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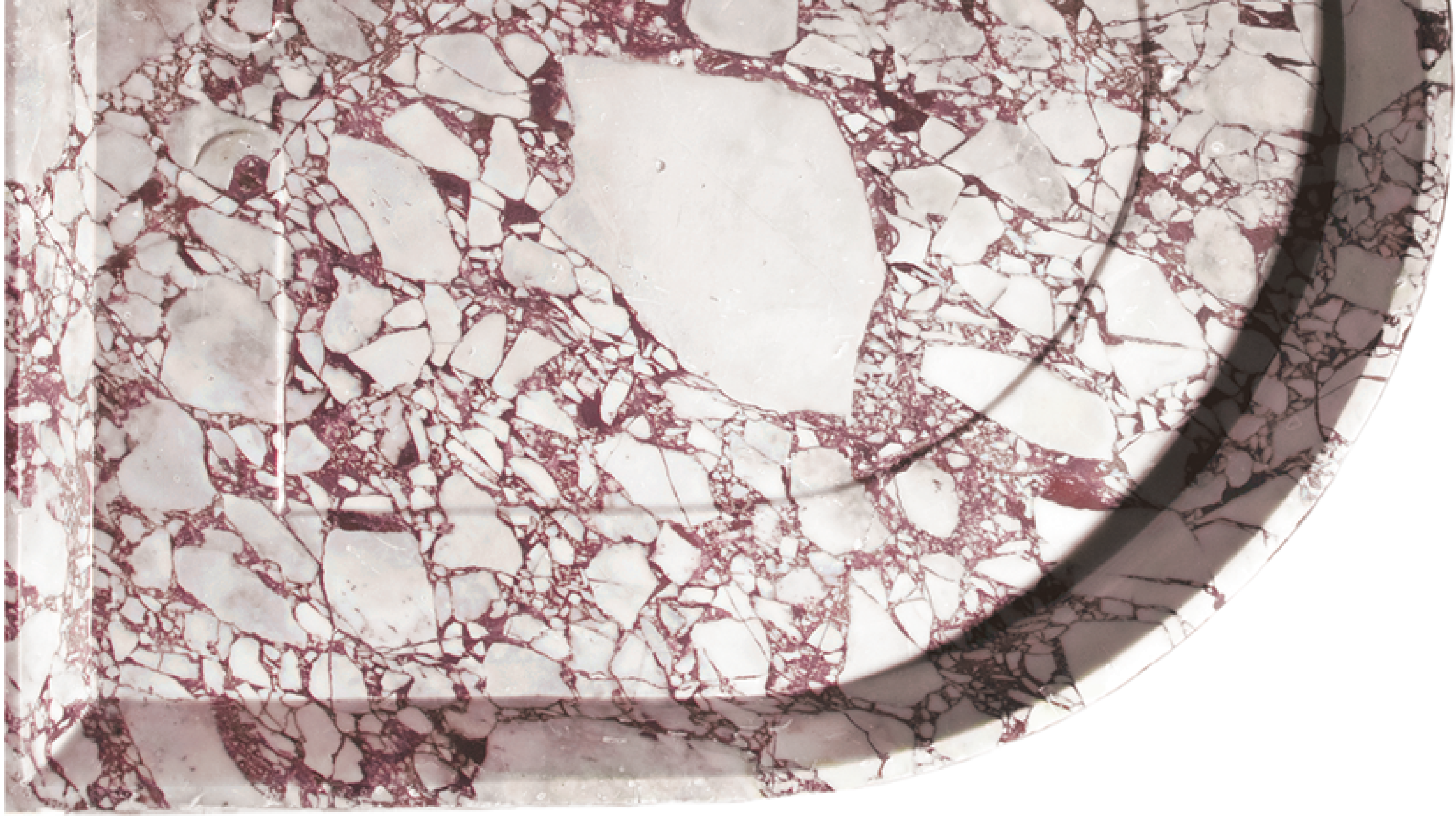

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CONTENT

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GREY AND GREYISH BANDED MARBLES FROM THE ESTREMOZ ANTICLINE IN LUSITANIA

Pilar Lapuente[1,3], Trinidad Nogales-Basarrate[2], Hernando Royo Plumed[1,3], Mauro Brilli[4] and Marie-Claire Savin[1,5]
[1] Petrology and Geochemistry, Earth Sciences Department, Zaragoza University, Zaragoza, Spain (plapuent@unizar.es; hroyo@icac.cat; marie-claire.savin@u-bordeaux-montaigne.fr)
[2] National Museum of Roman Art, Mérida, Spain (trinidad.nogales@mecd.es)
[3] Institut Català d´Arqueologia Clàssica (ICAC),Tarragona, Spain (plapuent@unizar.es; hroyo@icac.cat)
[4] Istituto di Geologia Ambientale e Geoingegneria, CNR, Area della Ricerca Roma1, Rome, Italy (mauro.brilli@igag.cnr.it)
[5] Institut de recherche sur les Archéomatériaux – Centre de Recherche en Physique Appliquée à l'Archéologie (IRAMAT-CRP2A), Université de Bordeaux Montaigne, Bordeaux, France (marie-claire.savin@u-bordeaux-montaigne.fr)

Abstract

This contribution addresses the characterization study of grey and greyish banded marbles from the Estremoz Anticline (EA) marble district in Portugal. During Roman times, the whole of *Lusitania*, including its capital *Augusta Emerita*, was supplied with white and coloured marbles from this district. Grey varieties were used mainly in architectural and epigraphic elements, funerary *stelae* and *arae*. The interest in the EA marbles focuses here on the grey types as they could have been used for trading beyond their provincial administrative domains, as was the case with the white marbles, in competition with other Iberian and classical materials, creating an increasing need to know how to identify them. A multi-method approach combining visual examination, spectrophotometry to quantify the greyscale tones, optical microscopy (OM), cathodoluminescence (CL) and stable C and O isotope analysis was applied to 25 quarry samples, which were collected from the south-eastern part of the EA structure.

Keywords
Estremoz Anticline quarries, Lusitanian grey marbles, provenance study

Introduction and aims

With few exceptions, when scholars talk about the use of the noble Roman material par excellence, they are referring to white statuary marble. However, in recent years, grey marbles used in antiquity have begun to attract great interest among the scientific community (LAZZARINI *et al.* 1999; LAZZARINI 2013) especially after the discovery of ancient quarries found in Asia Minor (ATTANASIO *et al.* 2009; BRUNO *et al.* 2012; YAVUZ *et al.* 2011, 2012). Several noble varieties also include those referred to as *bigio morato* and *bigio antico* in traditional literature, which have been scientifically studied in different archaeological papers (ATTANASIO *et al.* 2013; BRUNO *et al.* 2015). After comparisons with reference materials, there is a better perspective regarding the identification of their quarry sources (ATTANASIO *et al.* 2015, BRILLI *et al.* 2015; 2018). Additionally, banded or listed black and white varieties, and in particular the so-called *greco scritto* marbles, have been studied and their original sources have been redefined, either to the quarries near Ephesus, or those in North Africa (ANTONELLI *et al.* 2009; ATTANASIO *et al.* 2012; HERRMANN *et al.* 2012).

The perception of the use of grey materials in the Western Roman provinces is not comparable with the important role that they played in Rome or in other ancient locations related to the Aphrodisias workshops. However, in *Hispania,* grey marbles of different quality and features were extensively exploited and used for different purposes. In the majority of the Iberian marble districts, both white and coloured marbles were jointly extracted. But, it is clear that the workshops selected the materials according to their intended purpose. White marble was a preferable choice, not only for sculpture, as Pliny noted, but also for architectural elements using the polychrome types as part of the decorative programmes (FUSCO, MAÑAS 2006; NOGALES, GONÇALVES, LAPUENTE 2008; TAELMAN *et al.* 2013a). Most of these grey marbles could be classified, according to the traditional nomenclature, as *bigio antico*, as they vary from light to dark grey, almost black. But also the repertory includes marble banded in black and white or in patches which remind us of those of the *greco scritto* types. Here reference is made to grey and greyish banded marbles. Roman Lusitanian architectural ornamentation (columns, pavements and wall veneer slabs), funerary *stelae* and *arae*, *lapidae* and epigraphic elements are documented in different qualities of this material. Visigothic elements were also carved in white and grey veined marbles and in dark to very dark grey, many of them reused from Roman artefacts (VILLALÓN 2015).

Fig. 1a. One of the active quarries in the south of the EA district. b. Detail of the foliation developed on a grey marble outcrop

When grey marble pieces appear on an Iberian archaeological site, they are usually assigned to a local provenance if some supply quarry is relatively close. The problem arises when the location of the site is sufficiently far away from whatever area of regional supply, as imported materials were also brought into the Western provinces. It is in a case like this that archaeometric studies must distinguish local-regional or imported marble source.

In recent years, knowledge of the marbles used in antiquity in Hispania has made great progress, but much work still remains to be done. The situation regarding the use of each marble quarry is almost complete in some districts, while in others, several lithotypes are still waiting to be described and discriminated. With regards to the Lusitanian marbles of the EA, varieties of statuary quality have been defined, and their discrimination from other Hispanic marbles of wide use in other Roman provinces, such as the Almadén de la Plata marbles, has progressed thanks to the CL features. The updated analytical database of white marble has helped us better to recognize the white statuary marble of the EA used in artefacts found in *Augusta Emerita*, modern Mérida, the capital of Roman Lusitania. (LAPUENTE *et al.* 2014; NOGALES *et al.* 2015).

Once the updated database was put into implementation with relative success, the recognition of white statuary marble of the EA found in other Roman provinces advanced. Recently a white marble head (possibly of Tiberius) found in *Caesar Augusta* was identified as EA marble. This site located more than 700 km from the marble district of Estremoz and is currently the north-easternmost place from which this Portuguese material has been located (NOGALES *et al.* 2017; LAPUENTE *et al.* 2016).

In this context, this paper aims at highlighting the identifying characteristics of grey and banded marbles of the EA, combining visual examination, spectrophotometry to quantify the greyscale tones, OM, CL and stable C and O. A wider study with additional Iberian grey varieties, is being carried out, in which archaeological samples are also being analysed to check the consistency of the analytic quarry marble database generated.

Geological setting

The EA is one of the Variscan macrostructures in the Ossa Morena Zone, a major geological unit of the Iberian Massif in the SW of *Hispania*. During the Hercynian orogeny, this Zone underwent a complex structural-metamorphic evolution, governed mainly by transpressional tectonics with progressive and continuous deformation associated with regional metamorphism, generally of low grade (greenschists) but high temperature-low pressure thermal domes developed locally.

The EA, located in the Alto Alentejo province of Portugal, is an elongated NW-SE geological structure approximately 40 km long by 5-7 km wide. The stratigraphic sequence of the EA structure consists of a Precambrian detrital basement, a Lower Cambrian "Estremoz Dolomitic Formation", a Cambrian-Ordovician carbonate sequence referred to as the "Estremoz volcano-sedimentary carbonate Complex" (OLIVEIRA *et al.* 1991) and Silurian detrital deposits. According to CARVALHO *et al.* (2008), three main Variscan tectonic phases are usually distinguished. The D_1 episode produced isoclinal recumbent westward-verging folds with N-S oriented axes, schistosity with bedding transposition and low-grade regional metamorphism. With progressive deformation, the D_2 episode assigned to the Late Carboniferous age developed NW-SE trending fold axes and left-lateral shear zones, with discrete occurrence, showing deformation stages from ductile to brittle. Finally,

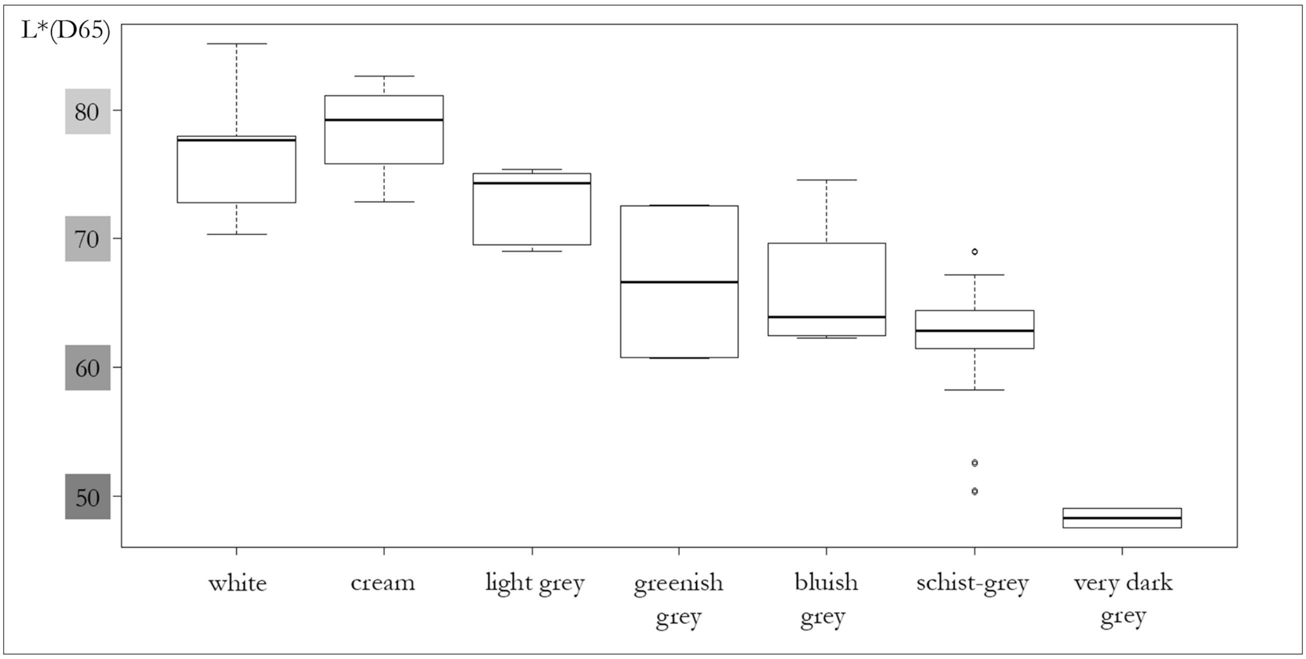

Fig. 2. Visual categories of grey (light, intermediate -greenish and bluish-, and dark to very dark grey according to the L*(65) parameter

during a late Variscan event, the EA was segmented by a network of NE-SW trending fractures, some associated with dolerite dikes.

The marbles form part of the Cambrian-Ordovician carbonate sequence whose age is still under discussion due to the lack of biostratigraphic and geochronological data. The metamorphic complex comprises a 300 m thick sequence of marbles and calc-schists with lenticular bodies of acid metavolcanites and metadolomites (CARVALHO *et al.* 2008; LAMBERTO, SÁ CAETANO, 2008). The light-coloured varieties, namely the white and cream coloured marbles, are found throughout the entire complex. The pink marbles, currently the most commercial materials, are associated with veined varieties interlayered with green metavolcanic rocks (TAELMAN *et al.* 2013b). Grey and dark grey marbles occur either in continuous levels at the top of the sequence or as lenses in the light-coloured unit showing alternating bands in white and grey (Figs. 1a, 1b). The current 400 marble quarries are concentrated next to the locations of Estremoz, Vila Viçosa and Borba, and particularly in the South, close to Pardais. For this study, 25 grey marbles were collected preferentially in the southern half of the EA structure.

Macroscopic description: Colour measurements and patterns

One of the macroscopic features is the colour, which was measured using a MINOLTA CM-2600d portable spectrophotometer whose SpectraMagic NX 2.5 software obtains different parameters. The instrument provides a direct display of CIE colour coordinates (X, Y, Z) along with CIELAB values (L*, a* and b*). In addition, the Whiteness index (WI) was calculated according to both CIE 1982 and the norm E313-96. Each test specimen was prepared with a flat unpolished surface, at least 2cm thick, to avoid any rough irregularity, and on which three small circles were drawn for each visual colour category (white, cream, light grey, greenish grey, bluish grey, dark schistose-grey and very dark grey). Three automatic measurements were taken in each of the small circles, using a window of 3 mm diameter. Other standard specifications were the D65 illuminant, and the 10º observer. Both modes, the specular reflectance excluded (SCE) and included (SCI), were simultaneously measured. In the SCE mode, only the diffuse reflectance is measured, producing a colour evaluation that correlates with the way the observer sees the colour of the specimen. When using the SCI mode, both the specular and the diffuse reflectance are taken into account during the measurement process, whose evaluation provides the total appearance independent of surface conditions.

In Fig. 2, the visual categories of grey are shown attending the L* parameter of Luminescence, which in most of the cases ranges from 60 to 75, excluding the very dark grey with values below 50. Light grey is well defined between 69 and 75. Intermediate grey tones (mostly in white/grey alternations) include some greenish grey ($60 < L^* < 73$), whose negative a*-axis represents the green shares of the spectrum, and bluish grey ($62 < L^* < 75$), whose negative b* -axis characterizes the blue portions. Many of the measured samples have a negative value of parameter b*, especially the dark greys, which determines

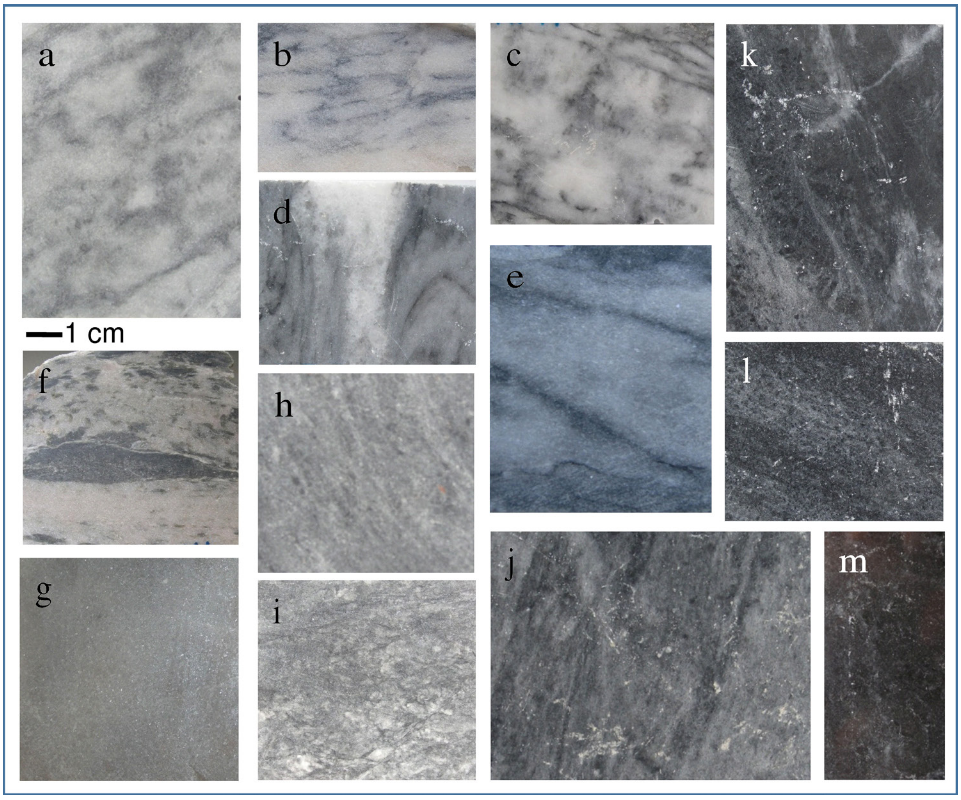

Fig. 3.
Different patterns on grey and greyish banded marbles. All on the same scale

a bluish tone not always visually evident. The L* measurements made in the schistose dark grey show a high range of values, the majority between 62 and 65, but with outliers at 50 and 69, due to the fine millimetre coloration, which is difficult to adjust to the size of the window. The L* parameter is quite well associated with the WI (ranging from 23 to 62; with values below 10 for the very dark grey).

The defined grey categories are differently combined, giving a high diversity of patterns with several visual aspects (Fig. 3). Three are the most common: -1) a diverse combination of white and grey in bands or in veins (Figs. 3 a, b, c, d, e, f), which show different grey tones, as sheath microfolds in dark grey (Fig. 3d), or lenticular greenish grey (Fig. 3f). They may all sometimes show differences in the width of bands and streaks; -2) homogeneous light grey or spotted in white (Figs. 3 g, h, i); and -3) dark to very dark grey (Figs. 3 j, k, l, m).

Mineralogical-petrographical and CL features

Several analytical techniques (XRD, OM, CL) were applied to characterize the mineralogical-petrographical and CL features. Experimental procedures were carried out according the methodology described elsewhere (LAPUENTE *et al.* 2014). Taking into account that foliated marbles were used by Romans in two orientations, thin-sections were prepared taken from 2 or 3 different sub-perpendicular alignments. This is particularly useful for the white and grey layered, veined and streaked samples, in which the foliation provides a different pattern with a white background crossed by spotted and irregular grey stripes, which in some cases could look like the so-called *greco scritto* marble.

XRD confirms calcite as the principal mineral in all the grey categories. Dolomite is present as an accessory mineral, always in aggregates of microcrystals. Quartz is relatively frequent, mono and polycrystalline, small granular but also in poikilitic and reabsorbed forms. K-feldspar, muscovite and chlorite are rare, while phlogopite appears quite often, especially in the greenish grey bands and occasionally in the dark grey samples, where graphite and Fe-bearing opaques are very common. One significant feature is the variation of the Maximum Grain Size (MGS) in the same sample depending on the thin-section orientation, which in many cases differs by 0.2-0.4 mm, though occasionally is in the order of 1 mm. However, the Most Frequent Size (MFS) is very fine in all samples, regardless of the orientation of the samples (Fig. 4). In most of the analyzed specimens, MGS is in the fine grain category (less than 2 mm long). When this parameter is in the coarse to very coarse grain, the texture is extremely heteroblastic, with a few isolated porphyroclasts in a very fine grain texture.

One of the most characteristic features of the EA marbles is the great variety of textures observed in the same thin-section. Depending on the orientation, the microstructural variation is even more evident, as Fig. 5 shows, where two sub-perpendicular thin-sections were taken from the same sample (on the left), or on the right,

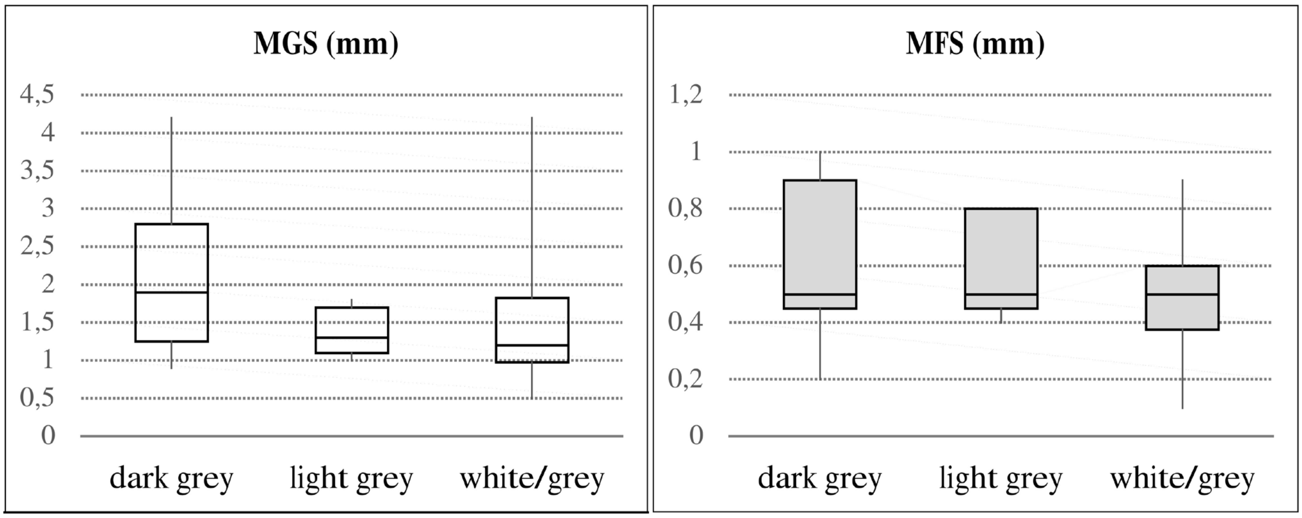

Fig. 4. Maximum grain size (MGS) and most frequent size (MFS) in the three categories of grey and greyish banded marbles from the EA, measured in 45 thin-sections. Quartiles and median are indicated in each box

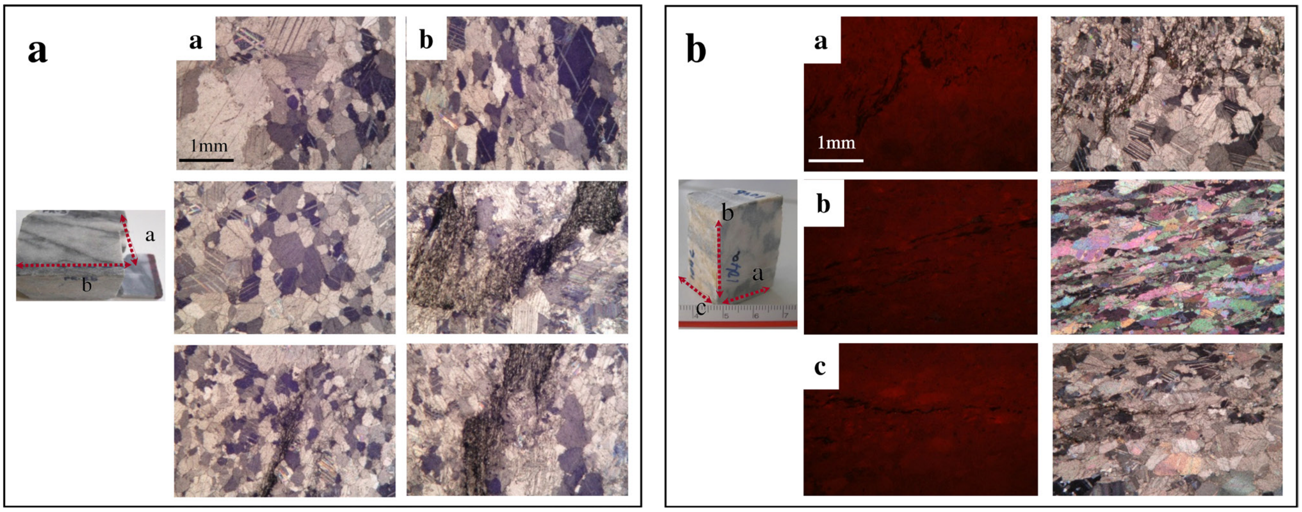

Fig. 5a. Three different microphotographs in each (a) and (b) thin sections of the same sample, where the variation is evident in texture (homeo/heteroblastic), fabric (iso-anisotropic), MGS and distribution of accessories. b. CL and petrographic microphotographs of three sub-perpendicular (a), (b) and (c) thin sections of the same sample

where both fabric and texture are quite different in view of three sub-perpendicular orientations. However, CL features are not affected by the thin-section orientation.

In the dark to very dark grey marbles (Figs. 6 a, b, c, d) it is common to find, in the same sample, a great variation of grain size combining zones with apparent isotropic and clearly anisotropic foliation. MGS ranges from 1 to 4.2 mm, but the MFS varies from 0/2 to 1 mm. (Fig. 4). High strained textures are frequent and signs of intracrystalline deformation are evident on the coarser grains of calcite (Fig. 6b). Lenticular and elongated aggregates of microdolomite with graphite are very common (Figs. 6a, 6d), but the carbonaceous matter is also dispersed in the dark grey type. Quartz, mono and polycrystalline, are frequent, especially in association with small white patches and very small white veins (Fig. 6c). Regarding CL, very dark grey marbles show medium luminiscence in reddish orange calcite, which occasionally exhibits an intracrystalline zoned CL of stronger intensity, with dispersed microdolomite, as shown in Fig. 6 d but also very faint to faint intensities are possible.

In the spotted light grey varieties (Figs. 6 e, f), microdolomite is dispersed (Fig. 6e), while the homogeneous light grey shows higher uniform CL intensity (Fig. 6f). Both subtypes of light grey are quite homeoblastic, as shown in Fig. 4 where the median of the MGS is 1.25 mm and the MFS is below 0.8 mm, with a median of 0.5 mm.

With the white and grey banded marbles (Figs. 6 g, h, i, j), two types have been distinguished; one is homeoblastic and extremely fine grained, with a heterogeneous

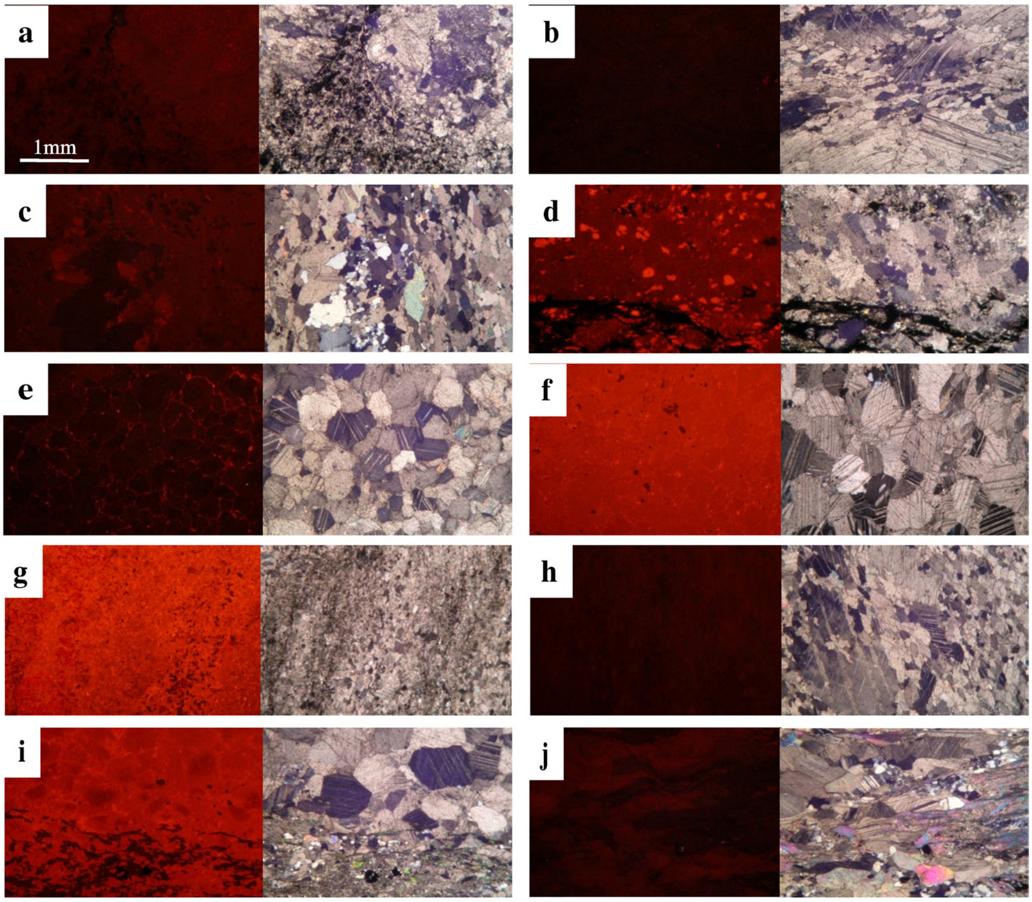

Fig. 6.
Selection of CL images and photomicrographs in crossed polarized light. Dark to very dark grey samples (a, b, c, d); light grey samples (e, f); white / grey samples (g, h); white / greenish grey samples (i, j)

distribution of CL in medium-high to high intensities (Fig. 6g), and a second heteroblastic with MGS ranging from fine to medium-coarse grained, in faint CL intensity (Fig. 6h). When the grey bands are schistose-greenish grey in white, phlogopite in lepidoblastic texture is developed and another two varieties have been differentiated. In the first, schistosity is defined by an alignment of platy small flakes (Fig. 6i) and in the second, by bands of massive phlogopite (Fig. 6j). CL shows heterogeneous distribution with medium to moderately strong intensities with intracrystalline zoning in the first, and faint intensity CL in the second.

Isotopic signature

Oxygen and carbon isotopes were determined by isotope ratio mass spectrometry with Finningan MAT 252 equipment following the standard procedures. The $d^{18}O$ and $d^{13}C$ (PDB) values are shown in Fig. 7 grouped by each lithotype. Most of them fall inside the EA isotopic field defined for the white marbles (LAPUENTE *et al.* 2014). Only 4 samples out of 25 are plotted outside. The isotopic $d^{18}O$ value is quite uniform, ranging mostly from -5.3 to -6.8‰, with two samples of lower values (close to -8‰). However, $d^{13}C$ varies from 0.3 to 3‰. The white and grey banded samples show a wide range of dC values and several samples from Pardais overlap the isotopic signature defined for the white marbles of Almadén de la Plata (AP) (Fig. 7a). Comparing the isotopic data with those from various Mediterranean sources of veined, streaked or banded, white and grey marbles, including the typical *greco scritto* from different authors (ANTONELLI *et al.* 2015, with data by ANTONELLI *et al.* 2009, 2010; ATTANASIO *et al.* 2012; HERRMANN *et al.* 2012), they fall into an area of the isotopic diagram with an extensive overlap (Fig. 7b). Therefore, the isotopes should be used in combination with other petrographic and CL parameters.

Conclusions

The differences in physical and/or compositional features of marbles collected at each quarry in the southern half of the EA district are minimal and depend largely on the specific collection of samples taken. The intra-quarry variation of features ranges from the macro to microscale. Moreover, the petrographic and CL variability found in samples from each quarry makes it difficult to highlight the marble fingerprints of each quarry. Consequently, instead of specifying characteristics of a single quarry, it is more reasonable to consider all the quarries of the EA district as one unique reference group.

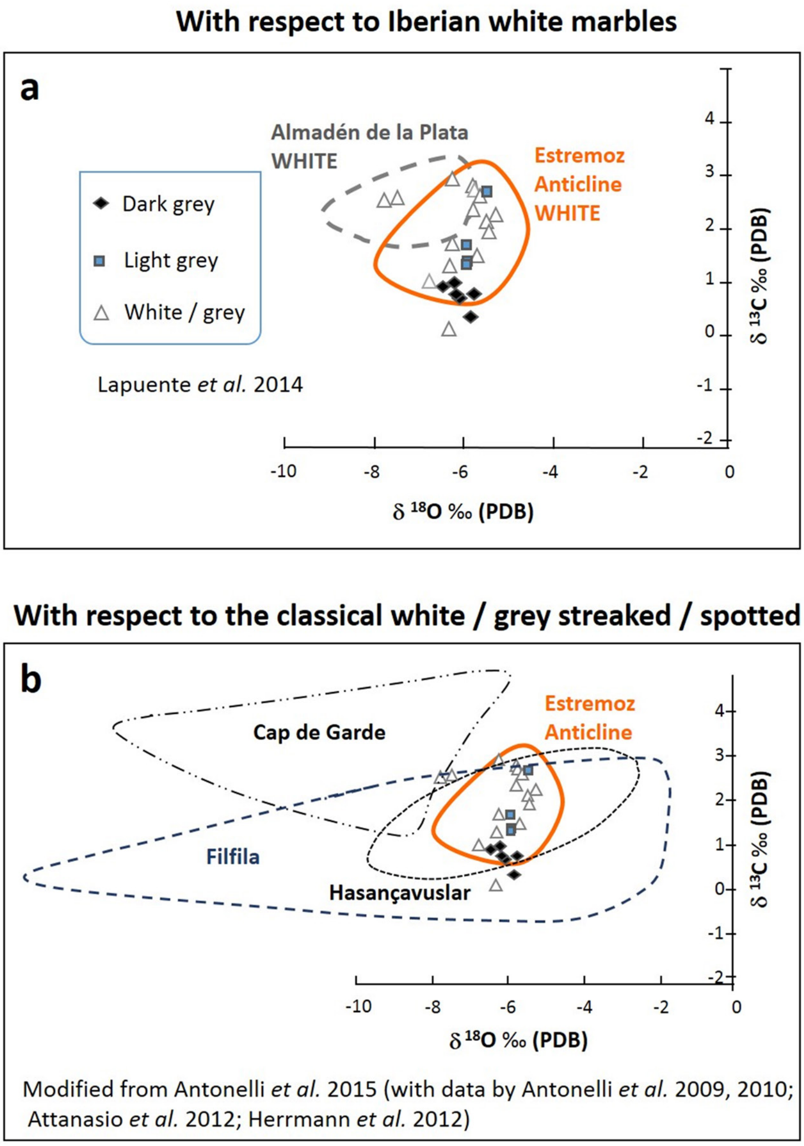

Fig. 7. Isotopic diagrams of the grey and greyish banded marbles from the Lusitanian EA district

Different lithotypes have been distinguished from the grey marbles of the EA according to the visual aspect, color, mineralogy, MGS, MFS, microstructure-texture and CL features along with the C and O isotopes. Regarding the visual appearance, three main varieties have been characterized, with subsequent subcategories:

Dark to very dark grey types (L* < 50): Macroscopically homogeneous, spotted or with small veins, in white. MGS ranges from fine to very coarse grain, but mostly below 2 mm; however MFS is very fine to fine grain size, below 1 mm.

Light grey types (69 < L*< 75): homogeneous or spotted in bluish dark grey, MGS ranges from 1 to 1,8 mm, while MFS is between 0,4 to 0,8 mm.

White/grey (intermediate tones, 60 < L* < 74) with a great diversity of patterns, either centimetric to millimetric bands in bluish grey, or in veins, spotted, sheath microfolds in dark bluish grey; or in schistose bands in greenish grey. MGS are in the fine grained range, some even in the very fine.

Petrographic features (microstructure-texture) are determined by the thin–section orientation. Signs of intracrystalline deformation are very common with occasional isotropic mosaic zones.

CL intensity varies from very faint to faint-medium intensity, but certain very fine grained samples in white/grey exhibit strong luminescence. The CL distribution is quite often homogeneous, but heterogeneous, patched and frequent zoned calcite is observed in areas of the same sample.

The isotopic signature of the grey and greyish EA marbles is quite uniform in the $d^{18}O$ value, but not in the $d^{13}C$ data. Altogether, almost all isotopic values are projected within the updated white EA isotopic field defined in LAPUENTE *et al.* 2014. Finally, extensive overlap exists with the isotopic values of the Mediterranean sources of white/grey marbles.

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