### Updated Characterisation of White Saint-Béat Marble. Discrimination Parameters from Classical Marbles

Royo Plumed, Hernando; Lapeunte, Pilar; Cuchí, José Antonio; Brilli, Mauro; Savin, Marie-Claire

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#### UPDATED CHARACTERISATION OF WHITE SAINT-BÉAT MARBLE. DISCRIMINATION PARAMETERS FROM CLASSICAL MARBLES

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#### Abstract

Saint-Béat marble (Central French Pyrenees) is considered one of the most famous Roman Gaul marbles by archaeomaterials researchers. White, grey and banded greyish varieties were exploited during Roman times, to be used mainly in sculpture (including sarcophagi), decorative architecture and epigraphy, primarily in Southern Gaul, but they were also distributed beyond the Hispanic border. A multi-method approach combining petrography, spectrophotometry, cathodoluminescence with EDX spectroscopy and stable C and O isotope analysis was applied to a new collection of samples from quarries along both banks of the Garonne River, next to Saint-Béat village. This work aims to highlight the most useful parameters to discriminate one of the Saint-Béat marble varieties (the coarse-grained white), from other Classical and Hispanic marbles of similar features. This updated characterisation has been successfully checked with Roman artefacts from different archaeological sites of the Hispania Tarraconensis Province.

Keywords

Pyrenean marble, petrological parameters, isotopic analysis

#### Saint-Béat marble. Archaeological and geological setting

Saint-Béat marble is one of the French ornamental stones exploited by the Romans and widely employed in Gaul. The quarries are located along both banks of the Garonne River near the village of Saint-Béat (Saint-Gaudens Arrondissement), in the Central French Pyrenees (Department of Haute-Garonne, Occitanie) and are well connected to the Atlantic Ocean by the large valley of the Garonne River. Although some authors studied this before for the purpose of archaeometric application (COSTEDOAT 1992, 1995; BLANC 1995), they focused on the differentiation of numerous quarry fronts, the main purpose being to identify from which of them were extracted the marbles found in Roman artefacts from regional archaeological sites. In contrast, the present paper aims to highlight which parameters are the most useful for the discrimination of the coarse-grained white variety of Saint-Béat marble from similar Classical and Hispanic marbles<sup>1</sup>.

This approach is crucial in order to check its Roman use outside of Gaul, and in particular on the Hispanic side of the Pyrenees. When the focus was on the Ebro Valley (ancient Conventus Caesaraugustanus, of the Hispania Tarraconensis Roman Province), its presence was not thoroughly checked. An initial archaeometric study of architectural and sculptural pieces from the Roman Theatre of Caesar Augusta revealed the use of Saint-Béat marble, but also some uncertainties as a set of samples of problematic Pyrenean origin (LAPUENTE, TURI, BLANC 2009). This was mainly caused by the lack of petrographic and isotopic data from French Pyrenean marble districts. Furthermore identification of marble of this type in marble artefacts found in Hispanic territories is a difficult task because the favourable geographical location of the Ebro Valley, open to the Mediterranean, enabled the entrance of Classical marbles from all around the Empire. Additionally, the Hispanic marble from the Estremoz Anticline district has been recently identified in Caesar Augusta in a portrait of Tiberius (LAPUENTE et al. 2016; NOGALES, LAPUENTE, RODÀ 2017).

<sup>1</sup> Which is part of the Ph. D thesis recently defended by H. Royo at Zaragoza University (Spain), aiming to update the characterisation of the Pyrenean marble quarries from an exclusively archaeometric standpoint (ROYO PLUMED 2016).

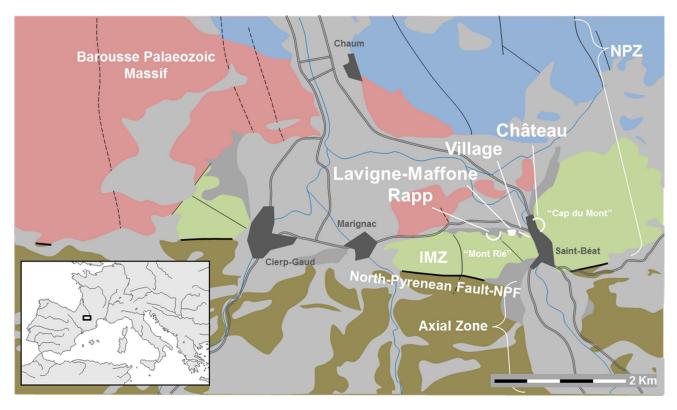


Fig. 1. Saint-Béat quarries location on a simplified geological map of the area (Carte géologique détaillée de la France à 1/50000 1072-Arreau (BRGM-SGN) modified). Pink: Barousse Palaeozoic Massif (Cambro-Ordovician). Dark green: Palaeozoic Axial Zone (Ordovician-Devonian). Blue: NPZ (non-metamorphic Mesozoic). Light green: IMZ-NPZ (undifferentiated Mesozoic). Grey and light grey: Quaternary

Therefore, the provenance study of Roman marble artefacts found in this territory required a clear distinction between Classical, Hispanic and French Pyrenean marbles.

An additional argument for making its discrimination parameters better knwown is because other Pyrenean marbles were also exploited near or within the area where Saint-Béat marble was distributed. This was the case dealt with in a previous paper (ROYO *et al.* 2015) where a comparative study of marbles from Arties quarry, located about 40km south of Saint-Béat, was carried out in order to identify both materials in different Roman archaeological pieces found in the Aran Valley (Spain).

From the geological point of view, Saint-Béat marble is part of certain carbonate series of Jurassic-Lower Albian age, affected by HT/LP (high temperature/low pressure) metamorphism of Cretaceous Upper Albian times. In addition to this so-called Pyrenean metamorphism, an intense penetrative deformation was developed as the result of several Alpine tectonic phases, responsible for the lifting of the Pyrenees. These undifferentiated carbonate series form the most important outcrops of Pyrenean marble located in the Internal Metamorphic Zone (IMZ) which intermittently outcrops in the south of the North Pyrenean Zone (NPZ), and is particularly well developed in its southern central part. The NPZ and IMZ, when exposed, are both bounded to the south by the North Pyrenean Fault (NPF) (Fig. 1), the particular geological history of which has a high influence on the observed macro and microstructure features.

With large and massive layers, more or less homogeneous, mostly of grey and greyish banded but also white marbles, the marble outcrops are situated on both banks of the narrow gorge opened by the Garonne River. Several quarries with their respective fronts became well known after studies published two decades ago (COSTEDOAT 1992, 1995). Their easy access to the river facilitated their distribution along the Garonne Valley. In the present paper, marbles from the Cap du Mont (right bank) were sampled inside and outside the Château quarry, while on the left bank (Mont Rié), marbles were collected outside the so-called Lavigne-Maffone underground quarries, in the Rapp quarry and in the outcrop next to the village (Fig. 1). Unfortunately, the important quarrying activity in the Saint-Béat district over the centuries has destroyed any vestige of ancient working (BESSAC, SABLAYROLLES 2002). The most ancient part of white marble extraction of Roman date is located at the top of the current front of Rapp, in Mont Rié (BLANC, BLANC 2012). In 1946, a landslide led to the discovery of a late 1st century BC front that was reused as a Gallo-Roman sanctuary during the 2<sup>nd</sup> century AD and where many small arae were also found. The site, known as Mailh déras Higouras, was destroyed shortly after it was sighted as a result of another landslide due to ongoing exploitation works (SAPÈNE 1946; FABRE, SABLAYROLLES 1995).

Different varieties of marbles are currently extracted from this district: white, bluish-white, grey, grey regular and irregular banded marbles, all of which were exploited by the Romans, mainly from the 1st to the 2nd century AD. Most of the Saint-Béat marbles are purely calcite. Some marbles with less than 50% in dolomite are in other quarries of the region (Boutx and Sost) whose Roman age has not been verified. Additionally, a dolomitic front (100% dolomite) close to the ancient quarry of the "Brèche des Romains" (La Pène-Saint-Martin), between Saint-Béat and Lez villages, was thought to be the site of ancient exploitation (GISBERT, GASPAR 2004)<sup>2</sup>. A recent general study (ROYO PLUMED 2016) visually identified several lithotypes by their macroscopic features; only the coarse-grained white is considered in the present study3.

#### Analytical methodology

Petrography, spectrophotometry, cathodoluminescence (CL) and carbon and oxygen stable isotope ratio determination ( $\delta^{13}$ C and  $\delta^{18}$ O) were carried out using a sequential approach. This combined method allows discrimination among different quarries, useful when the isotopic ratios overlap. Petrography is always the first step to understand the geological history and formation processes better. It has been systematically applied for studying themacroscopic and microscopic features of these marbles.

The relative grain size of the principal mineral components and main colour was registered by the naked eye. The colourimetric parameters were measured with a portable Minolta spectrophotometer in association with SpectraMagic NX 2.5 software<sup>4</sup>. This equipment enables the measurement of the grey-to-white scale with their CIELAB colour space parameters<sup>5</sup>, complementing the

- 3 An additional paper is being prepared with all marble varieties from the Haute-Garonne Valley.
- 4 The measurements conditions are explained in detail in another article (see LAPUENTE *et al.* in this volume).

colour data visually recorded. All these features make it possible to perform a first subdivision of the marbles.

Uncovered thin sections of each sample<sup>6</sup> were observed under a Nikon Eclipse 50iPOL Polarising Light Microscope (PLM). A set of mineralogical and textural parameters with a diagnostic significance for marble discrimination were measured (LAZZARINI et al. 1980; GORGONI et al. 2002). Special attention was paid to grain size, measuring the maximum grain size (MGS), and fabric, with the grain boundary shape (GBS). Additional features were observed with a deformation degree experiment based on calcite twin morphology (FERRILL et al. 2004) and the identification of intra-crystalline deformation microstructures. In addition, the main mineralogical composition (calcite and/or dolomite)<sup>7</sup> and the presence of accessory minerals were determined. Their optical recognition was supplemented through the application, point-by-point, of a chemical microanalysis energy dispersive X-ray spectroscopy (EDX), using an Amptek MCA 8000 X-ray detector added to the CL8200 Mk5 equipment. This equipment is also suitable to CL analysis and made possible the observation of the luminescent capacity of certain minerals, particularly from carbonates. The electron energy applied to the thin sections was 15-20 kV and the beam current was operated at 250-300 µA. CL Colour, intensity and distribution patterns were recorded taking digital photographs using an automatic digital Nikon Coolpix 5400 camera. This CL methodology has been applied to Classical and Hispanic marbles (BARBIN et al. 1989, 1992; LAPUENTE, TURI, BLANC 2000; LAPUENTE et al. 2014). Carbon and oxygen stable isotope signatures were measured on calcite marble samples with an isotope ratio mass spectrometer (IRMS) Finnigan MAT252 according to MC CREA (1950). The results were expressed in terms of the usual delta notation  $\delta^{13}$ C and  $\delta^{18}$ O in ‰ relative to the International Reference Standard PDB (Pee Dee Belemnite).

As one experimental method, the use of a spectrophotometer has been checked to be applied in specific problematic cases of discrimination, in order to check the existence of different colour parameters. To minimise the possible effects due to the state of the surface, all samples were prepared with a flat unpolished surface, at least 2 cm thick, to avoid any rough irregularity.

<sup>2</sup> However, this dolomitic marble quarry was opened in the fifties of the last century by the OMG society ("Onyx et Marbres Granules") after a personal communication by the administrative quarry staff.

<sup>5</sup> The three coordinates of CIELAB colour space represent the lightness of the colour (L\*), its position between red/magenta and green (a\*) and its position between yellow and blue (b\*).

<sup>6</sup> Prepared at the laboratory of "Preparación de Rocas y Materiales Duros", (Zaragoza University). The authors would like to acknowledge the use of this "Servicio General de Apoyo a la Investigación-SAI".

<sup>7</sup> By staining with Alizarin Red S stain.

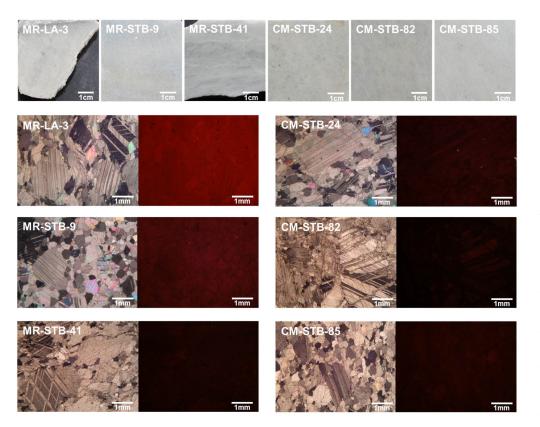


Fig. 2. Selection of samples taken as reference of the coarse-grained white marble of Saint-Béat (MR: Mont Rié quarry-fronts samples. CM: Cap du Mont quarry-front samples). Visual appearance (above) in which the light greyish white tone is evident, and pairs of photomicrographs with cross polarised light (left) and cathodoluminescence pattern (right)

#### Analytical results: characterisation of the coarsegrained white Saint-Béat marble

The studied reference set of coarse-grained white marble of Saint-Béat comprises fifteen samples. In agreement with their visual colour, they all show a light greyish-white hue, whose measurements using the spectrophotometer present a value range from 75.4 to 86.2 for lightness index (L\*) and from 56.0 to 69.0 for whiteness index (WI). The inequigranular calcite, easy to recognise by the naked eye, is the manifestation of their microscopic "core-and-mantle" structure<sup>8</sup>. Visually, the larger grains are usually evidenced by a lower lightness of greyish-white colour, with an appearance colloquially referred to as "rice grain" texture (a feature which is also common in the grey and banded Saint-Béat varieties).

All samples are calcite marbles<sup>9</sup> but rarely present less than 5% in dolomite, determined by staining (Alizarin Red S) and EDX analyses. As common accessory minerals, there are isolated crystals of quartz, phengitic muscovite and pyrite. Additionally, some aggregates of graphite and rare idiomorphic crystals of calcium-rich scapolite (meionite) are sporadically present. As a marble with a light greyish-white hue, the existence of graphite aggregates is quite diagnostic along with the presence of meionite. This type of scapolite is especially diagnostic in the grey banded Saint-Béat marbles, in which it becomes a subordinate mineral.

This coarse-grained white Saint-Béat variety presents an isotropic fabric, but depending on the sample orientation it can exhibit a slightly anisotropic fabric, marked by a dimensional preferred orientation. The heteroblastic texture is quite distinctive with a remarkable difference in the grain size (bimodal distribution). This texture consists of dispersed large crystals (porphyroclasts exhibit several microstructures of stress) surrounded by a set of smaller crystals (in different states of recrystallization), according to a typical "core-and-mantle" structure (Fig. 2).

With the focus placed on the individual calcite crystals, the maximum grain size (MGS) varies between 2.4 and 6.5 mm with a median of 2.9 mm, and grain boundary shape (GBS) can be sutured, embayed and sometimes curved. Intracrystalline deformation is variably developed, only present in scattered larger crystals by their deformed twins or by a subtle undulatory extinction. Calcite twins are tabular and thick, although some

<sup>8</sup> This terminology, applied to cases where the structure is developed by dynamic recrystallization, replaces the usual "mortar" texture, since this later has a genetic implication as "mechanically crushed rock" and therefore its use is not recommended in this particular case (PASSCHIER, TROUW 2005).

<sup>9</sup> According to the proposal of the IUGS Subcommission on the Systematics of Metamorphic Rocks (FETTES, DESMONS 2011).

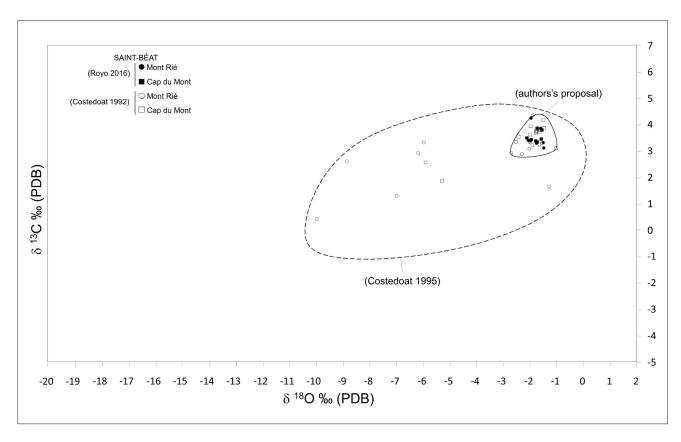


Fig. 3. Dispersion diagram with the specific isotopic field proposal for the coarse-grained white Saint-Béat marble, compared to the previous general Saint-Béat marbles field (COSTEDOAT 1995) and the available  $\delta^{13}$ C and  $\delta^{18}$ O isotopic values (COSTEDOAT 1992; ROYO PLUMED 2016) of samples from Mont Rié (Rapp and Lavigne-Maffone) and Cap du Mont (Château)

crystals show curved and tapered thick twins<sup>10</sup>. Differences among samples taken from the different outcrops, cannot be established by fabric, texture or deformation degree.

CL shows a very homogeneous luminescence with faint-medium intensity from dark reddish orange to reddish orange. The percentage of accessory minerals is so slight that its luminescence is not significant. Cap du Mont samples always show a faint CL, while some Mont Rié samples exhibit medium intensity (Fig. 2, MR-LA-3).

The isotopic signature of the coarse-grained white Saint-Béat marble are quite homogeneous, drawing a single cluster in the plot for all the considered quarry fronts. These isotopic data (ROYO PLUMED 2016) are grouped in a relatively small area of the isotopic diagram, within the large Saint-Béat isotopic distribution field previously published (COSTEDOAT 1995)<sup>11</sup>. In our proposal, this coarse-grained white Saint-Béat isotopic field encloses values from 2.8 to 4.3‰ of  $\delta^{13}$ C and values from -2.7 to -1.0‰ of  $\delta^{18}$ O, expressed in PDB units (Fig. 3).

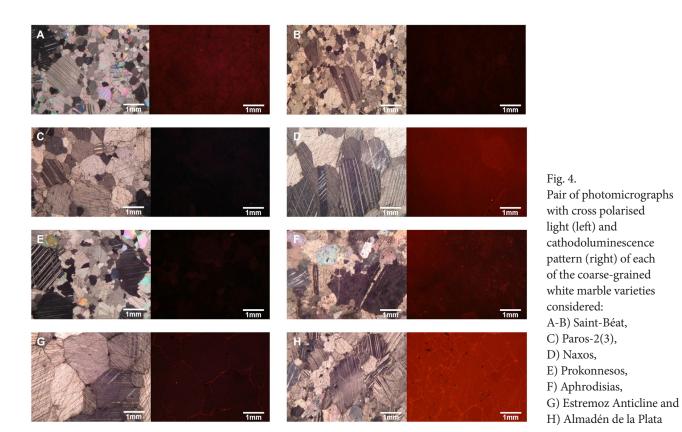
#### Discriminating from classical and hispanic marbles

Once the coarse-grained white marble from Saint-Béat has been characterised, its comparison with other Classical and Hispanic varieties is needed in order to highlight the parameters that offer the best discrimination. This step has been accomplished applying an identical methodology in our own quarry samples of Classic and Hispanic marbles.

With reference to the visual characteristics or macroscopic petrographic features, the peculiar "rice grain" visual aspect is also quite common in other heteroblastic white marbles, such as some marbles from Prokonnesos, Paros-2(3), Naxos and more rarely from the Estremoz Anticline in Lusitania. However, certain petrographic characteristics are rather specific and help to differentiate this coarse-grained white Saint-Béat variety from most other Classical and Hispanic marbles, especially its texture with bimodal distribution and the MGS (Fig. 4). Additionally when graphite aggregates and especially meionite crystals are present, both are clear parameters that support Saint-Béat marble identification. Only Prokonnesian marble exhibits a similar heteroblastic texture with an average size of MGS (2.5 mm), slightly less than the value determined for Saint-Béat (2.9 mm)

<sup>10</sup> Twins type II and III respectively (FERRILL *et al.* 2004).

<sup>11</sup> We want express our gratitude to Drs. A. Blanc and Ph. Blanc who facilitated the unpublished isotopic data from Costedoat Ph. D Thesis.



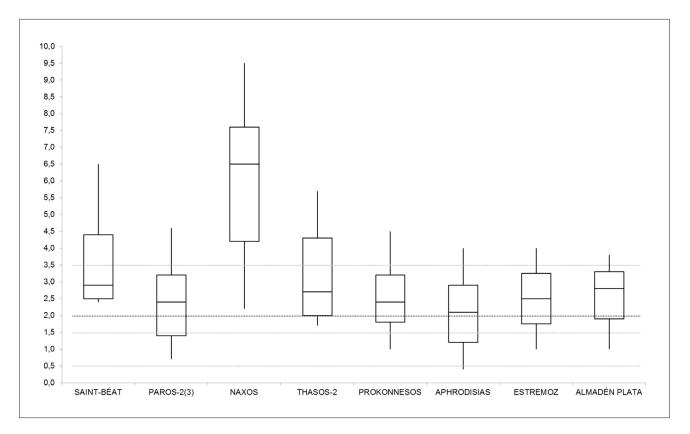


Fig. 5. Box and whisker diagram for the statistical distribution of MGS in the varieties of the coarse-grained white marble considered (LAPUENTE, TURI, BLANC, 2000; GORGONI *et al.* 2002; ATTANASIO, BRILLI, OGLE 2006; ORIGLIA *et al.* 2011; LAPUENTE et al. 2014; ROYO PLUMED 2016)

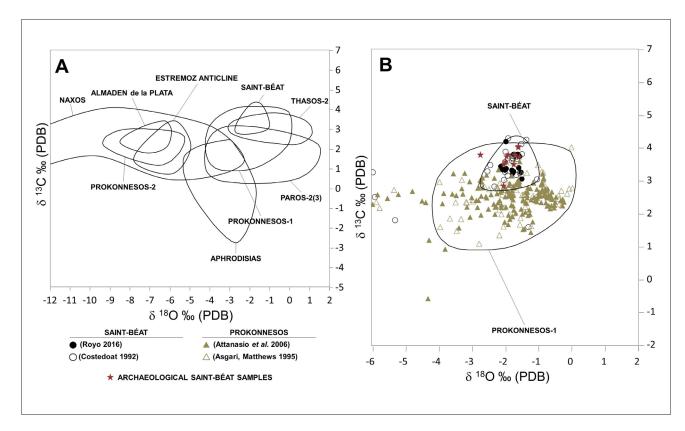


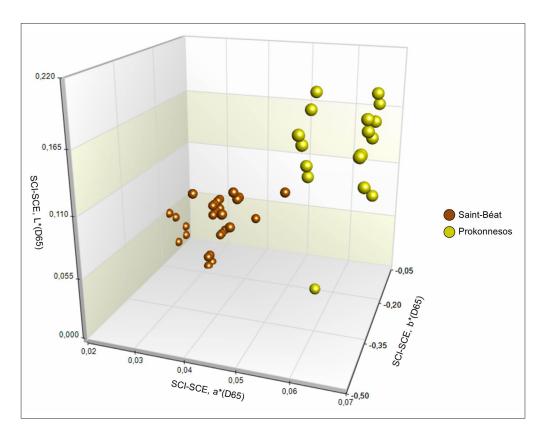
Fig. 6. A) Dispersion diagram with isotopic fields of coarse-grained white marble considered (GORGONI *et al.* 2002; LAPUENTE *et al.* 2014). B) Dispersion diagram with  $\delta^{13}$ C and  $\delta^{18}$ O isotopic values available for varieties of similar petrographic characteristics and overlapping isotopic fields (COSTEDOAT 1992; ASGARI, MATHEWS 1995; ATTANASIO, BRILLI, OGLE 2006; ROYO PLUMED 2016) and values of archaeological samples identified as coarse-grained white Saint-Béat marble in previous papers

(Fig. 5). A parameter with a clear value for the discrimination of Prokonnesos is the extremely faint intensity of its CL, with a dark blue tone. However this feature alone is not useful, for its discrimination from some marbles from Cap du Mont that exhibit a very faint luminescence (Fig. 4). Moreover, the isotopic data of white Saint-Béat fall inside the isotopic fields of several Classical marbles (Fig. 6, A), and especially problematic is its overlap with the Prokonnesian field. However, paying attention to specific values, those of white Saint-Béat are quite well grouped at the top of the isotopic field, helping to separate them from Prokonnesian isotope values (Fig. 6, B).

In order to increase the occasionally difficult discrimination between coarse-grained white Saint-Béat and Prokonnesos an additional method using the spectrophotometer has been tested on 48 white quarry samples from the two districts. Measurements of CIE 1976  $L^*a^*b^*$  colour space coordinates in both modes, the specular reflectance excluded (SCE) and included (SCI), have been simultaneously taken. In the SCE mode, only the diffuse reflectance is measured, producing a colour evaluation which 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. A 3D scatter diagram shows the subtractions between SCI and SCE modes of L\*a\*b\* values, which clearly allows their discrimination (Fig. 7). Although this spectrophotometric method seems to be quite promising, more analyses including archaeological samples must be tested to check its reliability.

#### Use in Tarraconense archaeological artefacts

At this point, it is appropriate to note that, since 2009, the sequential multi-method approach has been successfully applied to a wide range of Roman arte-facts from different archaeological sites of the *Conventus Caesaraugustanus (Tarraconensis* Province) on the southern slope of the Pyrenees. The results of these studies have been steadily published (LAPUENTE, TURI, BLANC 2009; LAPUENTE *et al.* 2011, 2012, 2015; ROYO 2010; ROYO *et al.* 2015, 2016; ANDREU *et al.* 2015). The isotopic values of the archaeological artefacts carved in this material fall inside the updated field with only one exception, which supports our proposal (Fig. 6B). The white coarse-grained Saint-Béat marble has been used in sculpture, *arae*, urns and ornamental elements as slabs and ornate slabs (with a possible pilaster capital among



#### Fig. 7.

Diagram with values of the subtractions between SCI and SCE modes for each colour space coordinates (L\*a\*b\*), corresponding to 14 samples of white Saint-Béat marble and 26 samples of white Prokonnesian marble

ARCHAEOLOGICAL SITE	SAMPLE NUMBER	INVENTORY NUMBER	ARTEFACT	CONTEXT	CHRONOLOGY	REFERENCE
Caesar Augusta, Zaragoza (Zaragoza)	05-Z-AA		Togated	Theatre	Early imperial period	Lapuente et al. 2009
	05-Z-AB		Sculpted fragment (rounded)	Theatre	Early imperial period	Lapuente et al. 2009
	05-Z-AC	2931	Sculpted fragment (heel)	Theatre	Early imperial period	Lapuente et al. 2009
	07-Z-CB	98782	Ornamental fragment (acanthus leaf)	Theatre	Early imperial period	Lapuente <i>et al.</i> 2009
"La Sinagoga", Sádaba (Zaragoza)	11-SNG		Slab fragment (9)		Indeterminate	Andreu <i>et al.</i> 2015
	4-SNG		Slab fragment	Terms	Indeterminate	Lapuente et al. 2011
	5-SNG		Slab fragment (edge)	Terms	Indeterminate	Lapuente et al. 2011
	6-SNG		Slab fragment (chip)	Terms	indeterminate	Lapuente et al. 2011
S.Úrbez, Nocito (Huesca)	16-HU	NIG.04133/ /NIG.03543	Head		2 <sup>nd</sup> cent. AD	Lapuente et al. 2015
Hospital de Benasque (Huesca)	60-HU	1	Ara			Lapuente et al. 2015
S.Esteban, Llert	1-LL		Ara (with inscription)			Lapuente et al. 2015
(Huesca)	2-LL		Ara			Lapuente et al. 2015
Montcorbau? (Lleida)	2-ART		Urn front		2 <sup>nd</sup> -3 <sup>rd</sup> cent. AD	Royo <i>et al.</i> 2015
Rectoría, Gausac (Lleida)	4-ART		Urn front		Mid-3 <sup>rd</sup> cent. AD	Royo <i>et al.</i> 2015
	5-ART	Inv.497	Urn front or Stele		2 <sup>nd</sup> -3 <sup>rd</sup> cent. AD	Royo et al. 2015

Fig. 8. Data collection of Roman artefacts made of coarse-grained white Saint-Béat, found in archaeological sites of *Tarraconensis* Province that have been published by this group and collaborators

them). Taken together they allow the deduction of a chronological interval of use from the mid-1<sup>st</sup> century AD until at least the mid-3<sup>rd</sup> century AD (Fig. 8).

#### Conclusions

The multi-method approach applied to coarsegrained white Saint-Béat marble allows us to verify the homogeneity of their petrographic and isotopic parameters, even in samples taken from different outcrops in the Saint-Béat district. Its particular metamorphic history based on dynamic recrystallisation processes affords its own petrographic characteristics, which are the "core-and-mantle" structure, the particular "rice grain" heteroblastic texture, the CL-pattern and the presence of accessory graphite and meionite which are identifying fingerprints.

Therefore, all of them allow a reliable discrimination from other Classical and Hispanic marbles of similar coarse grain features. In this sense, this studied Saint-Béat marble is easy to discriminate from most of the varieties that potentially may have been used together in Roman times. Rarely does the natural variability of certain petrographic features of an inequigranular marble result in confusions with other inequigranular varieties as Paros-2 and some Estremoz Anticline marbles. These confusions are solved by taking into consideration the sequential approach followed in this study. More complicated is its discrimination from Prokonnesian marble, whose parameters are quite similar. Some ways to differentiate the two marbles are being searched with the application of other techniques. The attempt based on the subtractions between SCI and SCE modes of L\*a\*b\* values seems to offer promising results. The potential of this discriminant parameter obtained with a non-destructive method should be reinforced with measurements in other marble sources, but at the moment it seems to be quite successful.

Coarse-grained white Saint-Béat was used with diverse purposes and had a wide distribution in the *Tarraconensis* Province. Furthermore, it was employed together with the most important Roman marbles. All of these facts underline its importance in the western part of the Roman Empire, but also they raise new questions as to why this marble was used and no other, how much marble was used, what the transportation routes were, what motivated the beginning of export to the *Hispania Tarraconensis* Province and what caused the end of it. These questions must be analysed in the light of the studies recently accomplished (ROYO PLUMED 2016) in this same territory showing the presence of other Pyrenean marbles.

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