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## ARCHAEOMETRIC ANALYSIS FOR PROVENANCE AND CONTENT OF ROMAN AMPHORAE FROM THE SITE OF SA MESQUIDA (MALLORCA, SPAIN)

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### ABSTRACT

This paper presents the results of an inorganic and organic approach to Roman amphorae found at the rural Roman site of Sa Mesquida (Mallorca). A set of amphorae were discovered in some rooms of the earlier phases of the villa. These amphorae were assigned to a general Tarraconensian and Italic provenance on a macroscopic level. In order to verify this and to accurately determine the provenance an archaeometric characterization was carried out on twelve amphorae. The analytical approach combined wavelength dispersive X-ray fluorescence (WD-XRF) for the chemical analysis, as well as powder x-ray diffraction (XRD) and optical microscopy by thin-section analysis (OM) for the mineralogical and petrographic characterization. In addition, organic residue analysis by gas-chromatography mass spectrometry (GC-MS) was applied to investigate the content of some of the amphorae for which wine was suggested on an archaeological basis.

The results have allowed for a better provenance determination identifying some amphorae from specific workshops or micro-regions within ancient Tarraconensis, as well as a better definition of the Italic materials. The organic analysis has confirmed the presence of wine in the analysed samples.

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**KEYWORDS:** chemistry, mineralogy, petrography, organic residues, amphorae, Balearics, Roman

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## 1. INTRODUCTION

The site of Sa Mesquida was discovered in 1984 in the town of Santa Ponça (Calvià, Mallorca) (Figure 1). The discovery of structures and abundant ceramic materials led to undertake in the 1980s and 1990s of the 20th century archaeological excavations (i.e., Vallespir *et al.*, 1987). These excavations uncovered a series of rooms articulated around a courtyard with a well, an industrial deposit and a ceramic kiln for common ware (Cau, 2008) (Figure 1). Few hundred meters away from the main ruins, separated by modern construction, a cistern used as a rubbish dump in Late Antiquity was also discovered (Orfila and Cau, 1994; Cau, 2003). New archaeological excavations undertaken in 2010 were concentrated on the rooms and the pottery kiln. The results of the excavation of the kiln suggest the abandonment of the structure at the end of the 2nd or beginning of the 3rd century AD, and a later use as a rubbish dump during Late Roman period (Mas Florit *et al.*, 2015).

The data obtained shows that the villa was occupied from the 1st century BC, with an important moment in the 1st and 2nd centuries AD. At the very end of the 2nd century or beginning of the 3rd century AD, the settlement suffered a traumatic event, since evidence of a fire is attested, with the consequent destruction of the main building (Mas Florit *et al.*, 2015). In any case, the site shows signs of a later occupation, as demonstrated by the rubbish dumps found both in the kiln, with ceramics dating to the last quarter of the 4th and the first quarter of the 5th century AD, and in the cistern, with materials dated mainly in the 5th century AD and with some later materials from the Byzantine period (Orfila and Cau, 1994; Cau, 2003).

The present contribution deals with the earliest phases of the villa focusing on the amphorae appeared in some of the rooms. The idea is to explore the provenance and content combining inorganic and organic analyses which have been normally addressed separately. To this end, a complete inorganic archaeometric characterization has been performed on a set of amphorae classified as Dressel 1B, Tarraconense 1 and Pascual 1, and organic residues analysis has been applied to some of them. The main aim is to determine the provenance and content of what in archaeological terms have been considered as Italic and Tarraconensian wine amphorae.

Dressel 1B amphorae were produced in a number of pottery workshops along the Tyrrhenian coast of Italy between the last quarter of the 2nd century BC until the last decade of the 1st century BC, and were widely distributed in urban and rural settlements in the western Mediterranean (Peacock and Williams, 1986; Tchernia, 1986).

Tarraconense 1 and Pascual 1 are the characteristic types of wine amphora production in north-eastern Spain (Hispania Citerior province, later called Tarraconensis under Augustus' territorial reorganisation). Tarraconense 1 was produced from mid-1st century BC, and especially around 50-30 BC in some pottery workshops located in the central Catalan coast, around the Roman city of Iluro/Mataró. This type includes a wide variety of profiles but, for the most part, they exhibit an everted rim, short neck and ovoid body. All the variants were produced to replace the first vessels used in the province after the Roman conquest for the transport of local wine, that is, the imitations of Italian Dressel 1 amphora. The type Pascual 1 was introduced from 40 BC in the workshops located near Iluro/Mataró and Baetulo/Badalona in the central Catalan coast. This type replaced the previous Tarraconense 1 variants and was produced on a large scale in the north-eastern territories of the province from the last third of the 1st century BC until mid-1st century AD. Pascual 1 was the amphora mainly used for trading local wine to Gallia and, from the Augustus reign, to Britannia and the military camps located in the Germanic limes (Martínez Ferreras, 2014).

The amphorae analysed were recovered in different archaeological stratigraphical units, but all of them have been dated to the same phase of occupation at the Roman villa of Sa Mesquida. The Italian Dressel 1B amphora was still predominant in the western territories until the early Augustan period. Therefore, these contexts should be dated around 40/20 BC, when the Tarraconense 1 and Pascual 1 types coincided and competed with the Italian Dressel 1 amphorae in the north-western Mediterranean markets as evidenced in the cargo of the Port Vendres 4 shipwreck (Martínez Ferreras *et al.* 2015) and in many sites such as Tarraco/Tarragona (Gibellí Borràs, 2015), Emporiae/Empúries (Tremoleda *et al.*, 2015) and Narbo Martius/Narbonne (Sanchez, 2015).

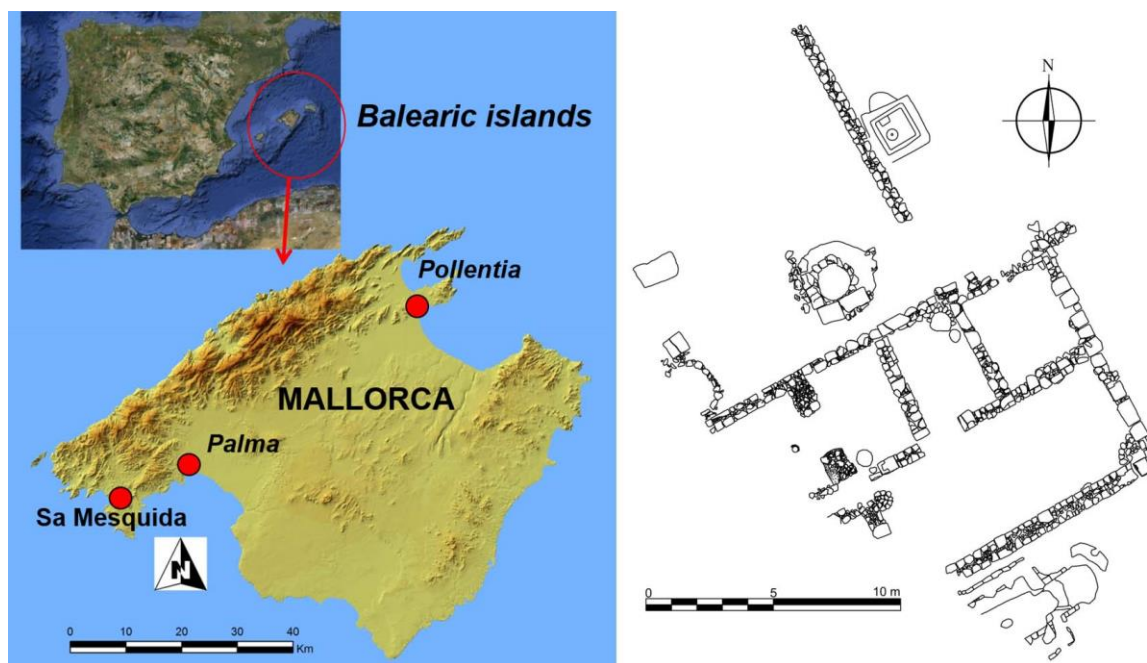


Figure 1. Map of Mallorca with the location of Sa Mesquida (left); general plan of the archaeological site (right)

2. MATERIALS AND METHODS

A total of 12 amphorae were randomly selected from several contexts from the same building of the Roman site of Sa Mesquida (Table 1; Figure 2). According to typological criteria and after a first examination with a stereomicroscope, two main provenances were identified. The amphorae of type Dressel 1B (five samples: MCF065, MCF066,

MCF067, MCF068 and MCF070) consist in several fabrics considered to have been produced in Italy (Figure 3). The rest of the amphorae are Tarracense 1 (MCF075) and Pascual 1 (MCF069, MCF071 to MCF073, MCF076 and possibly MCF074), for which a provenance in the current Catalan area was proposed.

Table 1. Amphora samples analysed in this study; SU=Stratigraphical Unit; Rim-handle=Rim with handle; XRF=WDX-Ray Fluorescence; XRD=X-Ray Diffraction; OM=Optical Microscopy; Organic=label of the samples in the organic residue analysis

Sample	SU	Type	Sampled part	XRF	XRD	OM	Organic
MCF065	3075	Dressel 1?	Spike	x	x	x	122
MCF066	3075	Dressel 1B	Rim-handle	x	x	x	125
MCF067	3075	Dressel 1B	Rim-handle	x	x	x	123
MCF068	3075	Dressel 1B	Rim	x	x	x	126-127
MCF069	3054	Pascual 1	Rim	x	x	x	-
MCF070	3073	Dressel 1B	Rim-handle	x	x	x	124
MCF071	3040	Pascual 1	Rim	x	x	x	-
MCF072	3022	Pascual 1A	Rim	x	x	x	-
MCF073	3005	Pascual 1A	Rim	x	x	x	-
MCF074	3087	Indet. (Pascual 1?)	Body sherd	x	x	x	-
MCF075	3087	Tarracense 1	Rim	x	x	x	-
MCF076	3087	Pascual 1	Rim	x	x	x	-



*Figure 2. Illustrations of the amphorae analysed in this study. In the photograph: MCF068 Italic amphora; MCF075 amphora from Tarraconensis; MCF065 Italic amphora*

The inorganic composition of these amphorae was characterized using wavelength dispersive X-ray fluorescence (XRF), thin-section petrographic analysis through optical microscopy (OM) and powder X-ray diffraction (XRD). Five Dressel 1B amphorae were subjected to organic residue analysis by gas-chromatography-mass spectrometry to check their use for wine trade or for any other type of product.

For WD-XRF and powder XRD analyses, about 15 g of each sample were pulverised and homogenised (after removing its superficial layers) using a mill with a tungsten carbide cell. Each specimen was subsequently dried in an oven at 105°C for 12 hours.

Quantitative information on the chemical composition of the individuals was obtained through WD-XRF, using a Panalytical-Axios PW 4400/40 spec-

trometer. This allowed for the identification of 29 major, minor and trace elements, including Fe<sub>2</sub>O<sub>3</sub> (as total Fe), Al<sub>2</sub>O<sub>3</sub>, MnO, P<sub>2</sub>O<sub>5</sub>, TiO<sub>2</sub>, MgO, CaO, Na<sub>2</sub>O, K<sub>2</sub>O, SiO<sub>2</sub>, Ba, Rb, Th, Nb, Pb, Zr, Y, Sr, Ce, Ga, V, Zn, Cu, Ni, Cr, Mo, Sn, Co and W. Of these, Mo and Sn were discarded due to their low counting statistics, while Co and W due to a possible contamination from the tungsten carbide cell of the mill used in the preparation of the samples. For the determination of major and minor elements, glass beads were prepared from 0.3 g of specimen in an alkaline fusion with lithium tetraborate (1/20 dilution). For the trace elements determination, powder pills were prepared, using 5 g of specimen mixed with binder (resin) Elvacite 2044 placed over boric acid in an aluminium capsule and pressed for 60 s at 200 kN. The quantification of the elements was obtained by using a calibration line performed with 60 International Geological Standards. The loss on ignition (LOI) was calculated from calcinations of 0.3 g of dried specimen, heated at 950°C for 3 hours.

For the mineralogical composition powder XRD analysis was performed using a Siemens D-500 diffractometer, equipped with a graphite monochromator in the diffracted beam, at 1.2 kW (40kV, 30mA), and working with the Cu K $\alpha$  radiation ( $\lambda=1.5406 \text{ \AA}$ ). Measurements were taken between 4 and 70°2 $\theta$ , with a step size of 0.05°2 $\theta$  and a step time of 3 s. Crystalline phases were assessed using the software HighScore Plus by PANalytical, including the Joint Committee of Powder Diffraction Standards (JCPDS) data bank.

Thin sections of the specimens were prepared for petrographic analysis. A transverse section of each sample was obtained, impregnated with epoxy resin, mounted using Loctite UV glue and sectioned using a Struers Discoplan TS. Each section was finished by hand using an abrasive powder until reaching a thickness of 30  $\mu\text{m}$  in which quartz presents a grey-white first order interference colour. Observations were made using a polarising microscope Olympus BX41, working under magnification between 20X and 200X. Microphotographs were taken with a digital camera Olympus DP70 attached to the microscope, using the software Analysis Five. The description followed the standards used in ceramic petrology (Whitbread, 1989, 1995; Quinn, 2013).

In order to assess the provenance of these amphorae beyond the established provincial adscription, a comparison with the geochemical and petrographic reference database from different production centres and micro-regions available at ERAAUB was performed. The database for comparative purposes includes Tarraconense 1 and Pascual 1 amphorae from twenty pottery workshops in Hispania Citerior-Tarraconensis. In fact, the reference data selected

for comparison correspond to the reference groups already identified for these types at the pottery workshops distributed around the main Roman cities and the river valleys (Martínez Ferreras, 2014; Vila, 2011). The archaeometric data of the Dressel 1B amphorae from Sa Mesquida were compared with samples of Italian origin found in the Port Vendres 4 shipwreck (Port-Vendres, south France), dated to 40/30 BC (Martínez Ferreras et al., 2015). Its cargo was constituted by a particular assemblage of Roman wine amphorae from the Tyrrhenian coast of Italy (Dressel 1B and Lamboglia 2 types) and north-eastern Spain (Pascual 1 type) destined for Gallic markets. It therefore represents a case study of trade in transit contemporary to the archaeological contexts from Sa Mesquida. In addition, compositional data regarding Italian amphorae productions obtained from bibliography was added for comparison (Thierrin-Michael et al., 2004; Thierrin-Michael, 2007; Olcese et al., 2013a, 2013b).

In most of the amphorae studied only the rim was preserved, which could not be sampled for residue analysis, as it likely was not in contact with the original content, but with the lid of the amphora. Only the five samples of Dressel 1B amphorae showed part of the neck and could therefore be sampled and subjected to organic residues analyses (Table 1). All of them come from the SU (stratigraphic unit) 3075, except for the individual MCF070 that comes from the SU 3073 (Table 1). They had been previously washed, except the sample MCF068 that arrived at the laboratory filled with earth; the analysis of this sediment, allowed for a control over any possible contamination caused by the burial environment. The amphorae were mechanically cleaned with a scalpel before sampling; the samples were pulverised and analysed by means of gas chromatography-mass spectrometry (GC-MS).

Different extractions were carried out: the total lipid extract and its hydrolysis (a, b) were obtained on 2 g of pulverised sample following Mottram et al. (1999) and Giorgi et al. (2010); the extraction for the identification of wine residues (c) was performed on 0,5 g of sample following Pecci et al. (2013), and the hydrolysis on the solid fraction (d) was obtained following Giorgi et al. (2010). All the extracts were derivatised by adding 25  $\mu\text{L}$  of N,O-bis(trimethylsilyl)trifluoroacetamide (BSTFA, Sigma-Aldrich) and heating at 70 °C for 1 h. Then, 75  $\mu\text{L}$  of hexane. 1  $\mu\text{L}$  was injected for the analysis. The analysis was performed using a Thermo Scientific TS GC ultra chromatograph (equipped with a silica capillary column of 30 m and 0.25  $\mu\text{m}$  film thickness) and a mass spectrometer Thermo Scientific ITQ 900, operated in the electron ionisation mode (70 eV). The mass range was 40-650 m/z. The GC oven tempera-

ture was held at 50°C for 1 min, then increased 5°C/min up to 310 °C and held isothermally for 10 min.

### 3. RESULTS AND DISCUSSION

#### 3.1. Petrographic analysis

The examination of the amphorae through thin section analysis points to eight different petrographic fabrics, four of which correspond to amphorae Dressel 1B (fabrics a to d) and four to the Tarraconense 1 and Pascual 1 types (fabrics e to h).

##### 3.1.1. Dressel 1B amphorae

The five Dressel 1B amphorae consist of medium-coarse fabrics; according to the petrographic composition they can be attributed to four different workshops or production areas located in the Tyrrhenian coast of Italy.

a) Fabric with acidic metamorphic rocks, chert and few volcanic inclusions: sample MCF065 (Figure 3a). Iron-rich clay matrix (with some carbonates) that displays optical activity. The fine fraction (<0.1 mm) comprises quartz, muscovite, feldspars and, in lower frequency, micritic calcite. The coarse fraction ( $\leq 0.5$  mm) is abundant, single-spaced, constituted by angular to subrounded grains. It contains predominant quartz, dominant plagioclase, K-feldspar and micritic calcite, frequent quartzite and chert, common micas (muscovite), sanidine and iron oxides, and few amphibole and clinopyroxene crystals.

The petrographic composition of Fabric a might be compatible with some areas of marginal volcanism in the Tyrrhenian coast of Italy, given the frequency of quartz and chert in addition to the few volcanic components. Although sanidine could be associated with the alkali-potassic volcanism of the Somma-Vesuvio (area of Lazio/Campania), the low presence of volcanic inclusions points to southern Tuscany or the littoral zone between southern Lazio and northern Campania as the possible areas of origin (Thierrin-Michael, 1991, 2007; Thierrin-Michael *et al.*, 2004; Olcese and Thierrin-Michael, 2009; Olcese, 2012; Cibecchini and Capelli, 2013; Olcese *et al.*, 2013a).

b) Fabric with sandstone, acidic metamorphic rocks and volcanic rocks: sample MCF066 (Figure 3b). The clay matrix is iron-rich and displays no op-

tical activity. The fine fraction (<0.1 mm) comprises predominant quartz, muscovite and, to a lesser degree, micritic calcite and inclusions derived from volcanic rocks. The coarse fraction ( $\leq 1$  mm, mainly  $\leq 0.5$  mm) consists of abundant, single-spaced inclusions, angular to subrounded in shape. Quartz is predominant followed by frequent quartz-mica schist, chert, plagioclase and K-feldspar. Calcareous inclusions (Ca-microfossils and micritic calcite), volcanic rock fragments (basalt and trachyte) and clinopyroxene crystals are common; opaque minerals and sub-rounded fragments of Fe-rich siltstone and fine-grained sandstone are rare.

Fabric b contains more aplastic inclusions from volcanic origin, especially volcanic rock fragments and clinopyroxene crystals. The area of provenance might be located between southern Tuscany and southern Lazio, excluding the Campania region because of the presence of acidic metamorphic rock fragments and sandstone (Thierrin-Michael, 1991, 2007; Thierrin-Michael *et al.*, 2004; Olcese and Thierrin-Michael, 2009; Olcese, 2012; Cibecchini and Capelli, 2013; Olcese *et al.*, 2013a).

c) Fabric with acidic metamorphic rocks and planktonic foraminifera microfossils: sample MCF067 (Figure 3c). Calcareous matrix, with some iron oxides; it displays moderate optical activity. The fine fraction (<0.1 mm) is very abundant, containing predominant quartz and muscovite, along with calcareous microfossils (foraminifera). The coarse fraction ( $\leq 0.75$  mm) is relatively abundant, single-spaced to double-spaced, oriented parallel to the margins of the amphora. It comprises angular to subrounded inclusions among which quartz, plagioclase and K-feldspar are predominant. Planktonic foraminifera, partially or totally decomposed during the firing process, are frequent; acidic metamorphic rock fragments, opaque minerals and micas (biotite and muscovite) are common, while clinopyroxene crystals are rare.

The general characteristics of Fabric c, in particular the high frequency of acidic metamorphic rocks and foraminifera microfossils, resemble some products from the Strait of Messina area (Calabria or north-eastern Sicily), this being a possible provenance hypothesis (Olcese, 2012; Cibecchini and Capelli, 2013).



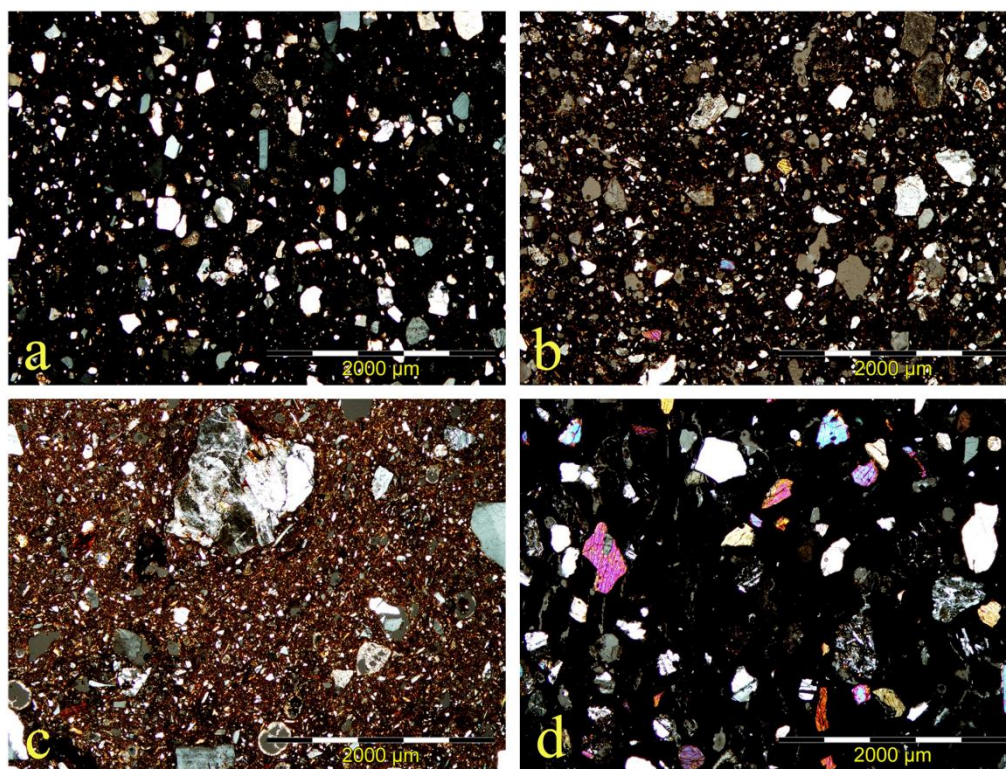


Figure 3. Microphotographs of thin sections of Dressel 1B fabrics, taken under crossed polars at the same magnification (40x). (a) MCF065. (b) MCF066. (c) MCF067. (d) MCF068

d) Fabric with predominant volcanic rocks and minerals (clinopyroxene, basalt, trachyte, sanidine): samples MCF068 and MCF070 (Figure 3d). Iron-rich matrix (with some carbonate concentrations), optically inactive in MCF068 or with moderate optical activity in MCF070. Frequent elongated voids are visible, oriented parallel to the ceramic walls. The fine fraction (<0.1 mm) is almost absent. The coarse fraction ( $\leq 0.5$  mm) is relatively abundant, single-spaced, constituted by angular to subrounded grains oriented slightly parallel. Inclusions consist of predominant clinopyroxene (augite and aegirine), volcanic rock fragments (trachyte and basalt) and quartz, dominant sanidine, frequent sedimentary rock fragments with Fe-rich cement and iron oxides, common micritic calcite, and few amphiboles.

The petrographic characteristics of Fabric d (MCF068 and MCF070) suggest that the coarse fraction was added as temper. The frequency and nature of the volcanic inclusions, associated with an alkaline volcanism, point to a provenance in southern Campania and, in particular, in the area of the Gulf of Naples (Olcese and Thierrin-Michael, 2009; Cibecchini and Capelli, 2013; Olcese et al., 2013a, 2013b; Martínez et al., 2015).

### 3.1.2. Tarraconense 1 and Pascual 1 amphorae

The seven amphorae of types Tarraconense 1 and Pascual 1 consist of coarse fabrics with large aplastic

inclusions among which fragments of acidic plutonic rocks and crystals derived from these rocks are prevalent. However, differences in the groundmass and the presence/absence and frequency of specific lithological types led to distinguish four diverse fabrics.

e) Calcareous fabric, with granitoids/metagranitoids and foraminifera microfossils: sample MCF071 (Figure 4a). Calcareous matrix with a reddish-brown colour in PPL. Bimodal distribution of the inclusions. The fine fraction (<0.1 mm) is abundant, with predominant quartz, feldspars, muscovite, and micritic calcite and calcareous microfossils partially or totally decomposed due to the firing process. The coarse fraction ( $\leq 1.5$  mm) is moderately abundant, single-spaced; the grains are subrounded to subangular and oriented slightly parallel to the margins of the amphora. Coarse inclusions comprise predominant granitoids, in addition to quartz, K-feldspar and (to a lesser degree) plagioclase, derived from these rocks; a low-grade metamorphism is observed in some fragments. Calcite and calcareous microfossils (foraminifera) semi-decomposed in micrite are frequent, while opaque minerals are common; phyllosilicates and sandstone fragments are rare.

Fabric e is compatible with the petrographic characteristics detected in amphora products from the northern Catalan coast. Specifically, it resembles



calcareous Pascual 1 amphorae produced in some pottery workshops located around the Roman cities of Iluro/Mataró, *Aquae Calidae*/Caldes de Montbui, *Blandae*/Blanes and *Emporiae*/Empúries, in which aplastic inclusions are constituted by (meta-) granitoids and micritic calcite (Martínez Ferreras 2014).

f) Border calcareous fabric, with granitoids/metagranitoids and biotite: sample MCF073 (Figure 4b). Iron-rich clay matrix, with a reddish colour in PPL, and a moderate calcareous content. The fine fraction (<0.1 mm) is abundant to very abundant and includes quartz, plagioclase, feldspar and micas (biotite and muscovite), as well as frequent semi-decomposed micritic calcite. The coarse fraction ( $\leq 2$  mm) is composed of single-spaced to double-spaced inclusions, subrounded to subangular. Granitoid fragments are predominant, usually showing low-grade metamorphism. Grains of quartz, plagioclase and K-feldspar derived from the granitoids are frequent; there are also abundant micas (dominant biotite), in addition to few micritic calcite and opaques.

The petrographic composition of Fabrics e and f is very similar, the main differences being related to the frequency of carbonates, which are rare in the latter. Therefore, the provenance of Fabric f might be fixed in the same littoral and pre-littoral area between the Roman cities of *Iluro* and *Emporiae* (Martínez Ferreras 2014).

g) Fabric with granitoids and micritic calcite nodules: sample MCF072 (Figure 4c). Iron-rich clay matrix with carbonates, reddish-brown coloured in PPL. The fine fraction (<0.1 mm) is very abundant, with predominant quartz, feldspar, muscovite and biotite, as well as common opaque minerals. The coarse fraction ( $\leq 1.5$  mm) comprises double-spaced to single-spaced inclusions, subrounded to subangular. Quartz crystals are pre-dominant, while nodules of micritic calcite –partially or totally decomposed during firing–, fragments of granitoids and crystals of plagioclase and K-feldspar are frequent. Biotite flakes and opaques are common, and metamorphic rocks (phyllite and schist) are rare.

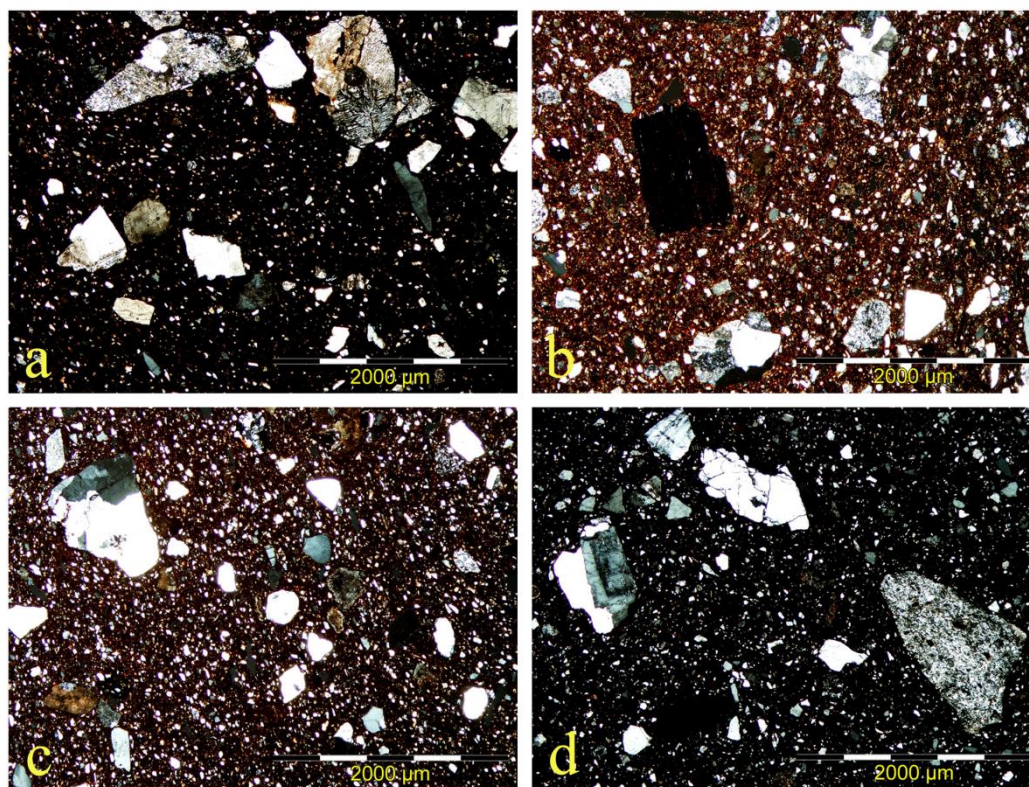


Figure 4. Microphotographs of thin sections of Tarraconense 1 and Pascual 1 amphorae, taken under crossed polars at the same magnification (40x). (a) MCF071. (b) MCF073. (c) MCF072. (d) MCF074

The Pascual 1 amphora MCF072 presents a petrographic composition that is common in some products from the central-northern Catalan coast, in which granitoids appear to be the prevalent aplastic inclusion (Martínez Ferreras 2014).

h) Low calcareous fabric, with predominant granitoids/metagranitoids and biotite: samples MCF069, MCF074, MCF075 and MCF076 (Figure 4d). These samples show the same fabric, with slight differences only. They present a low calcareous, iron-rich matrix, with a red colour in PPL. The fine fraction



(<0.1 mm) is moderately abundant and comprises predominant quartz, micas (biotite mainly), and feldspars. The coarse fraction ( $\leq 2$  mm) is composed of single-spaced inclusions, subangular to sub-rounded, oriented slightly parallel to the vessel margins. Granitoids and crystals derived from these rocks, specially quartz, plagioclase and K-feldspars are predominant. Other frequent components of these rock fragments are coarse laths of biotite; opaque minerals are common while muscovite and epidote are rare.

Based on their petrographic characteristics, the provenance of the four amphorae in Fabric h should be associated with a production zone in the Laietanian area, probably located around the Roman cities of *Iluro*, *Blandae* and *Aquae Calidae* (Martínez Ferreras, 2014). At least three known workshops in this zone (Ca l'Arnau, Can Notxa and Can Portell) produced Tarraconense 1 and Pascual 1 amphorae, using Quaternary river sediments that are mainly composed of igneous (granitoid) rock fragments derived from the Late Hercynian complex of the Catalan Coastal Range.

### 3.2. Inorganic Chemical analysis

The normalised chemical composition for the 12 amphorae analysed is given in Table 2. In order to obtain a first insight into the variability of the data set, the Compositional Variation Matrix (CVM) was calculated (Aitchison, 1986, 1992; Buxeda, 1999; Buxeda and Kilikoglou, 2003). The total variation value ( $tv = 1.96$ ) is quite high and consistent with the presence of different fabrics as identified by thin section analysis. The CVM provides information on the variation that each element introduces into the data set ( $\tau_i$ ); in this case, the highest values are related to CaO ( $\tau_{CaO} = 10.73$ ), Sr ( $\tau_{Sr} = 7.22$ ), Cu ( $\tau_{Cu} = 7.17$ ), Cr ( $\tau_{Cr} = 1.21$ ),  $P_2O_5$  ( $\tau_{P_2O_5} = 4.46$ ), Th ( $\tau_{Th} = 4.40$ ), Ni ( $\tau_{Ni} = 4.36$ ) and Y ( $\tau_Y = 4.13$ ). Conversely, the less variable element in the data set is  $Al_2O_3$  ( $\tau_{Al_2O_3} = 2.45$ ).

Although the formation of secondary calcite is a common phenomenon in archaeological ceramics which can affect the concentrations of CaO and other elements (i.e. Sr), the variations observed for CaO in this study seem more related to differences between amphorae made from Ca-rich pastes (MCF065 to MCF068, MCF070 and MCF071) and others produced using non-calcareous pastes (MCF069, MCF072 to MCF076) (Table 2), what accounts for the high  $\tau_{CaO}$  value. The high variation in Sr ( $\tau_{Sr}$ ) can also be explained by these differences since it is usually subjected to interferences caused by its correspondence with other alkaline-earth metals such as CaO. The high variability introduced by  $P_2O_5$  and Cu into the data set might be associated both with

differences in the initial composition of the amphorae or with possible perturbations due to alteration and/or contamination processes. In order to avoid problems with any possible compositional perturbation, these elements (in addition to others, such as Pb) were not considered for the statistical treatment of the data.

The high  $\tau_i$  values obtained for Y, Ni and Cr can be more probably related to differences in the initial composition of the amphorae, since the concentrations of these trace elements do not tend to be affected by alteration/contamination problems in archaeological ceramics. Some Italian amphorae exhibit the highest values in Ni (MCF067, MCF068, MCF070) and Cr (MCF068 and MCF070), while the Y values are especially high in four amphorae from Hispania Tarraconensis (MCF069, MCF074, MCF075, MCF076).

For the statistical treatment of the chemical data, the concentrations were transformed into additive log ratios (Aitchison, 1986; Buxeda, 1999); the subcomposition  $Fe_2O_3$ , MnO,  $TiO_2$ , MgO, CaO,  $Na_2O$ ,  $K_2O$ ,  $SiO_2$ , Ba, Rb, Nb, Zr, Y, Ce, Ga, V, Zn, Ni and Cr was used, using  $Al_2O_3$  as divisor for the alr transformations. A Cluster Analysis (CA), based on the alr transformed subcomposition, was performed, using the centroid agglomerative method and the squared Euclidean distance. The dendrogram (Figure 5), shows the amphorae distributed in two main groups, one including the five individuals of Dressel 1B type while the other one comprising the amphorae Pascual 1 and Tarraconense 1. This distribution supports the initial hypothesis of a provenance in two separate geographic regions for the amphorae analysed, that is, the Italian peninsula on one hand and the north-eastern Iberian peninsula on the other hand. The variable distribution of the individuals in the dendrogram is explained mainly by differences in CaO, MgO, Ni and Cr (Table 2). The Dressel 1B amphorae (MCF065 to MCF068, and MCF070), as well as one sample of Pascual 1 (MCF071), tend to present a higher CaO content than the other Pascual 1 and Tarraconense 1 amphorae. In addition, the Dressel 1B samples usually show higher concentrations of MgO, Ni and Cr, as observed especially in MCF068 and MCF070. These two amphorae exhibit similar chemical compositions suggesting probably the same provenance; they consist of calcareous pastes with relatively low  $SiO_2$  and higher concentrations of  $Fe_2O_3$ ,  $Al_2O_3$ , MnO,  $TiO_2$ , MgO, Ba, Nb, Ce, Ga, V, Zn, Ni and Cr than the rest of the samples of type Dressel 1B (Table 2). MCF065 shows low Ba, Y, Ga; MCF066 exhibits low  $Fe_2O_3$ , Ga and V, and MCF067 presents the lowest Ce value (Table 2). As for the Pascual 1/Tarraconense 1 amphorae, there are only two individuals with a low dissimilarity

level (MCF069 and MCF076), sharing a higher content of MgO, Zr and Ce, and lower of Ni and Cr (Table 2), than the rest of the samples in this group; the other amphorae (MCF071 to MCF074) show more heterogeneous compositions and are probably associated with more than one production centre.

Aiming to accurately assess the provenance of the Dressel 1B amphorae, the chemical data were compared with the Dressel 1B and Lamboglia 2 ampho-

rae from the Port-Vendres 4 shipwreck and with the reference groups previously identified (Thierrin-Michael *et al.*, 2004; Thierrin-Michael, 2007; Olcese *et al.*, 2013a, 2013b; Martínez Ferreras *et al.*, 2015). The available subcomposition (Fe<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>, MnO, MgO, CaO, SiO<sub>2</sub>, Nb, Zr, Y, Zn, Ni and Cr) was first transformed using TiO<sub>2</sub> as divisor and then a Principal Component Analysis was performed.

**Table 2. Normalised chemical results of the amphora samples obtained by XRF. Concentrations of major and minor oxides are in %, other minor and trace elements are in ppm**

Sample	Fe <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	MnO	P <sub>2</sub> O <sub>5</sub>	TiO <sub>2</sub>	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	SiO <sub>2</sub>
MCF065	5.3	15.7	0.08	0.16	0.65	2.2	8.0	1.3	2.7	62.7
MCF066	4.7	14.9	0.12	0.15	0.68	1.7	5.9	1.8	2.6	66.0
MCF067	5.3	16.1	0.08	0.22	0.66	2.1	10.1	2.0	2.9	58.7
MCF068	7.8	18.0	0.15	0.36	0.88	4.2	10.1	2.0	3.1	54.0
MCF069	7.4	17.8	0.12	0.15	1.06	2.0	2.3	1.8	3.3	64.8
MCF070	7.8	17.9	0.15	0.37	0.88	4.3	10.8	1.7	3.0	53.3
MCF071	5.0	15.7	0.07	0.14	0.62	1.3	9.7	1.0	2.9	60.9
MCF072	6.5	17.1	0.08	0.08	0.90	1.4	2.4	1.0	3.1	67.6
MCF073	4.5	15.5	0.05	0.12	0.68	1.3	4.3	1.2	3.9	67.0
MCF074	6.6	18.3	0.09	0.12	0.90	1.8	3.4	1.6	3.0	64.8
MCF075	7.9	18.4	0.11	0.20	1.06	2.0	2.9	1.6	3.6	64.2
MCF076	7.4	18.0	0.12	0.15	1.04	2.0	2.4	2.1	3.2	65.3

Sample	Ba	Rb	Th	Nb	Pb	Zr	Y	Sr	Ce	Ga	V	Zn	Cu	Ni
MCF065	388	119	11	19	29	179	24	285	76	17	101	76	24	38
MCF066	460	147	14	21	26	231	37	265	98	17	88	75	28	30
MCF067	629	111	10	16	21	170	26	426	59	19	112	71	24	42
MCF068	767	130	16	30	33	212	30	437	107	21	174	106	34	53
MCF069	553	135	23	21	27	266	47	134	117	21	112	97	8	18
MCF070	854	129	17	30	55	213	30	484	103	22	176	97	38	53
MCF071	613	111	11	14	21	198	27	168	68	17	84	65	16	23
MCF072	582	123	14	20	26	294	33	109	78	21	122	80	24	41
MCF073	628	176	13	16	27	234	33	173	75	18	92	88	19	29
MCF074	550	124	16	19	27	251	40	125	92	22	103	92	10	24
MCF075	603	142	21	22	39	245	47	124	111	24	135	110	20	25
MCF076	574	138	29	22	26	273	48	139	146	22	113	101	8	18

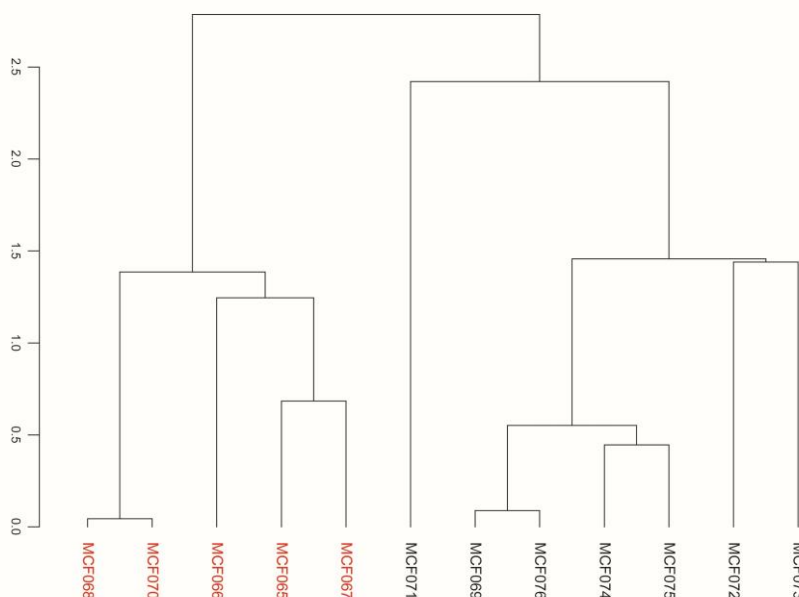


Figure 5. Dendrogram from cluster analysis (using the centroid agglomerative method and the squared Euclidean distance) on the  $\ln$  transformed subcomposition  $\text{Fe}_2\text{O}_3$ ,  $\text{MnO}$ ,  $\text{TiO}_2$ ,  $\text{MgO}$ ,  $\text{CaO}$ ,  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$ ,  $\text{SiO}_2$ ,  $\text{Ba}$ ,  $\text{Rb}$ ,  $\text{Nb}$ ,  $\text{Zr}$ ,  $\text{Y}$ ,  $\text{Ce}$ ,  $\text{Ga}$ ,  $\text{V}$ ,  $\text{Zn}$ ,  $\text{Ni}$  and  $\text{Cr}$ , using  $\text{Al}_2\text{O}_3$  as divisor. In red, Dressel 1B amphorae; in black, Tarraconense 1 and Pascual 1 amphorae

Figure 6 shows the best 2D projection, the horizontal and vertical axes respectively containing the first and second principal components, which represent the largest possible variance in the dataset (72.8% of the original variability). The first two principal components place most of the Dressel 1B amphorae from Sa Mesquida in two main groups constituted by amphorae from the shipwreck and other products from the central and southern Tyrrhenian coast of Italy (Figure 6). On the one hand, the two amphorae (MCF068 and MCF070) grouped in Fabric d are close to the  $\text{Fe}_2\text{O}_3$  and  $\text{MnO}$  arrows because they present higher concentrations in these two elements than the rest of the Dressel 1 amphorae examined. They also appear linked to the Lamboglia 2 amphorae of fabric L2B from Port-Vendres 4, for which an origin in the Gulf of Naples was established (Martínez et al., 2015). Moreover, all these containers are associated with the amphorae called “Eumachi Type” (TEUMCH), whose provenance has been accurately fixed at Pompeii (Thierrin-Michael, 2007).

On the other hand, MCF065 and MCF067 appear rather close because they are calcareous amphorae with low  $\text{Fe}_2\text{O}_3$  and  $\text{MnO}$  values and similar concentrations in some elements. However, as detected by thin section analysis, the two amphorae clearly correspond to two different products. The chemical composition of MCF065 resembles that of the Dressel 1B from fabric D1B and that of the Lamboglia 2 from fabric L2A identified in the cargo of the Port Vendres 4 shipwreck. Their provenance has been fixed in a marginal volcanic area of the Tyrrhenian coast (in

southern Tuscany, southern Latium or northern Campania), which is consistent with the data provided by the petrographic analysis. The chemical reference group attributed to Mondragone, in northern Campania, appears relatively close to all these amphorae.

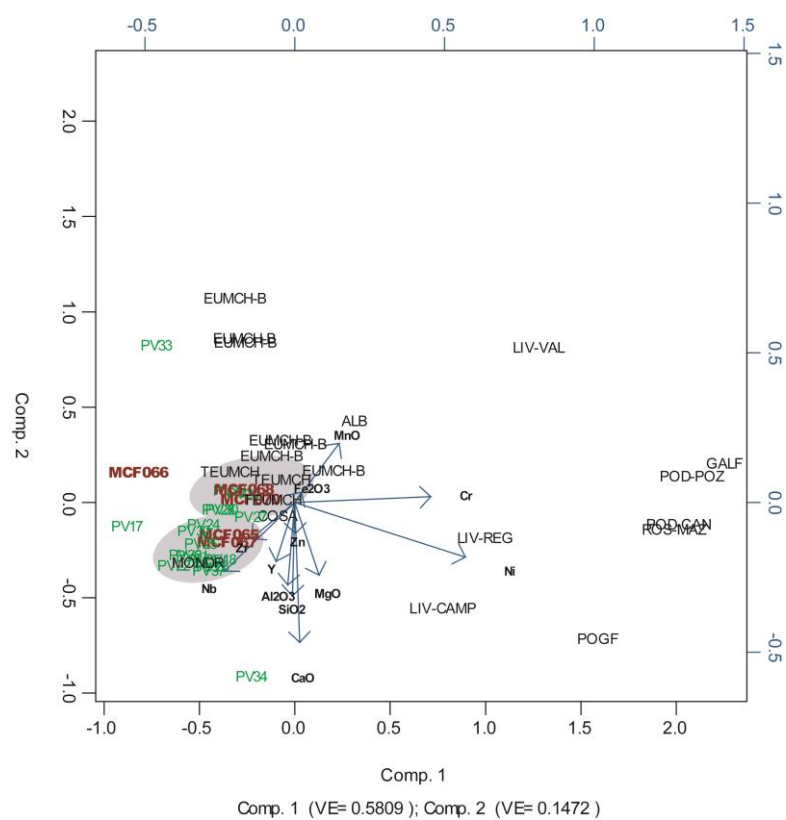
The amphora MCF066 exhibits the lowest  $\text{Fe}_2\text{O}_3$ ,  $\text{CaO}$ ,  $\text{V}$ ,  $\text{Ni}$  and  $\text{Cr}$  concentrations. It was produced in a different area in the Tyrrhenian coast although its provenance cannot be established by using the limited comparative chemical data available. Moreover, it should be noted that all the Dressel 1 amphorae analysed from Sa Mesquida significantly differ of the products from the northern Tuscany, which exhibit higher  $\text{Ni}$  and  $\text{Cr}$  contents.

In order to identify the specific pottery workshops or production areas of the Pascual 1 and Tarraconense 1 amphorae from Sa Mesquida, a PCA was performed on these and the reference groups from twenty different workshops in Hispania Citerior-Tarraconensis selected for comparison (Martínez Ferreras 2014; Vila 2011). The subcomposition  $\text{Fe}_2\text{O}_3$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MnO}$ ,  $\text{TiO}_2$ ,  $\text{MgO}$ ,  $\text{CaO}$ ,  $\text{K}_2\text{O}$ ,  $\text{SiO}_2$ ,  $\text{Ba}$ ,  $\text{Rb}$ ,  $\text{Nb}$ ,  $\text{Zr}$ ,  $\text{Y}$ ,  $\text{Ce}$ ,  $\text{Ga}$ ,  $\text{V}$ ,  $\text{Zn}$ ,  $\text{Ni}$  and  $\text{Cr}$  was first log-ratio transformed using  $\text{SiO}_2$  as divisor. Figure 7 shows the biplot of the first two principal components which account for 75.8% of the original variability. In this graph, the Tarraconense 1 (MCF075) and two Pascual 1 (MCF069 and MCF076) amphorae appear close to the low calcareous reference groups MOR-2 and MOR-3 from El Moré workshop, located in the territory of the Roman city of *Iluro* (Mataró). These

amphorae exhibit high  $\text{Fe}_2\text{O}_3$ ,  $\text{SiO}_2$ , Y, V and Zn content and low MgO and Ni values.

The Pascual 1 amphora MCF074 presents a certain similar chemical composition to the reference group ACM-B3 from Ca l'Arnau workshop, also located in the territory of *Iluro*. This group is characterised by low CaO and MgO values and high  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  contents; the chemical and petrographic composition of these amphorae resemble those from El Moré and therefore, the provenance of samples MCF069, MCF074, MCF075 and MCF076 should be established in the territory of the Roman city of *Iluro*. MCF071 matches with the reference group LLA-A from Llafranc workshop, placed in the territory of

*Emporiae*, in the northern coast. These amphorae exhibit calcareous pastes with low concentrations of MgO,  $\text{TiO}_2$  and Ce (Vila *et al.*, 2005). The Pascual 1 amphorae MCF072 and MCF073 exhibit low CaO and MgO contents and other compositional differences with the rest of the Pascual 1. They cannot be attributed to any of the reference groups considered in this study and, therefore, it is probable that they were not produced in any of the pottery workshops characterised so far and evaluated here. According to the petrographic examination, the latter could be associated to the productions of the Vallès Oriental area, where no pottery workshop has been completely characterised yet.



**Figure 6.** PCA of the *alr*-transformed chemical data for the amphora samples. Biplot of the two first principal components considering the compositional data of the Dressel 1B amphorae from Sa Mesquida (MCF) and the reference data regarding Italian amphorae



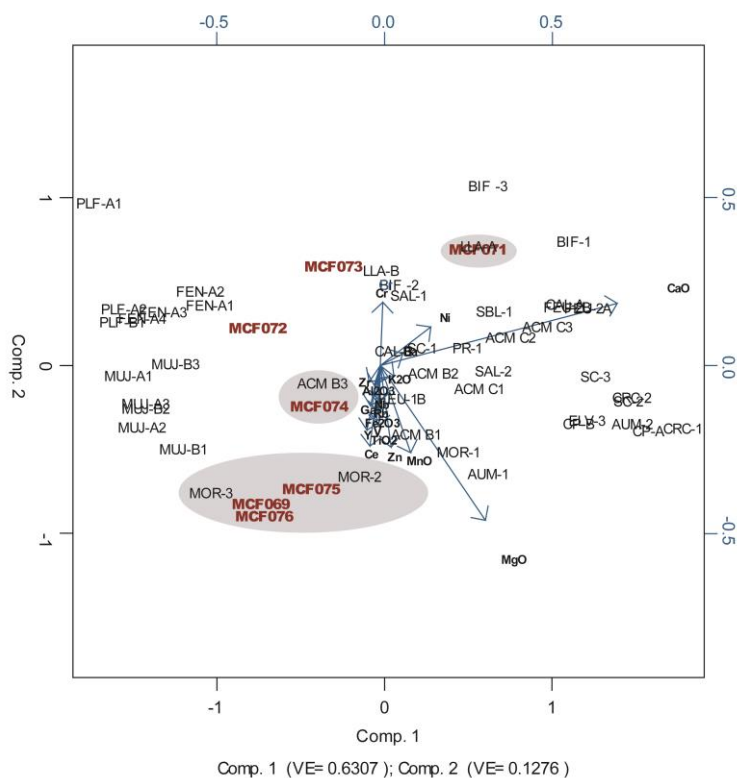


Figure 7. PCA of the alr-transformed chemical data for the amphora samples. Biplot of the two first principal components considering the compositional data of the Tarraconense 1 and Pascual 1 amphorae from Sa Mesquida and the reference data regarding wine amphorae in north-eastern Spain

### 3.3. XRD mineralogical analysis

The XRD analysis of the 12 amphorae from Sa Mesquida has enabled the identification of some technological processes of their manufacture. In addition to the information on the mineralogical composition of the amphorae, it has been possible to obtain an estimation of the equivalent firing temperatures (EFT) (see Roberts, 1963; Maggetti, 1982; Cultrone et al., 2001; Buxeda and Cau, 2004; Maggetti et al., 2011), as well as to determine that the firing atmosphere was mainly oxidising in all the samples (Table 3).

#### 3.3.1. Dressel 1B amphorae

The compositional differences observed in both the chemical and the petrographic analysis between the five samples of Dressel 1B analysed, are also associated with differences in the mineralogical phases identified through XRD. The samples MCF068 and MCF070, which exhibit the same chemical-petrographic composition, present slightly diverse mineralogical phases due to differences in their firing temperatures (Figure 8). MCF070 contains primary crystalline phases only (quartz, plagioclase, clinopyroxene, K-feldspar, sanidine, illite-muscovite, and few calcite), and hematite, suggesting an EFT under 800/850°C. In MCF068, calcite and illite-muscovite are totally decomposed, while the peaks

of plagioclase and K-feldspar show higher intensity, and those of sanidine and clinopyroxene present a lower intensity than in MCF070; the EFT for MCF068 would be  $\geq 950$ -1050°C. The diffractograms of the samples MCF065, MCF066 and MCF067 show the presence of primary phases only, with minor differences between them. They all contain quartz, illite-muscovite, calcite, plagioclase, K-feldspar and hematite (less in MCF067), in addition to clinopyroxene (MCF065, MCF066) and sanidine (MCF065). The estimated EFT for these individuals should be  $\leq 800$ /850°C.

#### 3.3.2. Tarraconense 1 and Pascual 1 amphorae

The calcareous amphora MCF073 exhibits only primary mineralogical phases such as quartz, K-feldspar, plagioclase, illite-muscovite, calcite, and also low hematite; the firing temperature should be estimated around  $\leq 800$ °C (Figure 8). The diffractogram of the border calcareous amphora MCF071 presents the same phases in addition to gehlenite and few diopside, which are considered firing phases. Illite-muscovite and calcite appear decreased while hematite is increased suggesting an EFT around 800-900°C. The rest of the amphorae are low calcareous and their diffractograms show an evolution of the phases according to the firing temperature. The EFT for the Tarraconense 1 amphora

(MCF075) should be fixed around 700-800°C since it contains primary phases only (quartz, K-feldspar, plagioclase, muscovite) and few hematite. In the diffractograms of the Pascual 1 amphorae MCF069 and MCF076 spinel is detected as a firing phase, hematite increases and illite-muscovite decreases. Therefore, the firing temperature has been estimated

around 900-950°C. This tendency is even more pronounced in the amphora MCF074, in which spinel is very intense suggesting an EFT of 950-1000°C. In the diffractogram of the amphora MCF072 illite-muscovite have disappeared and spinel has increased, what indicates an EFT between 1000-1100°C.

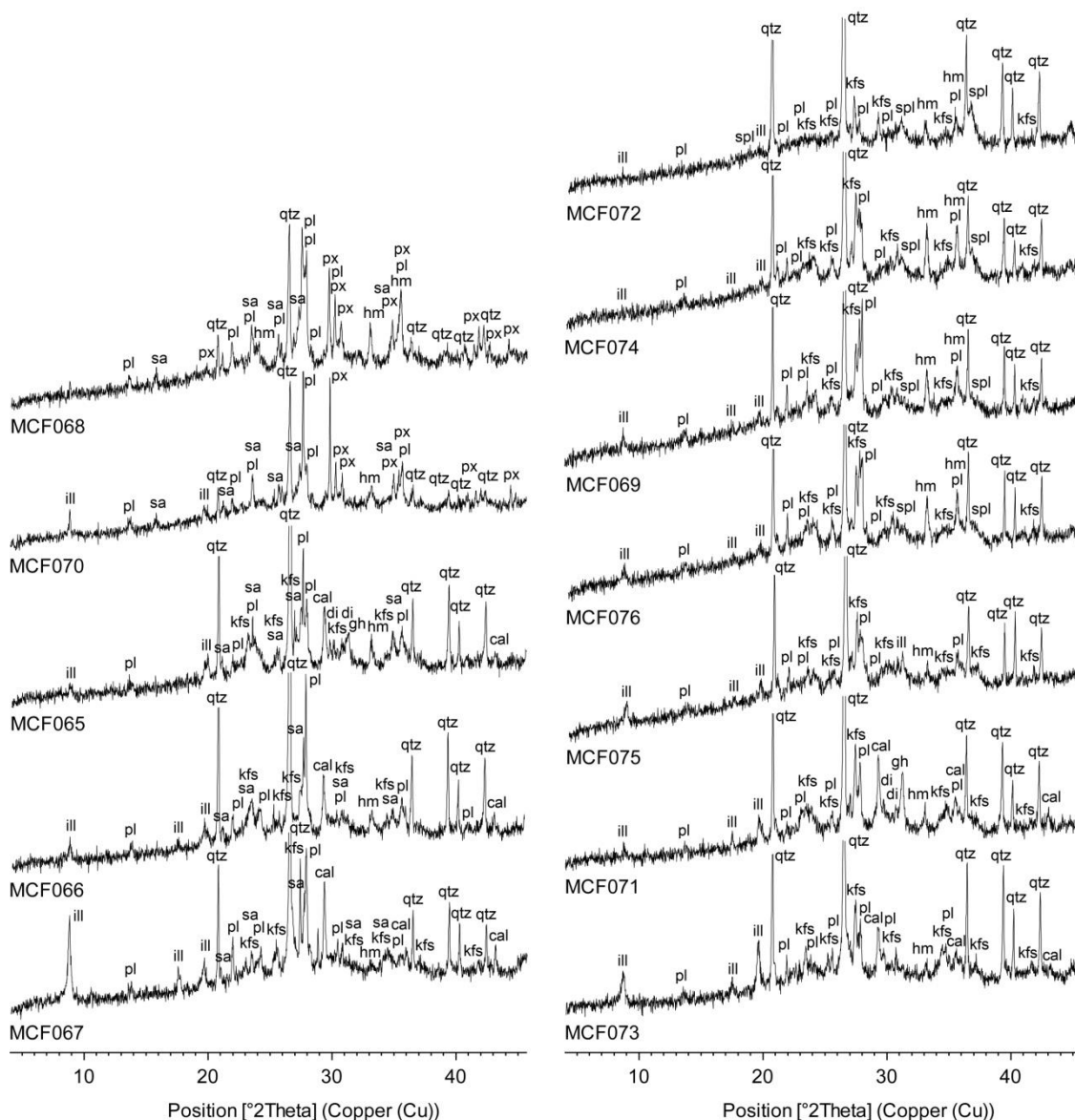


Figure 8. Diffractograms of the amphorae from Sa Mesquida; cal= calcite; px= pyroxene; di= diopside; gh= gehlenite; hm= hematite; ill= illite- muscovite; kfs= K-feldspar; pl= plagioclase; qtz= quartz; sa= sanidine; spl= spinel

### 3.4 Organic residue analysis of Dressel 1B amphorae

The chromatograms obtained through the analysis of the total lipid extract and the hydrolyses of the five samples indicate that *Pinaceae* products are present in all of them (Table 4; Figure 9).

In fact, dehydroabietic and 7-oxodehydroabietic acid are present in all the samples (Mills and White, 1977; Colombini *et al.*, 2005). In the hydrolysis of the

total lipid extract of sample MCF065 there is also methyl dehydroabietate, which is considered as a marker of pitch obtained directly from wood (Colombini *et al.*, 2005). These data indicate that all the amphorae were lined with *Pinaceae* products, that was surely pitch in the case of MCF065. This is consistent with the data obtained with the analyses of almost all the amphorae analysed until now in other case studies, regardless their content (Romanus *et*

al., 2009; Pecci and Cau, 2010, 2014; Pecci et al., 2010a, 2010b, 2016; Garnier et al., 2011).

As for the content of the amphorae, the analysis of the extract obtained following Pecci et al. (2013) (extraction c) allowed for the identification of tartaric acid, in four out of the five samples of Dressel 1B analysed (all but sample MFC070). Although other fruits, such as tama-

(Barnard et al. 2010), the scarce diffusion of these products in the investigated area, suggests that tartaric acid can be considered marker of grape (Guash Jané et al., 2004; Pecci et al., 2013, 2017; Garnier and Valamoti, 2016). The presence of succinic, malic, and fumaric acids is also compatible with the presence of wine.

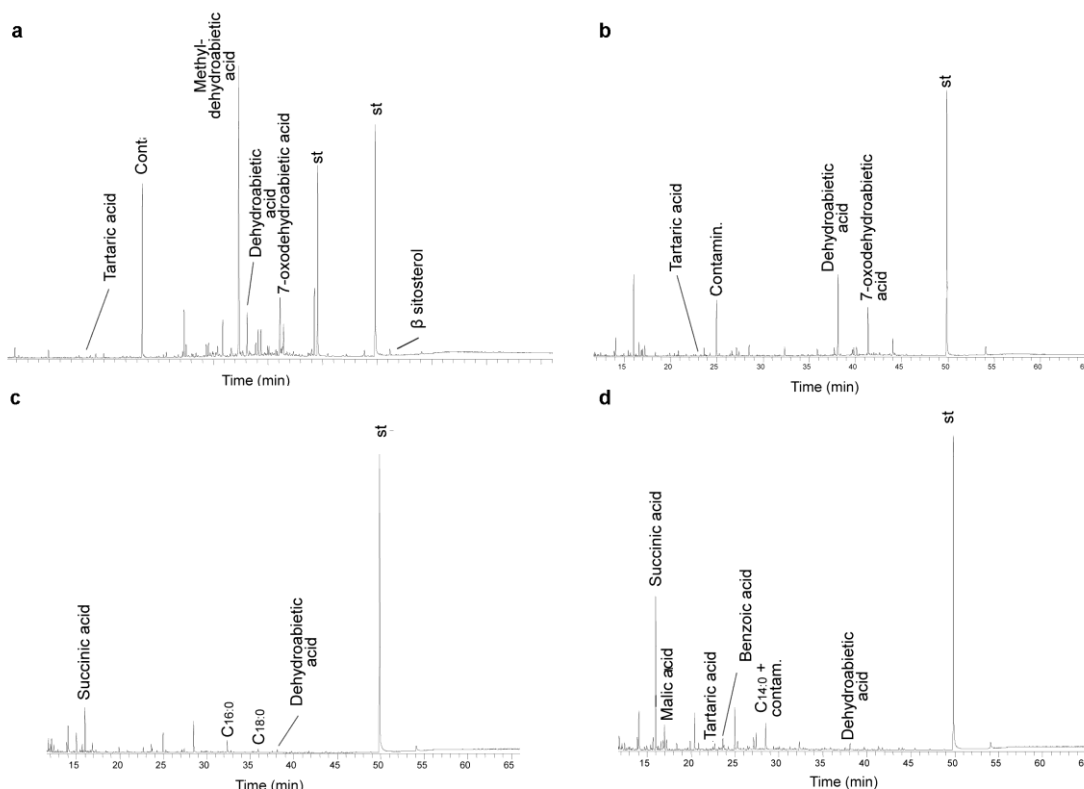


Figure 9. Chromatograms resulting from the organic residues analysis on Dressel 1B amphorae. (a) MCF065, hydrolysis of the total lipid extract. (b) MCF065 extract for the identification of wine residues; (c) MCF070, extract for the identification of wine residues (d) MCF066, extract for the identification of wine residues

Table 4. Samples and substances detected with the organic residue analysis

Sample	SU	Sampled part	Type	Pinaceae products	tartaric acid	succinic acid
MCF065	3075	Spike	Dressel 1?	pitch	x	x
MCF066	3075	Rim with handle	Dressel 1B	x	x	x
MCF067	3075	Rim with handle	Dressel 1B	x	x	x
MCF068	3075	Rim	Dressel 1B	x	x	x
MCF070	3073	Rim with handle	Dressel 1B	x		x

The analysis of the sediment of sample MCF068 indicate that there is no tartaric acid in the sample, confirming that it comes from the amphora content. As for sample MCF070, this is the only individual in which no tartaric acid is present. Here the succinic acid might suggest that the amphora contained a fermented beverage different from wine (Garnier and Valamoti, 2016).

#### 4. CONCLUSIONS

The archaeometric characterization of 12 amphorae of types Dressel 1B, Tarraconense 1 and Pascual 1 found in Sa Mesquida has allowed for a better identification of the provenance of these ceramics as well as for obtaining a first insight into their technology of production.

On the one hand, it has been possible to propose a provenance in the Tyrrhenian coast of Italy for all the Dressel 1B amphorae analysed. However, various production areas have been identified including southern Lazio or southern Tuscany (MCF065, MCF066), Campania (MCF068, MCF070) and the Strait of Messina – Calabria or north-eastern Sicily – (MCF067).

With regard to the Tarraconense 1 and Pascual 1 amphorae, the analysis points to a provenance in the north-eastern Iberian peninsula for all the samples, a variety of productive areas being represented in the analysed assemblage. Based on the chemical and petrographic composition, four amphorae of both types (MCF069, MCF074, MCF075 and MCF076) should be attributed to different pottery workshops located around the Roman city of *Iluro* (Mataró). Production of these amphorae has been attested in *figlinae* such as Ca l'Arnau (Cabrera de Mar), Can Portell and Can Notxa (Argentona) from the third or second quarter of the 1st century BC and at El Moré (Sant Pol de Mar) from the Augustan period. They consist in low-calcareous amphorae with coarse aplastic inclusions derived of igneous rocks (granitoids).

A Pascual 1 amphora (MCF071) seems to have been produced in one of the northernmost production centres of the Catalan coast. Specifically, it matches with one of the amphora groups from Llafranc (Palafrugell), where pottery manufacture started in the Augustan period. Amphorae produced in this workshop are characterized by clay pastes rich in iron oxides with an important carbonate component (due to the presence of foraminifera microfossils), along with large fragments of granitic rocks. No evidence on the specific provenance of the Pascual 1 amphorae MCF072 and MCF073, from the SU 30022 and SU 3005 respectively, was found. However, according to the petrographic characteris-

tics, a provenance in the central-northern area of the Catalan coast can be suggested.

The results of the organic residue analysis support the archaeological hypothesis related to the presence of wine in the Dressel 1B amphorae found in Sa Mesquida. Only one of them (MCF070) did not show evidence of wine, and could have contained another fermented substance. Residues related to the presence of *Pinaceae* products have been identified in all the samples; these products were likely used as an organic coating of the amphorae.

The information obtained from the archaeometric characterization of amphorae Dressel 1B from Italy, which showed wine markers, and Tarraconense 1 and Pascual 1 from the Hispania Tarraconensis province, which have always been considered wine amphorae, suggests the arrival of wine from various regions to the Roman villa of Sa Mesquida, during the same period. Most probably, the import of these amphorae from the wine production areas was not made directly, but through some port of the island where these products arrived from several parts of the Empire and were then redistributed between the settlements that occupied Mallorca in the Early Imperial period.

The combination of inorganic and organic analyses proves to be a good approach in order to tackle provenance and content studies at once. In addition, the existence of large comparative databases, including the reference groups for workshops in different regions, is essential to better define the provenance of the materials received in consumption centres. In this case, even with a relatively small sample of amphorae, this pattern association allows for a much better understanding of the provenance of the amphorae that arrived to this Roman settlement in the island helping to reconstruct the economies of the ancient Mediterranean.

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