rest of the ruins. With the exception of two small figures, a hand and a feline head found in the nymphaeum area, the large group of marble fragments was located within a stratum of abandonment and accumulation of rests, attested by the arrangement of the pieces, perfectly chopped up and placed on a relatively small surface, concentrated in the amortization space [8]. The site was abandoned at an undetermined moment in the 5th century, leading to a process of ruin that would culminate in the total concealment of the vestiges. After the abandonment of the villa, the ruins had some type of sporadic occupation, reusing many building materials. The looting of the nymphaeum must have occurred at the end of the 18th century, when the ruins were used as a quarry for the construction of the main house of the current building complex of Quinta das Longas.

Reasonably, the found sculptural ensemble does not correspond to the totality of the sculptures that originally adorned this area of the villa. There are numerous incomplete figures—humans such as a Venus and an anguiped male figure, as well as animals and vegetation elements—all found significantly fragmented. Some of the best preserved

pieces are illustrated in Figure 2. The whole was previously widely described from the iconographic and artistic point of view [2,8], emphasizing not only their similar typology to other well-known parallels, but also their high level of technique and style, raising the hypothesis of being linked with the Aphrodisian ateliers.

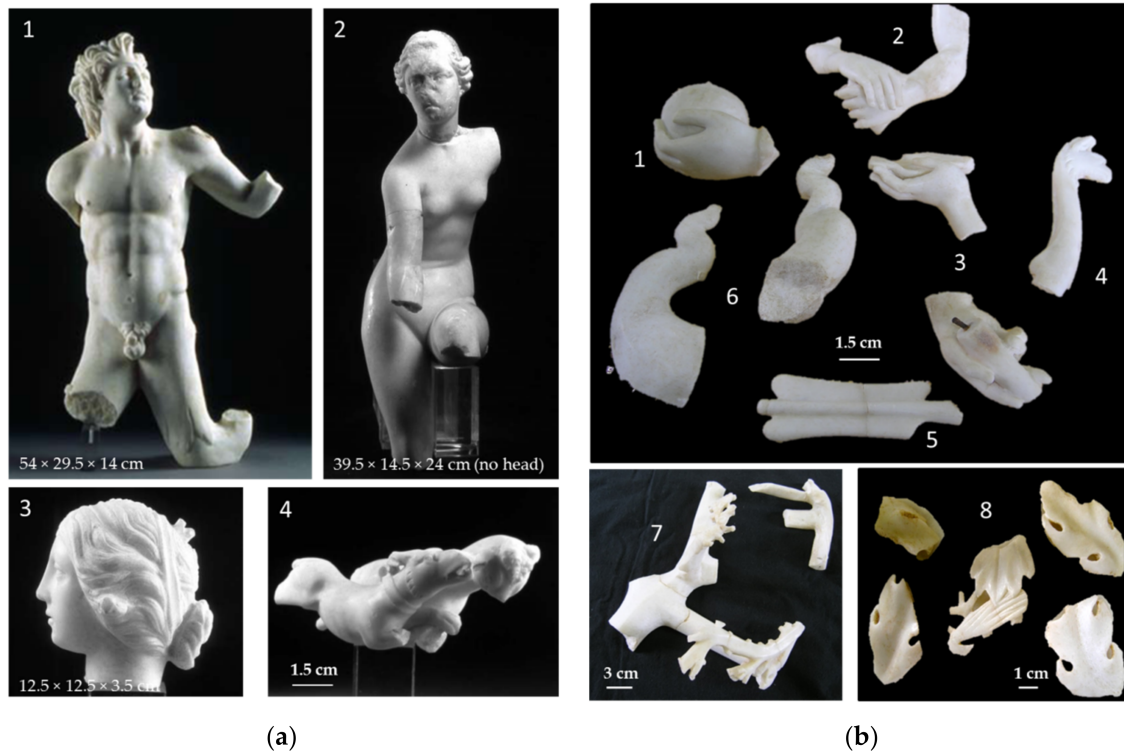


Figure 2. Some of the recovered sculptures: (a) (1) Anguiped male creature, (2) Venus holding a sandal, (3) ideal female head (nymph), (4) two hunting group animals; photographs by J. Pessoa, Instituto Portugues de Museos (IPM); (b) various fragments such as (1) Uranus muse hand, (2) female arm leaning on male arm, (3) two female hands holding objects, (4) gryphon head ship bow, (5) rudder tip, (6) two angled tail ends, (7) fragments of tree trunks and (8) leaves; photographs by M.P., Lapuente Mercadal.

At first, the marble identification of unknown artefacts can certainly appear to be too complex a task, considering all known quarry sites used in antiquity. Their authentication goes through a previous stage of multi-method analysis to try to find the markers or traces that serve to discriminate one marble from another. Moreover, to obtain reliable results, the comparison of the representative discriminant parameters must be accompanied by identical analytical protocols. However, the task is simplified if the comparative approach focuses on a set of quarries selected as likely sources on the basis of art history and archaeological information. The known classical fine-grained marbles exploited and traded during imperial times, especially for statuary sculpture, are Paros-lychnites, Afyon (Docimium), Pentelicon, Carrara and Göktepe [9]. Consequently, a priori in the case under consideration, the Göktepe quarries offer a very high probability of having been used by the Carian artists.

The recent discovery of ancient marble quarries near Göktepe (Muğla province, Western Turkey) [10] has transformed the landscape of archaeometric studies focused on fine-grained marble used in emblematic Roman sculptures. The richness of the materials exploited in them—with respect to both their diversity of chromatic tones and their high sculpturability, ranging from very fine- to fine-grained marble—undoubtedly indicates that right from the beginning, these were raw materials worthy for artistic works for the nearby Aphrodisian workshops [11].

As part of a research program developed a decade ago by a Spanish team concerning the identification of marble on sculptural pieces stored in the imperial Villa Adriana of Tivoli, Göktepe materials were identified without certainty [12] based on comparison with

the limited analytical references published up to that time. That archaeometric study demonstrated the complexity of identifying certain fine-grained white varieties found in the imperial Villa, since the parameters usually applied in provenance studies (petrography and C and O isotopes) do not properly discriminate certain white Göktepe from Luni-Carrara marble; consequently, it has become clear that, up to that time, it is very likely that countless pieces carved with this Turkish material might have been mistaken for Carrara marble [13].

Nevertheless, over the last few years, different efforts to find the discriminant parameters of Göktepe white marble quarry have greatly contributed to their correct identification in archaeological pieces [14–17], although not without controversy in some contributions [17–22]. However, all scholars are in agreement that a combination of multi-methods is needed to recognize white Göktepe in ancient sculptures. Additional papers, focused on the role of the application of specific techniques, have further contributed to the discrimination of Göktepe from other fine-grained marbles. Among others, these techniques include the use of CL parameters, either qualitative [23] or quantitative [24]; trace elemental composition, including rare earth elements [25]; and nuclear magnetic resonance attributes [26,27]. Consequently, using a combination of selected techniques, white Göktepe marble can now be identified with certainty in ancient artefacts [14,17,28]. This is particularly important when it comes to identifying very fine-grained marble from sculptural pieces of great stylistic quality, as is the case studied here.

2. Materials and Methods

In order to determine the origin of the white marble used in this sculptural ensemble, a well-established sequential multi-method analytical approach [29] was applied, including mineralogical and geochemical techniques [15,23,25]. This approach focuses on comparison with the analytical properties of a set of quarries, mentioned above, selected as likely sources on the basis of art history and archaeological information, from which the statuary white Göktepe is a priori the best candidate. In this sense, additional parameters obtained by other less common techniques in marble provenance studies, but known as discriminants for the recognition of Göktepe marble [14,17,24], were also applied. The results were compared with our own reference database and a review of the analytical data included in different publications related to the provenance of fine-grained marbles, as detailed in the discussion section.

All pieces were visually examined using a handheld magnifying lens, from which several fragments, marked as QL, were selected for the analyses (Figure 3a). Two thin sections were made with the two largest fragments (4251 and 4253); finding no differences between them, as expected, the other two fragments were reserved to perform the rest of the analyses based on a powdered sample. The high quality of delicate carving of this noble material is noteworthy; the material has the visual appearance of ivory (Figure 3b) and has a high transmittance for light, which was checked by directly applying an LED source to its surface (Figure 3c).

Polarized light optical microscopy and CL microscopy were applied on the thin sections made at the Laboratory of Central Services for the Research of the Earth Sciences department (UNIZAR, Zaragoza, Spain). A standard optical JENAPOL microscope with a NIKON D3400 photographic camera, available at the Petrology and Geochemistry Laboratory of UNIZAR, was used to examine the mineralogy, microstructure (including texture), Grain Boundary Shapes (GBS), Maximum Grain Size (MGS) and the Most Frequent Size (MFS), since they are well known to be useful for marble discrimination when combined with other analytical results [9,15]. A Technosyn CL8200Mk5-1, provided by Cambridge Image Technology Ltd. (Cambridge, UK), coupled to a Nikon Eclipse 50iPOL microscope at the ICAC laboratory of Tarragona was also used to take CL images of the same thin section, with an automatic digital Nikon Coolpix5400 camera. The electron energy applied was 15–20 kV, the beam current operated at 250–300 μ A and the vacuum was 0.17 mbar (17 Pa). The CL images taken were automatically recorded (29 mm focal length,

f/4.6 aperture, 1s exposure time, ISO-200). Three parameters were qualitatively observed from them: color, intensity and distribution of CL [23].

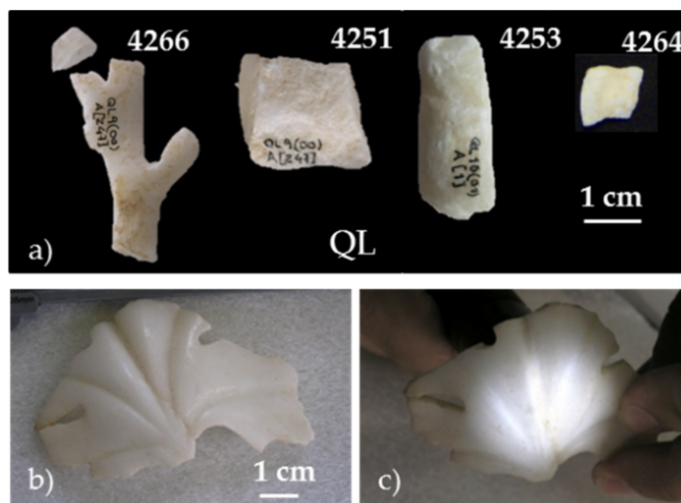


Figure 3. Small fragments of the same sculptural program: (a) selected pieces for analyses (QL); (b) given this statuary marble's high quality, brilliance and superficial fineness for carving, it could be mistaken for a piece of ivory; (c) it has a high transmittance for light. Photographs by M.P. Lapuente Mercadal.

To quantify the CL phenomenon, a spectrometer accessory for a Scanning Electron Microscope (CL-SEM) was used to register the CL emission spectrum and measure the characteristic peak intensities. This Zeiss Supra 55 VP SEM device, developed at the laboratory of the Université Pierre et Marie Curie (Sorbonne, Paris, France), consists of a parabolic mirror as a collector, coupled by a fiber to a grating spectrometer (Triax, Jobin Yvon) with an LNT cooled CCD. The system is able to perform from 250 nm to 1200 nm. Measures were registered between 300 nm and 750 nm by means of 3 spectra of 1 s, on 3 places of each compressed powder (3 mg). The analytical conditions were 20 kV, 40 nA and under 20 Pa vacuum. Intensities at the respective peak of 620 nm (from calcite) and the sum of intensities at the peaks emitted on the UV spectrum were measured in cps units. Further information about this technique, including sampling preparation and a database with the most important classical marbles, is reported in [24].

To check the absence of dolomite, XRPD was carried out using an automatic Philips PW 1130/00 diffractometer (CuK α radiation at 40 kV, 20 mA; data recorded in the 3–70° 2 θ range, 1°/min, 2 s/step). Oxygen and carbon isotopes were determined on the calcium carbonate QL sample by Isotope Ratio Mass Spectrometry (IRMS). This was performed in the Laboratory of the Istituto di Geologia Ambientale e Geoingegneria in Rome (Consiglio Nazionale delle Ricerche, CNR), by means of the usual acid digestion technique at 72 °C using a Thermo Gasbench II in line with a Delta+ mass spectrometer. The results are expressed in the usual delta notation ($\delta^{18}\text{O}$ and $\delta^{13}\text{C}$), which represents the relative deviation in parts per thousand of the heavy isotope/light isotope ratio of the sample from that of an international reference standard (V-PDB). The analytical precision was better than 0.1‰ for both isotopes.

A Varian Vista-PRO Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES), at the Earth Science Department of Sapienza University of Rome, Italy, was used for trace element determination. Sample aliquots were dissolved in solutions of HCl (3%) to measure Sr and Mn elemental concentrations (ppm). Deionized water (resistivity 18 M Ω cm $^{-e}$) obtained from a Milli-Q purification system was used to prepare all standard and sample solutions. Internal standards and the precision of the method are explained elsewhere [15,22].

Electron paramagnetic resonance (EPR) spectroscopy was carried out with an X-band Varian E-9 spectrometer equipped with a multipurpose rectangular cavity from the Istituto di Struttura della Materia in Rome (CNR). Usually, the marble EPR spectrum is the standard spectrum given by Mn^{2+} substitutionally diluted into the calcite lattice. The spectrum was recorded at room temperature on weighted amounts (ca. 30 mg) of finely ground marble powder packed in constant diameter (2.8 mm) quartz tubes, at the X-band frequency ca. 9.10 GHz; microwave power 12 db (12.5 mW); modulation amplitude 1 G; and time constant 0.1 s or 0.3 s, with corresponding recording times of 4 min or 8 min. According to experimental procedures and standardization methods described elsewhere [30,31], two variables were used: intensity (Intens) and EPR linewidth (W).

Sr isotope composition was measured in the Isotope Laboratory at the University of Poznań, Poland. About 50 mg of powdered sample was dissolved on a hotplate (~100 °C, overnight) in closed PFA vials using hydrochloric acid (0.75 N). Strontium was loaded with a $TaCl_5$ activator on a single Re filament and analyzed in dynamic collection mode on a Finnigan MAT 261 mass spectrometer. The analytical protocol and adjustment of $^{87}Sr/^{86}Sr$ values followed are described in [17].

3. Results and Discussion

The results of the mineralogical-petrographic examination and the main CL characteristics (qualitative and quantitative) are summarized in Table 1. Parameters obtained from EPR, isotopes, and Sr and Mn concentrations are displayed in Table 2. From here on, the results of each technique will be presented, and at the same time, the identification will be discussed, comparing them with the analytical data available for each of the techniques.

Table 1. Petrographic, Optical-CL and quantitative CL-SEM properties and mineralogical composition of sample QL.

MGS	MFS	GBS	Texture Fabric	Qualitative CL-Optical	CL-SEM Intensities Extrinsic (620 nm) /Intrinsic Sum	XRPD
0.4 mm	Extremely fine 0.2 mm	Curved to Straight, occasionally to Embayed	Homeoblastic Mosaic Slightly strained (Crystallographic Preferred Orientation)	Low CL with dull domains	1398/1470	Calcite

Table 2. Parameters of sample QL by conventional C and O isotopes and complementary techniques.

$\delta^{13}C$ ‰ (PDB-V)	$\delta^{18}O$ ‰ (PDB-V)	EPR Intens	EPR Width	Sr ppm	Mn ppm	$^{87}Sr/^{86}Sr$
+2.78	−2.44	0.040	0.534	800	6	0.707358

3.1. Petrography, XRPD and CL-Optical Observations

The QL sample consists of very fine-grained marble of pure calcite composition without any accessory mineral observable at the microscale. The absence of accessory minerals was checked by XRPD. Figure 4a,b illustrates the mosaic homeoblastic texture, where calcite crystals exhibit curved to straight, occasionally to embayed, GBS. Despite apparently being an isotropic fabric, the microstructure shows a very slightly strained fabric with evidence of crystallographic preferred orientation in some calcite. MGS is 0.4 mm but the MFS is ≤ 0.2 mm, following the same method explained in previous studies [15]. Both parameters reveal the extremely fine grain of this marble in concordance with the lithotype G white statuary assigned to Göktepe in the sculptures of Villa Adriana [12,13,15,28].

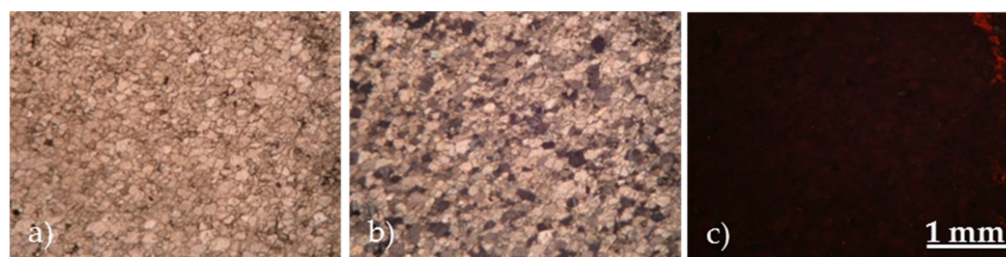


Figure 4. Photomicrographs of sample QL, all with the same scale: (a) in parallel polarized light; (b) in crossed polarized light; (c) CL image.

The CL pattern (Figure 4c) is also typical for this variety of white Göktepe—very dark reddish in color, with homogeneous low intensity and small areas with dull domains, which is in agreement with literature [15,17,23,24].

This extremely fine marble is significantly different from the rest of the white classical marble, in terms of MGS (Figure 5). The parameter measured in the QL sample (0.4 mm) matches well with that proposed for the lithotype G in [12,13,15], identified as Statuary White Göktepe in the pieces of the imperial Villa Adriana. It should be noted that although the MGS of 0.4 mm could be measured in certain Carrara marble, the value obtained for the MFS (≤ 0.2 mm) is significantly lower than the average obtained (≤ 0.5 mm) for a representative sample of Carrara marble using the same methodology [15]. In any case, other parameters will reinforce the QL identification as Göktepe, as seen below.

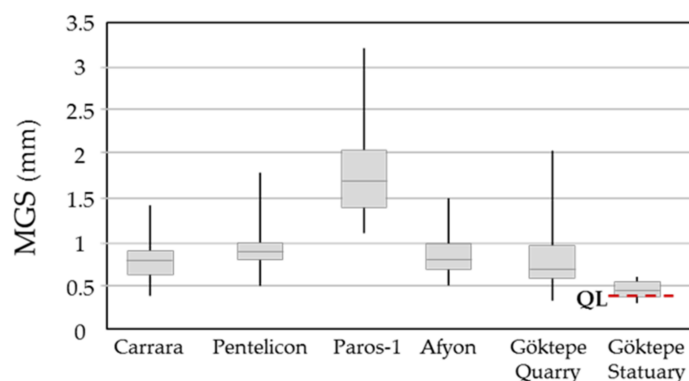


Figure 5. Maximum Grain Size of the white fine-grained marble. The dashed red line (MGS of 0.4 mm in sample QL) is in the 25th percentile of data measured in lithotype G identified in pieces of Villa Adriana [15].

3.2. Quantitative CL-SEM

In line with the qualitative observations made using the CL-optical image, the intensity measurements of the CL-SEM spectrometer are also characteristic of a marble with low extrinsic intensity (1298 cps) compared to that shown by the rest of classical fine-grained marble [24]. This value, representative of the peak due to the energy emitted by the free electron of Mn^{2+} in the visible spectrum of calcite, easily falls within the range observed for Göktepe marble (between 353 and 5504), which is in agreement with the low Mn concentration (6 ppm) (expressed in Table 2) measured in the sample QL. On the other hand, the sum of intensities for the intrinsic or UV emissions of calcite (peaks at 352 nm and 408 nm) are 1470 cps; this value is within the range between 848 and 2928 measured for white Göktepe marble [24].

The graphical representation of the CL intensity variation measured in the fine-grained marble helps to visualize the CL intensities measured in the marble under consideration (QL), which falls inside the ellipse assigned to Göktepe, as can be observed in Figure 6. However, other parameters are needed to ensure its identification, since the values of CL are close to the probability ellipses of other quarries.

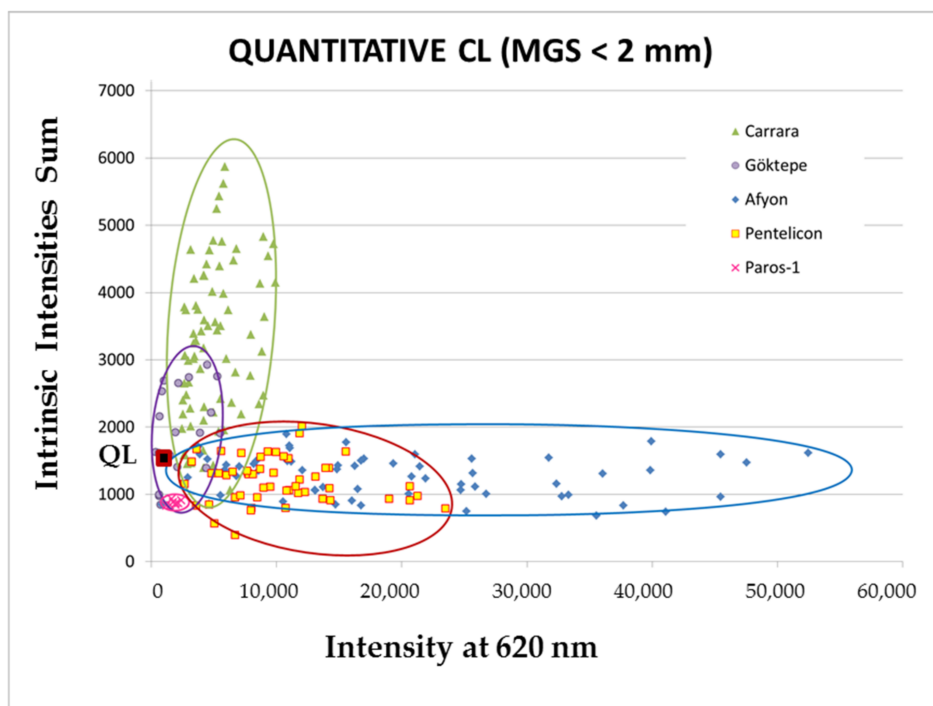


Figure 6. Representation of CL intensities measured in sample QL (red and black square) in the general quantitative CL diagram for the main classical fine-grained types of marble according to [24].

3.3. C and O Isotopes

Isotopic signatures were measured in the QL sample, whose compositions are -2.44% ($\delta^{18}\text{O}$) and $+2.78\%$ ($\delta^{13}\text{C}$), as shown in Table 2. They were plotted in the general isotopic reference graph for the fine-grained marble (Figure 7) with respect to different databases [5,9,14,15,17,30,32].

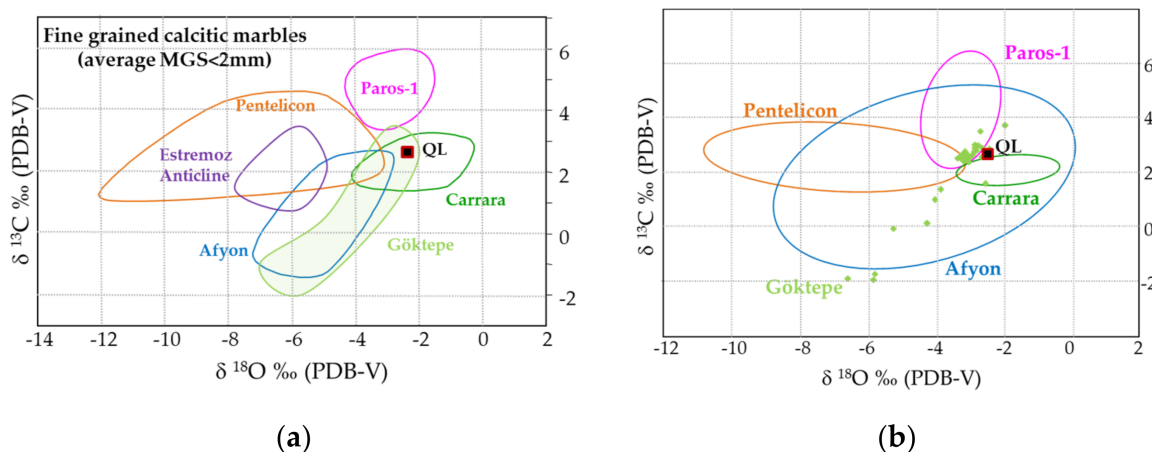


Figure 7. Isotopic C and O signature of the archaeological sample QL (red and black square) plotted on two different diagrams for classical fine grained marbles: (a) isotopic diagram adapted from [32] with data from [5,14,15,17]; (b) scatterplot of the C and O isotope compositions of the Göktepe white marble adapted from [15], with the probability distribution (99%) of isotope data represented by ellipses adapted from [30].

In Figure 7a, the QL sample falls inside the overlapping area of Carrara and Göktepe isotopic fields. However, in Figure 7b, QL follows the main cluster of the Göktepe marble quarries, in a narrow area approximately between -3% and -2% of $\delta^{18}\text{O}$ and $+2\%$ and $+3.5\%$ of $\delta^{13}\text{C}$, and also falls outside the probabilistic ellipse of the Carrara isotopic field. Of course, in either of the two graphs, the data of the C and O isotopes would not be sufficiently discriminant if it was not accompanied by the data obtained by other

techniques that reinforce the discrimination between Göktepe and Carrara marble sources. However, the comparison of Carrara (Fantiscritti) and Göktepe marble with respect to the isotopic data of $\delta^{13}\text{C}$ with those of the intensity of CL (Figure 8) helps to support the Göktepe assignment for QL.

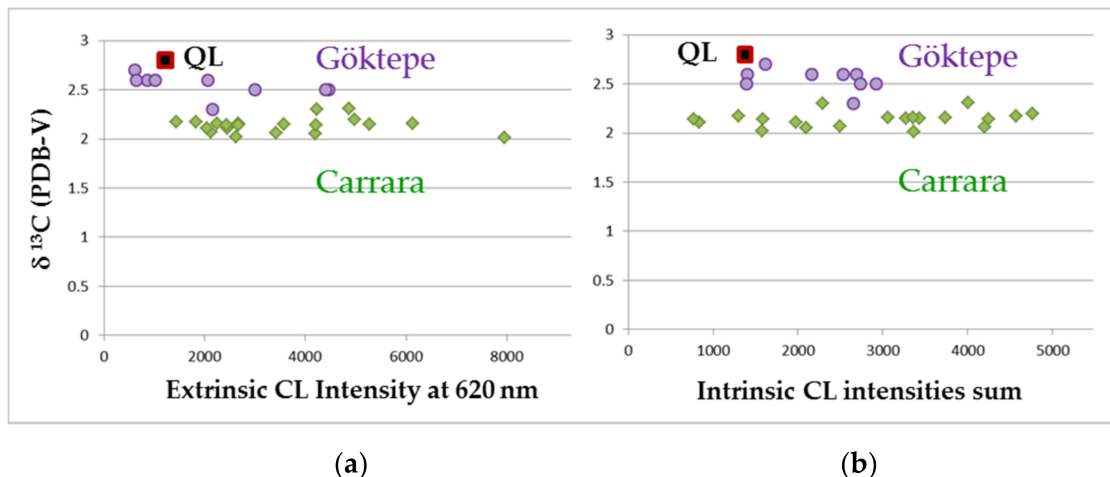


Figure 8. Isotopic $\delta^{13}\text{C}$ vs. CL intensities of the archaeological sample QL (red and black square) along with data from Göktepe and Carrara (Fantiscritti) quarries, according to [24]. (a) scatterplot of the extrinsic CL; (b) scatterplot of the UV emissions.

In fact, the graphic representation of the QL sample clearly approaches the cloud of points for Göktepe marble in both diagrams of Figure 8. Furthermore, although the sum of intensities of intrinsic CL might not be discriminant between certain Carrara (Fantiscritti) and Göktepe types of marble (see the plotting of those Carrara samples with lower CL values in Figure 8b), it is noteworthy that the lower extrinsic CL intensities (Figure 8a) discriminate both marble sources. This characteristic, together with the higher $\delta^{13}\text{C}$ values, strengthen the Göktepe assignment of this QL marble.

3.4. Sr and Mn Concentration

The recent paper [25], which focused on trace elements for quarry provenance investigation of ancient marble, has revealed the potential for using several trace element concentrations at ppm level as markers for marble identification. The implementation of laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS), with high-precision measurement of elements in given points of calcite crystals, allows one not only to prevent any disturbances during the pre-treatment of sampling, but also spurious results related to the possible inclusion of accessory mineral phases. Certain elements—common substitutes of calcium in the calcite crystal lattice, such as Y, +REE, Mn, Th, and many others with remarkably low contents—were recognized as good markers for marble identification. In fact, Mn^{2+} is an important cation at the basis of widely used techniques for marble provenance such as CL and EPR.

On the other hand, as already explained in different papers [14,15,25], Göktepe marble has an aragonitic protolith feature that allows strontium to easily substitute calcium in the aragonite crystal lattice, whereas manganese rarely does so; consequently, Göktepe marble contains more Sr and less Mn in comparison with other marbles of calcitic precursors. This makes the combination of both elements a complementary good marker for Göktepe marble, as has been recognized in those papers. However, it should be taken into account that the range of concentrations possible in Göktepe varies slightly depending on the analysis technique, even on the analytical protocol for the acid attack [21,22]. In addition, although there is no doubt that among ICP techniques, LA-ICP-MS guarantees a better approximation for the concentration of each trace element (since it is carried out on the calcite crystals themselves), it should be recognized that in most geochemistry laboratories,

this particular technique is not accessible (unlike others such as ICP-AES, which is used in many papers) [14–16,21,22]. Therefore, using a standard method of analysis (ICP-AES), the concentration of these two elements in the QL sample has been measured (Table 2) to tentatively compare it with the data provided in the literature. At first, the comparison is based on the concentration data obtained by LA-ICP-MS [25], as represented in Figure 9. In addition, QL is plotted in a scatterplot of Sr versus Mn with Göktepe data (Figure 10) obtained by different techniques [15,17,25].

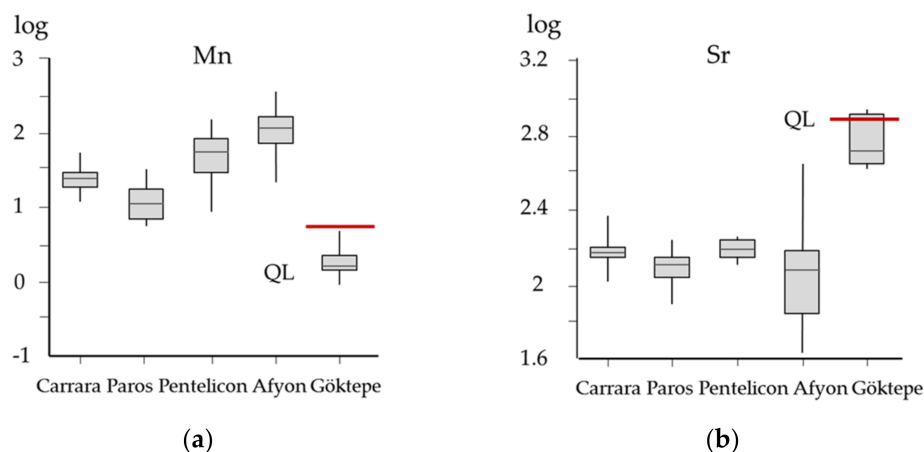


Figure 9. Box and whisker plots of Mn and Sr log-transformed concentration data from fine-grained marbles and the representation of the archaeological sample QL (red line), adapted from [25]. (a) Mn concentration; (b) Sr concentration.

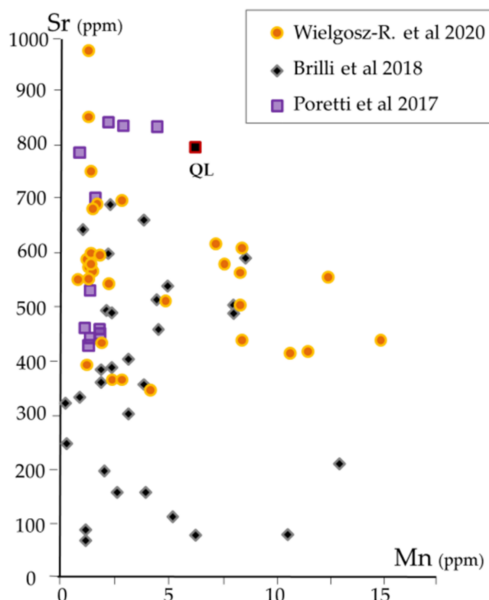


Figure 10. Scatterplot of the concentration of strontium and manganese (units in ppm) of the white Göktepe marble modified from [15], with additional data from [17,25] and the QL sample under study (red and black square).

As can be seen in Figure 9a, despite the fact that the concentration obtained for Mn in the QL sample is slightly above the values measured for Göktepe by LA-ICP-MS, the value of 6 ppm obtained is still low enough to be compatible with Göktepe, in line with the quantitative CL data discussed above.

Regarding the concentration of Sr (Figure 9b), the value of 800 ppm obtained in QL falls within the range expressed by the log-transformed data of 25th and 75th percentiles for Göktepe in [25].

Finally, in Figure 10, using the modified diagram published in [15] with data for Göktepe measured by the same method used here (ICP-AES) and data via LA-ICP-MS from [25] and via ICP-OES from [17], the Sr versus Mn concentration of QL is visually compatible with the dispersion obtained for Göktepe marble.

3.5. EPR Parameters

In order to verify the assignment to Göktepe with the technique commonly used by the scientific team that discovered these Turkish quarries [10,11,14], the EPR was carried out in the QL sample, whose values are expressed in Table 2 and represented in Figures 11 and 12.

In Figure 11, the QL sample clearly falls inside the area of Göktepe data points compiled from [14,30], next to those represented by Paros. Both Göktepe and Paros have a lower EPR intensity among the fine-grained marbles, in agreement with the quantitative CL data obtained, as previously shown in Figure 6. An additional feature to highlight is the relatively wide dispersion of the Göktepe data compared to the distribution of Paros in the scatterplot of EPR parameters. This characteristic was also detected in the diagrams of quantitative CL proposed in [24] (see Figures 6 and 7 therein), where the set of Parian samples analyzed showed similar very homogeneous intrinsic CL values, corresponding to the sum of intensities emitted at 352 nm and 408 nm UV peaks.

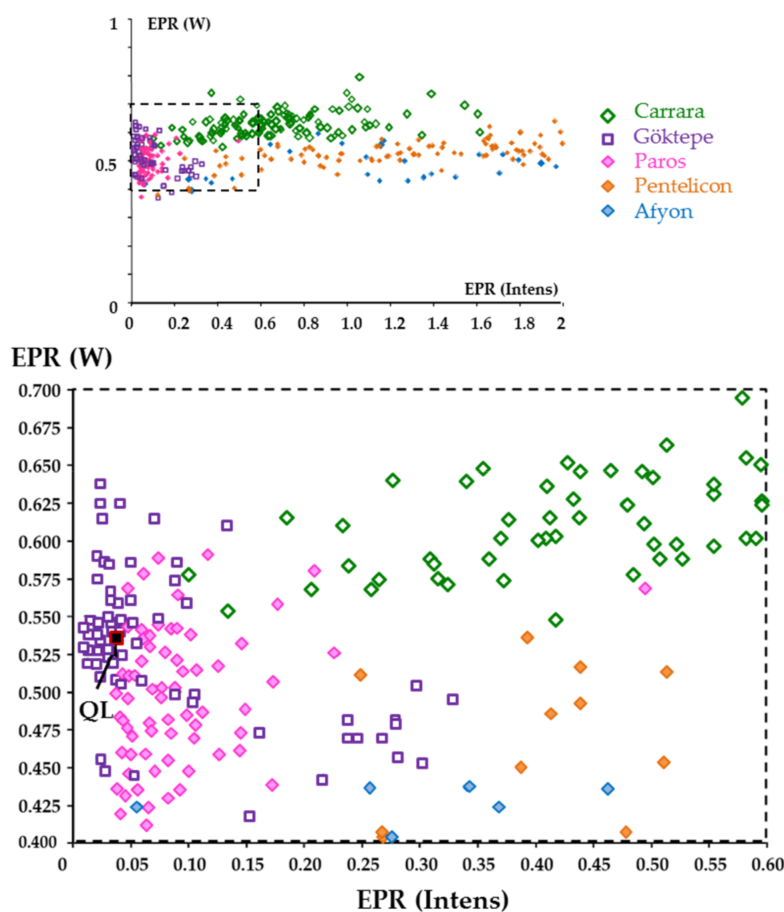


Figure 11. Scatterplot and close-up view (area with dashed line) of EPR parameters W versus Intens of the archaeological sample QL (red and black square) with data compiled from [14,30] for the most important fine-grained marble quarries.

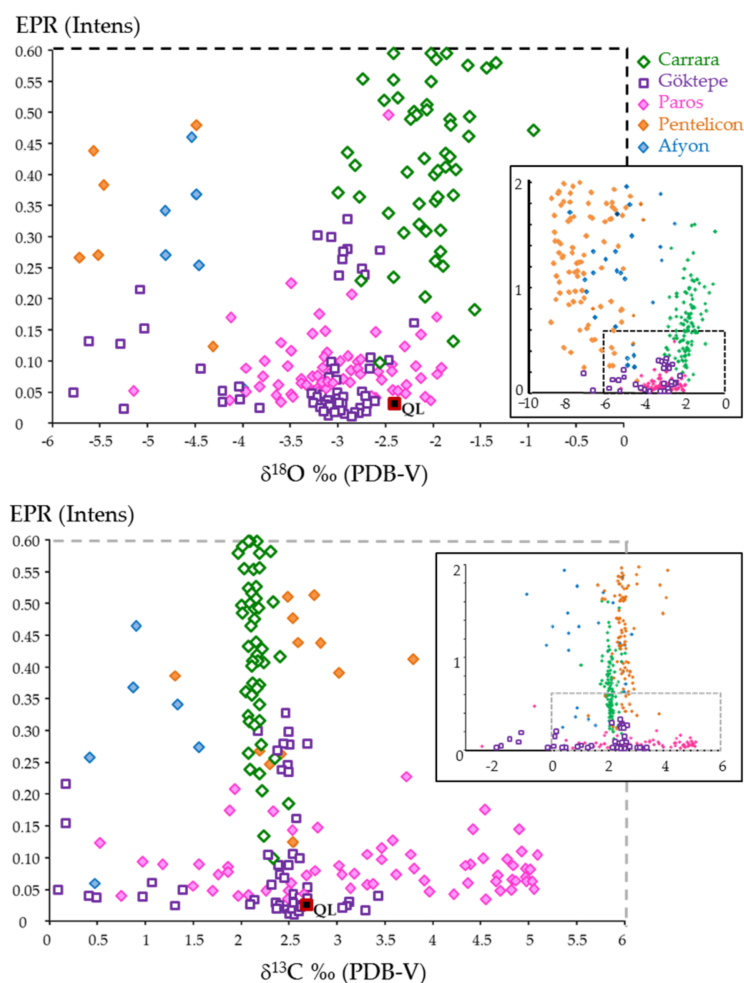


Figure 12. Scatterplot and close-up view (area with dashed line) of EPR (Intens) versus O and C isotopic data of the archaeological sample QL (red and black square) with data compiled from [14,30] for the most important fine-grained marble quarries.

Regarding the representation of the intensity values of EPR with respect to the data achieved from C and O stable isotopes (Figure 12), it is noteworthy that the QL sample is better associated with Göktepe in the binary diagram of EPR Intensity versus $\delta^{13}\text{C}$ than in the diagram versus $\delta^{18}\text{O}$, showing in both cases a close approximation to the data obtained for the Parian marbles.

3.6. $^{87}\text{Sr}/^{86}\text{Sr}$ Parameter

An additional contribution concerning Göktepe marble [17] has reported the utility of Sr isotope measurements in combination with certain geochemical parameters, improving its discrimination from Carrara and other fine-grained white marbles. In the same paper, the identification of several archaeological pieces of Villa Adriana that we had previously identified as Göktepe with our own multi-technique protocol was checked. They confirmed the Göktepe assignment through $^{87}\text{Sr}/^{86}\text{Sr}$, highlighting the remarkably low variability in values from both types of samples (quarry samples and Villa Adriana artefacts), ranging between 0.70724 and 0.70741 (mean = 0.70732, sd = 0.00004).

The Sr isotope measurements of the sample QL, obtaining the value of 0.707358 (Table 2), perfectly match with Göktepe marble, as shown in Figure 13. In both diagrams proposed in [17], the isotopic data of Sr combined with those of C and O clearly discriminate Göktepe and Carrara marbles. In them, the representation of two Villa Adriana samples, previously identified as Göktepe marble in [12,13,15] using a multi-analytical

protocol (petrography, CL-optical, CL-SEM and C and O isotopes), have also been plotted, reinforcing the Göktepe assignment by the $^{87}\text{Sr}/^{86}\text{Sr}$ composition.

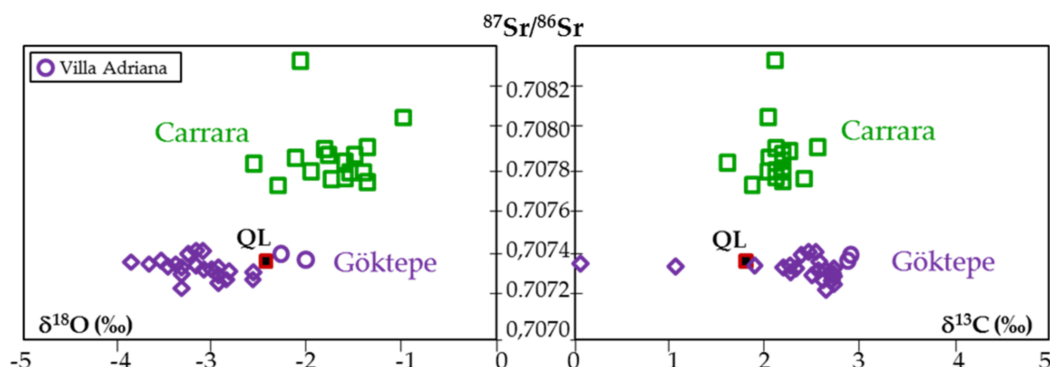


Figure 13. Plots of Sr isotope composition vs. $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ obtained in the QL sample (red and black square), based on the diagrams proposed in [17], allowing for unequivocal discrimination between Göktepe and Carrara white marble, both quarry samples and artefacts from Villa Adriana.

4. Archaeological Considerations

The exceptional group of sculptures of the villa of Quinta das Longas, dating from the 4th century AD, is an excellent example of how rural private environments emerged as flourishing clientele for marble artifact trade. During late Roman times, the villae became one of the main platforms for the social, cultural and economic prestige of their owners.

The identification of Göktepe marble from the region of Aphrodisias as the material used in this sculptural program is not surprising; on the contrary, it confirms the starting hypothesis. One aspect that caught our attention right from the beginning was the high quality of the marble used. As has already been seen, it is a very fine and compact marble, which allowed the artist to make an extremely refined work. Its whiteness purity and its highest technical quality are not usual in Hispanic marbles, an assertion that we can make after the experience of working with them for several decades, both in quarry materials and in archaeological pieces [5,33]. Certainly, the local types of marble of Estremoz Anticline that were extensively exploited during high imperial times, although of remarkable quality for large-format statuary, do not allow this level of refinement. Neither do other Hispanic marbles which were exploited for statuary with regional character. Therefore, from the beginning, the hypothesis of being an imported marble was raised—a hypothesis that is confirmed by the analyses carried out.

On the other hand, the uniformity of style—all the figures being of medium–small format linked to domestic decorative cycles associated with garden spaces and a strong presence of water—along with their high level of sculpture technique and style, led us to associate them with skilled craftsmen from workshops specialized in mythological art works. The way of working the anatomies, especially the athletic musculature in the male body or the softness of the features in the female figures, follow models associated with the school of Aphrodisias [2,8]. Indeed, we observed a similar typology to other well-known parallels spread throughout the Empire, such as the Venus of the Villa de St. Georges de Montagne, the Diana of Bordeaux or the Diana of Cherchel, among other similar cycles, all of which are linked to the production of oriental workshops. The most immediate Iberian parallel is found in the sculptural cycles of the Villa of Valdeterres de Jarama (Madrid). The iconographic types of the anguiped creatures in Valdeterres and Quinta das Longas are very close, although in Valdeterres, the white and black marbles are combined, creating a chromatic effect that recalls in colors and style that of the Silataraga group [2]. The detailed analysis of the materials shows that both sculptural ensembles have very close stylistic and technical character. Moreover, although the Valdeterres marble has not yet been analyzed, it is visually identical to the black and white marble studied from the Göktepe quarries [15].

Could this association suggest the presence in Hispania of itinerant artists who travelled where skilled sculptors were in demand for ambitious projects? In the case of the pieces of Quinta das Longas, given their medium–small format, they could have been effortlessly transported, so they probably could have been sold already manufactured. There were art works for select customers which, properly packaged, could be easily disseminated throughout the Empire.

What is clear is that the discovery of these sculptural pieces, their style and mythological cycles, as well as the present testimony of having used a marble imported from the ancient Caria, suppose a manifestation of great interest for the better knowledge of the exploitation, trade and distribution of marble in the Roman Empire. They help to obtain a better perspective of the role played by certain local elites in Hispania and rural owners which incorporated productions of oriental origin for the ornamentation of their impressive villas, and the identification of the Turkish marble in these productions definitely certifies the artistic and commercial connections between East and West.

5. Conclusions

A multi-method approach to unambiguously determine the provenance of white marble used in archaeological pieces is always essential, and the case under consideration—a very fine-grained white marble used in the Lusitanian sculptural program of Quinta das Longas—was not an exception.

Provenance identification was carried out using a sequential approach, taking into account petrography and CL pattern as the first step, combined with C and O stable isotopes. Complementary techniques, quantitative CL, EPR parameters, ICP-AES for the Sr and Mn concentration and Sr isotopes were used to confirm the Göktepe marble source. The results all point to the best quality white statuary Göktepe marble, confirming the stylistic connection to the ancient Carian sculptors.

In this sequential approach, the initial petrographic analysis, especially the MGS (0.4 mm) and MFS (≤ 0.2 mm) parameters, indicated right from the beginning that this is a raw material of very high technical quality for sculpture. Similar petrographic variety with low CL intensity had been detected in certain small marble pieces from Villa Adriana studied a decade ago, where the lithotype was unambiguously assigned as being of Göktepe origin.

Finally, the importance of having identified this type of Turkish marble in the westernmost Roman province of the Empire must be emphasized. Even more so considering the existence of local marble from the Anticline of Estremoz, which were highly appreciated by the Roman inhabitants not only from Lusitania but also from the adjoining Roman provinces, including North Africa. This is significant for two reasons; on the one hand, it highlights the decline of the exploitation of these Lusitanian marbles whose maximum splendor corresponds to the high imperial period. On the other hand, the use of Göktepe associated with oriental workshops, located in an inland rural location of Iberia, emphasizes the richness of the decoration of the Quinta das Longas villa in line with the most ostentatious standards of the time.

Author Contributions: Conceptualization, M.P.L.M., T.N.-B. and A.C.; methodology, M.P.L.M.; software, M.P.L.M.; validation, M.P.L.M., T.N.-B. and A.C.; formal analysis, T.N.-B. and A.C.; investigation, M.P.L.M., T.N.-B. and A.C.; resources, M.P.L.M., T.N.-B. and A.C.; data curation, M.P.L.M., writing—original draft preparation, M.P.L.M., T.N.-B. and A.C.; writing—review and editing, M.P.L.M.; funding acquisition, M.P.L.M., T.N.-B. and A.C. All authors have read and agreed to the published version of the manuscript.

Funding: This research was partially funded by the Spanish Government, Ministerio de Ciencia e Innovación, and is part of the objectives of the projects “Sulcato marmore ferro. Canteras, talleres, artesanos y comitentes de las producciones artísticas en piedra en la Hispania Tarraconensis” (PID2019-106967GB-I00).

Data Availability Statement: Data is contained within the article.

Acknowledgments: The authors would like to express their gratitude to M. Brillì, D. Attanasio, M. Bojanowski, Ph. Blanc and O. Boudouma for their respective laboratory facilities and to the reviewers for their constructive and useful remarks. They also acknowledge the use of Servicio General de Apoyo a la Investigación-SAI, Universidad de Zaragoza for technical support.

Conflicts of Interest: The authors declare no conflict of interest.

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