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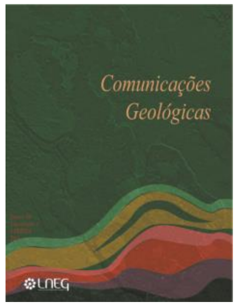

The Ossa-Morena marbles used in the Classical Antiquity: review of their petrographic features and isotopic data

Mármores da Zona de Ossa-Morena utilizados na Antiguidade Clássica: revisão das características petrográficas e isotópicas

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Abstract: The use of marbles during the Classical Antiquity, namely in the Roman Period, was a common practice, being extended during the 1st century. During this historical period, Hispania (Iberian Peninsula) was not an exception and there are several places where marbles were exploited with ornamental or architectonic purposes. In places like the Estremoz Anticlinal, Viana do Alentejo region, Trigaches-São Brissos, Alconera and Almadén de la Plata, evidence of ancient exploitation, attributed to the Roman Period, were recognized. In this work, these marbles are petrographically characterized and the published isotope data, namely regarding the $\delta^{13}C$ - $\delta^{18}O$ pair and $^{87}Sr/^{86}Sr$ ratio, are analyzed and discussed in the context of the geological processes and their application in provenance studies of materials used during Roman Period. The available data show that the application of both isotopic and petrographic studies allows the distinction of the several marbles of the Ossa-Morena Zone from each other, therefore ascribing a probable provenance for the classical marbles of this paleogeographic zone.

Keywords: Marbles, Ossa-Morena Zone, classical antiquity, isotope geology, petrography.

Resumo: A utilização de mármores na Antiguidade Clássica, nomeadamente durante a Época Romana, é uma prática comum e ampliada durante o século I d.C.. Durante este período histórico, a Hispania (Península Ibérica) não foi exceção e vários foram os locais onde estas rochas foram exploradas com fins ornamentais ou arquitetónicos, nomeadamente no Anticlinal de Estremoz, na região de Viana do Alentejo, em Trigaches-São Brissos, em Alconera e em Almadén de la Plata. Nestes locais foram encontradas evidências de exploração em épocas remotas, e atribuídas à Época Romana. Neste trabalho, caracterizam-se petrograficamente estes mármores e apresentam-se os dados isotópicos publicados por vários autores, nomeadamente o par $\delta^{13}C$ - $\delta^{18}O$ e a razão $^{87}Sr/^{86}Sr$, e comparados com dados obtidos em peças arquitetónicas. Estes dados são interpretados e discutidos no contexto dos processos geológicos atuantes e da sua aplicação a estudos de proveniência de materiais utilizados durante a Época Romana. Os dados mostram que a aplicação destes estudos isotópicos e o cruzamento com os dados petrográficos permitem distinguir os diversos mármores da Zona de Ossa-Morena, e assim atribuir proveniências prováveis para os mármores desta zona paleogeográfica.

Palavras-chave: Mármores, Zona de Ossa-Morena, antiguidade alássica, geologia isotópica, petrografia.

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1. Introduction

The isotopic characterization of marbles has been used as a distinguishing tool of the marbles used during the Classical Antiquity, namely for the Roman Period. The isotopic signatures, integrated with other methodologies (*e.g.* petrography, cathodoluminescence or elemental geochemistry), have been used to establish the fingerprint and the geological source of the marbles. Several papers compare the isotopic signatures of marbles collected in geological context (Cabral *et al*., 1992, 2001; Lapuente and Turi, 1995; Perez *et al*., 1998; Lapuente *et al*., 2000; Morbidelli *et al*., 2007; Origlia *et al*., 2011; Taelman *et al*., 2013a; Moreira *et al*., 2019) with marbles used in archaeological-architectural pieces (*e.g.* Lapuente *et al*., 2000; Cabral *et al*., 2001; Origlia *et al*., 2011; Taelman *et al*., 2013b), to trace the probable provenance of the marble's raw-materials.

This paper presents a synthesis of the published isotopic data from the Ossa-Morena Zone (OMZ) marbles, their comparison with data obtained in archaeological pieces, a discussion about the geological processes that can modify the isotopic contents in the OMZ marbles and the application of petrographic and isotopic data, namely $\delta^{13}C$ - $\delta^{18}O$ and $^{87}Sr/^{86}Sr$, as a provenance indicator of the OMZ marbles used during the Roman Period.

2. Use and exploitation of marbles in Iberia during Roman times

Marbles were considered a raw-material during the Classical Antiquity, particularly during the Roman Period, being closely linked to the culture and aesthetic sense of the Roman society (*e.g.* Fusco and Mañas, 2006; Antonelli *et al*., 2009). This is evident throughout the Iberian Peninsula, where marbles have been used in a diverse array of architectural and artistic elements (*e.g.* Encarnação, 1984; Cabral *et al*., 2004; Fusco and Mañas, 2006; Origlia *et al*., 2011; Taelman *et al*., 2013b; Lapuente *et al*., 2014). During the Roman Period, the marbles had diverse provenances, with several evidence of exploitation throughout the empire (*e.g*. Iberia, Italian, Balkan and Anatolia Peninsulas or North Africa; *e.g.* Lapuente *et al*., 2000; Russel, 2013a, b) and with the Mediterranean Sea used as a link for marble and other natural resources dispersion (*e.g*. Russel, 2013b; Moreira and Lopes, 2019; Terpstra, 2019). The intensive use of marble in the Roman Empire begins with Augustus (27 BC to AD 14) and, in the southern domains of the Iberian Peninsula (Hispania), two major marble exploitation domains are well documented (*e.g*. Lapuente *et al*., 2000; Fusco and Mañas, 2006; Mañas and Fusco, 2008; Beltrán *et al*., 2012; Russel, 2013a) (Fig. 1A):

(1) Betic Belt, in south-eastern Spain, more specifically in Málaga (e.g. Mijas) and Alméria (e.g. Macael) districts;
(2) OMZ, namely the Estremoz Anticline, Viana do Alentejo, Trigaches-São Brissos (Portugal), Alconera, San Pedro-Carija and Almadén de la Plata (Spain).

The previous mentioned OMZ sites present evidence of exploitation activities during Roman Period (*e.g.* Justino Maciel, 1998; Fusco and Mañas, 2006; Nogales *et al*., 2008; Beltrán *et al*., 2012; Mañas, 2012), although most of the archeological records must have been erased by the recent quarrying activity (Fusco and Mañas, 2006). As examples, remains of Roman exploitation were found in the Estremoz Anticlinal (Fig. 1B; sarcophagus and incipient sculptures were found in Herdade da Vigária; *e.g.* Justino Maciel, 1998) or in Almadén de la Plata (Fig. 1C; Roman ancient quarry in Cerro de Los Covachos; Beltrán *et al*., 2012); among those extractive poles in the OMZ, the previous mentioned places may have been the main proto-industrial extractive centers of Hispania (Fusco and Mañas, 2006; Nogales *et al*., 2008; Beltrán *et al*., 2012; Carneiro, 2014, 2019; Moreira and Lopes, 2019).

Alconera and Almadén de la Plata are located in the Hispania Baetica Province, while the remaining OMZ exploitation poles belonged to the Hispania Lusitania Province (Fig. 1A). The geographical dispersion of OMZ marbles along the Roman Empire was highly dependent on transportation and communication routes (the Viae Iberiae and the waterways), and consequently some marbles were locally dispersed (e.g. Alconera and Trigaches- São Brissos marbles), while others were scattered throughout all the Hispania provinces, and were also used outside Hispania (*e.g.* Estremoz Anticline and Almadén de la Plata marbles; Fusco and Mañas, 2006; Origlia *et al*., 2011; Antonelli *et al*., 2015).

3. Ossa-Morena Zone Marbles used during Roman Period: geological framework and petrographic features

3.1. Geological framework

The Iberian Massif is composed of pre-Mesozoic rocks and is divided into several paleogeographic zones, including the OMZ, which represents the southernmost zone of the Iberian Terrain (*e.g.* Ribeiro *et al*., 2007; Dias *et al*., 2016). The stratigraphic, magmatic and metamorphic features of the OMZ are closely related its geodynamic evolution during the Variscan Cycle (*e.g.* Oliveira *et al*., 1991; Ribeiro *et al*., 2010; Moreira *et al*., 2014a); those features allow the definition of lithostratigraphic successions, ranging from Neoproterozoic to Carboniferous (*e.g.* Oliveira *et al*., 1991; Araújo *et al*., 2013). The analysis of the OMZ lithostratigraphic successions records four marine carbonated sedimentation episodes during Palaeozoic times (*e.g*. Oliveira *et al*., 1991; Robardet and Gutiérrez-Marco, 2004) among which the Early Cambrian episode is the better represented episode; its age (Series 2) is constrained by the fossil content in the Alconera and Pedroche successions (*e.g*. Liñan, 1984; Creveling *et al*., 2013). This episode is characterized by a shallow marine carbonate platform succession, with limestones and dolostones and subordinate volcanic and siliciclastic rocks (Liñan, 1984; Oliveira *et al*., 1991). However, the primary textural features and the fossil content of these carbonates have often been completely obliterated by the Variscan tectono-metamorphic events, resulting in calcite and dolomite marble-rich successions (Estremoz, Viana do Alentejo, Trigaches-São Brissos and Almadén de la Plata successions). In these cases, the age assignment is based on lithostratigraphic correlations (*e.g*. Oliveira *et al*., 1991; Araújo *et al*., 2013), sometimes supported by geochemical data (*e.g*. Puelles *et al*., 2018; Pereira *et al*., 2012; Moreira *et al*., 2019).

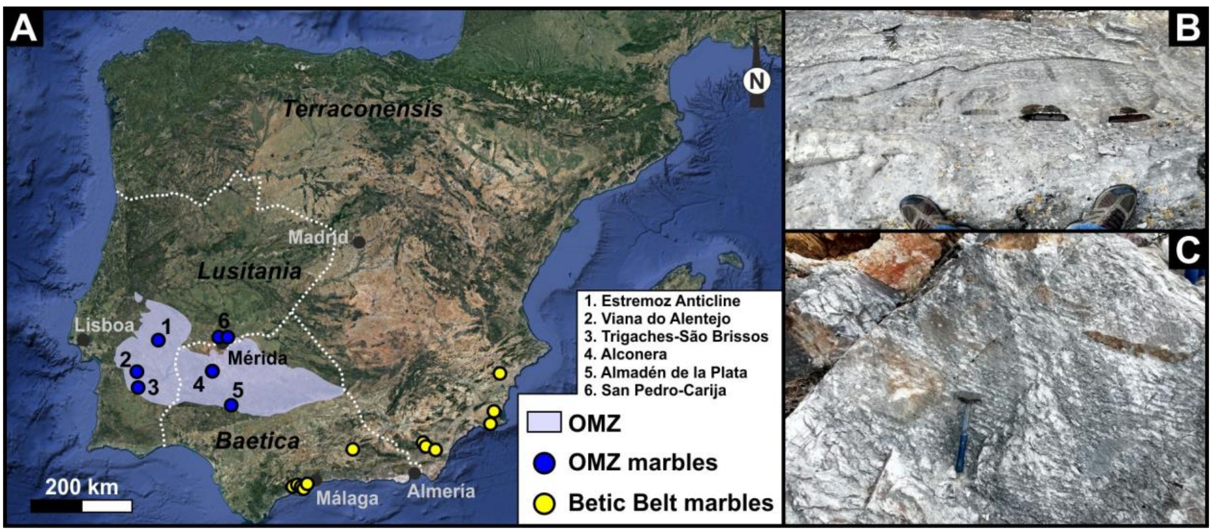

Figure 1. Classical marbles exploitations from the southern domains of the Iberian Peninsula (Hispania): A – location of recognized marble quarries during the Roman Period, emphasizing the Betic belt and Ossa-Morena Zone clusters (data base from Russel, 2013a); B – wedges and groove marks from Herdade da Vigária, Estremoz Anticline (currently exposed in the Vila Viçosa Castle); C – roman exploitation relicts from ancient quarry in Cerro de Los Covachos, Almadén de la Plata.

Figura 1. Explorações de mármores clássicos nos domínios meridionais da Península Ibérica (Hispânia): A – localização das explorações de mármore durante o período romano, enfatizando os clusters da cordilheira bética e da Zona de Ossa-Morena (adaptado da base de dados de Russel, 2013a); B – marcas da cunha em mármores da Herdade da Vigária, Anticlinal de Estremoz (atualmente exposto no Castelo de Vila Viçosa); C – marcas de exploração romana na Pedreira de Cerro de Los Covachos, Almadén de la Plata.

3.2. Petrographic features

Some OMZ Cambrian marbles were exploited during the classical period (probably since the 1st century; Fusco and Mañas, 2006; Russell, 2013a; Moreira and Lopes, 2019 and references therein). In the following section, a brief picture of the geological framework and petrographic features for each of these marbles is presented.

- *Alconera*

The "Alconera Marbles" are fine-grained and include a set of distinctive lithotypes, with calcite and dolomite varieties (sometimes with marls and recrystallized carbonates), being affected by very low temperature metamorphism (López-Munguira and Nieto García, 2004). The macroscopic features are a key-piece in the identification of these marbles: they are heterogeneous in color, from white and pale grey to dark grey, pink and purple specimens (the most distinctive ones) and preserve their primary textures and fossiliferous content (Fig. 2A), such as archaeocyatha and stromatolite reef facies, as well as microbial and algae structures (Liñan, 1984).

- *Estremoz Anticline*

In the Estremoz Anticline, two marble units were identified: the dolomitic basal unit - without ornamental interest - and the top unit composed of calcite marbles, from which the Estremoz Marbles are exploited. The Estremoz Marbles are generally homogeneous, ranging from white and cream (most common), pink (often with green stripes) and dark-grey to black varieties (*ruivina*); sometimes the marbles present compositional stripes, providing chromatic patterns. These marbles are calcite-rich (usually > 95%; Fig. 2B), with accessory mineralogy composed of quartz, biotite, muscovite/sericite, chlorite, feldspars s.l., dolomite, oxides and pyrite (Casal Moura *et al*., 2007; Lopes and Martins, 2015; Moreira *et al*., 2019). They are medium to fine-grained marbles (average values between 0.5 to 1mm, although grain size may vary between 0.2-4.0 mm; Fig. 2B), with granoblastic to granolepidoblastic texture (Lapuente and Turi, 1995; Menningen *et al*., 2018).

- *Trigaches-São Brissos*

These marbles are calcite-rich and have distinctive features, such as their dark grey to bluish tones (sometimes with dark grey stripes) and their coarse-grained homogeneous mosaic texture (generally greater than 1 mm; Fig. 2C; Carvalho *et al*., 2013). The calcite-rich marbles are the most common variety, however Ferreira *et al*. (2013) describes impure lithotypes, also with very coarse-grained calcite, but with the presence of olivine (forsterite) and other ferromagnesian minerals. Another particularity of these marbles, is the foul odor when struck, due to the presence of dispersed organic matter.

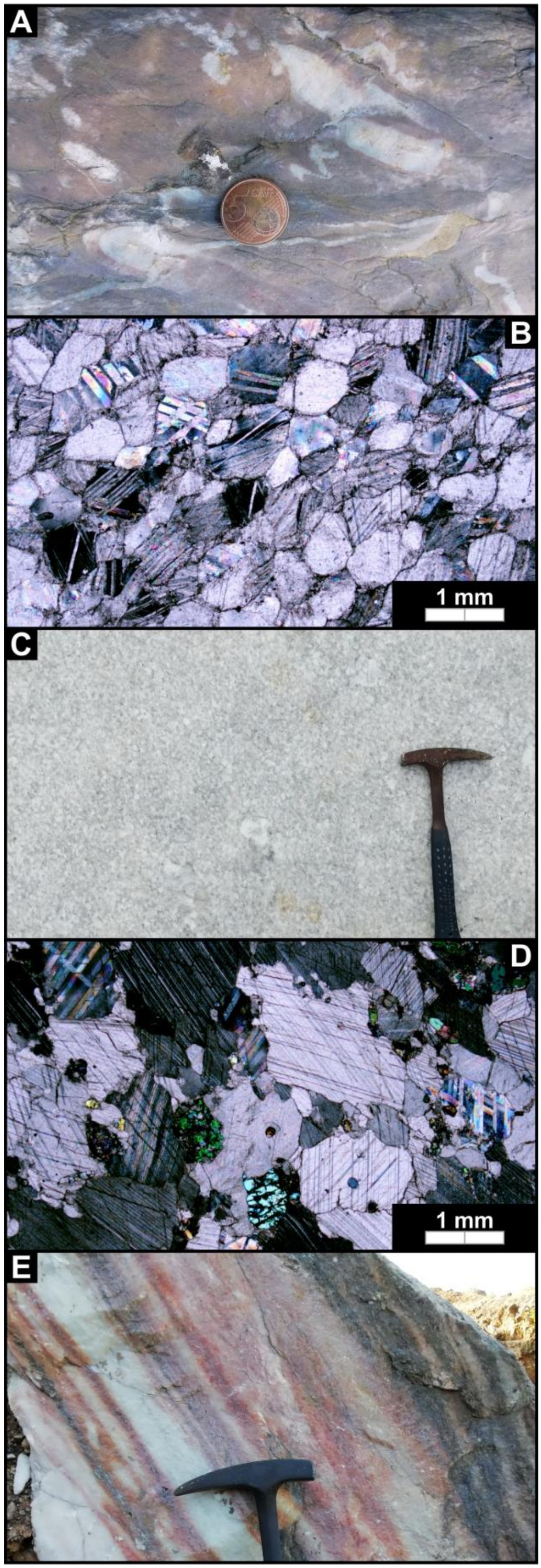

Figure 2. Macro and microscopic features of Ossa-Morena Zone marbles: A – Alconera Marble showing their typical pink-purple tones and their primary fossiliferous content; B – Estremoz Anticline medium-grained calcite marble, showing granoblastic texture; C – Trigaches-São Brissos coarse-grained calcite marble; D – Viana do Alentejo coarse-grained calcite marble, with granoblastic texture and olivine crystals; E – Almadén de la Plata coarse-grained calcite striped marble, with whitish to pink tones.

Figura 2. Características macro e microscópicas dos mármores da Zona de Ossa-Morena: A – mármore de Alconera mostrando as tonalidades rosa-violeta típicas e as características texturais e fossilíferas primárias; B – mármore calcítico de grão médio do Anticlinal de Estremoz, mostrando textura granoblástica; C – mármore calcítico de grão grosseiro de Trigaches-São Brissos, de elevada pureza mineralógica; D – mármore calcítico de grão grosseiro, com cristais de olivina e textura granoblástica; E – mármore calcítico de grão médio-grosseiro de Almadén de la Plata, mostrando bandado de cores claras a rosadas.

- Viana do Alentejo

These marbles have distinctive features, namely their green massive to green ornate variety (the so-called "Verde Viana"), clearly distinct from the other OMZ marbles, though presenting medium to coarse-grained texture (commonly greater than 1mm; Fig. 2D). However, other varieties are also representative of this region, such as the calcite-rich varieties with light tones (quite similar to Almadén de la Plata marbles). These marbles generally show granoblastic texture, although sometimes striped marbles, with granolepidoblastic texture, are described, presenting a well-marked mineralogical and textural foliation (Gomes and Fonseca, 2006). These marbles are dolomite or calcite-rich, with a diverse accessory mineralogy, including (Gomes and Fonseca, 2006; Rosas *et al*., 2008; Moreira *et al*., 2019): quartz, olivine (Fig. 2D), serpentine, wollastonite, biotite, tremolite, diopside, feldspars *s.l.*, chlorite, epidote, talc, garnet and sphene; sometimes iron oxides (magnetite and hematite) and sulfides are also described.

- Almadén de la Plata

These marbles are medium to coarse-grained (crystal sizes higher than 1mm) with white to pink tones (most common; Fig. 2E), generally with stripes, although black-grey and greenish varieties are also described (Ontiveros *et al*., 2012; Puelles *et al*., 2018); there are varieties of high chromatic uniformity (white and dark ones). Usually, they are calcite-rich (80-97%), but dolomite phase can be higher than 10%; subordinate dolomite-rich varieties are also present in this succession (Puelles *et al*., 2018). These marbles show granoblastic texture and diverse accessory mineralogy (mainly in the green and grey varieties), although its abundance is lower than 5%, consisting of quartz, olivine, diopside, biotite, spinel, titanite, muscovite, serpentine, feldspars *s.l.*, tremolite, talc, iron oxides and chlorite (Ontiveros *et al*., 2012; Puelles *et al*., 2018). It is common the presence of more than one carbonate generation in dolomitic marbles with heterogeneous grain size and recrystallization degrees (Puelles *et al*., 2018).

4. Isotope Data from Ossa-Morena Zone Marbles

The first studies for the isotopic characterization of OMZ marbles used the ^{13}C and ^{18}O stable isotope pair. The $\delta^{13}C$ and $\delta^{18}O$ pair obtained in the OMZ marbles collected in geological context (Cabral *et al*., 1992, 2001; Lapuente and Turi, 1995; Perez *et al*., 1998; Lapuente *et al*., 2000; Lopes *et al*., 2000; Origlia *et al*., 2011; Morbidelli *et al*., 2007) were compiled (including new data – Tab. 1) and a statistical analysis of $\delta^{13}C$ and $\delta^{18}O$ pair from Alconera, Estremoz Anticline, Viana do Alentejo, Almadén de la Plata and Trigaches-São Brissos marbles is now performed (Tab. 2), thus allowing the review of the isotopic fingerprint and the identification of $\delta^{13}C$ and $\delta^{18}O$ anomalous values.

The $\delta^{18}O$ values are quite similar in marbles from Estremoz Anticline, Viana do Alentejo, Almadén de la Plata and Trigaches-São Brissos, while the Alconera marbles present a distinct signature, presenting systematically lower $\delta^{18}O$ values (Tab. 2; Fig. 3A). If the outliers are excluded, the Alconera and Estremoz Anticline marbles data decrease their dispersion and the Estremoz Anticline slightly differ from the Viana do Alentejo and Almadén de la Plata marbles, both with slightly lower values and great dispersion of $\delta^{18}O$ values. On the other hand, the $\delta^{13}C$ is quite similar in all the marbles; the Alconera marbles have lower data dispersion, while the highest spread of data is obtained in the Viana do Alentejo marbles (Fig. 3A and Tab. 2).

Table 1. δ13C and δ18O data obtained in Ossa-Morena Zone marbles (this work) and their mineralogy (*Laboratório de Isótopos Estáveis - Departamento de Geologia da Faculdade de Ciências da Universidade de Lisboa).

Tabela 1. Dados δ13C e δ18O obtidos em mármores da Zona de Ossa-Morena (este trabalho) e a sua mineralogia (*Laboratório de Isótopos Estáveis - Departamento de Geologia da Faculdade de Ciências da Universidade de Lisboa).

	Locality	Typology	Main carbonate phase	Accessory mineralogy	$\delta^{13}C$*	$\delta^{18}O$*
ETZ-2	Estremoz Anticline	White marble	calcite	quartz + muscovite	2.74	-5.78
ETZ-3	Estremoz Anticline	White marble	calcite	dolomite + quartz + muscovite	2.87	-6.06
ETZ-5	Estremoz Anticline	Dark-grey marble	calcite	quartz + muscovite + organic matter	1.49	-7.31
VIA-1	Viana do Alentejo	Green marble	calcite	quartz + olivine + epidote + titanite + brucite + clay minerals	2.18	-7.87

Table 2. Statistical data summary table of $\delta^{13}C$ and $\delta^{18}O$ data (including new data and data from: [1] Lapuente *et al*., 2000; [2] Morbidelli *et al*., 2007; [3] Origlia et al., 2011; [4] Perez *et al*., 1998; [5] Cabral *et al*., 1992; [6] Cabral *et al*., 2001; [7] Lapuente and Turi, 1995; [8] Lopes *et al*., 2000).

Tabela 2. Tabela sumária da análise estatística dos dados de $\delta^{13}C$ e $\delta^{18}O$ (inclui novos dados e dados de: [1] Lapuente *et al*., 2000; [2] Morbidelli *et al*., 2007; [3] Origlia *et al*., 2011; [4] Perez *et al*., 1998; [5] Cabral *et al*., 1992; [6] Cabral *et al*., 2001; [7] Lapuente and Turi, 1995; [8] Lopes *et al*., 2000).

REF	**Alconera** [1] [2]		**Estremoz Anticline** [1] [2] [3] [4] [5] [6] [7]		**Viana do Alentejo** [1] [2] [3]		**Almadén de la Plata** [1] [2] [3] [4]		**Triagaches** [8]	
	$\delta^{13}C$	**$\delta^{18}O$**	**$\delta^{13}C$**	**$\delta^{18}O$**	**$\delta^{13}C$**	**$\delta^{18}O$**	**$\delta^{13}C$**	**$\delta^{18}O$**	**$\delta^{13}C$**	**$\delta^{18}O$**
n	21		229 +3		36 +1		36		1	
MIN	1.40	-15.18	0.10	-9.80	-1.69	-10.37	0.10	-11.04	-	-
MIN NO	2.40	-12.30	0.20	-7.70	-1.69	-10.37	1.10	-11.04	-	-
Q1	2.60	-11.60	1.36	-6.40	0.20	-8.20	2.05	-8.18	-	-
MED	2.73	-10.90	1.66	-5.86	0.87	-7.30	2.62	-6.43	1.67	-4.64
Q3	2.90	-10.20	2.19	-5.51	2.34	-6.35	2.87	-5.96	-	-
MAX NO	3.10	-9.10	3.20	-4.56	3.00	-5.79	3.50	-5.24	-	-
MAX	3.10	-9.10	3.20	-4.56	3.00	-5.79	3.50	-5.24	-	-
n outliers	3	1	1	13	0	0	3	0	-	-
$\bar{x} \pm \sigma$	2.60 ± 0.47	-11.05 ± 1.27	1.74 ± 0.58	-6.08 ± 0.83	1.24 ± 1.26	-7.43 ± 1.21	2.37 ± 0.82	-7.16 ± 1.66	-	-
$\bar{x} \pm \sigma$ (without outliers)	2.77 ± 0.18	-10.84 ± 0.90	1.74 ± 0.58	-5.94± 0.62	1.24 ± 1.26	-7.43 ± 1.21	2.56 ± 0.56	-7.16 ± 1.66	-	-

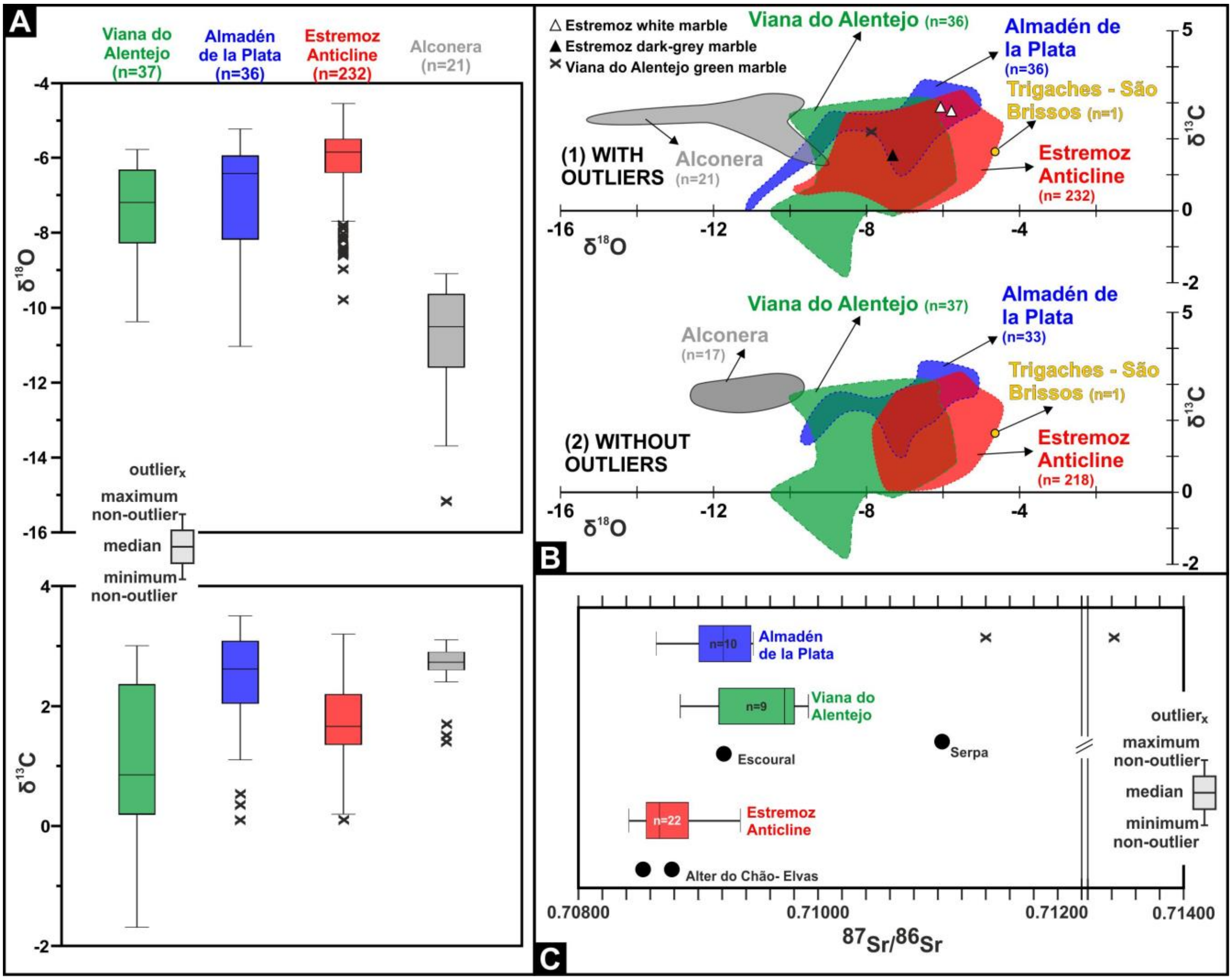

Figure 3. Isotopic features of Ossa-Morena Zone marbles: A – Statistical characterization of the $\delta^{13}C$ and $\delta^{18}O$ data (database includes new analyses and data from: Cabral et al., 1992, 2001; Lapuente and Turi, 1995; Perez et al., 1998; Lapuente et al., 2000; Lopes et al., 2000; Origlia *et al*., 2011; Morbidelli *et al*., 2007); B – $\delta^{13}C$ vs $\delta^{18}O$ cartesian projections and definition of typical scattered areas of Ossa-Morena Zone marbles, with and without outliers; C – Box-plot diagram for $^{87}Sr/^{86}Sr$ data (database includes data from: Morbidelli et al., 2007; Taelman *et al*., 2013a; Moreira *et al*., 2019).

Figura 3. Caracterização isotópica dos mármores da Zona de Ossa-Morena: A – Caracterização estatística dos dados de $\delta^{13}C$ e $\delta^{18}O$ (base de dados inclui novos dados e os dados de: Cabral *et al*., 1992, 2001; Lapuente and Turi, 1995; Perez *et al*., 1998; Lapuente *et al*., 2000; Lopes *et al*., 2000; Origlia *et al*., 2011; Morbidelli *et al*., 2007); B – Projeção cartesiana do par $\delta^{13}C$ vs $\delta^{18}O$, com definição das áreas típicas para cada um dos mármores da Zona de Ossa-Morena, incluindo e excluindo os valores anómalos; C – Diagrama de bigodes com a razão isotópica $^{87}Sr/^{86}Sr$ (base de dados inclui dados de: Morbidelli *et al*., 2007; Taelman *et al*., 2013; Moreira *et al*., 2019).

Two $\delta^{13}C$ - $\delta^{18}O$ diagrams were presented (Fig. 3B): (1) including all the stable isotopes pairs obtained in the OMZ marbles collected in geological context and (2) excluding all the outliers from database. The new data from Estremoz and Viana do Alentejo are similar to those published in previous works (Cabral *et al*., 1992, 2001; Lapuente and Turi, 1995; Perez *et al*., 1998; Lapuente *et al*., 2000; Lopes *et al*., 2000; Origlia *et al*., 2011; Morbidelli *et al*., 2007). The exclusion of outliers from the database clearly allows to reduce the scattered area defined for each of the OMZ marbles. This isotopic approach is unable to clear distinguish most groups of OMZ marbles from each other (Fig. 3B). Indeed, analysis of the $\delta^{13}C$ - $\delta^{18}O$ pair diagram shows a partial overlap of the Estremoz Anticline, Viana do Alentejo, Almadén de la Plata and Trigaches-São Brissos marble stable isotope signatures; generally, the Estremoz Anticline have slightly higher values of $\delta^{18}O$ and the Viana do Alentejo marbles display some negative $\delta^{13}C$ values. The Alconera isotope data clearly define an autonomous cluster, without overlapping the other OMZ marbles clusters, due to their lower $\delta^{18}O$ (Fig. 3B).

More recently, the $^{87}Sr/^{86}Sr$ ratio were also used to characterize and differentiate marble types. This ratio on marbles reflects the seawater isotope signature, considered stable and uniform at a certain moment in the geological time (McArthur *et al*., 2012) and acquired during the limestones genesis; however, they can suffer minor changes due to the geological processes acting during their petrogenetic evolution (Moreira *et al*., 2018, 2019). The analysis of strontium ratio published data, from the Estremoz Anticline, Almadén de la Plata and Viana do Alentejo marbles (Morbidelli *et al*., 2007; Taelman *et al*., 2013a; Moreira *et al*., 2019) show that the $^{87}Sr/^{86}Sr$ ratios are consistently higher in the Almadén de la Plata and Viana do Alentejo marbles when compared to those obtained for the Estremoz Anticline marbles, where the isotopic signature shows small variation (Fig. 3C; Tab. 3). The overlap between the data of the Estremoz marbles and the Almadén de la Plata and Viana do Alentejo ones is quite low (overlapping ca. 50% of the data; Tab. 3). The statistical study of the sample set, indicates the presence of two abnormally high results (outliers) in Almadén de la Plata (Fig. 3B).

Table 3. Statistical data summary table of $^{87}Sr/^{86}Sr$ data (data from: [1] Morbidelli *et al*., 2007; [2] Taelman *et al*., 2013a; [3] Moreira *et al*., 2019).

Tabela 3. Tabela sumária da análise estatística dos dados de $^{87}Sr/^{86}Sr$ (base de dados inclui: [1] Morbidelli *et al*., 2007; [2] Taelman *et al*., 2013a; [3] Moreira *et al*., 2019).

$^{87}Sr/^{86}Sr$	Estremoz Anticline	Viana do Alentejo	Almadén de la Plata
REF.	[1] [2] [3]	[1] [3]	[1]
n	22	9	10
MIN NO	0.70842	0.70885	0.70865
Q1	0.70856	0.70917	0.70901
MED	0.70867	0.70972	0.70921
Q3	0.70892	0.70980	0.70944
MAX NO	0.70949	0.70992	0.70946
n outliers	0	0	2
$\ddot{x} \pm \sigma$	0.70881± 0.00032	0.70951± 0.00039	0.70977± 0.00141

5. The use of petrographic and isotopic features on archeological marbles: an assay

The $\delta^{13}C$ - $\delta^{18}O$ studies for the isotopic characterization of OMZ marbles allowed, in many cases, the differentiation between the OMZ and some Mediterranean Classical marbles, such as Carrara (Italy), Thassos or Paros (Greece) marbles (Lapuente and Turi, 1995; Antonelli and Lazzarini, 2015). However, several marbles with petrographic similarities with Estremoz Anticline or Almadén de la Plata marbles, as those from the Macael (Spain), Aphrodisias, Dokimeion (Turkey) or Pentelikon (Greece), generally shows similar $\delta^{13}C$ - $\delta^{18}O$ values (Fig. 4A; Lapuente and Turi, 1995; Morbidelli *et al*., 2007; Antonelli and Lazzarini, 2015). Nevertheless, the $^{87}Sr/^{86}Sr$ isotopic signature of OMZ marbles seems to be a better approach to differentiate the OMZ marbles from each other and from Mediterranean marbles. Indeed, this approach clearly distinguishes the Estremoz Anticline, Viana do Alentejo or Almadén de la Plata marbles (with higher $^{87}Sr/^{86}Sr$ values), from those of Carrara, Paros, Macael, Aphrodisias or Dokimeion (Fig. 4B; Betic and Mediterranean marbles database from Morbidelli *et al*., 2007 and Rondolino *et al*., 2020). The $^{87}Sr/^{86}Sr$ ratios of the OMZ marbles overlap the values reported from Pentelikon and Thasos; however, it should be noticed that wider ranges are obtained in the two last places, where the marbles can have values much lower than those recorded by OMZ samples.

Several archeological pieces (*e.g.* capitals, reliefs, statues, and sarcophagus) were considered as created with marbles from the OMZ, based on macroscale similarities or petrographic and geochemical studies (*e.g.* Encarnação, 1984; Beltrán, 2013; Taylor *et al*., 2017; Beltrán *et al*., 2018). In some cases, even without isotopic analysis, the study of macroscopic features, the micro-textures and mineralogy seem to be adequate to identify the provenance of OMZ marbles, namely:

(1) The Alconera marbles (Fig. 2A), due to their clear distinctive macroscopic features, such as their tones, textures and their fossiliferous content, were recognized in Regina or Itálica (e.g. Beltrán, 2013), showing local dispersion.

(2) The Viana green marble were locally used (Encarnação, 1984), being possible their dispersion along Roman Empire (Cardoso *et al*., 2011). However, similar green marbles are reported in other places with evidence of exploitation during Roman Period (*e.g.* Almadén de la Plata).

(3) The macroscopic features of Trigaches-São Brissos marbles (Fig. 2C), namely as their grey-bluish tones and coarse-grained textures, allowed to sign this provenance to several marbles used in architectural pieces. Their local dispersion is noticeable, with several archeological pieces spread in southern Lusitania (*e.g.* Beja, Mértola or Alcácer do Sal; Encarnação, 1984, 2015; Cardoso *et al*., 2011), however their dispersion could not be fully accounted and may be undervalued.

When the macro and microscopic features do not allow to discriminate the geographical and geological sources (e.g. varieties of white and dark-grey marbles), it is necessary perform isotopic studies in archeological pieces, in order to trace their possible provenance. Several authors used the $\delta^{13}C$ - $\delta^{18}O$ pair (Lopes *et al*., 2000; Cabral *et al*., 2001, 2004; Justino Maciel *et al*., 2002a, b, 2003, 2006; Justino Maciel and Cabral, 2008; Antonelli *et al*., 2009, 2015; Royo *et al*., 2010; Origlia *et al*., 2011; Lapuente *et al*., 2014; Álvarez *et al*., 2017; Soutelo *et al*., 2018) to trace the marble provenance, while other authors applied the $^{87}Sr/^{86}Sr$ isotope ratio approach (Taelman *et al*., 2013b).

The $\delta^{13}C$ - $\delta^{18}O$ results obtained in white and dark-grey marbles from archeological pieces seem to demonstrate that the OMZ marbles use spills out of Iberia. Indeed, the interconnected use of the petrographic and $\delta^{13}C$ - $\delta^{18}O$ isotopic studies suggest that Estremoz Anticline marbles were highly dispersed along the Lusitania (Mérida, Évora, Alcoutim, Mértola and W Portugal), Baetica (Regina) and Terraconensis (Asturias, Tui and Toledo) provinces of Iberia, as well as in North Africa (Banasa and Volubilis) (Fig. 4A). Similar studies indicate that Almadén de la Plata marbles were also dispersed in the Lusitania (Mérida) and Baetica provinces and in North Africa (Banasa, Thamusida and, perhaps, in Volubilis) (Fig. 4A). However, the discrimination between Estremoz Anticline, Almadén de la Plata and some Mediterranean marbles, namely those from Aphrodisias, Dokimeion or Pentelikon, based in petrographic and $\delta^{13}C$ - $\delta^{18}O$ studies, is often difficult, although the geographic proximity to the Estremoz Anticline and Almadén de la Plata leads to consider these sources as probable (*e.g.* Cabral *et al*., 2001, 2004; Justino Maciel *et al*., 2002a, 2006; Lapente *et al*., 2014). Although there is a limited number of works, the combined use of petrographic and $^{87}Sr/^{86}Sr$ isotope studies seems to be more accurate to identify the marble sources. The study performed by Taelman *et al*. (2013b) in archeological marble pieces from Ammaia (Lusitania province) and the comparison with Betic and Mediterranean marbles database (Morbidelli *et al*., 2007; Rondolino *et al*., 2020), pointed the Estremoz Anticline as the probable source of all the analyzed marbles, although there is some overlap with the strontium signatures from Pentelikon and Almadén de la Plata marbles.

6. Geological constraints and final remarks

Isotopic studies used to establish the provenance of classical marbles, must be always accompanied/complemented by detailed macroscopic description and petrographic studies. Only this careful control of macro and micro-textural features and mineralogical content allows a robust interpretation of the isotopic data, in order to assess the influence of post diagenetic processes on the isotopic signatures of marbles, such as high temperature metamorphism/metassomatism or secondary dolomitization (Moreira *et al*., 2018, 2019). Indeed, the uniformity of the $\delta^{13}C$ and $\delta^{18}O$ values obtained in the Estremoz, Viana do Alentejo, Almadén de la Plata and Trigaches-São Brissos marbles (Fig. 3B) may result from tectono-metamorphic and metasomatic processes during the Variscan Cycle, thus explaining the differentiation between the Alconera marbles and the other classical marbles which experience very low-grade metamorphism (Fig. 3B).

In fact, the Trigaches-São Brissos, Viana do Alentejo and Almadén de la Plata marbles (all located in the southern domains of the OMZ) have textural and petrographic features that contrast with the Estremoz and Alconera ones (Fig. 2). The southern

OMZ marbles were generally affected by high temperature metamorphism/metasomatism, responsible for grain size increasing (medium to coarse-grained marbles) and mineralogical diversity (Gomes and Fonseca, 2006; Rosas *et al*., 2008; Ontiveros *et al*., 2012; Puelles *et al*., 2018; Moreira *et al*., 2019), which includes pyroxene and olivine, a common feature in high temperature marbles (Bucher and Grapes, 2011). The high temperature metamorphism would be responsible for the modification of the primary $^{87}Sr/^{86}Sr$ ratio, as result of the interaction between high temperature metamorphic/metasomatic fluids, with crustal sources, and marbles (Moreira *et al*., 2019). Therefore, it is expected that the marbles from the southernmost sectors of OMZ affected by high temperature metamorphism (including the Trigaches-São Brissos Marbles), display higher $^{87}Sr/^{86}Sr$ values when compared to the primary isotopic signature of the Estremoz Marbles (Moreira *et al*., 2019). This interpretation agrees with the already obtained $^{87}Sr/^{86}Sr$ ratios in marbles from southern OMZ, such as those from Serpa and Escoural (Fig. 3C), which present similar geological framework (*e.g*. Chichorro *et al*., 2008; Moreira *et al*., 2019). The Alconera marbles, with very low-grade metamorphism (López-Munguira and Nieto García, 2004), have clearly distinctive macroscopic features, allowing their differentiation from other OMZ marbles. It is expected that these marbles show a $^{87}Sr/^{86}Sr$ ratios close to the primary signature identified not only in Estremoz, but also in the Elvas Carbonate Formation, which is stratigraphically correlated with the Alconera Marbles (Oliveira *et al*., 1991; Moreira *et al*., 2014b, 2019).

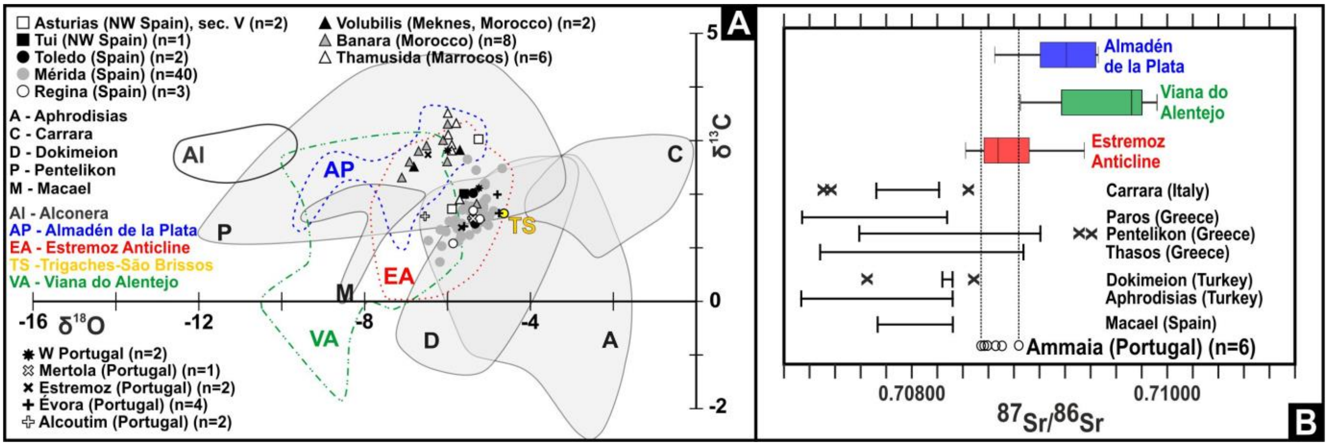

Figure 4. Ossa-Morena Zone marbles isotopic features and its comparison with data obtained from Roman archaeological marble pieces: A – $\delta^{13}C$ vs $\delta^{18}O$ data (archaeological marble pieces database includes data from: Lopes *et al*., 2000; Cabral *et al*., 2001, 2004; Justino Maciel *et al*., 2002a, b, 2003, 2006; Justino Maciel and Cabral, 2008; Antonelli *et al*., 2009, 2014; Royo *et al*., 2010; Origlia *et al*., 2011; Lapuente *et al*., 2014; Vidal *et al*., 2017; Soutelo *et al*., 2018); B – $^{87}Sr/^{86}Sr$ data (archaeological marble pieces database includes data from Taelman *et al*., 2013b).

Figura 4. Caraterísticas isotópicas dos mármores da Zona de Ossa-Morena e a sua comparação com os dados isotópicos obtidos em peças arqueológicas de mármore de Idade Romana: A – $\delta^{13}C$ vs $\delta^{18}O$ (dados isotópicos em peças arqueológicas de mármore de: Lopes *et al*., 2000; Cabral *et al*., 2001, 2004; Justino Maciel *et al*., 2002a, b, 2003, 2006; Justino Maciel and Cabral, 2008; Antonelli *et al*., 2009, 2014; Royo *et al*., 2010; Origlia *et al*., 2011; Lapuente *et al*., 2014; Vidal *et al*., 2017; Soutelo *et al*., 2018); B – $^{87}Sr/^{86}Sr$ (dados isotópicos em peças arqueológicas de mármore de Taelman *et al*., 2013b).

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