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Provenance of sculptural limestones in protohistoric Provence (SE France): insights from carbonate rock petrography and stable isotope geochemistry

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Abstract

The combination of a detailed analysis of microfacies, diagenetic features and isotopic signatures (carbon and oxygen) of limestones from protohistoric statues and the comparison with substantial regional petrographic and geochemical databases have proven to be successful for constraining the provenance and providing insights into the production of statuary materials in Provence during the Iron Age. The limestones forming the sculptures from Roquepertuse

(*twin-headed "Hermes"*, *the lintel with horses* and the *warrior seated cross-legged*) and Rognac (*warrior seated cross-legged*) are interpreted to derive from lower Barremian carbonate formations from La Fare Massif, likely from the vicinity of Coudoux, based on an analogy in 1) micropalaeontologic markers (orbitolinids), 2) grain size, texture and grain composition (oolithic, oobioclastic and bioclastic grainstones), 3) dissolution features affecting ooid cortices and 4) carbon and oxygen isotope signatures. The bryozoan-echinoderm association and the occurrence of *Amphistegina* suggest that the *bust of warrior* from La Cloche oppidum is made of a Miocene limestone from La Couronne formation. Its isotope composition suggests a provenance from quarries located at Anse du Verdon or possibly further to the west. This demonstrates a use of La Couronne limestone as a statuary material in protohistoric times (3rd to 1st century BC). The *fragment of scapulary tunic* found in a disturbed archaeological context dated of 6th to 2nd century BC on the Baou de Saint-Marcel oppidum, in segobrige territories east of Massalia, is made of a vuggy limestone whose clotted peloidal-fenestral fabric and isotope signature are consistent with local, lower Pleistocene travertines. The *young male head* from the Ziem Museum revealed to be made of a planktonic foraminiferal, quartz-free limestone with *Heterohelix* for which no analogue is known in the region, thus suggesting that its place of manufacture is likely to be located outside Provence. Finally, the analyzed set of limestones from protohistoric sculptures, and of certain Provençal provenance, derive from stone production sites geographically close (<20 km) from the places of discovery. A set of non-invasive in situ techniques has been also used to discriminate among the possible sources (magnetic susceptibility, X-ray fluorescence chemical analysis, sound velocity, photogrammetry), but they appeared non discriminant for the present case.

Key-words: sculptural limestones, provenance, protohistory, petrography, stable isotopes, non-destructive methods, Provence, Gaul.

1.Introduction

In the last decades, following major discoveries of statues in Germany and France in particular (e.g. Frey and Hermann, 1997; Menez et al., 1999), European protohistoric sculpture has re-emerged as a topic of interest in archaeology and art history. Provence represents one of the most prolific regions for its corpus of protohistoric sculptures (Duceppe-Lamarre, 2002; Arcelin and Rapin, 2003). Stylistic analyses and detailed characterization of the archaeological setting of these statues constitute major inputs for positioning protohistoric Provence within a European cultural framework. In this context, provenance analyses allow to discriminate a local or regional origin for the statuary material and to identify potential importations. Such studies are particularly useful when the place of discovery of statues is uncertain or their archaeological setting poorly constrained. Additionally, such provenance studies provide significant insights into stone artefact production and trading in ancient times. Following the terminology used by Flügel and Flügel (1997), local material is defined as material extracted at the site where statues were discovered, regional material derives from outcrops located within a distance of several tens of kilometers while the term 'imported' should refer to lithic material brought from far distances. Microfacies analysis combined with micropaleontological and/or geochemical approaches on chips detached from artefacts have revealed to provide a means of distinguishing between local, regional and imported materials (e.g. Attanasio et al., 2000; Capedri et al., 2001; Sanmarco et al., 2015; Brilli et al., 2011). Moreover, non-invasive in situ techniques are more and more demanded to avoid sampling patrimonial cultural objects. They proved for example to be effective for the study of rocks such as obsidian, granite, sandstones and greywacke (Frahm et al., 2013; Williams-Thorpe et al., 2009; Rochette et al. 2022; Ushida et al, 2021). The present work focuses on the provenance of statuary limestones of a set of six protohistoric

statues from various localities in Provence and hosted at the Musée d'Histoire de Marseille (MHM) (**Fig. 1 A-F**). Additionally, a male head sculpture (**Fig. 1G**) hosted in the Ziem Museum of Martigues and whose site of discovery is unknown, has been integrated in the present work. The objective of the provenance study performed on this set of protohistoric statues is mainly to answer the following questions: 1) did the sculptor used local, regional or imported limestone? 2) can the statuary limestone material be traced to specific areas and production sites? 3) do production sites of sculptural limestones coincide with those of building stones?

2.Archaeological and geological setting of protohistoric sculptures

2.1 Roquepertuse oppidum

The Roquepertuse oppidum (**Fig. 2A**) is a protohistoric site located in the municipality of Velaux which yielded one of the most important statuary group from Provence (Gérin-Ricard, 1927; Boissinot, 2011, Py 2011). The set of sculptures analyzed in the present work, a *warrior seated cross-legged* (**Fig. 1A**), a *lintel with horses* (**Fig. 1B**) and the *twin-headed "Hermes"* (**Fig. 1C**), were discovered on a terrace located in the southern flank of the oppidum, and they were first interpreted as elements of a sculptural monument known as a "portique" dated to the 3rd-2nd century BC. The age of the statues remains very controversial. Although the stratigraphic position of most of the statues discovered during early excavations is poorly constrained, a revision of the chronology of the site was undertaken, following excavations carried out between 1994 and 1999, leading to assign the habitat to the first half of the 3rd century BC while the lapidary material (lintels, seated warriors), found as fragments scattered within the whole settlement had to be considered older (Boissinot et al., 2000). Stylistic studies have contradicted this attribution and have suggested an age from the transition between the First and Second Iron

Ages to the beginning of the Second Iron Ages, i.e. from 525 to 375 BC (Arcelin and Rapin, 2003; Rapin, 2004).

The Roquepertuse oppidum rests on the flanks and on top of a flat-topped hill composed of nodular and marmorized continental limestones, latest Campanian to earliest Maastrichtian ("Calcaire de Rognac" formation) in age (**Fig. 3A**).

2.2 Rognac

The *warrior seated cross-legged* from Rognac (**Fig. 1D**) was discovered at the beginning of the 20th century in a dry stone wall at a place called "Plan du Clapier" (Clerc, 1914; Py (2011), located north of the town of Rognac (**Fig. 2A** and **Fig. 3B**). This statue having been reused as a building material, its initial place of use is unknown but it may derive from a neighboring oppidum, such as Le Castellas oppidum located 3 km to the south-east, or Roquepertuse (5 km to the north-east). The stylistic analysis of the statue has led to compare the Rognac statue with the seated warriors from Roquepertuse and then to assign them an age ranging from 525 to 375 BC (Arcelin and Rapin, 2003).

The place of discovery is located on a plain covered with late Pleistocene colluvium, while the neighbouring Le Castellas oppidum rests on uppermost Campanian to lowermost Masstrichtian continental limestones ("*Calcaire de Rognac*" formation) (**Fig. 3B**).

2.3 La Cloche oppidum

A set of fragments belonging to the same statue was discovered at La Cloche oppidum, at Les Pennes-Mirabeau (**Fig. 2A** and **Fig. 3C**) in 1975, which allowed reconstructing a bust of warrior (**Fig. 1E**). La Cloche oppidum is a settlement of the first half of the 1st century BC, while a prior ritual deposit dated of the end of the 3rd-2nd century BC (Chabot, 1996, 2005) was discovered at the top of the site. The fragments were found on a street at the entrance area of the 1st century

BC settlement, but could come from the cultural area. Its fragmentation is believed to be related to the final destruction of the oppidum by Caesar's troops in -49 (Chabot, 1983, 2004). Regarding stylistic studies, while Chabot (2005) attributes to the head a massaliote influence, Arcelin and Rapin (2003) suggest a much older age, i.e from the end of the 4th century to the middle of the 3rd century BC. The oppidum rests on tight and fractured Barremian limestones with rudists (Urgonian facies) (**Fig. 3C**).

2.4 Baou de Saint-Marcel oppidum

The Baou de Saint-Marcel oppidum is a fortified Celto-Ligurian, Segobrige settlement site, located in the eastern districts of Marseille (**Fig. 2A** and **Fig. 3D**), founded at the end of the first quarter of the 6th century BC and abandoned in the last quarter of the 2nd century BC (Agostini, 1972; Guichard and Rayssiguier, 1993). It rests on the top of a flat-topped hill made of Pleistocene calcareous tufa (**Fig. 3D**). The studied material from Baou de Saint-Marcel oppidum is an element of the edge of the dorsal part of a rectangular and rigid scapular tunic (**Fig. 1F**) belonging to a limestone statue. This fragment has been found in a reworked context, interpreted as metal deposit, located under the collapse of the southern bastion of the eastern rampart (Rayssiguier,1989 ; Rayssiguier and Guichard, 1989 ; D'Ovidio and Rothé, 2005 ; Dedet and Gantès, 2014), containing also metal furniture, ceramic and a silver obolus from Marseille which, in spite of the dominance of artefacts from the 6th century BC, give a broad dating ranging from the 6th to the 2nd century BC. However, according to Py (2011), the scapular tunic from Baou de Saint-Marcel closely resembles that of the seated warriors of Roquepertuse and could be of a neighboring age, i.e. 3rd century BC.

2.5 Ziem Museum

The Ziem Museum in Martigues acquired from a local collector a limestone juvenile male head evaluated as being a work of high empire style and probably sculpted in a workshop from the south of Gaul (D. Terrier, CCJ, Ziem Museum internal document) (**Fig. 1G**) but without any documentation regarding the site of discovery. Despite its great interest it is not exposed due to the uncertainty of its provenance.

3. Database and methods

3.1 Sampling

For laboratory characterization of the statuary material (thin-section preparation and stable isotope measurements), we were allowed by the museums to detach a small chip (at most one gram) from areas without significant sculpted surface, i.e. previous fractures limiting excavated fragments. We obtained three samples from different objects of the Roquepertuse provenance (see above), and one sample per statue from the other four provenances. A number of potential source local and regional material have been sampled on geological outcrops to be compared with statuary material (see **Table 1**). We also based our methodology on our database derived from numerous petrological and petrophysical studies of regional limestones.

3.2 Thin-section database and petrographic analysis

The petrographic study of the seven limestone sculptures has been performed by means of macroscopic direct observations of lapidary collections at the MHM (Musée d'Histoire de Marseille) and at the Ziem Museum and analysis of thin-sections under polarized-light

microscopy. Thin-sections were prepared from the seven chips extracted from the sculptures. An existing thin-section database of regional limestones from outcrops (**Table 1**), hosted at CEREGE laboratory (Marseille), has been integrated in this study for qualitative and quantitative comparison with the statuary material. Thin-sections were also prepared from local limestones sampled at or at the vicinity of the oppida. Additional thin-sections of ancient building stones from the Hellenistic harbor of Massalia, housed at the CICRP have been also integrated. Thin-sections have been studied for microfacies characterization of limestone including texture, grain composition, biotic assemblages and diagenetic features. Additionally, a selection of thin-sections of both sculptures and outcrops were point-counted on the basis of 300 points to quantify the composition of the sand-grained fraction of limestones by using J-microvision® software.

3.3 Scanning Electron Microscopy (SEM)

In order to identify the microstructure of limestone at a micrometer-sale and to check the elemental composition of minerals, small fragments (~5 mm in diameter) from the statuary limestones were imaged with the Philips XL 30 Environmental Scanning Electron Microscope (ESEM) of the Plateforme de Recherche Analytique Technologique et Imagerie (PRATIM, Aix-Marseille University, FSCM, France), equipped with a LaB6 filament and Energy Dispersive X-Ray analysis (EDAX Genesis equipped with an Apollo 10 SDD detector). The analytical settings used were: 20 keV; back-scattered electron mode BSE; no carbon-coating; pressure of 0.2 Torr.

3.4 Stable isotopes

Carbon and oxygen stable isotope ratios ($\delta^{13}C$ and $\delta^{18}O$) have been measured on bulk carbonate powders from statuary limestones, local limestones from Roquepertuse, Le Castellas

(Rognac), La Cloche and Baou de Saint-Marcel oppida and selected limestones from regional outcrops and from building stones belonging to the hellenistic Massalia harbour. The carbon and oxygen stable isotope database used for this study is summarized in **Table 2** while the measurements are displayed in the **Appendix**.

Measurements on statuary and local limestones as well as on regional limestones from Coudoux have been performed using a Dual-Inlet Isotope Ratio Mass Spectrometer (DI-IRMS Delta V Plus,Thermo Scientific) coupled to an automated carbonate preparation line (Carbonates Device Kiel IV, Thermo Scientific) at CEREGE platform for stable isotope analysis (PANISS). Carbonate powders from Upper Barremian (« Pierre de Cassis »), Upper Burdigalian-Langhian (« Pierre de La Couronne ») limestones and Pleistocene calcareous tufa were carried out at the GeoZentrum Nordbayern department, Friedrich-Alexander-Universität Erlangen-Nürnberg (Germany) through reaction with phosphoric acid at 70°C using a Gasbench II connected to a Thermo Fisher Scientific DELTA V Plus mass spectrometer.

All measured isotopic values are normalized against NBS-19 and are expressed relative to the V-PDB standard. Mean external reproducibility is better than 0.05‰ and 0.07‰ for $\delta^{13}C$ and $\delta^{18}O$ respectively.

3.5 Non-invasive techniques

We tested several techniques that were applicable directly on the statues in the museum without surface preparation:

1) Magnetic susceptibility (K) measurements were performed on statues using the SM30 sensor whose detection volume is a 5 cm diameter cylinder below the coil. Sensitivity

is a few 10^{-6} SI, depending on the presence of surrounding metal. Contribution to the signal decreases with depth and 69% of it derives from the first cm. Corrections are needed to account for surface unevenness and curvature as the sensor is calibrated for a perfectly flat surface (Rochette et al., 2022). For geological samples, the most sensitive instrument MFK1 Kappabridge, was used.

2) Chemical analysis using portable X-ray fluorescence instrument. We have used the Bruker tracer IV, in major trace element mode with a counting time of 90 s for each mode. It has an analysis spot size of the order of 1 cm and a penetration depth in the 10-1000 μm depth range depending on the elements analysed and thus the energy of re-emitted X-rays. It shows a sensitivity on trace elements such as Rb and Sr contents of the order of 5 ppm while light major elements have a much higher detection limit: e.g. near 2 and 4-5% for Al and Mg, respectively. We used several measurement spots per statue or sample (usually 5) and calculate the average results. The methodology for such measurements has been recently detailed in Triantafyllou et al. (2021).

3) Velocity of sound (Vp). In limestones Vp determination is a good proxy for porosity and pore structure, that may be used to discriminate among different formations and outcrops. We used a PUNDIT PL 200, with a peak frequency of 54 kHz.

4) Bulk density and porosity estimate. Bulk density ρ_b was obtained by weighting the statues or the geological samples cut as parallelepipeds, and determining their volume using photogrammetry and edge dimensions, respectively. As we deal with nearly pure limestones, porosity estimate (Φ) was computed by the formula $\Phi = (1- \rho_b/\rho_c)x100$, where ρ_c is the density of pure calcite, i.e. 2.71 10^3 kg/m^3.

3.6 Photogrammetry

The *Twin-headed "Hermes"*, the *Bust of Warrior* from La Cloche and the *Young Male Head* from the Ziem Museum have been scanned in 3D by close-range photogrammetry (**Fig 1H**). For acquisition, we have set up each statue in several positions on a table, and we have captured around 100 pictures per position at different angles in a convergent way. The camera used is a high-grade DSLR Sony Alpha 7 R4 with a FE 50mm F1.2 Zeiss lens. The sensor is full-frame, 9504x6336 pixels BSI-CMOS with 63 megapixels. Pictures were processed with Capture One v20 software. We made a color calibration with the ColorChecker® Classic chart from X-Rite and its software ColorChecker Camera Calibration v2.2.0, allowing to create a custom ICC color profile for each lighting condition. Photogrammetric processing was undergone with Agisoft® Metashape v1.8 software using a standard workflow (Brunier et al., 2016). All photos were aligned in separate chunks for each statue position. We obtained a very high-resolution point cloud, with density 5 pts/mm2, and low accuracy value with RMSE lower than 1 mm for each processing. Triangular meshes have been interpolated from point cloud, and texture of photos were projected on them. The meshes have been used to derive volume and area of the statue.

3.7 Provenance assessment methodology

We will show that the used non-destructive techniques revealed to be not effective to discriminate among the different limestones investigated. Therefore, provenance assessment of statuary limestones studied here will be based on the following workflow:

1- Detailed petrography (depositional facies and diagenetic features) of statuary limestones, stable isotope (C and O) signatures and stratigraphic attribution using available microfossils;

2- Consistency between facies, diagenetic features, isotope signatures and age of statuary limestones and local limestone. If consistent, a local origin is privileged otherwise a regional origin must be investigated.

3- Identification, from published literature, of regional formations sharing the following properties: 1) similar geological age to that of the statuary material, 2) similar depositional facies and environments, 3) mechanical properties compatible with use for sculpture.

4- Creation of a database of thin-sections and stable isotope signatures for these identified formations from existing or newly acquired field data and laboratory measurements.

5- From the available database, identification of intervals and localities where depositional facies (characterized quantitatively), diagenetic features and stable isotope signatures are analogous to those of statuary limestones. Such intervals and localities will be considered as probable areas of provenance of the statuary material.

6- Comparison of such potential localities with ancient sites of stone production and critical evaluation of provenance assessment within the archaeological context.

4. Results

4.1 Petrographic analysis of statuary limestones

4.1.1 Roquepertuse oppidum

4.1.1.1 The *twin-headed "Hermes*"

Macroscopically, the *twin-headed "Hermes"* from Roquepertuse is made of a homogeneous fine-to-medium grained limestone with a slightly reddish patina, and chalky aspect on fresh sections. Observations under optical microscopy reveal that the *twin-headed "Hermes"* (**Fig.**

4A) is composed of a well-sorted grainstone, largely dominated by ooids (78%), with minor proportion of bioclasts (12%) and peloids (9%). The biota includes benthic foraminifers (miliolids and orbitolinids), echinoids and sparitized fragments of molluscs and green algae. Grain size averages 300 µm. The ooid cortices were partly leached and ooid nuclei appear commonly suspended in the middle of moldic cavities created by leaching (**Fig. 4B-C**). Such dissolution molds are partially filled with sparse equigranular sparry calcite crystals (20-50 µm) (**Fig. 4D-E**). Ooids are surrounded by a thin (~20 µm) isopacheous rim of palisade calcite cement (**Fig. 4D-E)** while the intergranular space is completely occluded by coarser (>50 µm) blocky calcite cement. Most of the grains are strongly micritized and exhibit significant microporosity between micrite particles under SEM microscopy (**Fig. 4D**). The outer surface of peloids commonly shows evidences of dissolution having led to the development of a perigranular porosity.

4.1.1.2 The Warrior seated cross-legged

Macroscopically, the *warrior seated cross-legged* from Roquepertuse is made of a homogeneous fine-to-medium grained limestone with a light beige patina, slightly chalky on fresh sections. Observations under optical microscopy reveal that the statuary material is made of a well-sorted grainstone (**Fig. 5A**) mainly composed of bioclasts (44%), ooids (29%) and peloids (27%). Grain size averages 200 µm. The bioclastic association is similar to that of the *twin-headed "Hermes"* limestone and is dominated by benthic foraminifers such as miliolids and orbitolinids, together with echinoids, pieces of molluscs and green algae. In both samples, the outer parts of ooids and micritized grains are significantly leached and similarly to the *twin-headed "Hermes"* grainstone, the resulting perigranular moldic cavities are partially occupied by sparse equigranular sparry calcite crustals (**Fig. 5B-C**). Most of the grains (benthic

foraminifers, ooids and peloids) are strongly micritized and exhibit, under SEM observation a significant microporosity between micrite particles.

4.1.1.3 The lintel with horses

The *lintel with horses* is made of a homogeneous, medium to coarse-grained limestone with light beige patina. The microfacies consists in a well-sorted bioclastic grainstone (**Fig. 5D**) dominated by bioclasts (75%) including rounded and sparitized fragments of rudists and coral, benthic foraminifers (miliolids and orbitolinids) and peloids (25%). The outer part of most of the grains is significantly micritized (micrite envelope) and/or leached (**Fig. 5D-E**). The intergranular space is partly occluded by an isopachous rim of bladed calcite cement postdated by coarser equigranular calcite cement (**Fig. 5E**). Intergranular porosity may be partly preserved in large pores (**Fig. 5E**). The microfacies differs from that of the *twin-headed "Hermes"* and to the *warrior seated cross-legged* from Roquepertuse in being slightly coarser (average grain size: 500 µm) and devoid of ooids.

4.1.2 Rognac

The *warrior seated cross-legged* from Rognac is made of a medium to coarse-grained, homogeneous, light beige limestone. Similarly to Roquepertuse statuary limestones, fresh sections exhibit a slightly chalky texture. Observations under optical microscopy (**Fig. 5F**) show that the Rognac limestone is a bioclastic grainstone (average grain-size: 500 µm) with very similar composition to that of the *Lintel with horses* from Roquepertuse. Bioclasts (71 %), including miliolids, orbitolinids, mollusk fragments and echinoderms dominate the grain association, while peloids (29%) are subordinate components. Additionally, diagenetic features are identical to those evidenced in Roquepertuse statuary limestones: significant perigranular

porosity around grains (**Fig. 5G**), partial infills of molds by equigranular calcite and blocky calcite cements.

4.1.3 La Cloche oppidum

The *bust of warrior* from La Cloche oppidum is macroscopically made up of a fairly heterogeneous, vuggy, bioclastic limestone exhibiting a pinkish patina. Thin section observations under polarized-light microscopy point towards a bioclastic packstone-grainstone (**Fig. 6A-B**) whose biological assemblage is largely dominated by bryozoans (46%) and echinoderms (39%), the subordinate biota being mainly composed of benthic foraminifers (*Amphistegina*), coralline algae and mollusks. The matrix is composed of very fine peloids (<100 µm) and occupies partially the intergranular space. The residual intergranular space is partially filled by equigranular sparry calcite cement. Poikilotopic cements may occlude some large intergranular pores.

4.1.4 Baou de Saint-Marcel oppidum

Macroscopically, the fragment of scapulary from Baou de Saint-Marcel oppidum is made of a limestone with a slightly yellowish patina, exhibiting abundant microvugs (<1 mm). Observations under optical microscope (**Fig. 6C-D**) show that the sample consists of an assemblage of peloids of irregular shape connected to each other by micritic bridges (= clotted peloids) encasing fenestral pores (typically 100-500 µm in diameter). In addition to this dominant microfabric, peloidal encrustations around plant stems are also present (**Fig. 6D**), the stems being preserved as voids. The clotted peloidal fabric, the dominant fenestral porosity as well as encrustations around plants allow identifying a calcareous tufa.

4.1.5 Ziem Museum statue

The *Young male head* statue from the Ziem Museum is made of very homogeneous, very tight limestone with a dark beige patina. Under polarized-light microscopy, this limestone is a very-fine grained grainstone-packstone composed dominantly of fragments of planktonic foraminifera (**Fig. 6E**) including biserial (*Heterohelix*: **Fig. 6F**) and planispiral (cf. *Hedbergella*) taxa. Observations under SEM reveal that the matrix is partly to entirely composed of fragments of coccoliths (**Fig. 6G**).

4.2 Petrographic analysis of local limestones

4.2.1 Roquepertuse oppidum

The continental upper Cretaceous deposits outcropping at the Roquepertuse oppidum are dominantly composed of nodular, marmorized limestones. The microfacies consists of a pisolithic packstone with numerous cracks and root traces (**Fig. 7A**) indicative of pedogenic limestones.

4.2.2 Le Castellas oppidum

The continental upper Cretaceous (Maastrichtian) deposits outcropping at Le Castellas (Rognac) oppidum are dominantly composed of tight, fractured and karstified limestones. The microfacies consists of bioclastic wackestones with characean gyrogonites and stems (**Fig. 7B**) as well as common gastropods. Early pedogenic features are common (root traces and circumgranular cracks) thus suggesting a deposition in palustrine environments.

4.2.3 La Cloche oppidum

Barremian limestones outcropping at La Cloche oppidum are dominantly tightly cemented bioclastic and peloidal packstone to grainstones (**Fig. 7C**). The biota is dominated by rudist fragments and benthic foraminifera including miliolids and orbitolinids.

4.2.4 Baou de Saint-Marcel oppidum

Pleistocene calcareous tufa from Baou de Saint-Marcel oppidum are characterized by clotted-peloidal fabrics as well as by micritic and sparitic encrustations around phytoliths or peloids (**Fig. 7D**). Larger (>1 mm) oncoids are also common.

4.3 Stable isotopes

Stable isotope results ($\delta^{13}C$ and $\delta^{18}O$) on bulk carbonate powders from the selected statuary limestones are displayed in **Fig. 8A**. Roquepertuse and Rognac statues display very close values of $\delta^{13}C$ (-0.71 to -0.07 ‰) and $\delta^{18}O$ (-5.93 to -5.05 ‰). Measurements on the *Bust of Warrior* from La Cloche are significantly ^{13}C and ^{18}O-depleted compared to Roquepertuse and Rognac statues with $\delta^{13}C$ and $\delta^{18}O$ averaging -2.51‰ and -6.28 ‰ respectively. The fragment of scapulary from Baou de Saint-Marcel exhibits both highly negative $\delta^{13}C$ and $\delta^{18}O$ values (-5.97 and -6.40 ‰ respectively) which are consistent with continental carbonates (Deocampo, 2009). Results from the Ziem Museum statue stands out from other measurements by positive $\delta^{13}C$ values (averaging +0.78 ‰) and slightly ^{18}O-enriched composition with $\delta^{18}O$ averaging -4.29 ‰. The comparison between statuary and local limestone isotope composition (**Fig. 8A**) suggests a non-local origin for the statues of Roquepertuse, Rognac and La Cloche. In contrast, the very ^{13}C and ^{18}O-depleted values of the calcareous tufa from Baou de Saint-Marcel outcrops are consistent with those of the scapulary fragment.

Additionally, an isotopic database ($\delta^{13}C$ and $\delta^{18}O$) of limestones from Provence, likely to provide statuary material, has been achieved in order to make the attributions of provenance of the different statues more robust. Such a database integrates original measurements (**Table 2** and **Appendix**) as well as published data (Léonide *et al*., 2014; Aubert *et al*., 2020). The Upper Barremian "*Pierre de Cassis*" ("*Cassis Stone*"), a tight limestone which is very widely used in the region as a building stone or in sculpture (Tréziny, 2009) is characterized by positive $\delta^{13}C$ values ranging from +0.17 to +3.01‰ (**Fig. 8B**). In contrast, chalky Upper Barremian limestones from Martigues are significantly ^{13}C and ^{18}O-depleted compared to time-equivalent facies from Cassis. The Lower Barremian "*Pierre de Calissane*" is a chalky limestone which was extensively quarried in La Fare Massif area and which is characterized by dominantly negative $\delta^{13}C$ values and by a rough positive covariant trend between $\delta^{13}C$ and $\delta^{18}O$ (**Fig. 8B**). Samples from an abandonned quarry located in Coudoux show slightly negative $\delta^{13}C$ values (from -1.04 to -0.70‰) while $\delta^{18}O$ range from -5.23 to -5.11‰. The Upper Burdigalian-Langhian "*Pierre de La Couronne*" is a regionally famous building stone used since Antiquity which makes up most of the hellenistic buildings from the Massalia harbour (Pédini, 2013). On $\delta^{13}C$- $\delta^{18}O$ cross-plot (**Fig. 8B**), "*Pierre de La Couronne*" measurements cover a wide domain with dominantly negative $\delta^{13}C$ values (-5.30 to 0.29‰) and $\delta^{18}O$ ranging from -6.93 to -1.02‰. Although $\delta^{13}C$- $\delta^{18}O$ domain of Upper Burdigalian-Langhian limestones partially overlaps that of Barremian limestones, isotope composition of "*Pierre de La Couronne*" samples are typically more ^{13}C-depleted for a given $\delta^{18}O$ value. Additionally, the present "*Pierre de La Couronne*" isotope database reveals a decreasing trend in $\delta^{18}O$ from east (Tamaris section) to west (Anse du Verdon section). Isotope signatures of building stones from the hellenistic Massalia harbor are consistent with the western area (Anse du Verdon) isotope composition. Finally, the isotope signature of Pleistocene calcareous tufa stand out very clearly by their very negative $\delta^{13}C$ values (-9.61 to -4.53‰).

446

447 **4.4 Non-destructive analyses**

448 In the course of our study, it appeared that the studied material presents a number of
449 unfavorable characteristics to obtain reliable results with the deployed techniques. Statues are
450 fragmentary and have been reassembled using cement and steel bars. They also show numerous
451 holes and fractures. Such characteristics will obviously bias the sound velocity measurements
452 as well as magnetic measurements. Magnetic susceptibility measurements on the statues were
453 hardly measurable: K always below 2 10^{-5} SI, apart from spots affected by the steel bars. This
454 is close to noise level in the museum conditions and typical of un-metamorphosed platform
455 carbonates, with negligible magnetite and limited clay contents, and does not allow any
456 discrimination. On geological samples, K was negative, i.e. diamagnetic. Sound velocity
457 measurements on the statues show strong variability, from 2 to 4.5 km/s (**Table 3**) and cannot
458 obviously be used for lithological determinations.

459 Porosity estimates (**Table 3**) vary from 5 to 25%. The value measured for the *twin-headed*
460 *"Hermes"* from Roquepertuse (17.4 %) is in the high range observed for Coudoux outcrop
461 samples, likely possibly due to the alteration of the statues. The three pieces of the la Cloche
462 statue yield coherent porosities, with an average of 20.4 ±2.1%, identical to the value measured
463 in La Couronne limestone at Baou Tailla quarry (west of Anse du Verdon). The Ziem head has
464 the lowest porosities of the studied statues (12.6 %) in agreement fine the petrographic
465 observations.

466 Chemical analysis by pXRF may be contaminated by products pasted on the statue surfaces
467 after their fabrication. This may be intentional (painting from the in-use period or unfortunately
468 applied by early 20th century archeologists; artificial patina applied in the museum to
469 homogenize color or fill holes) or incidental: contamination by soil within which the material

was buried, or by dust accumulated in the surface porosity since discovery. pXRF results (**Table 3**) are obviously affected by the state of the statue surface: determined CaO content varies from 37 to 62 wt.%, while is always higher than 82 % (and mostly in the 91-95% range) for geological equivalent samples, measured on sawn surfaces. To compensate this effect we normalized other elements to CaO. MgO/CaO ratios are in the range 90 to 240 ‰, on the statues as well as on geological samples. These high values are in contradiction with the petrographic observations that do not show the presence of dolomite. However, Mg quantification is problematic with the pXRF instrument. Al_2O_3/CaO, SiO_2/CaO, ratios are systematically higher on statues (up to 70 and 180 ‰, respectively) than on geological samples (around 20 ‰ for both ratio). The same is true for Fe/Ca and K/Ca. This clearly signs a contamination, either by soil or cement applied in the museum. In all geological samples (except Baou-Tailla) Fe/Ca is below 4 ‰ confirming diamagnetic magnetic susceptibility. Significant contents of Ti, Zn, Pb are sometimes observed only on the statues (not reported in Table 3), again pointing toward a contamination (modern painting, urban pollution?).

5. Discussion

5.1 Provenance assessment of protohistoric statuary limestones from Provence

5.1.1 Roquepertuse and Rognac statues

The studied statuary material from Roquepertuse and Rognac exhibits common sedimentological, diagenetic and geochemical characteristics: 1) chalky character of the limestone linked to significant microporosity, 2) depositional texture (grainstone) and faunal association composed predominantly by miliolids, orbitolinids and rolled fragments of rudists, 3) the presence of perigranular dissolution features around micritized grains and 4) a very

494 homogeneous carbon and oxygen isotope signature with values ranging from -5.0 to -5.9‰ in
495 $\delta^{18}O$ and from -0.1 to -0.7 ‰ in $\delta^{13}C$.
496 Members of the larger benthic foraminiferal family Orbitolinidae occurred from the early
497 Cretaceous to the Oligocene (Boudagher-Fadel and Price, 2019) and in Provence, they are
498 reported from the Berriasian (Virgone, 1997) to the Coniacian (Babinot and Tronchetti, 1983).
499 The observed microfacies (**Fig. 4** and **5**) suggest outer carbonate platform environments
500 (Leonide et al., 2012). Both age and depositional environments contradict a local origin of the
501 statuary material, the oppida of Roquepertuse and Le Castellas (Rognac) being located on upper
502 Cretaceous continental limestones. Regionally, chalky, microporous grainstones from the inner
503 to outer Barremian carbonate platform have been reported in Martigues area, La Fare Massif as
504 well as in Mont-de-Vaucluse (Borgomano *et al*., 2013; Léonide *et al*., 2014). As a consequence,
505 based on 1) the Cretaceous age inferred from faunal associations, 2) the shallow carbonate
506 platform environments suggested by microfacies and 3) the chalky, microporous nature of the
507 statuary material, a regional Barremian origin is hypothesized for the limestone composing both
508 Roquepertuse and Rognac statues.
509 To validate such a hypothesis, a regional thin-section and stable isotope database (**Table 1** and
510 **3**) has been integrated and an additional field section has been surveyed at Coudoux (**Fig. 1** and
511 **Fig. 9**), which represents the closest locality to the studied oppida with Barremian outcrops (3.6
512 km from Roquepertuse and 7 km from Rognac). At Coudoux the upper 50 meters of the lower
513 Barremian chalky limestones crop out in the southern flank of the La Fare Massif, where they
514 are sharply truncated at top and overlain by late Cretaceous deposits (**Fig. 9B**). This carbonate
515 succession consists of an alternation of four depositional facies (**Fig. 9A**): 1) oolitic grainstones
516 (Facies BF1: ooid proportion>50%), 2) oobioclastic grainstones (Facies BF2: ooid
517 proportion<50%), 3) bioclastic grainstones (Facies BF3: no ooid and bioclast content>50%),
518 and 4) bioclastic rudstones (Facies BF4). Examinations of thin sections revealed that the facies

association of Coudoux is analogous with that of the statuary limestones from Roquepertuse and Rognac regarding depositional texture, grain-size, allochemical composition and diagenetic features (**Fig. 10**). The COU-A sample from Coudoux is a fine-to-medium grained oolitic grainstone where ooids represent 59% of the grains while subordinate components are bioclasts (31%) and peloids (10%), the bioclastic assemblage being composed of rolled sparitized rudists, benthic foraminifers (mainly miliolids and orbitolinids) and echinoderms (**Fig. 10A**). Additionally, the cortex of ooids exhibit similar dissolution features (**Fig. 10D**) as those evidenced in Roquepertuse statues (**Fig. 4B-C** and **5B-C**). As a consequence, the COU-A sample is quantitatively and qualitatively extremely close from the fine-to-medium grained oolitic grainstone of the Twin-headed "Hermes" from Roquepertuse (ooids: 66%; bioclasts: 21%; peloids: 13%). Similarly, the sample COU-B (oobioclastic grainstone: facies BF2) is an analogue (**Fig. 10B**), regarding texture, grain-size (fine-to-medium), diagenetic features (leached cortex of ooids) and grain composition (ooids: 41%; peloids: 26%; bioclasts: 33%) of the statue of warrior from Roquepertuse (ooids: 41%; peloids: 26%; bioclasts: 33%). Finally, the *lintel with horses* from Roquepertuse (peloids: 28%; bioclasts: 72%) and the *warrior seated cross-legged* from Rognac (peloids: 36%; bioclasts: 64%) are bioclastic grainstones (Facies BF3) of similar composition with the COU-C sample from Coudoux (peloids: 32%; bioclasts: 68%) (**Fig. 10C**). These results are therefore consistent with a common provenance, within the lower Barremian chalky limestones of La Fare Massif, for the studied statues from Roquepertuse and Rognac. Such a provenance is also supported by carbon and oxygen isotope signatures of the statuary limestones which fall within the $\delta^{13}C$-$\delta^{18}O$ domain of chalky lower Barremian limestones from La Fare Massif and which are extremely close from values measured in Coudoux samples (**Fig. 11A**). West of Coudoux, the lower Barremian chalky limestones passes laterally into dominantly bioclastic grainstone facies and ooids are almost lacking as evidenced in ancient quarries at Calissanne (Borgomano et al., 2013). As a

consequence, even if a common provenance, at Coudoux, for the 4 sculptures from Roquepertuse and Rognac is very probable, it is not excluded that the *Lintel with horses* from Roquepertuse and the *warrior seated cross-legged* from Rognac, both made of bioclastic grainstone (BF3), come from a more western sector of the La Fare Massif (distant up to 15 km from Roquepertuse). The petrographic and geochemical analyses of the three statues of Roquepertuse confirm the intuition of Gérin-Ricard (1927) according to which the material of the sculptures of Roquepertuse should derive from neighbouring outcrops at Coudoux. More recently, Lescure and Werth (2000) compared the X-ray diffraction spectra of Roquepertuse and Coudoux samples and have shown that both limestones are almost purely calcitic.

We note that porosity estimates are coherent between the Coudoux geological samples and the Roquepertuse and Rognac statues.

5.1.2 The bust of warrior from La Cloche

The occurrence of the benthic foraminifera *Amphistegina* in the *bust of warrior* from La Cloche suggests a Miocene age for the statuary limestone since this taxa has been reported in Provence from the Aquitanian (Anglada et al., 1974) to the Burdigalian-Langhian (Descote, 2010). This age together with the microfacies identified on thin sections (bryozoan-echinoderm grainstone-packstone: **Fig. 6A-B**) rule out the hypothesis of a local origin (Barremian rudist-bearing peloidal-foraminiferal packstone-grainstone: **Fig. 7C**). The light pinkish patina of the statue leads to the hypothesis that the statuary material derives from La Couronne limestone formation, late Burdigalian to early Langhian in age. Other Burdigalian limestones with similar mechanical properties as La Couronne limestones crop out South of Martigues (Ponteau Limestones) and are believed to have been used since the Antiquity as building stones (Tréziny, 2000). However, microfacies studies by Maurel-Ferrandini (1976) did not evidence bryozoan-dominated pack-grainstones within the Ponteau section, thus excluding this provenance. To

validate such a hypothesis, a detailed petrographic and stable isotope study has been performed on three sections located near La Couronne (**Fig. 1B** and **12**) across the Plan-de-Sausset (middle Burdigalian) and La Couronne (late Burdigalian-early Langhian?) formations. Quantitative analysis of grain composition of limestones revealed an excellent match between the statuary material from La Cloche (bryozoan: 46%; echinoderms: 39%) and the bryozoan-dominated limestones from the base of La Couronne formation (bryozoan: 45%; echinoderms: 41% in COU06 sample) at the Anse du Verdon section. Such a correspondence is strongly confirmed by carbon and oxygen stable isotope results (**Fig. 11B**): bulk $\delta^{13}C$ and $\delta^{18}O$ values measured for La Cloche statue fall within the $\delta^{13}C$-$\delta^{18}O$ domain of limestones from Anse du Verdon section, while these are significantly outside the isotopic domains identified for Tamaris and Beaumaderie sections. Such a lateral change in stable isotope signature within the Plan-de-Sausset and La Couronne formations likely result from lateral changes in cementation and neomorphic patterns from east to west. Interestingly, the stable isotope signature of La Cloche statue is also extremely close to that of building stones from the Hellenistic Massalia harbor (**Fig. 11 B**) which may support a common provenance, at the vicinity of Anse du Verdon and La Couronne locality (see location **Fig. 2B**) or potentially further to the west (Carro). In addition, the average estimated porosity values calculated from the different parts of the statue (20.5%) is very close to that measured on outcrop samples from Baou Tailla (see location **Fig. 2B**), west of Anse du Verdon (20.4%). Complementing the existing petrographic and stable isotope database by performing a systematic sampling in the whole set of identified ancient quarries from La Couronne is likely to constrain even more the exact location of the extraction site. Finally, our results show that the statue from La Cloche is made of a limestone whose place of provenance is located approximately 19 km from the oppidum.

5.1.3 The fragment of scapulary tunic from Baou de Saint-Marcel

The clotted peloidal fabric, the dominant fenestral porosity as well as the encrustations around plant stems allowed identifying the limestone from the Baou de Saint-Marcel statue as a calcareous tufa. A local origin for the statuary limestone is therefore plausible since the Baou de Saint-Marcel oppidum is located on top a hill composed of Pleistocene calcareous tufa, displaying similar depositional fabrics (**Fig. 7D**). Additionally, the carbon and oxygen isotope composition of the statuary limestone fall within the $\delta^{13}C$ and $\delta^{18}O$ range of Pleistocene calcareous tufa from Provence, including samples from Baou de Saint-Marcel hill (**Fig. 11C**). However, given the presence of various hills made up of calcareous tufa in the vicinity of the Baou de Saint-Marcel oppidum (**Fig. 3D**) and more widely within the Marseille basin, the origin of the statuary material could be not strictly local but located in other neighbouring continental Pleistocene limestone outcrops from the Marseille basin (within 12 km from Baou de Saint-Marcel).

5.2 The Ziem Museum head: a regional or imported statue?

One striking feature of the *male head* statue from the Ziem Museum is the dominance of planktonic foraminifera, particularly *Heterohelix* whose stratigraphic distribution covers the Upper Cretaceous and the Paleogene. The depositional facies of the Ziem Museum statue suggests open marine environments. From the available literature, the only planktonic-rich limestones known in Provence from the Upper Cretaceous-Paleogene interval occur in the upper part of the Fontblanche formation (Fontblanche 2 unit: latest Cenomanian) in the Beausset basin, 20 km southeast of Marseille (Rineau et al., 2021). From the available thin-section database the closest depositional facies has been found in samples from the Collongues slump (Fontblanche 2 unit: latest Cenomanian), 2 km north-east of Cassis (**Fig. 1A**). In these samples, *Heterohelix* and *Hedbergella* dominate the biotic assemblage but the relative

frequency of glauconite and quartz grains differ very strongly from that of the Ziem Museum statue. Additionally, no recent or ancient quarry is known in the Fontblanche 2 carbonate unit since these rocks do not have the mechanical qualities required to be used as building stone or sculpture because of their friability and disintegration into cm-thick plates. As a consequence, with the current knowledge on carbonate rocks and stratigraphy of Provence, it seems unlikely that the statuary limestone forming the Male Head from the Ziem Museum has a regional origin. Its porosity estimate is low compared to the other studied statues.

The nearest outcrops of such planktonic-rich carbonates with dominant *Heterohelix* are known in the Cenomanian-Turonian series from the Vocontian basin, southeast France (Oudet, 2013) and northern Africa (Morocco: Ettachfini and Andreu, 2004, Jati et al., 2011; Tunisia: Saïdi et al, 1995), as well as around the Cretaceous-Tertiary boundary from northern Italy (Luciani, 1997).

5.3 Production of statuary limestone in protohistoric Provence

The results of the integrative approach implemented in the present work provide strong evidences that the three studied statues of Roquepertuse and that of Rognac are sourced from the Lower Barremian, chalky limestones outcropping in the Massif de la Fare between Saint-Chamas and Coudoux. The uniqueness and exact position of the stone production site are, however, more questionable. The oolitic facies of the *twin-headed "Hermes"* and of the *seated warrior* from Roquepertuse, becoming significantly rare towards Calissanne and passing laterally into bioclastic grainstones ("Pierre de Calissanne"), leaves little doubt about the location of the stone production site at the vicinity of Coudoux, thus confirming the intuition of Gérin-Ricard (1927). Even though an ambiguity persists concerning the *Lintel with horses* which displays a more ubiquitous bioclastic facies, present in both Coudoux and Calissanne area, a common origin with the two other statues remains however to be privileged, taking into account the greater distance from this last site compared to the oppidum (12 km compared to 4

643 km). The existence, on the hills north of Coudoux, of various ancient artisanal quarries (**Fig.**
644 **9C**), small in size (<10 m) and of undetermined period of activity, is consistent with past stone
645 extraction activity within the Coudoux limestone. The results of our study show that the
646 exploitation of the Coudoux limestone began as early as the 3rd century BC and possibly as
647 early as the last quarter of the 6th century BC if we take into account the ages of Roquepertuse
648 statuary suggested from stylistic approaches (Arcelin and Rapin, 2003).

649 The implications of the provenance analysis of La Cloche material on the production of
650 protohistoric statuary material are strongly dependent on the age attributed to the statue, which
651 remains controversial. If the age proposed by Arcelin and Rapin (2003), which is based on
652 stylistic analysis is confirmed, this would imply that La Couronne limestone was already
653 exploited at the vicinity of Anse du Verdon, and used for sculpture, as early as the end of the
654 3rd – 2nd century BC. This would suggest an early use of La Couronne limestone for sculpture
655 by Celto-Ligurians, prior to the earliest Greek massaliote settlements and associated Hellenistic
656 quarries documented near La Couronne (2nd century BC after Chausserie-Laprée, 2005) and
657 prior to the beginning of stone exportation toward the Massalia harbor (Pedini, 2009). While
658 superficial excavations of limestone slabs in the area have been reported as early as the 5th
659 century BC, earliest evidences of significant stone exploitations within the substrate of La
660 Couronne limestone are extractions of monolithic sills in the 4th century BC (Chausserie-
661 Laprée, 2005).

662 A later age for the bust of warrior from La Cloche, 1st century BC, as suggested by the
663 archaeological setting and stylistic considerations by Chabot (2005), would imply that the statue
664 was sculpted coevally with stone extraction by Greeks. Interestingly, the similarity in carbon
665 and oxygen stable isotope composition between the warrior from La Cloche and stones from
666 the fresh-water basin and southern tower of the hellenistic harbor of Massalia (2nd century BC)
667 suggests that these building materials likely derive from the same area as the statue, in the

vicinity of Anse du Verdon. Such a result is consistent with an extraction of the La Cloche statuary limestone in a Hellenistic quarry. Additionally, such an interpretation would be also consistent with the Massaliote affinity of the statue as attributed by Chabot (2005).

A transport by land of the statue across La Nerthe massif (~20 km) is possible since a protohistoric winch press counterweight, made of La Couronne limestone, is known from the Entremont oppidum (Coutagne, 1993) at a greater distance (~40 km) from the site of extraction. However, a transport by sea of the statuary block from the coastal cliffs of La Couronne to the Etang de Berre shoreline or to the Massalia harbour would have minimized the distance of land transport (7 and 13 km respectively).

The early Pleistocene calcareous tufa (or *travertines*) from the Marseille basin are reputed to been used as building stones or sculpture material since Antiquity (Vacca-Goutoulli, 2020). Nicod (1974) mapped a set of ancient and recent quarries within the *travertines* from the eastern districts of Marseille. Travertine blocks have been used in the Greek archaic construction (Excavations of the Tunnel Major: Conche and Maufras, 2005) and in the reconstruction of the ramparts at the 4th century BC (Treziny, 2009). Its use for sculpture is also attested during late antiquity (Vacca-Goutoulli, 2020). The present results from Baou de Saint-Marcel material indicate that the use of local *travertines* for sculpture started as early as the protohistoric times in Segobrige territories east of Massalia.

6. Conclusions

An integrated approach including carbonate microfacies analysis, characterization of diagenetic features and stable isotope geochemistry provided new insights into the provenance and the production of statuary limestones in protohistoric Provence:

1) The *twin-headed "Hermes"*, the *lintel with horses* and the *warrior seated cross-legged* from Roquepertuse are made of lower Barremian limestones from La Fare Massif, likely from a same locality located at the vicinity of Coudoux, as suggested by a very close analogy in micropalaeontologic markers (orbitolinids), grain size, texture and grain composition (oolithic, oobioclastic and bioclastic grainstones), diagenetic features (dissolution affecting ooid cortices) and stable isotope (carbon and oxygen) signatures. Porosity values are also consistent with those measured in lower Barremian microporous limestones from La Fare Massif. This also attests that the production of Barremian limestones in La Fare Massif began as early as the 3rd century BC and possibly earlier.

2) The *warrior seated cross-legged* from Rognac displays petrographic features (occurrence of orbitolinids, bioclastic grainstone facies, development of perigranular porosity) and stable isotope signatures which are consistent with a common provenance with that of the Roquepertuse statues.

3) The bust of warrior from La Cloche oppidum is made of late Burdigalian-early Langhian (?) limestones from La Couronne formation as suggested by the occurrence of *Amphistegina* and the bryozoan-echinoderm biotic association. In addition, the carbon and oxygen isotope composition strongly suggest a provenance from quarries located at Anse du Verdon or possibly further to the west. This testifies the use of La Couronne limestone in protohistoric sculpture (3rd to 1st century BC).

4) The fragment of *scapulary tunic* from Baou de Saint-Marcel revealed to be made of local early Pleistocene calcareous tufa (=*travertines*), as suggested by the clotted-peloidal and fenestral microfabric and the similar carbon and oxygen isotope signature. This fragment records an early use of these limestones for protohistoric sculpture in segobrige territories bordering Massalia.

5) The young *male head* from the Ziem Museum (Martigues) is made of a tight limestone with planktonic foraminifera (*Heterohelix*) which indicate a late Cretaceous to Paleogene age and for which no exact microfacies analogue exists in Provence. The site of discovery as well as the stone provenance which is probably not located in Provence, remain unknown.

The study therefore shows that for all the statues for which the place of discovery is certain, the distance to the site of provenance of the limestone is less than 20 km. Finally, from a methodological point of view, this work demonstrates the potentiality of integrating detailed microfacies, diagenetic and stable isotope analyzes of carbonate rocks for determining the provenance of statuary limestones. However, the success of such an approach lies mainly on the existence of substantial petrographic and geochemical databases on a regional scale. On the other hand, the tested in situ non-invasive techniques proved to be inapplicable for the studied limestones. Provenancing of limestone statues is seldom reported (e.g. Middleton and Bradley, 1989), probably because such rocks are so common that comparison techniques are tedious. In the present case we benefited a lot from the existing petrographic, paleontologic and isotopic databases on Provence limestones.

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Figure caption

Fig.1: A-G: Photographs of the studied set of protohistoric statues: **A)** *warrior seated cross-legged* (Roquepertuse), Musée d'Histoire de Marseille (MHM), **B)** *lintel with horses* (Roquepertuse), MHM, **C)** *twin-headed "Hermes"* (Roquepertuse), MHM, **D)** *warrior seated cross-legged* (Plan des Clapiers, Rognac), MHM, **E)** *bust of warrior* (La Cloche), MHM, **F)** Fragment of scapulary tunic (Baou de Saint-Marcel), MHM, **G)** Young *male head*, Ziem Musem. **H**) Photogrametric image of the *twin-headed "Hermes"* (Roquepertuse), MHM.

Fig.2: **A)** Simplified geological map of Provence and location of the places of discovery of the studied statues. **B)** Close-up on La Couronne area and location of the studied sections and position of ancient quarries (after Pedini, 2013).

Fig. 3: Geological map around the places of discovery of the protohistoric statues: **A)** Velaux and Roquepertuse oppidum, **B)** Rognac and Le Castellas oppidum: the yellow star locates the place of discovery of the warrior seated cross-legged (Plan du Clapier), **C)** La Cloche oppidum, **D)** Baou de Saint-Marcel oppidum.

Fig. 4: Petrographic features of the *twin-headed "Hermes"* from Roquepertuse: **A)** thin-section microphotograph under polarized light showing the oolitic grainstone facies (BF1); Orb.: Orbitolinid, white arrows: ooids; **B-C)** close up on ooids showing the dissolution of the cortex and partial infill with equigranular sparry calcite under polarized (**B**) and polarized-analyzed light (**C**); **D-E)** SEM images of an ooid; *Pal.*: isopachous palissage calcite cement; *Equ*.: equigranular sparry calcite crystal occupying the dissolution void within the cortex of the ooid; *Bl*.: blocky calcite cement.

Fig. 5: Petrographic features of **A-B-C**) the *warrior seated cross-legged* from Roquepertuse, **D-E**) the *lintel with horses* from Roquepertuse and **F-G**) the *warrior seated cross-legged* from Rognac. **A**) thin section microphotograph under polarized light showing the oo-bioclastic

grainstone facies (BF2); white arrows: ooids; **B-C)** close up on ooids showing the dissolution of the cortex and partial infill with equigranular sparry calcite under polarized; **D** and **F**) thin section microphotograph under polarized light showing the bioclastic grainstone facies (BF3), *Orb*.: Orbitolinid; **E** and **G**) SEM images; black arrows: dissolution features around grains, white arrows: partially preserved intergranular pores.

Fig. 6: **A-B**) *Bust of warrior* from La Cloche: thin section microphotograph under polarized light showing the bioclastic packstone-grainstone dominated by bryozoan (Bryo.) and echinoids (Ech.); Amph.: *Amphistegina*; **C-D**: fragment of scapulary tunic from Baou de Saint-Marcel: thin section microphotograph under polarized light showing the clotted peloidal microfabric; white arrow: calcitic encrustation around plant stem; **E-G**) The *male head* from Ziem Museum: thin section microphotograph under polarized light showing the planktonic foraminiferal packstone (white arrow: sections of planktonic foraminifers); **F**) SEM image of a section of *Heterohelix;* **G**) SEM image showing fragmented coccoliths (white arrow) within the micrite matrix.

Fig.7: Thin section microphotograph under polarized light of outcrop samples: **A**) local limestone from Roquepertuse oppidum ("Calcaire de Rognac" formation, latest Campanian to earliest Maastrichtian); *rt*: root traces, *pis*.: vadose pisoids; **B)** local limestone from Castellas oppidum ("Calcaire de Rognac", latest Campanian to earliest Maastrichtian), *gyr*.: characean gyrogonite, *rt*: root traces; **C)** local limestone from La Cloche oppidum (Barremian, Urgonian facies), *Orb*.: orbitolinid, **mil.**: miliolids; *Ech*.: echinoderms; **D)** local limestone from Baou de Saint-Marcel oppidum, calcareous tufa (early Pleistocene) with calcite encrustations around stems (arrow), **E)** Planktonic-dominated packstone from the Collongues slump, north-east of Cassis (Fontblanche 2 formation, latest Cenomanian), *pf*: planktonic formainifer, *Qz*: quartz grain, *gl*.: glauconite.

Fig.8: A) $\delta^{13}C$-$\delta^{18}O$ cross-plot for statuary and local limestones, **B)** $\delta^{13}C$-$\delta^{18}O$ cross-plot for the regional limestone database.

Fig. 9: **A)** Textural log and vertical facies changes of the upper part of Lower Barremian succession, north of Coudoux (see location in **B** and **C**); **B)** Geological map of the southern flank of La Fare Massif, north of Coudoux and position of the section logged in **A**; **C)** Aerial view of the logged section and position of abandoned quarries.

Fig.10: A) Thin section microphotograph of sample COU-A from Coudoux section, showing the oolitic grainstone facies (BF1) and comparison of carbonate grain composition between COU-A and the *twin-headed "Hermes"* from Roquepertuse; **B)** Thin section microphotograph of sample COU-B from Coudoux section, showing the oobioclastic grainstone facies (BF2) and comparison of carbonate grain composition between COU-B and the *warrior seated cross-legged* from Roquepertuse; C) Thin section microphotograph of sample COU-C from Coudoux section, showing the bioclastic grainstone facies (BF1) and comparison of carbonate grain composition between COU-A, the *lintel with horses* from Roquepertuse and the *warrior seated cross-legged* from Rognac; **D)** Detail of an ooid with leached cortex and equigranular sparry calcite infill, under polarized (left) and polarized-analyzed light (right), COU-A sample, Coudoux.

Fig. 11: $\delta^{13}C$-$\delta^{18}O$ cross-plots: **A)** Statuary limestones from Roquepertuse and Rognac and regional Barremian limestones; **B)** Bust of warrior from La Cloche and regional Burdigalian-early Langhian (?) limestones ("Pierre de la Couronne"); **C)** Fragment of scapulary tunic from Baou de Saint-Marcel and regional Pleistocene calacareous tufa.

Fig.12: A) Log section of La Couronne/Anse du Verdon (see location in **Fig. 1B**) and bioclastic composition of limestones determined from point counting on thin-section; **B)** Photograph of the outcrop (eastern coast of Anse du Verdon) showing the upward transition from Plan de

Sausset (Middle Burdigalian) and La Couronne (Upper-Burdigalian-early Langhian?) formation and the position of ancient quarries on top; **C)** Comparison of microfacies and grain composition between COU6 sample from La Couronne and the *bust of warrior* from La Cloche; Bryo: bryozoan; Ech.: echinoderms.

Table 1: Thin-section database of limestones from Provence.

Table 2: Carbon and oxygen isotope database of selected limestones from Provence.

Table 3: Results of non-destructive methods: pXRF measurements using Bruker instrument expressed as ratio to CaO or Ca content (in ‰), density and porosity estimates.

Appendix: Carbon and oxygen isotope measurements

FIGURE 1

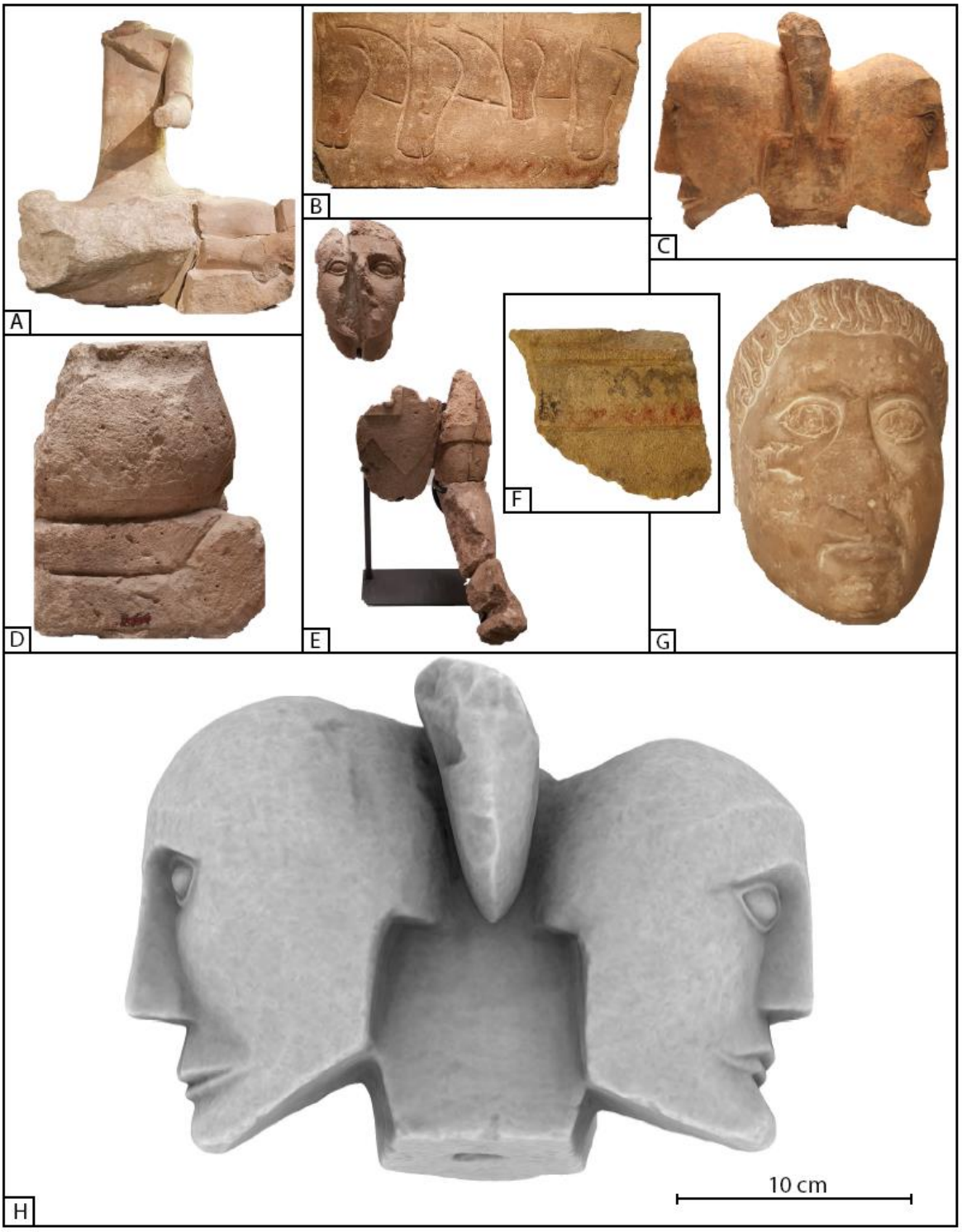

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FIGURE 2

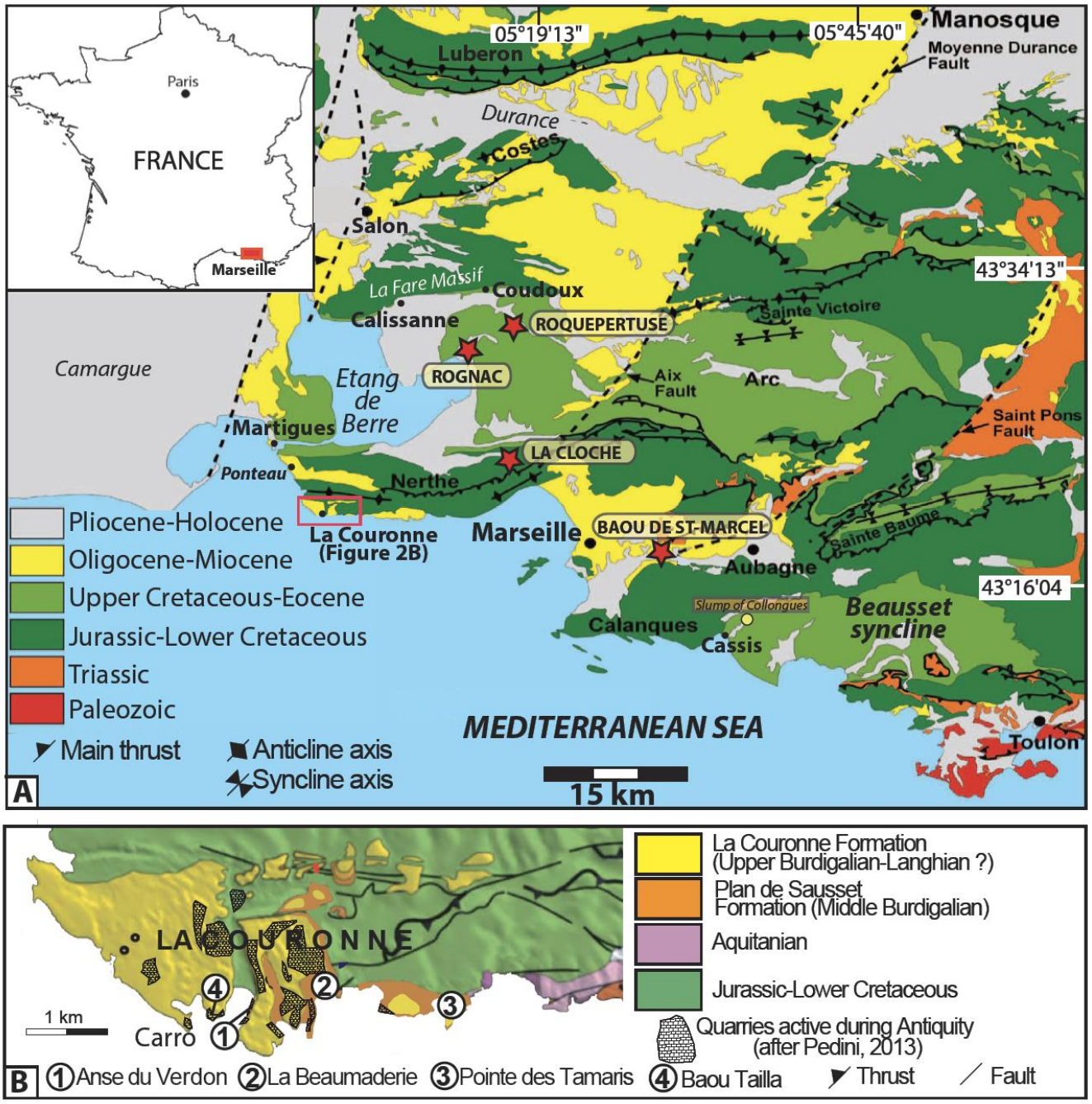

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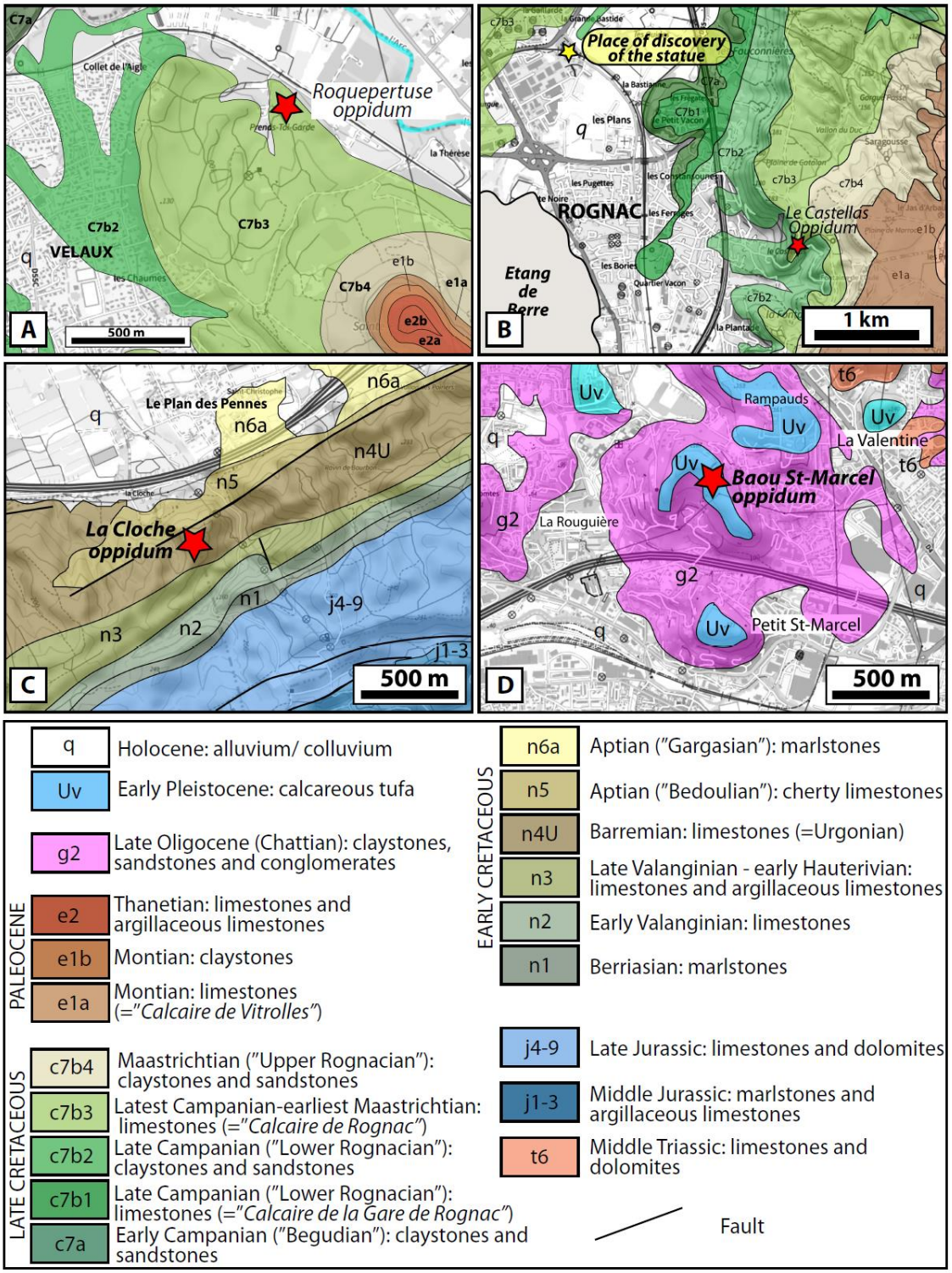
FIGURE 3



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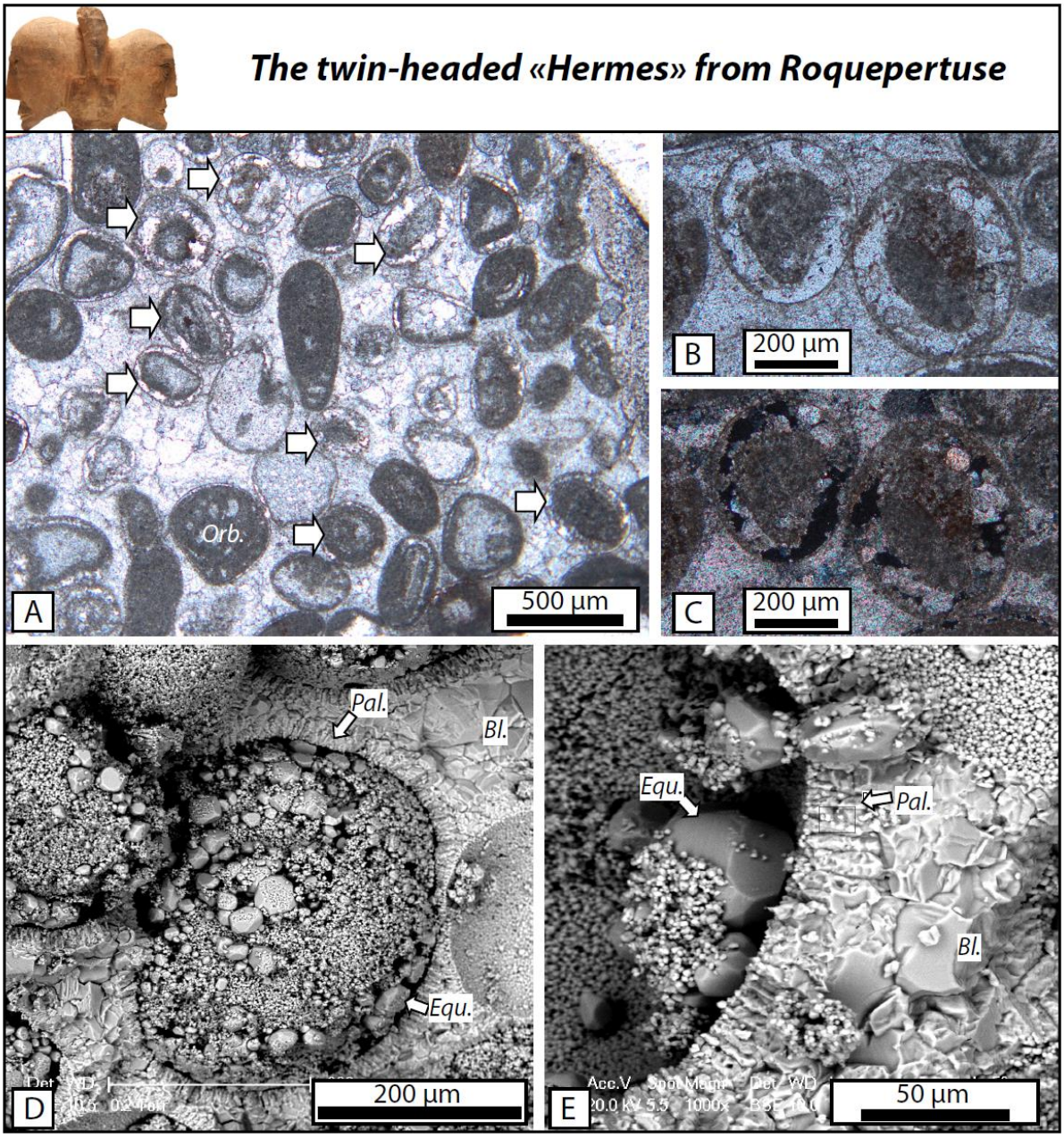

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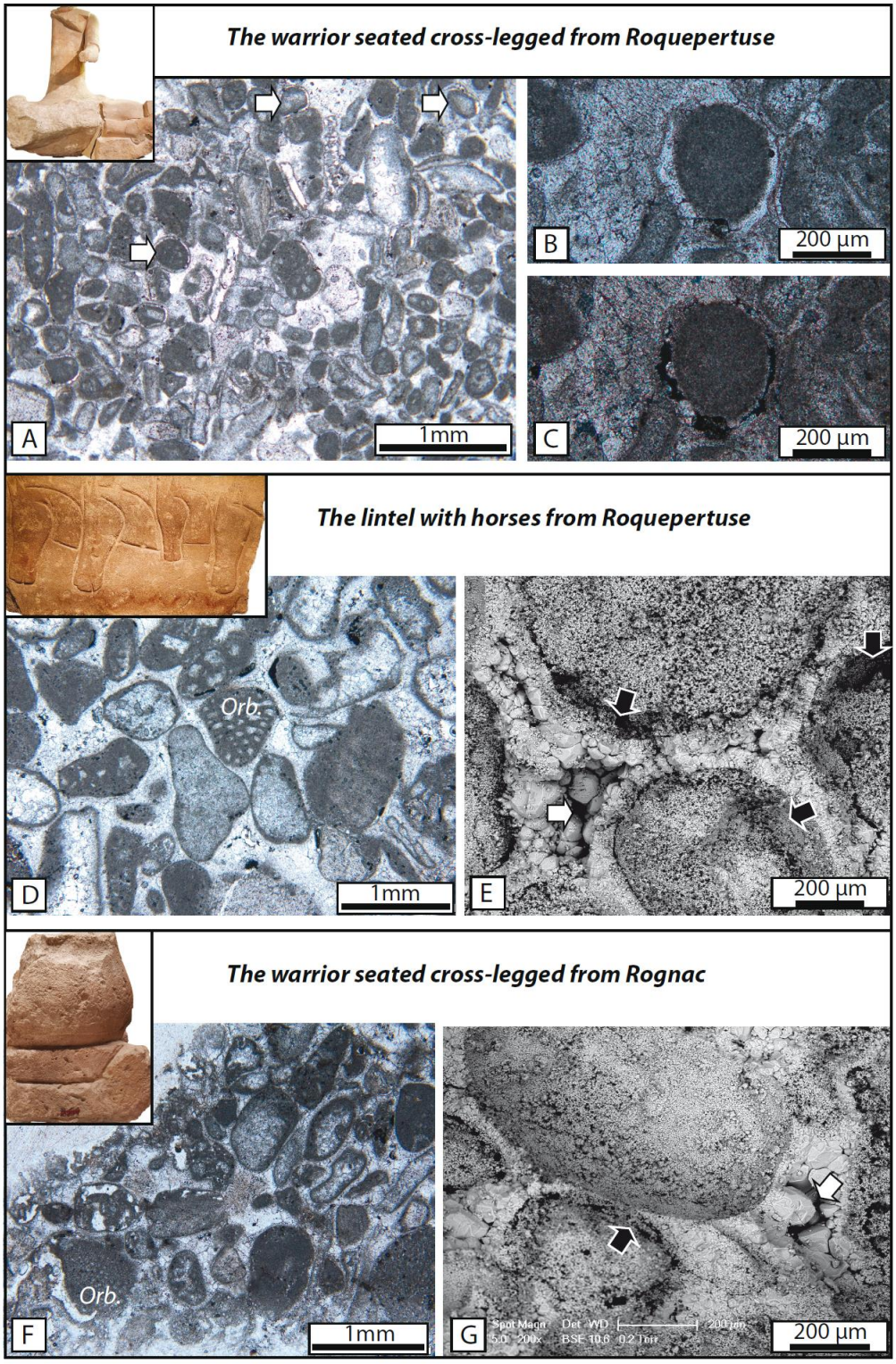

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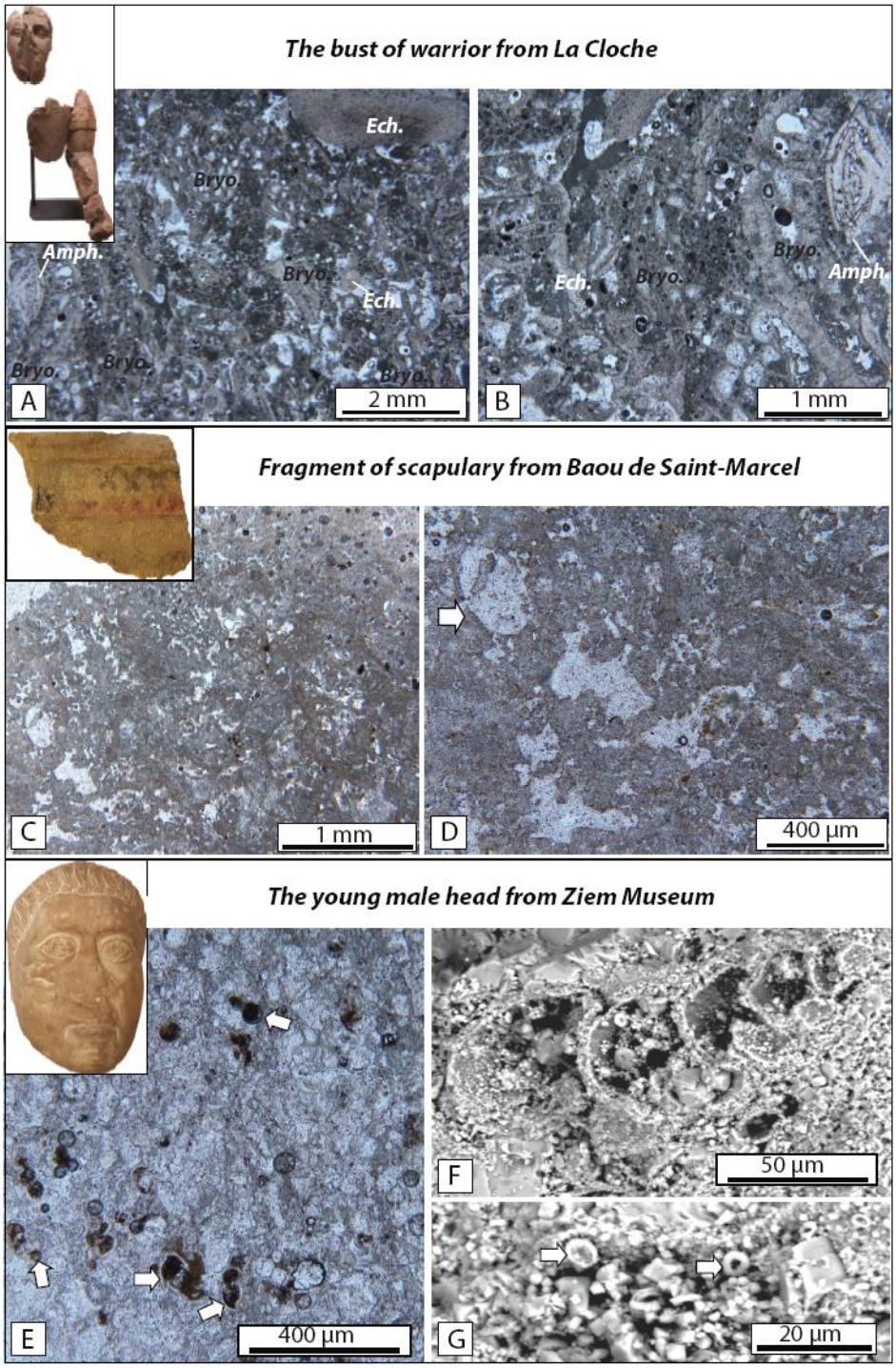

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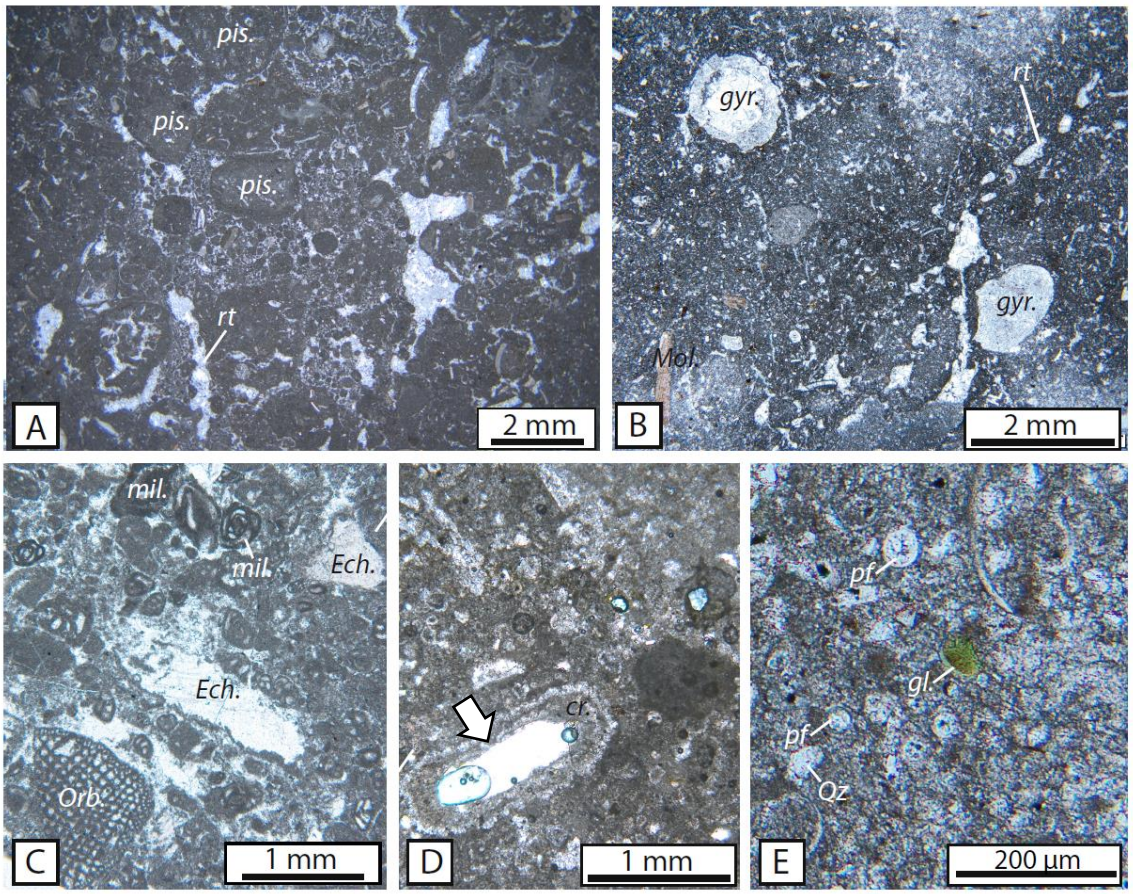
FIGURE 7



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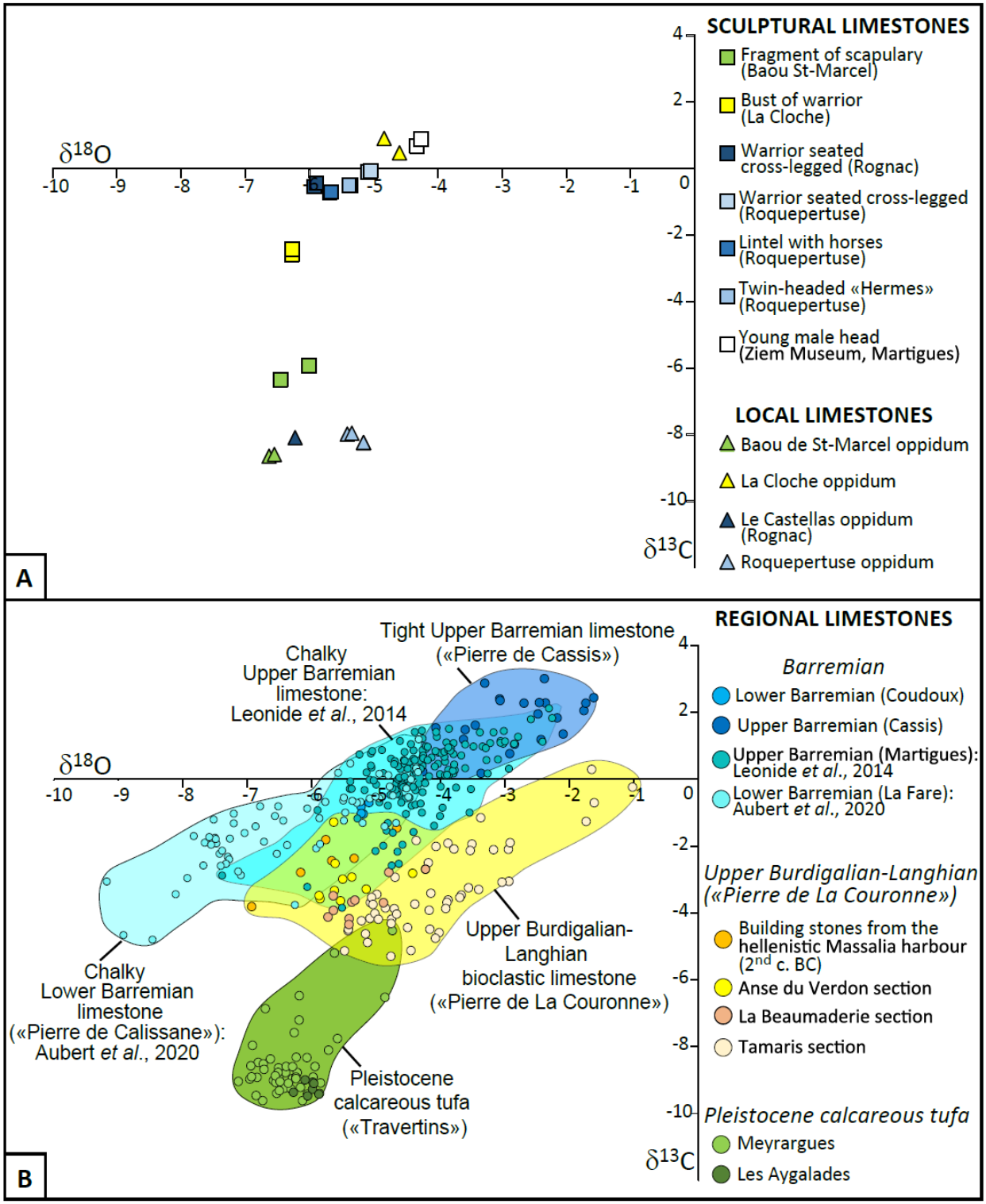

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FIGURE 9

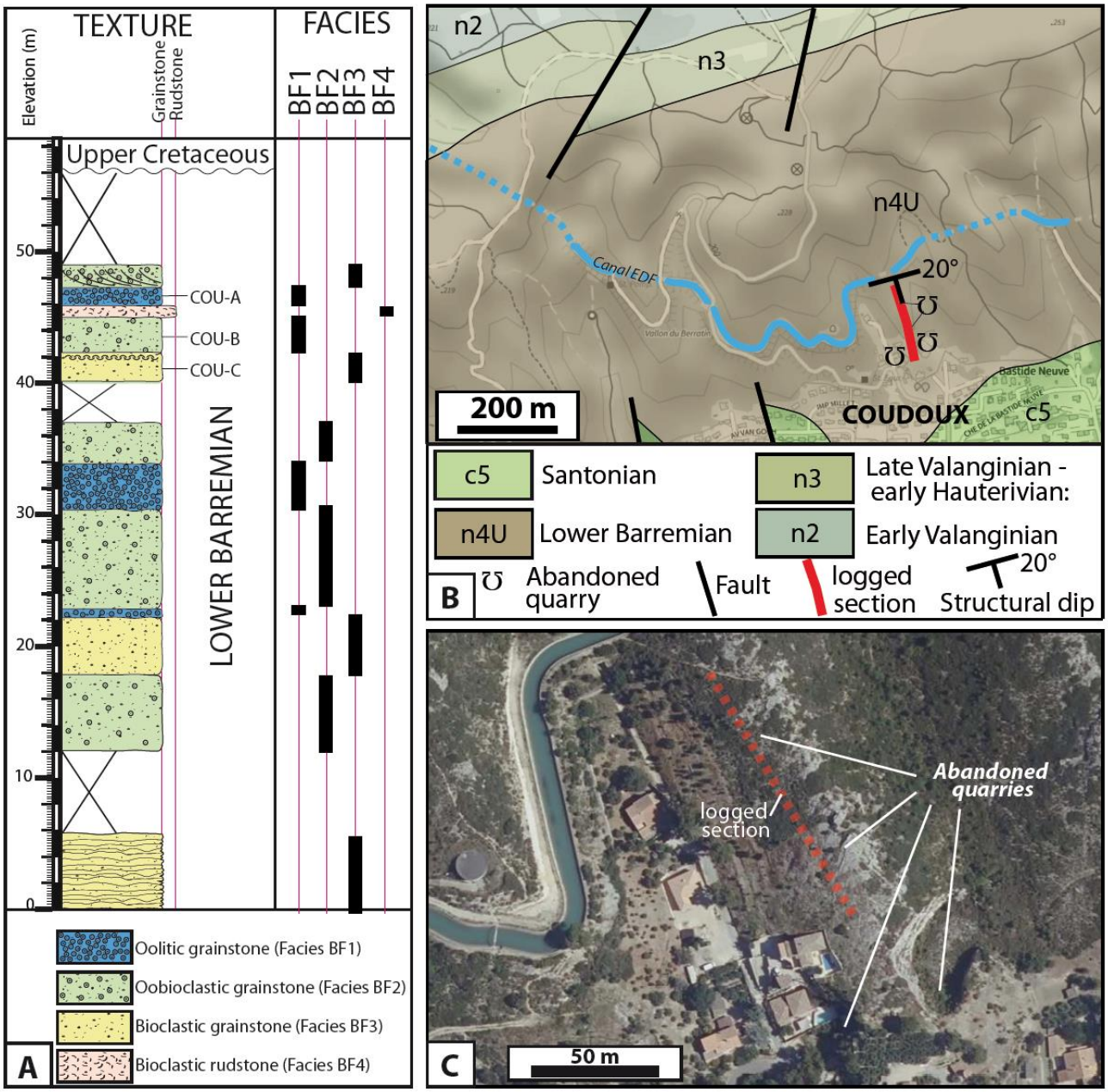

Figure 9: **A)** Textural log and vertical facies changes of the upper part of Lower Barremian succession, north of Coudoux (see location in **B** and **C**); **B)** Geological map of the southern flank of La Fare Massif, north of Coudoux and position of the section logged in **A**; **C)** Aerial view of the logged section and position of abandoned quarries.

FIGURE 10

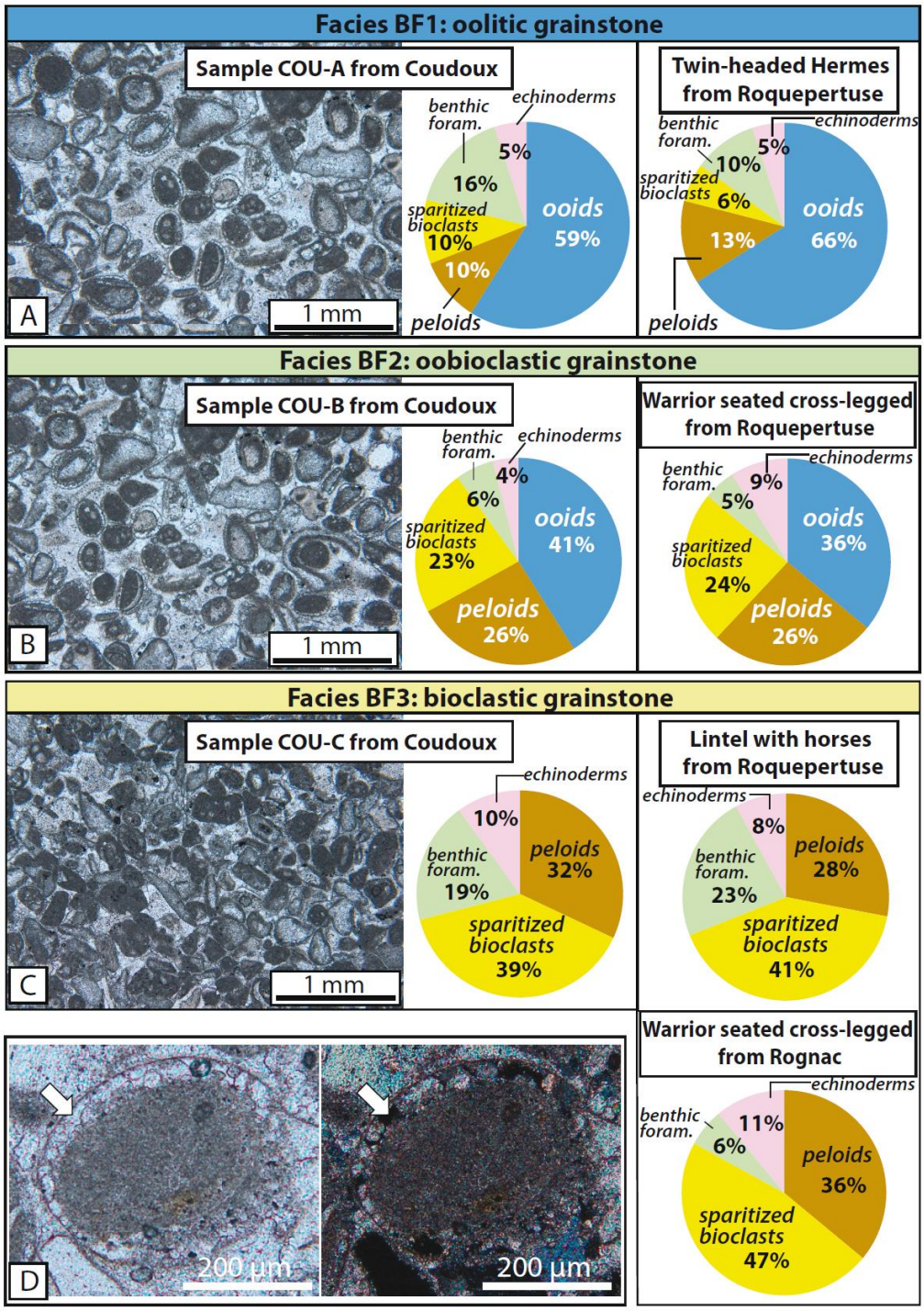

Figure 10: A) Thin section microphotograph of sample COU-A from Coudoux section, showing the oolitic grainstone facies (BF1) and comparison of carbonate grain composition between COU-A and the *twin-headed Hermes* from Roquepertuse; **B)** Thin section microphotograph of sample COU-B from Coudoux section, showing the oobioclastic grainstone facies (BF2) and comparison of carbonate grain composition between COU-B and the *warrior seated cross-legged* from Roquepertuse; C) Thin section microphotograph of sample COU-C from Coudoux section, showing the bioclastic grainstone facies (BF1) and comparison of carbonate grain composition between COU-A, the *lintel with horses* from Roquepertuse and the *warrior seated cross-legged* from Rognac; **D)** Detail of an ooid with leached cortex and equigranular sparry calcite infill, under polarized (left) and polarized-analyzed light (right), COU-A sample, Coudoux.

FIGURE 11

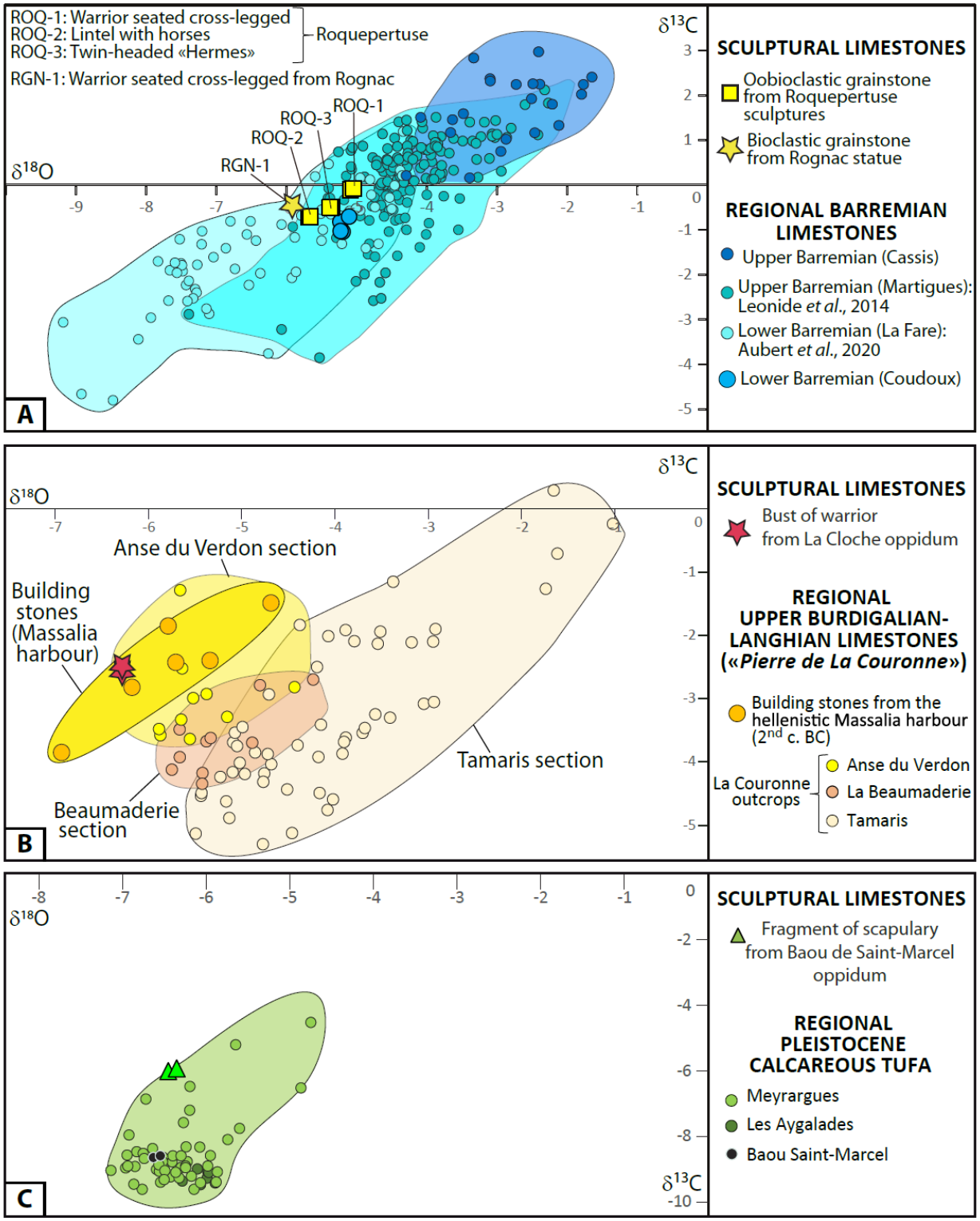

Figure 11: $\delta^{13}C$-$\delta^{18}O$ cross-plots: **A)** Sculptural limestones from Roquepertuse and Rognac and regional Barremian limestones; **B)** Bust of warrior from La Cloche and regional Burdigalian-early Langhian (?) limestones ("Pierre de la Couronne"); **C)** Fragment of scapulary tunic from Baou Saint-Marcel and regional Pleistocene calcareous tufa.

FIGURE 12

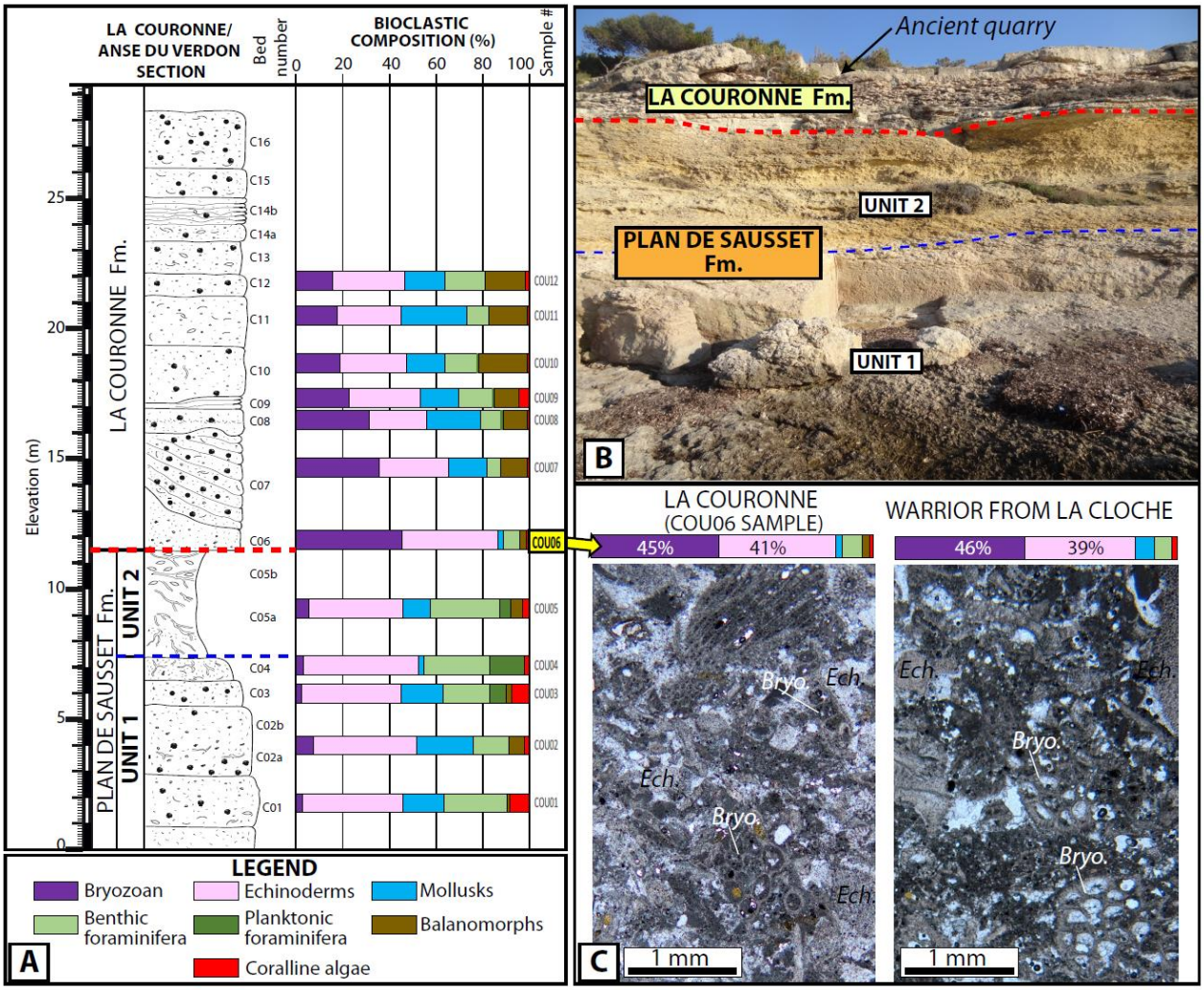

Fig.12: A) Log section of La Couronne/Anse du Verdon (see location in **Fig. 1B**) and bioclastic composition of limestones determined from point counting on thin-section; **B)** Photograph of the outcrop (eastern coast of Anse du Verdon) showing the upward transition from Plan de Sausset (Middle Burdigalian) and La Couronne (Upper-Burdigalian-early Langhian?) formation and the position of ancient quarries on top; **C)** Comparison of microfacies and grain composition between COU6 sample from La Couronne and the *bust of warrior* from La Cloche; Bryo: bryozoan; Ech.: echinoderms.

TABLE 1

Statuary limestone database	**Number of thin-sections**
Twin-headed Hermes (Roquepertuse): ROQ-3	1
Lintel with horses (Roquepertuse): ROQ-2	1
Warrior seated cross-legged (Roquepertuse): ROQ-1	1
Warrior seated cross-legged (Rognac): RGN-1	1
Bust of warrior (La Cloche); CL-1	1
Fragment of scapulary tunic (Baou de Saint-Marcel): BSM-1	1
Male head (Ziem Museum): ZM-1	1
Local limestones	
Roquepertuse oppidum	3
Le Castellas oppidum	3
La Cloche oppidum	2
Baou Saint-Marcel	2
Regional limestones	
Lower Barremian limestones from La Fare Massif	>200
Upper Barremian limestones from Martigues	50
Cenomanian limestones from Le Beausset bassin	50
La Couronne limestone (Burdigalian-Langhian)	69
Pleistocene calcareous tufa from Provence	30
Building stone limestones (Hellenistic harbor of Massalia)	
Western edge of the freshwater basin (2nd c. BC): MJV3	1
Western quay of the harbour horn: MJV4 (1st c. AD)	1
Interior facing of the south tower (2nd c. BC): MJV5	1
Paving of the freshwater basin (2nd c. BC): MJV6-7	2

Table 1: Thin-section database of limestones from Provence

TABLE 2

Statuary limestone database	**Number of C and O stable isotope measurements**
Twin-headed Hermes (Roquepertuse): ROQ-3	2
Lintel with horses (Roquepertuse): ROQ-2	2
Warrior seated cross-legged (Roquepertuse): ROQ-1	2
Warrior seated cross-legged (Rognac): RGN-1	2
Bust of warrior (La Cloche); CL-1	2
Fragment of scapulary tunic (Baou de Saint-Marcel): BSM-1	2
Male head (Ziem Museum): ZM-1	2
Local limestones	
Roquepertuse oppidum	4
Le Castellas oppidum	1
La Cloche oppidum	2
Baou Saint-Marcel	2
Regional limestones	
Lower Barremian limestones from Cassis ("Cassis Limestone")	22
Lower Barremian limestones from La Fare Massif (Aubert et al., 2020)	66
Upper Barremian limestones from Martigues (Léonide et al., 2014)	163
La Couronne limestone (Burdigalian-Langhian)	78
Pleistocene calcareous tufa from Provence	56
Building stone limestones (Hellenistic harbor of Massalia)	
Western edge of the freshwater basin (2nd c. BC): MJV3	1
Western quay of the harbour horn: MJV4 (1st c. AD)	1
interior facing of the south tower (2nd c. BC): MJV5	1
paving of the freshwater basin (2nd c. BC): MJV6-7	2

Table 2: Carbon and oxygen isotope database of selected limestones from Provence.

TABLE 3

	MgO/ CaO	Al_2O_3/ CaO	SiO_2/ CaO	Fe/Ca	K/Ca	density (10^3 kg/m3)	porosity (%)	P-wave velocity (m/s)
Young Male Head (Ziem Museum)	120	48	96	4	14	2.37	12.6	5032
Roquepertuse: seated warrior (ROQ 1)	90	70	150	9	17			4574
Roquepertuse: lintel with horses (ROQ 2)	160	63	150	7	14			2595
Roquepertuse: Twin-headed Hermes (ROQ 3)	170	46	300	9	20	2.24	17.5	1978
La Cloche: Bust of Warrior (CL1)	140	47	180	5	15	2.16±0.06	20.4 ±2.1	4166
Rognac: seated warrior (RGN1)	240	nd	32	3	11			2558
Coudoux outcrop: COU A	130	17	13	0.4	9	2.56	5.6	
Coudoux outcrop: COU B	160	18	13	0.5	9	2.24	17.3	
Coudoux outcrop: COU C	110	19	15	0.4	10	2.49	7.8	
Roquepertuse outcrop: ROQ C	140	23	18	2	9	2.32	14.3	
Roquepertuse outcrop: ROQ D	110	17	19	2	10	2.4	11.3	
La Cloche outcrop (CLA)	110	19	15	0.3	9			
Rognac-Le Castellas outcrop (RGNA)	nd	23	24	3	10	2.67	1.5	
Mauribas, near Velaux outcrop (MAU A)	90	22	16	4	9	2.41	11	
Baou Tailla (La Couronne)	nd	23	54	9	1.5	2.16	20.4	

Table 3: Results of non-destructive methods: pXRF measurements, density and porosity estimates and P-wave velocity.

APPENDIX

Carbon and oxygen isotope measurements

	Sample identifier	d18O vs PDB (‰)	d13C vs PDB ‰
STATUARY LIMESTONES			
	BSM	**-6,45**	**-6,01**
	BSM	**-6,35**	**-5,93**
	CL 1	**-6,28**	**-2,58**
	CL 1	**-6,27**	**-2,44**
	RGN 1	**-5,93**	**-0,55**
	RGN 1	**-5,89**	**-0,44**
	ROQ 1	**-5,09**	**-0,10**
	ROQ 1	**-5,05**	**-0,07**
	ROQ 2	**-5,68**	**-0,71**
	ROQ 2	**-5,66**	**-0,70**
	ROQ 3	**-5,36**	**-0,50**
	ROQ 3	**-5,38**	**-0,50**
	ZIEM	**-4,33**	**0,68**
	ZIEM	**-4,26**	**0,88**
LOCAL LIMESTONES			
	BSM A	**-6,63**	**-8,65**
	BSM A	**-6,55**	**-8,60**
	CL A	**-4,60**	**0,47**
	CL A	**-4,84**	**0,91**
	RGN A	**-6,23**	**-8,09**
	ROQ C	**-5,41**	**-7,98**
	ROQ C	**-5,34**	**-7,96**
	ROQ D	**-5,16**	**-8,25**
	ROQ D	**-5,16**	**-8,23**
REGIONAL LIMESTONES			
Lower Barremian limestone, Coudoux			
	COUA	**-5,23**	**-0,82**
	COUA	**-5,11**	**-0,70**
	COUB	**-5,20**	**-1,04**
	COUB	**-5,23**	**-1,03**

Upper Barremian limestone, Cassis ("Pierre de Cassis")

CAS-1	**-1,78**	**2,05**
CAS-2	**-1,75**	**2,27**
CAS-3	**-2,49**	**1,95**
CAS-4	**-2,37**	**2,28**
CAS-5	**-3,08**	**2,40**
CAS-6	**-2,47**	**2,29**
CAS-7	**-3,31**	**2,87**
CAS-8	**-2,39**	**3,01**
CAS-9	**-3,62**	**1,48**
CAS-10	**-4,28**	**0,23**
CAS-11	**-4,07**	**1,54**
CAS-12	**-1,63**	**2,44**
CAS-13	**-2,71**	**2,27**
CAS-14	**-3,46**	**1,62**
CAS-15	**-3,37**	**0,17**
CAS-16	**-3,08**	**2,35**
CAS-17	**-2,44**	**1,19**
CAS-18	**-2,10**	**1,35**
CAS-19	**-3,64**	**1,19**
CAS-20	**-2,85**	**1,04**
CAS-21	**-2,93**	**0,77**
CAS-22	**-2,19**	**1,83**

"Pierre de La Couronne" (Burdigalian-Langhian)

Building stones: hellenistic Massalia harbour (2nd c. BC)

MJV 2	**-4,68**	**-1,46**
MJV 3	**-5,34**	**-2,37**
MJV 4	**-6,93**	**-3,81**
MJV 5	**-6,17**	**-2,79**
MJV 6	**-5,69**	**-2,44**
MJV 7	**-5,78**	**-1,81**

Tamaris section

TP 01	**-5,13**	**-4,89**
TP 02	**-5,16**	**-4,62**
TP 03	**-4,72**	**-3,87**
TP 04	**-4,76**	**-4,18**
TP 05	**-3,88**	**-3,61**
TP 05-OR	**-2,94**	**-3,05**
TP 06	**-4,77**	**-4,31**
TP 06-OR	**-5,44**	**-4,54**
TP 07	**-4,51**	**-4,44**
TP 07-OR	**-5,49**	**-5,13**
TP 08	**-4,18**	**-3,92**
TP 08-OR	**-3,68**	**-3,46**
TP 09	**-4,68**	**-4,04**
TP 09-OR	**-5,23**	**-4,24**
TP 10	**-5,10**	**-4,16**
TP 10-OR	**-5,43**	**-4,49**
TP 11	**-4,40**	**-3,74**
TP 12	**-3,05**	**-3,08**
TP 13	**-2,95**	**-2,10**
TP 13-OR	**-1,62**	**-0,70**
TP 14	**-1,74**	**-1,26**
TP 14-OR	**-1,66**	**0,29**
TP 15	**-3,41**	**-3,30**
TP 16	**-3,57**	**-3,25**
TP 17	**-3,90**	**-3,68**
TP 18	**-3,18**	**-2,11**
TP 19	**-3,70**	**-3,54**
TP 20	**-4,02**	**-4,59**
TP 21	**-4,46**	**-5,12**
TP 22	**-4,08**	**-4,76**
TP 23	**-3,89**	**-1,91**
TP 23-OR	**-3,38**	**-1,15**
TP 24	**-4,07**	**-2,01**
TP 24-OR	**-4,38**	**-1,84**
TP 25	**-3,51**	**-1,94**
TP 26	**-3,54**	**-2,13**
TP 26-OR	**-4,19**	**-2,50**
TP 27	**-3,79**	**-2,09**
TP 28	**-3,92**	**-3,87**
TP 29	**-4,17**	**-4,49**

	TP 30	**-2,92**	**-1,90**
	TP 30-OR	**-1,02**	**-0,24**
	TP 31	**-4,15**	**-3,41**
	TP 32	**-4,77**	**-5,30**
	TP 33	**-5,09**	**-3,69**
	TP 33-OR	**-4,99**	**-3,45**
	TP 34	**-5,07**	**-3,54**
	TP 34-OR	**-4,71**	**-2,93**
	TP 35	**-4,86**	**-3,85**
	TP 36	**-5,05**	**-3,75**
	TP 37	**-5,02**	**-4,01**
	TP 38	**-4,97**	**-4,19**
Beaumaderie section			
	BP 01	**-4,89**	**-3,69**
	BP 01 OR	**-5,67**	**-3,49**
	BP 02	**-5,75**	**-4,12**
	BP 02 OR	**-5,42**	**-4,18**
	BP 03	**-5,66**	**-3,92**
	BP 03 OR	**-5,42**	**-4,35**
	BP 04	**-5,38**	**-3,67**
	BP 04 OR	**-5,33**	**-3,62**
	BP 05	**-4,80**	**-2,79**
	BP 05 OR	**-4,23**	**-2,70**
Anse du Verdon section			
	V 01	**-5,87**	**-3,58**
	V 01 OR	**-5,55**	**-3,64**
	V 02	**-5,89**	**-3,48**
	V 02 OR	**-5,65**	**-3,33**
	V 03	**-5,15**	**-3,29**
	V 03 OR	**-5,52**	**-2,99**
	V 04	**-5,64**	**-2,52**
	V 04 OR	**-5,66**	**-1,29**
	V 05	**-5,38**	**-2,93**
	V 05 OR	**-4,43**	**-2,82**
Calcareous tuffa from Provence (Pleistocene)			
Meyrargues			
	M 8	**-6,24**	**-8,32**
	M 9	**-6,22**	**-8,77**
	M 10	**-6,52**	**-8,64**
	M 11	**-6,51**	**-8,94**
	M 12	**-5,95**	**-8,59**
	M 13-A	**-6,41**	**-8,78**
	M 13-B	**-6,30**	**-8,95**
	M 19	**-6,08**	**-8,33**
	M 18	**-6,96**	**-8,97**
	M 27	**-5,86**	**-9,08**
	M 38	**-6,43**	**-9,24**
	M 39	**-6,59**	**-9,01**
	M 40-A	**-6,52**	**-8,77**
	M 41	**-5,98**	**-8,91**

	M 42	**-6,17**	**-9,22**
	M 43	**-5,74**	**-8,10**
	M 44-A	**-6,24**	**-8,91**
	M 14-A	**-6,72**	**-6,86**
	M 14-B	**-6,19**	**-6,48**
	M 15-A	**-6,83**	**-8,47**
	M 15-B	**-6,94**	**-8,57**
	M 16-A	**-4,86**	**-6,52**
	M 16-B	**-5,65**	**-5,20**
	M 17	**-7,14**	**-9,04**
	M 29-A	**-5,88**	**-9,40**
	M 30	**-6,32**	**-9,27**
	M 34	**-6,56**	**-9,07**
	M 1	**-6,20**	**-7,21**
	M 2	**-6,64**	**-8,37**
	M 3-4	**-6,40**	**-8,59**
	M 3-B	**-6,75**	**-8,52**
	M 4	**-5,60**	**-7,76**
	M 5	**-6,56**	**-8,87**
	M 6	**-6,54**	**-9,40**
	M 7	**-6,30**	**-8,60**
	M 20	**-6,36**	**-9,10**
	M 21	**-6,74**	**-8,68**
	M 22	**-6,85**	**-8,92**
	M 23	**-4,75**	**-4,53**
	M 24	**-6,49**	**-9,04**
	M 25	**-6,24**	**-8,95**
	M 26	**-6,50**	**-9,32**
	M 28	**-6,76**	**-9,61**
	M 31	**-6,14**	**-9,61**
	M 32	**-6,93**	**-8,90**
	M 33	**-6,07**	**-9,42**
	M 35	**-6,92**	**-7,96**
	M 36	**-6,27**	**-7,58**
	M 37	**-6,86**	**-9,46**
Les Aygalades			
	A1a	**-5,99**	**-9,28**
	A1b	**-6,28**	**-9,37**
	A2	**-6,06**	**-9,47**
	A3	**-5,98**	**-9,06**
	A5	**-6,10**	**-8,99**
	A6	**-5,89**	**-9,42**
	A7	**-5,97**	**-9,10**