

Vega-Maeso Cristina (ORCID ID: 0000-0003-3641-8815)

Gallelo Gianni (ORCID ID: 0000-0003-3641-8815)

CERAMIC PRODUCTIONS AND HUMAN INTERACTIONS DURING THE EARLY BRONZE AGE IN NORTHERN IBERIA.

Cristina Vega Maeso¹

Gianni Gallelo^{2,3}

Susana Palmero⁴

Begoña Ferrari⁵

Miguel Ángel Sánchez Carro⁶

Manuel González Morales⁷

Igor Gutiérrez Zugasti⁷

Mirco Ramacciotti^{2,8}

Agustín Pastor⁷

¹ Facultad de Humanidades y Ciencias Sociales. Universidad Internacional Isabel I de Castilla. C/ Fernán González, nº 76, 09003 Burgos. Spain.

² Department of Prehistory, Archaeology and Ancient History, University of Valencia, Avda. Blasco Ibáñez, 28, 46010, Valencia, Spain.

³ Department of Archaeology, University of York, King's Manor, York, YO17EP, UK.

⁴ Facultad de Ciencias. Universidad de Burgos. Pza. Misael Bañuelos s/n.09001 Burgos.

⁵ Instituto de Cerámica y Vidrio, CSIC, c/ Kelsen 5, 28049, Madrid.

⁶ Dpto. Ciencia e Ingeniería del Terreno y de los Materiales. Universidad de Cantabria. Avd. de los Castros s/n, 39005, Santander.

⁷ Instituto Internacional de Investigaciones Prehistóricas de Cantabria (Universidad de Cantabria, Gobierno de Cantabria, Grupo Santander), Universidad de Cantabria, Edificio Interfacultativo, Avda. Los Castros s/n. 39005. Santander, Spain.

⁸ Department of Analytical Chemistry, University of Valencia, 50 Dr. Moliner Street, 46100 Burjassot, Valencia, Spain.

⁹ Servicio de Innovación y Difusión. Dirección General de Patrimonio Cultural. Consejería de Cultura y Turismo. Junta de Castilla y León.

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process which may lead to differences between this version and the Version of Record. Please cite this article as doi: 10.1111/arc.12605

ABSTRACT

The Early Bronze Age ceramic collection found into the caves of La Llana and El Toral III in Asturias (Spain) presents common decoration like those found in the centre of Cantabrian Spain from the same period, which resemble others found in the Ebro valley and Atlantic Europe. Therefore, the main objective of this work it is to identify the raw material origin and understand the pottery production process during the Early Bronze Age in Cantabrian region. A methodological approach based on the chemical and mineralogical analysis of vessels and experimentally fired clay samples collected all over the centre of this region was developed. Furthermore, the post-depositional processes affecting the sherds composition have been evaluated employing the rare earth elements (REE) as markers. The results showed that the studied assemblage have important similarities with the raw materials of the surrounding area, which support the hypothesis of a regional mobility.

KEYWORDS

Pottery, Chemical-mineralogical characterisation, rare earth element, raw material, post-depositional processes, human mobility

1. INTRODUCTION

During the 3rd millennium BC the archaeological phenomenon called Bell Beaker, spread all over the Western Europe and the Northern Africa. Multiple studies have been carried out during the last decades to better know the nature of this archaeological culture especially identifying the provenience of ceramic raw materials. The analyses carried out in Western Europe by Salanova et al. (2016) employing different analytical techniques such as X-ray diffraction (XRD) and thin-section petrographic analysis showed that most of the studied ceramics were made from local raw materials, indicating that there was a low degree of ceramic circulation at a regional and extra-regional scale. Following the same research line, some works showed that the pottery assemblages of Neolithic and Chalcolithic in the Cantabrian region were compatible with the clay sources found in the surroundings of the sites (Vega Maeso 2012; Cubas 2013; Smith et al. 2014). Furthermore, the Beaker practice was related with population changes in Europe as confirmed by DNA studies (Szécsényi-Nagy et al. 2017; Olalde et al. 2018). But also, strontium isotopes data by Vander Linden (2007) showed that the mentioned population changes in Europe were probably related with marriage circuits.

Nevertheless, the Cantabrian region seems, relatively, isolated from the Bell Beaker cultural phenomenon. Thus, the beaker presence in this area could be defined as *“scarce, sporadic and marginal perhaps with the only exception of the region eastern corner”* including the Basque country area (Ontañón Peredo 2005). In fact, except for the Maritime-style Beaker sherds found in the Aramo mine in western Asturias (Blas 20015), the “early styles” are limited to the easternmost part of the region. In this area the corded-zoned Maritime variety (CZM) vessels are associated with megalithic burials and the All Over Corded (AOC) would be associated with non-burial sites (Ontañón Peredo 2003b). There is no ceramic evidence of this global phenomenon in the centre of Cantabrian region where only some “beaker metalwork “and ornaments have been found.

The archaeological record shows that while the Bell Beaker has evolved into the so-called ‘regional’ or ‘epi-Bell Beaker’ styles, in the centre of the Cantabrian region a similar ceramic style has also been documented. This pottery style, traditionally known as “Trespando”, is characterised by a decoration pattern based on small incisions or impressions combined with incised lines (Arias Cabal et al. 1986; Arias Cabal and Armendariz Gutiérrez 1998; Toledo Cañamero 1999) which present carinated, truncated conical or hemispherical forms. These fine vessels, usually are found high fragmented together with coarse smooth bowls and storage jars and are associated with funerary contexts using caves as ceremonial space (Vega Maeso 2017). Due to their characteristics this pots could be associated to the Pont-Long group in the north-western Pyrenees (Marembert 2000) or to the materials attributed to the North-east Arbolí group in the Ebro valley and Catalonia (Maya and Petit 1986).

Therefore, in order to shed light on the human interaction dynamics in the centre of Cantabrian region during the Early Bronze Age, the main objective of this work is to identify the provenance of the raw materials and understand the manufacturing processes of the ceramic objects found at La Llana and El Toral III caves.

For this purpose, a methodological approach was developed sherds collected in the two sites (La Llana and El Toral III) and clays found in different outcrops also located in the studied region have been sampled. Then, petrographic, mineralogical and elemental analyses were carried out, and the obtained data were finally processed by multivariate statistics. The results have been cross-referenced with archaeological data for solving provenance issues, and post-depositional processes have been taken into account to avoid misinterpretations, testing for the first time the capability of rare earth elements (REE) as markers of ceramic diagenesis processes taking action during their burial.

1.1 Geological characteristics of the area surrounding the sites

The sites of La Llana and El Toral III are located in Andrín (Llanes, Asturias, northern Spain) nearby a coastal landscape characterized by dunes, beaches, cliffs, and small estuaries where short rivers flow. The main feature of the surrounding relief is the presence of a perched marine abrasion surface, affecting between 1 and 5 km inland, at an altitude between 20 and 300 metres above the sea level (a.s.l.), locally known as '*rasa asturiana*'. This surface has undergone river erosion and processes of karstification in the areas where limestone outcrops (Flor 1983) (Figure 1).

Ordovician to Lower Cretaceous rocks (Figure 1) are present in the surrounding area of the studied caves (El Toral III and La Llana) while Quaternary materials have also a relative importance and Tertiary levels are located about 10 km east from the sites.

The bottom of the sequence consists of Ordovician quartzitic rocks. A terrigenous Devonian sequence was deposited over these, composed of sandstones, conglomerates, clays, and coal levels. Carbonate rocks were deposited during the Carboniferous and contain red nodular limestones with radiolarites, dark laminated limestones and white bioclastic limestones. Cretaceous materials deposited during the Aptian and the Albian ages are represented by an alternation of limestones and sandstones in discordant contact with the Paleozoic units. The carbonate units consist of biomicritic limestones, and the detrital units of sandstones, silts, clay, and sandy clay with coal remains (Instituto Geológico y Minero de España 1981).

Regarding the Quaternary and Tertiary deposits, it is worth noting that one of the most important geomorphological characteristics is the aforementioned perched marine abrasion surface. This flat surface was formed during the Oligocene coastal erosion (Flor and Flor-Blanco 2014) and uplifted by tectonic deformation during the Miocene and Pleistocene periods. In the studied area, Tertiary deposits, from the Middle and Lower Eocene, are represented by sandy limestones, dolomites, and limolitic or sandy marls. Quaternary deposits are instead associated with Ordovician quartzite outcrops and decalcified clay deposits in karstic depressions, talus cone and beach deposits

1.2 Archaeological context

La Llana and El Toral III sites are two significant examples of Bronze Age sites in the Cantabrian region, where surface contexts without stratigraphy are common. Near to

these sites, there are others with similar ceramic assemblages: La Cabañina, Trespando, Bricia, El Bufón and Arangas (Martínez Santa-Olalla 1930; Arias Cabal et al. 1986; Arias Cabal and Pérez Suárez 1995; Rasilla Vives 2011). The pottery set from La Llana had already been included in the original group of “Trespando” (Arias Cabal et al. 1986), but not El Toral III one, unknown at that time. Specifically, the shapes and decorations of the Asturian assemblages have a special resemblance to the ceramics recovered from the Bay of Santander. In this area, carinated vessels with decorations based on incised zig-zag lines are also common in the collections of El Mapa and El Ruso (Ruiz Cobo and Serna González 1990). Furthermore, we have found the same decorative syntax of one of the vessels of La Llana in the ceramic assemblages of other sites, such as Cubrizas, El Abrigo de la Castañera and El Pendo (Morlote Expósito 2001; Vega Maeso 2017).

Despite the small size of the pottery collections, their importance is due to the fact that those materials, were well recorded during the excavations and their surface distribution well identified. Therefore, we know both their position and their relationship with the other elements of the sites.

1.2.1 El Toral III

In 2009, an emergency excavation was performed in El Toral III. The site was divided in two areas: a southern area, called Zone A, and a northern one, called Zone B. Differences between the two zones are related to the human activity impact, the formation conditions and the erosion processes. Thus, Zone A has a complex stratigraphic sequence due to water circulation in the area, while Zone B was silted up by over a metre of highly fragmented stone material (Noval Fonseca 2013).

Most of the potsherds were recovered in Zone B at the surface level together with other archaeological materials such as remains of fauna, shells, lithic artefacts, ornamental elements (v-perforated button) and human skeletal remains with different radiometric dates. The archaeological evidence suggests that the cave was used as a funerary pantheon from the Mesolithic to the Bronze Age. (Table S1)

The ceramic collection is composed of 32 handmade potsherds, grouped according to their technological and morphological characteristics, which allowed to distinguish a Minimum Number of Vessels of eight (Millett 1979) (Table S2). The typological studies have shown that most of the pieces were liquid transport and storage vessels (Vessels 2, 4, 5, 6 and 8), while the vessel 3 was classified as Trespando type.

1.2.2 La Llana

The cave of “La Llana” was excavated between 1983 and 1985 (González Morales 1995). There are two main areas where archaeological remains were identified: a small niche with a human skeleton (Zone A), and a Mesolithic shell midden deposit (Zone B).

During the excavation, on the surface of the Mesolithic shell midden, objects identified as waste of different kinds of technological processes (potsherds, a small metal piece, a polished wild boar tooth, and three bevel-cut bones) were found. However, no domestic *in situ* activities related to them have been identified (González Morales 1995). At La Llana, radiocarbon dates were obtained from the human skeleton, and from a fragment of bevel-cut bone. Both remains were dated to the middle of the second millennium cal BC. (Table S1)

The pottery from La Llana constitutes a homogeneous assemblage of 199 fragments: 188 sherds and 11 rims. A Minimum Number of Vessels of at least four has been identified (Table S2). Two of them (Vessels 1 and 2) present profuse incised decoration and so can be classified within the Trespando group. In addition, there are two more vessels: one small, non-decorated and almost complete container (Vessel 3) (Figure 2B.3) and another fragmented (Vessel 4) which could not have been typologically identified. The profile of Vessels 1 and 2 was reconstructed from the overlapping of several sherds (Figure 2B.1 and 2).

2. MATERIAL AND METHODS

2.1 The Samples

The sampling strategy and the multi-instrumental approach have been developed in order to provide reproducible and comparable results taking into account both typological and technological characteristics and the amount of available materials. Each ceramic sample (C) was identified as “T” or “LL” plus the number of vessel (n) the number of samples (n) and percentage of analysed fragments (Table S3). In addition, a set of 15 clay were sampled in different outcrops in the centre of the Cantabrian region to be compared with the pottery samples. These clays were identified as “A” and when fired “AC” (Table S3).

Fifteen experimental fired clay samples and eighteen samples from the ten studied vessels (four from Llana and five from El Toral III) have been taken to carried out Inductively coupled plasma mass spectrometry (ICP-MS) and X-ray fluorescence

(XRF) (Table S3). When possible (seven out of ten vessels), two different samples have been analysed to observe possible internal differences in their elemental composition (major elements, trace elements and rare earth elements, REE) in order to enhance the interpretation of the results. X-ray diffractometry (XRD) have been employed to determine the mineralogical profile of the ten vessels and the fifteen clay samples analysed twice, before (A) and after being fired (AC).

Petrographic studies are particularly interesting in archaeological ceramics to observe morphological characteristics of the clay and interpret manufacturing processes. However, an important part of the sherd need to be employed to make a thin section for carrying out these analyses. Therefore, four representative vessels of the studied collection were selected for petrographic analysis according to their typological and technological characteristics: LL4C1 (not-decorated pot), LL1C4 and T3C5 (decorated pots), and T4C6 (storage pot).

The selection of clays has been carried out following a preliminary study based on a model recently developed by Vega Maeso (2017). According to this model, the presence or absence of possible deposits is based on observations made employing the archaeological record and the physical data of the studied area. The Sextante program integrated into gvSig OA Digital Edition 2010 has been used to design the model. The parameters that have been used are the following:

- Predictors: Variables of physical type (elevation, slope or others derived from the combinations) and archaeological type variable (density of cave burial sites).
- Points of presence: Settlements in the study area
- Method: Average distance has been used (Shennan, 1992).

These places and Bronze age settlements have been taken as reference locations of clay sources, according to criteria established by the Site Catchment Analysis (García Sanjuán 2005).

The collected clay were experimentally fired at 700°C (AC) simulating the ceramics firing temperature estimated from the presence of calcite tempers (Fabbri et al. 2014).

2.2. Analysis

2.2.1 Mineralogical and Petrographic study

The full set of the studied samples were analysed by X-ray diffractometry (XRD), using a Siemens D5000 diffractometer 216 (Germany) with a generator KRISTALLOFLEX 710 ($K\alpha$ (Cu) $217 \lambda = 1.5405 \text{ \AA}$; 40 kV; 30 mA; $2\theta = 5\text{--}60$).

This technique allowed us to identify crystalline phases and estimate the firing temperature of the samples, based on the presence of neoformed minerals (García Heras and Olaetxea 1992). Identification and semi-quantification of crystalline phases were carried out using the computer software BRUKER's DiffraC.suite eva. Phase abundances are obtained applying the Reference Intensity Ratio (RIR) method.

The selected pottery were previously consolidated using an epoxy resin to made tin section in order to guarantee the integrity of the components. After consolidation, samples were lapped and polished to obtain 30 microns thickness, suitable for polarized transmitted light optical microscopy. The thin-sections were viewed under a Kiowa Biopol 2 microscope. The micrographs were taken with a Canon EOS 450D fitted to the trinocular.

For the petrographic description and interpretation of the thin-sections, some standards have been adapted focused on the description of the inclusions, the clay matrix and the voids. The mineralogical identification has been made addressing the main characteristics: colour, pleochroism, zonation, twinning, exfoliation, and extinction (Whitbread 1989; Perkins and Henke 2002; Melgarejo 2003; Quinn 2013).

2.2.2 Major elements and trace elements analysis

A representative amount of each sample was powdered and homogenised through an agate mortar and pestle, and directly analysed by using X-ray fluorescence spectroscopy (XRF) to obtain major elements concentrations. Instrument spectra were obtained using a portable model S1 Titan energy dispersive X-ray fluorescence spectrometer from Bruker (Kennewick, Washington DC, USA) equipped with a Rhodium X-ray tube and X-Flash® SDD detector. For instrument control S1RemoteCtrl (Geochem-trace programme) and S1Sync software from Bruker were employed to measure the percentage of Al_2O_3 , SiO_2 , CaO , Ti , Fe and the ARTAX software from Bruker was used for spectra treatment.

Inductively coupled plasma mass spectrometry (ICP-MS) has been carried out for trace elements analysis. The total sample digestion was developed following the method reported by Ramacciotti et al.(2019)

The prepared dilutions were analysed by ICP-MS with Perkin Elmer Elan DRCII (Concord, Ontario, Canada). Calibration standards concentrations ranged between 1 and 600 µg/L for all analyzed trace elements Rh was used as internal standard for ICP-MS analyses and soil NIM GBW07408 was used as standard reference material.

2.2.3 Data analysis

Statistical analysis was not carried out on the full sample set since the outliers (AC3, AC4, AC7 y AC8) were not included. Major elements (Al_2O_3 , SiO_2 , CaO, Ti, Fe), trace elements as Ba, Bi, Cd, Cr, Co, Cu, Pb, Li, Mn, Mo, Ni, Sr, Tl, V, Zn, U, Th, REE, Sc and Y, were used as variables for principal component analysis (PCA) modelling. 28 samples and 38 variables (Major elements, trace elements, REE, Sc and Y) have been employed to run PCA.

Data analysis was carried out using the PLS Toolbox 8.2 for Eigenvector Research Inc., (Wenatchee, WA, USA) running in Matlab R2016b from Mathworks Inc., (Natick, MA, USA).

The normalised ratios of La_{PAAS}/Yb_{PAAS} , La_{PAAS}/Gd_{PAAS} , La_{PAAS}/Sm_{PAAS} and Sm_{PAAS}/Yb_{PAAS} were used to determine relative enrichment or depletion of Light REE (La-Nd, LREE), Medium REE (Sm-Ho, MREE), and Heavy REE (Er-Lu, HREE). Ce and Eu anomalies were used to evaluate differences between measured concentrations of Ce or Eu and the concentration expected based on neighbouring REE. The anomalies were calculated following the equations recently employed by Gallelo et al. (2019):

$$\text{Ce anomaly} = 3C_{ePAAS} / (2L_{aPAAS} + N_{dPAAS})$$

$$\text{Eu anomaly} = E_{uPAAS} / (S_{mPAAS} \times G_{dPAAS})^{1/2}$$

PAAS subscripts indicates Post-Archean Australian Shale normalised values (Taylor and McLennan 1985)

3. RESULTS

3.1. Mineralogical characterisation

The mineralogical results showed crystallographic similarities among the analysed samples (Table 1). In fact, all of them present quartz (29% to 87%), aluminosilicates of K and Na, such as orthoclase and albite (7% to 22%), and illite as the only clay mineral.

However, a small amount of kaolinite was detected in some of the clays "A" that disappeared after firing.

Calcite amount, and the presence of iron oxides and calcium carbonates are marking the main differences among the studied samples. Most of the archaeological samples were characterised by high amounts of calcite. It is over 40% in LL1C4, T4C6, T8C12 and T6C10, and ranges between 10% and 20% in LL4C1, LL3C2, LL2C3 and T3C5. Only two samples, T1C7 and T7C11, have an amount of CaCO_3 similar to the clay samples. It must be noticed that the presence of calcite in most of the clay samples was not detected.

In clays, the iron minerals documented were different: jacobsonite in sample A4 and AC4; pyrite in AC2 and AC3 and cubanite in AC6. In addition, AC3 also contains barite. However, in the archaeological samples only iron oxides were identified, normally magnetite except in two samples from El Toral III where hematite was reported (T7C11 and T8C12). Magnetite was observed in clay samples A2, A3, A11, A14 and A15. In all the cases, except for A14, it is transformed into hematite during firing, probably because the experiment was conducted in an oxidising atmosphere.

A further interesting issue is the presence of different calcium carbonate compounds in some studied samples. A small percentage of $\text{CaMn}(\text{CO})_3$ was found in T1C7 while in LL1C4, T4C6, LL3C2 and in clay samples A14, A5 and A4, $\text{CaMg}(\text{CO})_3$ was found.

The petrographic analysis did show large differences among the samples. All of them have grog tempers, quartz, opaque minerals and organic matter. None of the clay matrices showed evidence of vitrification, since they have an important index of birefringence, which reveals that they were fired at low temperatures. Although all the archaeological samples are characterised by a remarkable porosity there are differences that need to be highlighted: samples LL1C4 and T4C6 have a large amount of limestone with angular morphology among the non-plastic inclusions, (Figure 3A, 3B, 3I, 3J, 3K and 3L). In sample T3C5 carbonate with rounded morphologies and small muscovite crystals together with tempers common to all of them were observed. Grog seems to be the only material added to the clay matrix. (Figure 3C). As for sample LL4C1, among the non-plastic inclusions, there are sporadic clasts of limestone and other carbonates (Figure 3E and 4F) with an angular morphology that were added to the clay matrix (Figure 3G and 3H). Samples T4C6 and T3C5 have a large amount of secondary limestone in their pores (Figure 3D).

3. 2. Chemical elements

The chemical analysis confirms what was observed in the petrographic and mineralogical studies. There are significant differences in the majority of elements, especially in Al_2O_3 , SiO_2 and CaO . In fact, this last element is over-represented in some archaeological items. This over-representation was observed above all in the samples taken from storage vessels whose thin section showed the existence of large limestone tempers, something that is also confirmed by mineralogical and macroscopic examination. However, LL1C4 did not stand out particularly for CaO in the chemical analysis, although a significant amount of calcite was also observed. On the contrary, T7C11 y T7C18 showed significant concentration of CaO , despite no calcite was detected. CaO amount ranges between 0.06% and 1.5% in the clay samples, with the exception of sample A13 where a value of 9.09% was measured. In the case of the archaeological materials, CaO showed concentrations higher than 1.8% (Table S4). If we also observe trace elements and REE results, the differences are greater. To interpret these differences a statistical treatment of the data is necessary. In order to avoid biased results, the outlier values A3, A4, A7 and A8 were omitted.

According to the PCA, eight components explain 90% of the variance, with eigenvalues with values over 1. Through a biplot is possible to see that the first two principal components account for over 56% of the variance in the data. Three groups can be identified by comparing the clay samples and the archaeological ones (Figure 4). One cluster is composed only of clays (AC1, AC2, AC5, AC6, AC9, AC10, AC11 and AC12). On the other hand, samples T1C7, LL1C4, T4C6 and T3C5 would form another group. The final group includes most of the archaeological samples and the clays closest to the sites: AC13, AC14 y AC15.

As mentioned in the previous section, REE values were normalised to Post-Archaean Australian Shales (PAAS) (Taylor and McLennan 1985). The REE/PAAS profiles were reflecting the variations in REE concentrations listed above. The storage vessels show a strong enrichment in LREE over HREE, while it is possible to observe depletion of LREE over MREE in samples from La Llana and some from El Toral III. On the other hand, samples T7C11 and T7C18, the only samples from Zona A of El Toral III, show

a strong enrichment of MREE over HREE, while in T4C6, T3C16 y T1C7 the enrichment of these elements is lower (Table S5).

The positive anomaly of Eu is present in all the archaeological and natural samples, although the intensity is different (Table S5. Figure S1). This must be related to the presence of aluminosilicates, as the positive Eu anomaly is associated with sediments that contain feldspars, especially plagioclases (Prajith et al. 2015).

Most of the samples present a small negative Ce anomaly (Table S5 and Figure S1). However, there are some exceptions like AC11, AC6, both decalcification clays linked to Lower Cretaceous limestones. In the case of ceramics, Ce anomalies seem to be related to the amount of CaO.

4. DISCUSSION AND CONCLUSIONS

4.1. Manufacture and post-depositional process

Archaeological ceramics are the result of a combination of production and post-depositional processes that may modify their original clay composition.

The mineralogical analysis reveals that the samples were fired at a low temperature because no vitrification is observed in the ceramic matrix by optical microscopy or neoformed mineral by XRD. Another sign of low firing temperature is the presence of illite as the main clay mineral and limestone. Illite disappears between 800-900 °C (García Heras 1998) and limestone over 700-750°C (Quinn, 2013). Therefore, these pieces were fired in a reducing atmosphere, or at least a not completely oxidising one. XRD results of clays showed that magnetite, naturally present in some clays (A2, A3, A11, A14 and A15), becomes hematite when fired in an oxidising atmosphere. Hematite is only observed in two archaeological samples while the others present magnetite. Also, the dark colour of the thin-sections confirm a not completely oxidising environment during ceramic firing.

The human groups manipulated clay according to the function of the container they wanted to manufacture. This is perceptible through the grog that has been documented in all thin-sections studied, and in the limestone fragments. The use of calcite and grog as a temper is not new and is documented in the Cantabrian region since the Neolithic (Cubas 2012, Cubas et al. 2013) and, also during the Chalcolithic and Bronze Age (Olaetxea 2000; Smith et al. 2014).

Petrography has shown that the large amounts of calcite are not only natural components of the raw material used or an intentional addition, as in LL1C4, LL4C1

and T4C6, but also the result of a post-depositional process. Calcite is a mineral inclusion susceptible to be attacked by groundwater circulation or by the soil acidity (Buxeda i Garrigós and Cau Ontiveros 1995; Cau Ontiveros et al. 2002; Fabbri et al. 2014; Sánchez et al. 2016). This is the case of sample T3C5, in which some voids show the shape of the original inclusion that have been infilled with crystals of secondary calcite. These calcareous deposits are also visible on the surface of the sample T4C6 and XRD analysis revealed phases of modified calcite (Mg and Mn) in some samples (LL1C4, LL3 C2, T4C6 y T1C7).

In addition, when it was possible to conduct two chemical analyses of the same pot the obtained results were different probably due to the heterogeneity of the vessels related to both manufacturing and post-depositional processes (Schwedt et al. 2004). This can be observed in some major elements (CaO), in some trace elements (Ba, Bi, Cr, Co, Mn, Cu, Pb, Zn, Sr, V, Tl), and in REE ratios and Ce anomalies. Furthermore, T4C6 and T3C5 samples are associated by statistics with T1C7 and LL1C4 samples, which are characterized by a lower MREE over HREE enrichment and a positive Ce and Eu anomaly. These REE parameters results can be related to a limestone dissolution process, in fact, their characteristics are very similar to the REE and Y concentrations observed in the final step of an experimental sequential leaching of carbonate-rich sediments and rocks (Tostevin et al. 2016). This could be related with the amount of CaO found in T4C6, T3C5 and T1C7 that is below the mean, in fact, they are the lowest values of the whole set studied. Furthermore, these values do not agree with the amount of CaCO₃ observed in the XRD, which in the first two cases is higher than 40%. However, this may be associated with the results of the petrographic study of samples T4C6 and T3C5 that are affected by a process of dissolution of calcite and precipitation of secondary carbonates.

The strong enrichment of MREE over HREE of T7C11 and T7C18 are probably linked to water activity too. These samples were located in El Toral III, at zone A, where an important flow of water has affected the archaeological record. This pattern is similar to that exhibited by the soil pore water (Soyol-Erdene and Huh 2013).

4.2 Ceramic and clay provenance

The mentioned post-depositional processes have altered the chemical composition of the samples and in fact, this is reflected in the clustering distribution obtained in the PCA (Figure 4). However, there is a clear tendency to associate archaeological ceramics with the nearest clay outcrops, while the rest of the clay samples tend to cluster apart. This tendency is also visible in the distribution of two relatively

immobile elements in aqueous solutions such as uranium and titanium (Plant et al. 1999). Therefore, the samples distribution is just partially affected by post-depositional processes and some clay samples could be associated to the archaeological vessels (Figure 5). Also, XRD data showed similarities between vessels and the clay samples A14 and A15. Magnetite is documented in both samples and in A14 there is also a small amount of dolomite.

Petrographic analysis showed the existence of at least three different clay sources. However, the scatterplot Ti/U clusters the ceramic samples around four clay sources: AC13, AC14, AC15 and AC1, although the last one is more than 150 km away. Thus, T8C8 and T8C12 samples tend to be grouped with a sample, AC14, of Eocene origin from a clay outcrop 20 km away (Group 1). These results are reinforced by the geological data of the area confirming that Eocene materials emerge around 10 km away from the sites and even closer to some possible habitat areas.

Furthermore, petrographic analysis of LL1C4 and T4C6 shows characteristics that suggest similar production process and a common raw material. These samples may be associated to T1C7, T6C9, T6C10, and AC15 in the U/Ti scatterplot (Group 2). In addition, XRD analysis shows that the only iron mineral present in these samples is magnetite, and limestone is present in all of them, except for T1C7. In some cases, a small amount of modified calcite was found: $\text{CaMn}(\text{CO})_3$ in T1C7 and $\text{CaMg}(\text{CO})_3$ in LL1C4 and T4C6. These samples are grouped with Clay A15, from Carboniferous outcrops known as "Mountain Limestone".

Finally, Group 3 is composed by the samples LL2C3, LL2C13, LL3C2, LL3C15, LL4C1, T3C5, T3C16, T7C11, T7C18, AC1 and AC13. Although AC1 comes from a very far area, its geological origin is Lower Cretaceous. Since sediments from this chronology are common in this area this can explain why ceramics and clay samples are so similar. In the other hand, sample A13 is one of the closest but it is still more than 15 km away from the sites. This is also visible in the petrographic study, which shows that LL4C1 and T3C5 were made with different raw material. In addition, trace element results of LL4C1 reveals a high content of Zn and Pb, as in sample AC1. This could be related with the materials from the Cretaceous that outcrop in this area in which mineral related to these elements are common.

4.3. Behind the pots

The production dynamics for Trespando ceramics and other pots found in these contexts seem to be similar. All vessels were made using a simple manufacturing procedure within a domestic economy's framework.

Although both are two funerary contexts, the possible raw material sources are not very far away, at least no more than 15-20 km. Similar distance from potential sources was already observed in pottery assemblage from Cueva 3167 (Smith et al. 2014) and it is interpreted as a dissociation between the use and production contexts. We have not identified evidence of pottery production in the studied sites or in the two possible nearby settlements, Sierra Plana de la Borbolla and El Mazo (Ontañón Peredo 2003a; Gutiérrez Zugasti et al. 2018). Nevertheless, the employed housing suitability model indicates locations of appropriate housing development near the studied sites where pottery could be made (Vega 2017).

In this kind of context, ceramic assemblages are a sign of consumption pattern outside domestic spaces not directly linked to physical survival. Therefore, they should be assumed as evidences of habits of another nature, always within ceremonial activities. In other areas of Europe and North Africa commensality practices are related to Bell Beaker phenomenon (Dietler 1995; Aranda Jiménez and Esquivel Guerrero 2006; Garrido Pena et al. 2011). In short, a social framework in which feasts or social acts were carried out being the consumption of goods the central axis of ritualised acts (Hayden 1995). However, there were no evidence of this type of social behavior in this area until the Early Bronze Age. In addition, it is necessary to point out that, despite being a sign of identity of the Cantabrian Early Bronze Age, such potteries do not appear in all contexts and they are not exclusive of this area. In fact, there are similarities between those and other peninsular and extra-peninsular ceramic manifestations, including those found in the south-west of France and the Ebro River Valley (Maya and Petit 1986; Gardes 1993; Blanc et al. 1997). Data are similar to studies carried out in ceramics from different sites of Catalonia (Aigües Vives, Cova Frare, Les Maiolles and Mas Pla), whose results are coherent with the geological environment. Furthermore, this production process is very similar to that documented in this work with reducing-atmosphere and low-temperature fired ceramics assemblages, although with few intentionally added tempers (Clop i García 2007).

The formal and technical similarities among sets of pottery lead us to explain the local production in social terms, through contacts that seem to exclude the exchange of these products, at least as a main reason. Therefore, explanation would be more in line with the mobility of ideas and/or people. DNA studies focused around the 2000 cal BC show that *"the lineages common in Copper Age Iberia (I2, G2, and H) were almost*

completely replaced by one lineage, R1b-M269. These patterns point to a higher contribution of incoming males than females " (Olalde et al. 2019). Something similar happened in Germany where combining genome-wide, isotopic, anthropological and archaeological data revealed a social organization accompanied by patrilocality and female exogamy (Mittnik et al. 2019).

In the lineage society, as the Early Bronze Age in the Cantabrian region, the distribution of tasks for each member of the clan depended on their sex and age (Carmona Ballestero 2013). At least at the end of Bronze Age and the beginning of Iron Age, the pottery production was still included in the domestic sphere, being also the women in charge (Padilla Fernández and Dorado Alejos 2017). This is supported by ethnographic observation (González Ruibal 2005). In these circumstances, the training passes from mothers to daughters who repeat the acquired technical processes. So in our opinion this is one of the most plausible hypothesis, that directly relates formal and technical concomitances on pottery with the mobility of women through matrimonial circuits, in a restricted area, which would link these regions (Meillassoux 1977; Abarquero Moras 2005; Garrido Pena et al. 2011; Carmona Ballestero 2013). We think that our results are coherent and support a model where women travelled, but also transmitted their "know-how" and they were the ones who acted as a link between all these populations, transmitting their knowledge to the next generation. This hypothesis should be confirmed in future studies using ancient DNA and isotopic studies.

Acknowledgments

Gianni Gallelo acknowledges the financial support of the European Commission (Project H2020-MSCA-IF-2015-704709-MATRIX) and the Beatriz Galindo Fellowship founded by the Ministry of Science, Innovation and Universities. This research was also performed as part of the project HAR2016-75605-R, funded by the Spanish Ministry of Science, Innovation and Universities.

REFERENCES

- Abarquero Moras, F. J., 2005, *Cogotas I. La difusión de un tipo cerámico durante la Edad del Bronce*, Junta de Castilla y León, Valladolid.
- Aranda Jiménez, G., and Esquivel Guerrero, J. A., 2006, Ritual funerario y comensalidad en las sociedades de la Edad del Bronce del Sureste Peninsular: la Cultura del Argar, *Trabajos de Prehistoria*, **63**, 117–33.

- Arias Cabal, P., Martínez Villa, A., and Perez Suarez, C., 1986, La cueva sepulcral de Trespando (Corao, Cangas de Onís, Asturias), *Boletín del Instituto de Estudios Asturianos*, **119**, 805–31.
- Arias Cabal, P., and Pérez Suárez, C., 1995, Excavaciones arqueológicas en Arangas, Cabrales (1991-1994). Las Cuevas de los Canes, el Tíu Llínés y Arangas, In *Excavaciones Arqueológicas en Asturias (1991-1994)*, 79–92, Oviedo.
- Arias Cabal, P., and Armendariz Gutiérrez, A., 1998, Aproximación a la Edad del Bronce en la región cantábrica, In *A Idade do Bronce en Galicia: Novas perspectivas* (ed. R. Fábregas Valcarce), 47–80, Cuadernos do Seminario de Sargadelos, La Coruña.
- Blanc, C., Escude-Quillet, J.-M., and Marembert, F., 1997, Les tumulus en Béarn, In *Archéologie en Béarn, Actes du IV^o Colloque d'Arzacq*, 15–75, Ségur.
- Buxeda i Garrigós, J., and Cau Ontiveros, M. Á., 1995, Identificación y significado de la calcita secundaria en cerámicas arqueológicas, *Complutum*, 293–309.
- Carmona Ballester, E., 2013, *El Calcolítico en la Cuenca Media del Arlanzón. Comunidades campesinas, procesos históricos y transformaciones*, BAR International Series 2559, Archaeopress, Oxford.
- Cau Ontiveros, M. A., Day, P. M., and Montana, G., 2002, Secondary calcite in archaeological ceramics: evaluation of alteration and contamination processes by thin section study, *Modern Trends in Scientific Studies on Ancient Ceramics. Paper presented at the 5th European Meeting on Ancient Ceramics*, BAR International, Athens.
- Clop i García, X., 2007, *Materia prima y sociedad: la gestión de los recursos minerales para manufacturar cerámicas del 3100 al 1500 ANE en el noreste de la Península Ibérica*, Vol. ;1660, International Bar Series, :Archaeopress, Oxford .
- Cubas, M., 2012, La utilización de desgrasantes en las manufacturas cerámicas del V^o milenio cal B.C. en el Norte de la Península Ibérica, **5**, 375–82.
- Cubas, M., 2013, *La aparición de la tecnología cerámica en la Región cantábrica* Archaeopre ed., Oxford.
- Cubas, M., Bolado del Castillo, R., Pereda Rosales, R. M., and Fernández Vega, P., 2013, La cerámica en Cantabria desde su aparición (5000 cal BC) hasta el final de la Prehistoria: técnicas de manufactura y características morfodecorativas, *Munibe (Antropología-Arqueología)*, **64**, 69–88.
- Dietler, M., 1995, Feast and commensal politics in the Political Economy: Food, Power, and Status in the Prehistoric Europe, In *Food and the stauts quest. An interdisciplinary perspective* (eds. P. Wiessner, and W. Schiefenhövel), Berghanm

Books, Providence, Oxford.

Fabbri, B., Gualtieri, S., and Shoal, S., 2014, *The presence of calcite in archeological ceramics*, Vol. 34.

Flor, G., 1983, Las rasas asturianas: ensayo de correlación y emplazamiento., *Trabajos de Geología.*, **13**, 65-81.

Flor, G., and Flor-Blanco, G., 2014, Raised Beaches in the Cantabrian Coast., In *Landscapes and Landforms of Spain*, 239–48, Springer.

Gallelo, G., Ferro-Vázquez, C., Chenery, S., Lang, C., Thorton-Barnett, S., Kabora, T., Hodson, M. E., and Stump, D., 2019, The capability of rare earth elements geochemistry to interpret complex archaeological stratigraphy., *Microchemical Journal*(148), 691–701.

García Heras, M., and Olaetxea, C., 1992, Métodos y análisis para la caracterización de cerámicas arqueológicas. Estado actual de la cuestión., *Archivo Español de Arqueología*, **65**, 263–89.

García Heras, M., 1998, *Caracterización arqueométrica de la producción cerámica numantina.*, Archaeopress Series;, (BAR International 692)., Oxford.

García Sanjuán, L., 2005, *Introducción al reconocimiento y análisis arqueológico del territorio*, Ariel Prehistoria, Barcelona.

Gardes, P., 1993, Les urnes carénées de l' Age du Bronze Ouest-Pyrénéen français . Problèmes chronologiques, *Munibe (Antropologia-Arkeologia)*, **45**, 133–41.

Garrido Pena, R., Rojo Guerra, M., García-Martínez de Lagrán, I., and Tejedor-Rodríguez, C., 2011a, Drinking and eating together: the social and symbolic context of commensality rituals in the Bell Beakers of Interior of Iberia, In *Guess who's coming to dinner. Festing rituals in the Prehistoric societies of Europe and the Near East* (eds. G. Aranda Jiménez, S. Montón-Subias, and M. Sánchez Romero), 109–29, Oxbow Books, Oxford.

Garrido Pena, R., 2013, Alcohol, prestigio y poder: ritos de comensalidad en el Campaniforme del interior peninsular, In *De la cocina y sus ingredientes a la mesa y sus rituales. Desde los orígenes hasta las tradiciones populares en la Península Ibérica*, 47–59, Boletín de la Asociación Española de Amigos de la Arqueología, ISSN 0210-4741, N° 47.

González Morales, M. R., 1995, Memoria de los trabajos de limpieza y toma de muestras en los yacimientos de las cuevas de Mazaculos y El Espinoso (La Franca, Rivadesella) y La Llana (Andrín, Llanes) en 1993, In *Excavaciones Arqueológicas en Asturias 1991-1994*, 65–78.

- González Ruibal, A., 2005, Etnoarqueología de la cerámica en el oeste de Etiopía, *Trabajos de Prehistoria*, **62**(2), 41–66.
- Gutiérrez Zugasti, I., Cuenca Solana, D., González Morales, Manuel R. García Escárzaga, A., Salazar Cañarte, S., Teira, L. C., and Agudo Pérez, L., 2018, Intervención arqueológica en la cueva de El Mazo (Andrín, Llanes). Campañas de 2013, 2014, 2015 Y 2016, In *Excavaciones arqueológicas en Asturias. 2013-2016* (ed. C. de E. y Cultura), 133–42, Ediciones Trabe S.L.
- Hayden, B., 1995, Feasting in prehistoric and traditional societies, In *Food and the status quest. An interdisciplinary perspective* (eds. P. Wiessner, and W. Schiefenhövel), 127–47, Berghman Books, Providence. Oxford.
- Instituto Geológico y Minero de España, 1981, *Mapa Geologico de España. 1:50000. Llanes.*
- Vander Linden, M., 2007, What linked the Bell Beakers in third millenium BC Europe?, *Antiquity*, **81**, 343–52.
- Marembert, F., 2000, Un faciès original : le groupe du Pont-Long au cours des phases anciennes de l'Age du Bronze dans les Pyrénées nord-occidentales [article], *Bulletin de la Société préhistorique française Année*, **97**(4), 521–38.
- Martínez Santa-Olalla, J., 1930, Cerámica incisa y cerámica de la cultura del vaso campaniforme en Castilla la Vieja y Asturias, *Anuario de Prehistoria Madrileña*, **I**, 15–9.
- Maya, J. L., and Petit, M. A., 1986, El grupo del Nordeste. Un nuevo conjunto de cerámicas con boquique en la Península Ibérica, *Anales de prehistoria y arqueología*(2), 49–73.
- Meillassoux, C., 1977, *Mujeres, graneros y capitales: economía doméstica y capitalismo*, Antropología, Siglo XXI, México D.F.
- Melgarejo, J. C., 2003, *Atlas de asociaciones minerales en lámina delgada.*, Universitat de Barcelona. Fundació Folch, Barcelona.
- Millett, M., 1979, *Pottery and the Archaeologist*, UCL Institute of Archaeology Publications, Taylor & Francis, London.
- Mittnik, A., Massy, K., Knipper, C., Wittenborn, F., Friedrich, R., Pfrenkle, S., Burri, M., Carlichí-witjes, N., Deeg, H., Furtwängler, A., Harbeck, M., Heyking, K. Von, Kociumaka, C., Kucukkalipci, I., Lindauer, S., Metz, S., Staskiewicz, A., Thiel, A., and Wahl, J., 2019, Kinship-based social inequality in Bronze Age Europe, **734**(November), 731–4.
- Morlote Expósito, J. M., 2001, Los depósitos arqueológicos de la Edad del Bronce

- (Zona 8/96), In *La cueva de El Pendo. Actuaciones arqueológicas 1994-2000* (eds. R. Montes Barquin, and J. Sanguino Gonzalez), 249–60, Consejería de Cultura, Educación y Deporte del Gobierno de Cantabria, Santander.
- Noval Fonseca, M. A., 2013, Excavación arqueológica en la Cueva de El Toral III (Andrín, Llanes), In *Excavaciones arqueológicas en Asturias 2007-2012 en el centenario del descubrimiento de la caverna de la Peña de Candamo* (ed. G. del P. de Asturias), 381–5.
- Olaetxea, C., 2000, *La tecnología cerámica en la protohistoria vasca.*, Sociedad de Ciencias Aranzadi (Munibe suplemento 12)., San Sebastián.
- Olalde, I., Brace, S., Allentoft, M. E., Armit, I., Kristiansen, K., Booth, T., Rohland, N., Mallick, S., Szécsényi-Nagy, A., Mittnik, A., Altena, E., Lipson, M., Lazaridis, I., Harper, T. K., Patterson, N., (...), and Reich, D., 2018, The Beaker phenomenon and the genomic transformation of northwest Europe, *Nature*, **555**, 190.
- Olalde, I., Mallick, S., Patterson, N., Rohland, N., Villalba-mouco, V., Silva, M., Duliás, K., Edwards, C. J., Gandini, F., Pala, M., Soares, P., Ferrando-bernal, M., Adamski, N., Broomandkhoshbacht, N., and Cheronet, O., 2019, The genomic history of the Iberian Peninsula over the past 8000 years, **1234**(March), 1230–4.
- Ontañón Peredo, R., 2003a, *Caminos hacia la complejidad. El Calcolítico en la región cantábrica*, Servicio de Publicaciones de la Universidad de Cantabria, Santander.
- Ontañón Peredo, R., 2003b, El campaniforme en la Región Cantábrica: un fenómeno arqueológico en el seno de las sociedades calcolíticas del norte de la Península Ibérica, *Trabajos de Prehistoria*, **60**(1), 81–98.
- Ontañón Peredo, R., 2005, El campaniforme en la región cantábrica, In *El Campaniforme en la Península Ibérica y su contexto europeo* (eds. M. A. Rojo Guerra, R. Garrido Pena, and I. García-Martínez de Lagrán), 227–45, Universidad de Valladolid, Valladolid.
- Padilla Fernández, J. J., and Dorado Alejos, A., 2017, *Lo que la cerámica esconde : continuidad y cambio social a finales del Bronce y comienzos de la Edad del Hierro en el castro de las Cogotas (Cardeñosa , Ávila)*, Vol. 28.
- Perkins, D., and Henke, K. R., 2002, *Minerales en lámina delgada.*, Pearson Educación., Madrid.
- Plant, J. A., Simpson, P. R., Smith, B., and Windley, B. F., 1999, Uranium ore deposits. Products of the radioactive Earth, In *Uranium : mineralogy, geochemistry and the environment* (eds. P. C. Burns, and R. Finch), Mineralogical Society of America, Washington, DC.
- Prajith, A., Rao, V. P., and Kessarkar, P. M., 2015, Controls on the distribution and

fractionation of yttrium and rare earth elements in core sediments from the Mandovi estuary, western India, *Continental Shelf Research*, **92**, 59–71.

Quinn, P. S., 2013, *Ceramic Petrography. The Interpretation of Archaeological Pottery & Related Artefacts in Thin Section*, Archaeopress, Oxford.

Ramacciotti, M., Gallelo, G., Pastor, A., Castillo, A. D., and Puchol, O. G., 2019, Chert Nucleus and Cortex Characterization for Archaeological Provenance Study Tested in the Prebaetic System Region (Valencian Community, Spain), *Lithic Technology*, **44**(3), 166–80.

Rasilla Vives, M., 2011, *La cueva de El Sidrón (Borines, Piloña, Asturias): investigación interdisciplinar de un grupo neandertal*, Gobierno del Principado de Asturias, Oviedo.

Ruiz Cobo, J., and Serna González, M. R., 1990, Cerámicas incisas en cuevas de Cantabria, *Veleia*, **7**, 61–78.

Salanova, L., Prieto-Martínez, M. P., Clop-García, X., Convertini, F., Lantes-Suárez, O., and Martínez-Cortizas, A., 2016, What are large-scale Archaeometric programmes for? Bell beaker pottery and societies from the third millennium BC in Western Europe, *Archaeometry*, **58**(5), 722–35.

Sánchez, M. A., Cubas, M., Castro, R., Gutiérrez Medina, M., Bruschi, V., Bolado del Castillo, R., Ontañón, R., and Arias Cabal, P., 2016, Petrographic study of Iron Age pottery samples from Alto de la Garma hillfort (North Spain): assessment of the calcite dissolution caused by weathering, In *Euro-American Congress REHABEND 2016*, 138–44, Burgos.

Smith, P., Cubas, M., Corrin, J., Tapia, J., Pedro, I. de, Cobo, J. R., and Rosales, E. M. P., 2014, De arriba abajo : estudio integral de la cerámica prehistórica de la cueva 3167 (Matienzo , Cantabria , Norte de España)(3), 99–116.

Soyol-Erdene, T.-O., and Huh, Y., 2013, Rare earth element cycling in the pore waters of the Bering Sea Slope (IODP Exp. 323), *Chemical Geology*, **358**, 75–89.

Szécsényi-Nagy, A., Roth, C., Brandt, G., Rihuete-Herrada, C., Tejedor-Rodríguez, C., Held, P., García-Martínez-de-Lagrán, Í., Arcusa Magallón, H., Zesch, S., Knipper, C., Bánffy, E., Friederich, S., Meller, H., Bueno Ramírez, P., Barroso Bermejo, R., (...)and Alt, K. W., 2017, The maternal genetic make-up of the Iberian Peninsula between the Neolithic and the Early Bronze Age, *Scientific Reports*, **7**(1), 15644.

Taylor, S. R., and McLennan, S. M., 1985, *The Continental Crust: Its Composition and Evolution.*, Boston, MA.

Toledo Cañamero, M. C., 1999, Panorama científico sobre las investigaciones sobre la

producción cerámica de la Edad de Bronce en Cantabria, In *Regio Cantabrorum* (eds. J. A. Muñiz Castro, and J. M. Iglesias Gil), 13–20, Caja Cantabria, Santander.

Tostevin, R., Shields, G. A., Tarbuck, G. M., He, T., Clarkson, M. O., and Wood, R. A., 2016, Effective use of cerium anomalies as a redox proxy in carbonate-dominated marine settings, *Chemical Geology*, **438**, 146–62.

Vega Maeso, C., 2012, The ceramics of El Mirón Cave: production, morphology and discard, In *El Mirón Cave, Cantabrian Spain. The site and Its Holocene archaeological record* (eds. L. G. Straus, and M. R. González Morales), 372–425, University of New Mexico Press, Albuquerque.

Vega Maeso, C., 2017, *La cerámica inciso-impresa en el tránsito del III al II milenio cal. B.C. en la región Cantábrica* Nadir Edic ed., Santander.

Whitbread, I. K., 1989, A proposal for the systematic description in thin sections towards the study of ancient ceramic technology, In *Archaeometry: Proceedings of the 25th International Symposium*, 127–38, Elsevier, New York and Amsterdam.

Table 1. Semiquantitative XRD results showing percentages (%) of minerals.

SAMPLE	CRYSTALLINE PHASES											
	Quartz	Illite	Kaolonite	Albite	Orthoclase	Calcite Calcite	Dolomite Dolomite	Lime Lime Lime	Manganocalcite	Baryte	Jacobsite	Pyrite
LL1C4	29	16	0	1	7	42	4	0	0	0	0	0
LL2C3	55	13	0	3	9	20	0	0	0	0	0	0
LL3C2	71	7	0	4	4	10	3	0	0	0	0	0
LL4C1	62	8	0	14	5	9	0	0	0	0	0	0
T1C7	87	5	0	2	2	0	0	0	3	0	0	0
T3C5	60	9	0	6	14	10	0	0	0	0	0	0
T4C6	43	7	0	1	8	40	1	0	0	0	0	0
T6C10	36	10	0	4	0	48	0	0	0	0	0	0
T7C11	72	20	0	4	0	3	0	0	0	0	0	0
T8C12	32	9	0	3	0	54	0	0	0	0	0	0
A1	93	3	0	2	1	0	0	0	0	0	0	0
AC1	93	2	0	1	2	0	0	0	0	0	0	0
A2	39	28	0	0	0	7	0	0	0	0	0	0
AC2	35	15	0	0	0	0	0	0	0	0	0	18
A3	27	7	1	0	0	5	0	0	0	0	0	54
AC3	30	12	0	0	0	0	0	0	0	30	0	0
A4	38	7	0	0	0	2	49	0	0	0	4	0
AC4	51	13	0	7	0	0	5	0	0	0	5	0
A5	52	15	5		10	7	3	0	0	0	0	0
AC5	59	18	0	0	9	5	0	0	0	0	0	0
A6	83	10	3	2	2	0	0	0	0	0	0	0
AC6	82	8	0	4	4	0	0	0	0	0	0	0
A7	67	14	6	7	2	0	0	0	0	0	0	0
AC7	75	7	0	4	5	0	0	0	0	0	0	0
A8	63	30	0	0	3	0	0	0	0	0	0	0
AC8	59	26	0	0	8	0	0	0	0	0	0	0
A9	72	18	4	0	3	0	0	0	0	0	0	0
AC9	83	9	0	0	6	0	0	0	0	0	0	0
A10	74	15	3	1	5	0	0	0	0	0	0	0
AC10	77	7	0	3	10	0	0	0	0	0	0	0
A11	78	8	3	0	3	4	0	0	0	0	0	0
AC11	89	7	0	0	2	0	0	0	0	0	0	0
A12	67	9	8	0	6	0	0	0	0	0	0	0

Accepted Article

AC12	64	20	4	0	11	0	0	0	0	0	0	0
A13	61	8	0	0	7	21	0	0	0	0	0	0
AC13	62	8	0	0	5	2	0	22	0	0	0	0
A14	49	22	4	0	12	0	3	0	0	0	0	0
AC14	55	23	0	0	11	3	0	0	0	0	0	0
A15	74	13	0	3	5	1	0	0	0	0	0	0
AC15	88	5	0	0	3	0	0	0	0	0	0	0

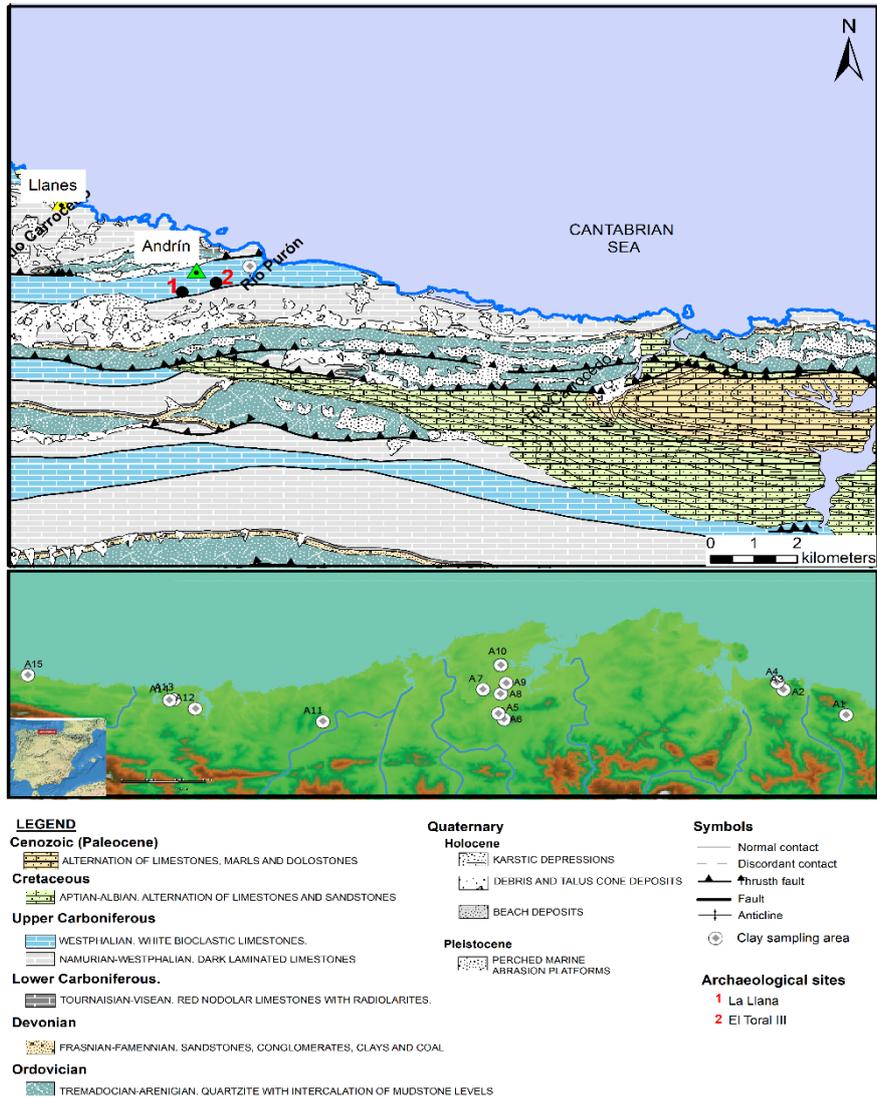


Figure 1. Geological map and location of sites and samples.

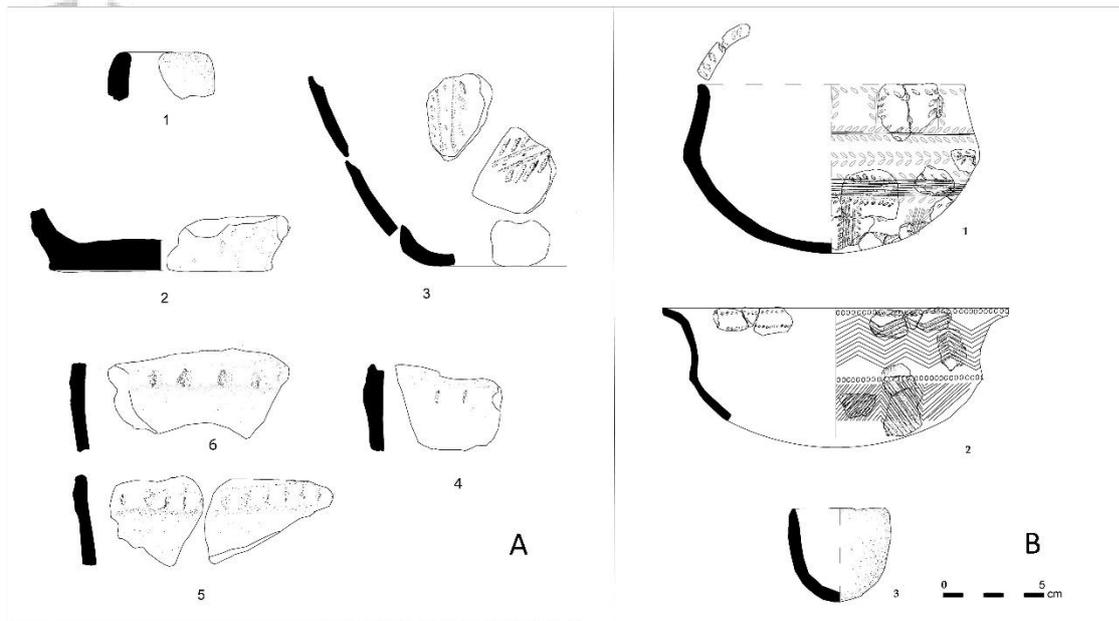


Figure 2. Significant vessels from El Toral III (A) and La Llana (B).

Accepted A

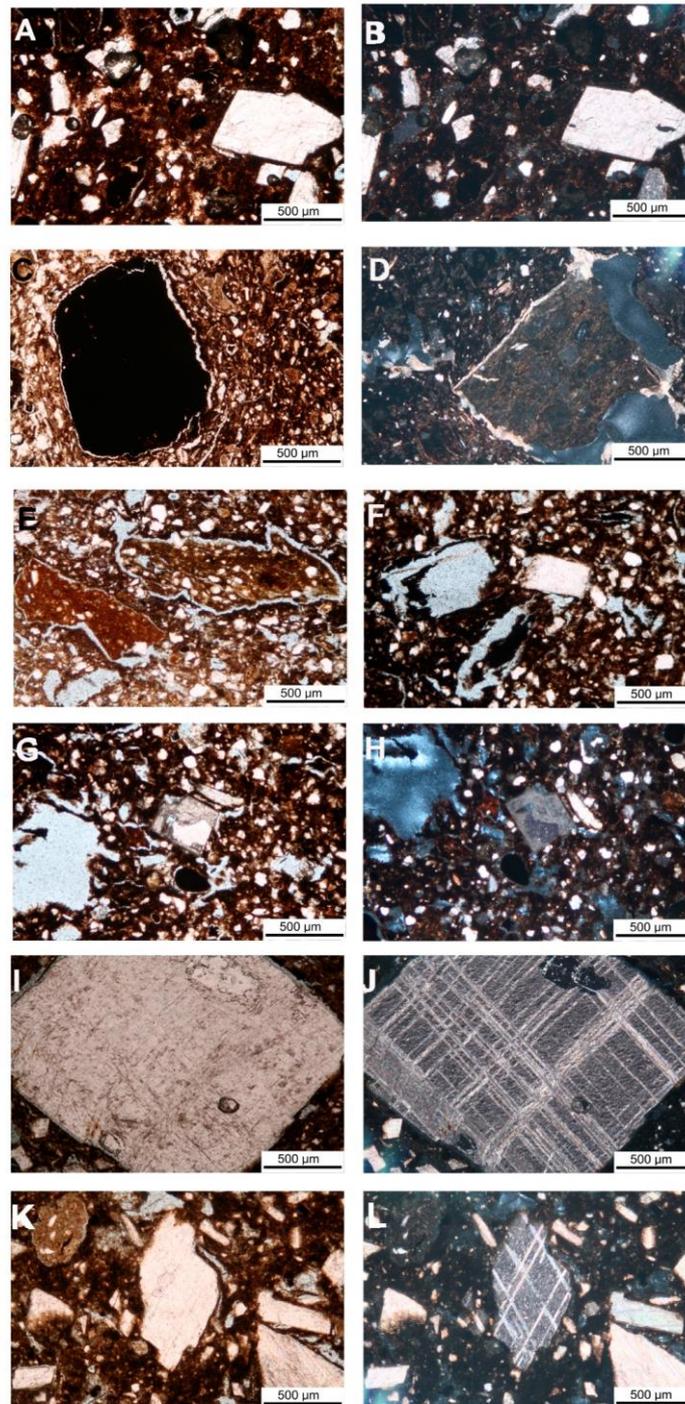


Figure 3. Microphotograph images: A and B - Textural aspect, Sample LL1C4; C - Opaque mineral, Sample T3C5; D - Secondary calcite, Sample T3C5; E - Grog-tempered pottery, Sample LL4C1; F - Organic matter, Sample LL4C1; G and H - Textural aspect, Sample LL4C1; I and J - Calcite-tempered pottery, Sample T4C6; K and L - Textural aspect, Sample T4C6. Plane-polarised light (A, C, E, F, G, I, K) and cross-polarised light (B, D, H, J, L).

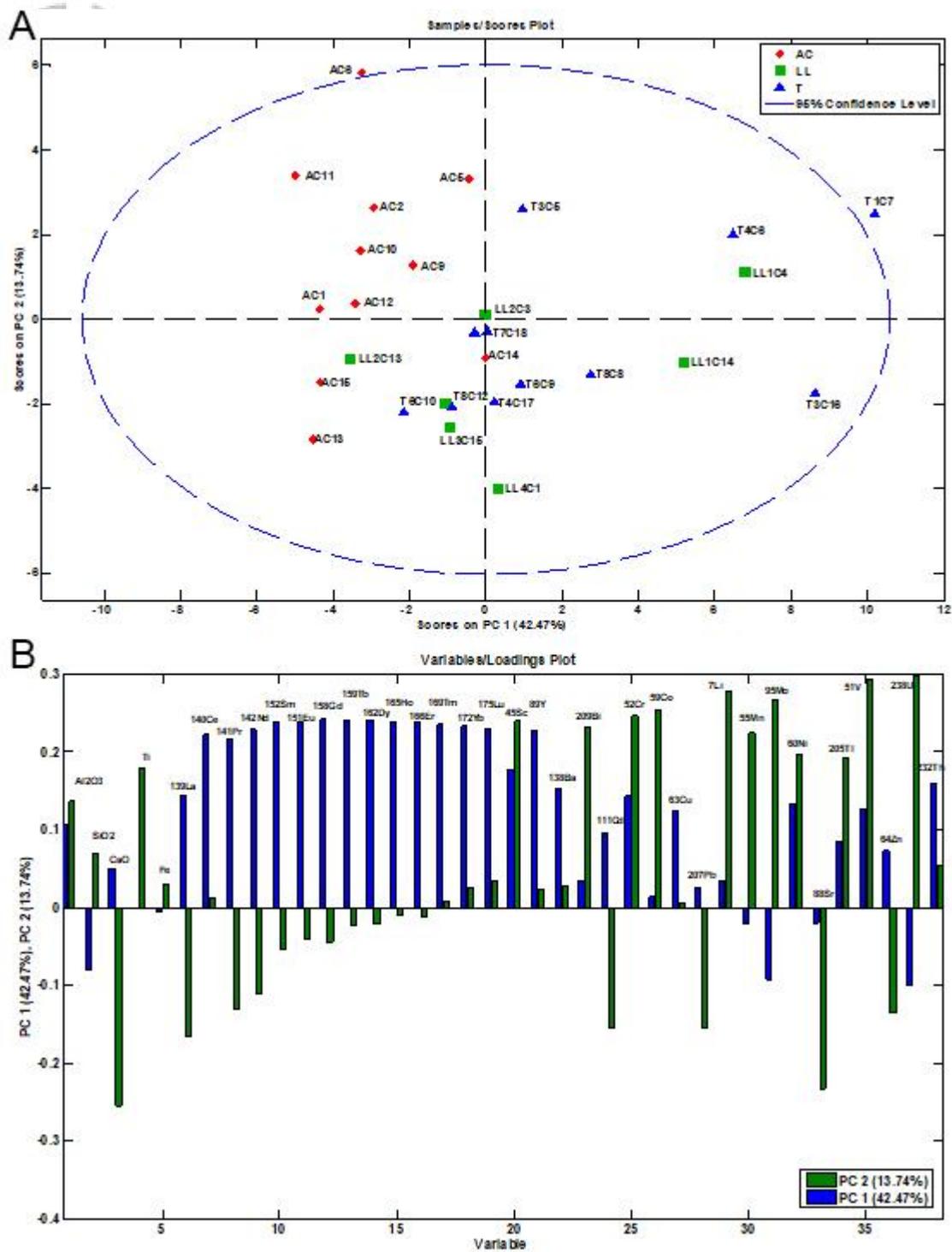


Figure 4. Results of PCA. Score plot (A) and loading plot (B) of PCs 1 and 2

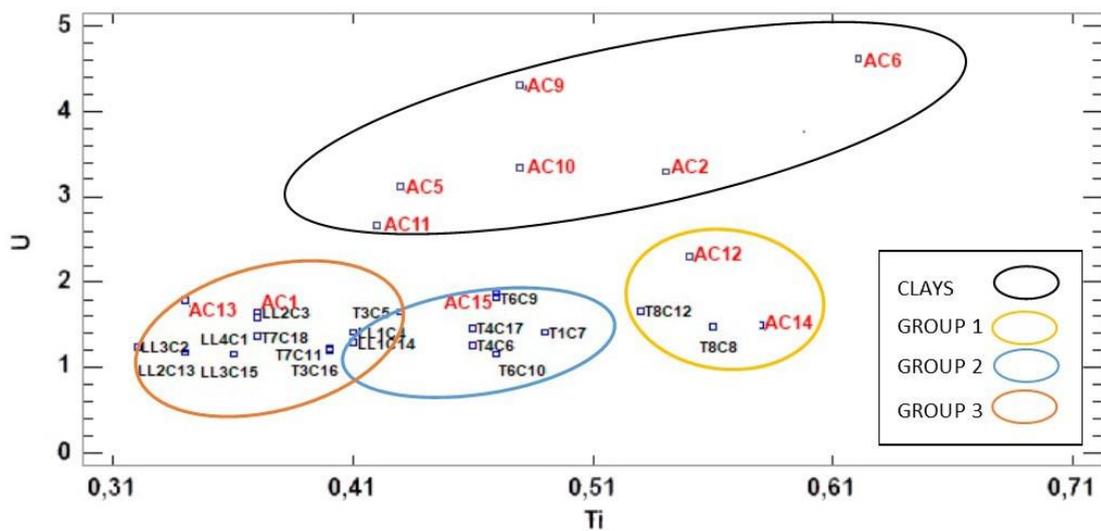


Figure 5. Scatterplot U versus Ti

Accepted A