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Research paper

Provenance and circulation of Bell Beakers from Western European societies of the 3rd millennium BC: The contribution of clays and pottery analyses

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ABSTRACT

This work intends to contribute for the discussion of beaker's social role in Western Europe, by studying Central - South Portugal evidences, establishing provenance and therefore pottery transactions between sites and/or regions, emphasizing the circulation /diffusion of this kind of pottery, and their impact on the European societies of the 3rd millennium BC. Ceramics from four relevant Chalcolithic - early Bronze Age archaeological sites of Central and Southern Portugal are studied, based on compositional paste analysis, confronting the bell beakers with other typologies, complemented with local/regional clays characterization. A broader spatial relationship is established, especially with other Iberian sites, and in the European context. Compositional studies were done by instrumental neutron activation analysis (INAA) and X-ray diffraction (XRD). Results for the four sites emphasize that some bell beakers have chemical composition similar to the other typologies, associated with local raw materials close to the archaeological site, pointing to local productions. On the other hand, the outliers identified are mainly comprised of bell beakers, assuming an exogenous nature. Thus, bell beakers are a complex material expression, where local productions are in relation with interregional systems of circulation of ideas and materials.

1. Introduction

Bell Beakers occur all over Europe and North Africa, and depending on the location, they belong to the Final Copper Age, or the Early Bronze Age (Fokkens and Nicolis, 2012; Kristiansen, 2011; Linden, 2004; Salanova, 2004; Turek, 2013). Classical archaeological studies regarding the Bell Beaker phenomenon together with laboratory research has been providing considerable important results (Dias et al., 2002; Hejcman et al., 2013; Lantes-Suárez et al., 2015; Prieto-Martínez et al., 2015) concerning some of the usual foremost questions posed by archaeologists studying this kind of pottery. These questions are related with production technology, raw materials exploitation strategies, provenance and distribution networks, therefore accomplishing a clue to mechanisms of circulation. These mechanisms of circulation are of significant importance, as bell beakers present considerable regional differences in style and context, which combined with the existence of an interregional system of shared ideas and social needs, contributes to a wider reading of the phenomenon.

The problem of the circulation versus provenance is mainly focused in the establishment of local production and importation of pottery. The potter was more tied to the local resources for his production than nowadays, which influenced pottery production and distribution, as it is more likely that the potter would have adapted local resources to answer specific needs, technical, functional, communicational and aesthetic. Being clays the major constituents of a pot, it is fundamental to know clays composition, the leftovers in the clay-making process and the chemical traces of both pots and potential raw materials, which can give a clue to the geographic origin of clay materials. Therefore, to understand the origin of the components or the materials, which make up pottery, chemistry/mineralogy and geology are of great help, achieving in this way the chemical identity and geological-mineralogical identities of the different components of a pot.

The first archaeometric study of Portuguese bell beakers was done in the eighties, with a chemical and mineralogical characterization of sherds from the "pre-beaker" and "beaker" levels of Porto Torrão Chalcolithic settlement located at Ferreira do Alentejo (Beja, South Portugal). Clay materials from the area were also analyzed (Cabral et al., 1988). Later, more southern and also central Portuguese bell beakers, from Fraga da Pena (Fornos de Algodres) (Dias et al., 2005a, 2002, 2000a) and from Monte do Tosco (Mourão) (Dias et al., 2013),

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Fig. 1. Location of the “bell beakers archaeological sites” in the Iberian Peninsula. 1. Fraga da Pena; 2. Perdigões; 3. Monte do Tosco; 4. Porto Torrão.

were studied, as well as clay materials from the corresponding areas (Dias, 2013; Dias et al., 2003, 2000b; Marques et al., 2010).

In the last years, the interdisciplinary Portuguese team (C^2TN and Era Arqueologia SA) and the Spanish team from Seville University worked together to better define the Middle Guadiana River Basin Copper Age pottery consumption and distribution patterns (Odriozola et al., 2009a, 2009b, 2008a, 2008b, 2007). In these works, the flow patterns of bell beakers are thought to mirror social dynamics and boundaries throughout the bell beaker production and consumption patterns across landscape. Supported in other evidences as settlement spatial patterning, and spatial distribution of ideological and symbolic related goods.

In this work, we aim to study the ceramic record from four archaeological sites (Fig. 1) of Central (Fraga da Pena) and Southern Portugal (Porto Torrão, Monte do Tosco and Perdigões), adding new evidences based on compositional paste analysis, stressing the bell beakers in relation to the other typologies, thus inferring provenance and circulation issues. In addition, local/regional clays for all those sites will be studied. This approach will allows working with large data sets in order to better define pottery production and consumption patterns in the transition Chalcolithic - early Bronze Age in southwest Iberia and its impact on the societies of the 3rd millennium BC. The methodological approach will include chemical composition by instrumental neutron activation analysis (INAA) and mineralogical composition by X-ray diffraction (XRD) applied to both ceramics and local/regional clays. Petrographic analyses are only applied in some cases that together with XRD will contribute to better define production technology, like firing temperature and admixing raw materials recipe. The combined use of this variety of techniques constitutes a key approach in our understanding of the technology and provenance of the materials examined here. This approach will generate reference pottery compositional groups that can discriminate between locally and non-locally produced groups, in order to establish pottery provenance and therefore pottery transactions between sites and/or regions, with special emphasis in the bell beakers phenomenon, thus contributing to the discussion of the circulation/diffusion of this kind of pottery.

2. Experimental

Two separate pieces ~2 cm^2 in area were removed from the shards (~2 g potsherd) for mineralogical and chemical analyses of the ceramic paste (whole sample) by XRD and INAA. To obtain powder samples of ceramics for XRD and INAA the inner and outer surfaces of the shard were scraped using a drill burr made of tungsten carbide. More details are given in Prudêncio et al., 2006. Clay raw materials and ceramics were grinded in a planetary agate mortar into a fine powder (< 53 mm). Sub-samples were taken for XRD and INAA.

To obtain the chemical data used in the compositional characterization of clay materials and ceramics, chemical analysis was done by means of INAA. Irradiations were done in the core grid of the Portuguese Research Reactor (Sacavém), as neutron source. GSD-9 (sediment) and GSS-1 (soil) of the “Institute of Geophysical and Geochemical Prospecting” (IGGE) were used as reference materials. The reference values were taken from data tabulated by Govindaraju (1994).

All powdered samples were prepared for irradiation by weighing 200–300 mg of powder into cleaned high-density polyethylene vials. Long irradiations (7 h) on specimens were performed on batches of 20 unknowns along with four standards in the core grid of the Portuguese Research Reactor at a thermal flux of 3.96×10^{12} n cm^{-2} s^{-1}; ϕth/ϕepi = 96.8; ϕth/ϕfast = 29.8, obtaining the concentration of the elements: Na, K, Fe, Ca, Sc, Cr, Co, Zn, Ga, As, Br, Rb, Zr, Sb, Cs, Ba, La, Ce, Nd, Sm, Eu, Tb, Yb, Lu, Hf, Ta, Th and U. Also short irradiations were performed, during 2 min, at a flux of 4.4×10^{12} n cm^{-2} s^{-1} obtaining the concentration of Mn and Dy. Nuclear radiation from activated products was measured by a high-resolution γ -ray spectrometry, which includes detectors made from high purity germanium associated with preamplifiers, amplifiers and multichannel analyzers. More details in Dias and Prudêncio, 2007; Dias et al., 2010; Prudêncio et al., 2009. Relative precision and accuracy are, in general, to within 5%, and occasionally within 10%.

The corrections for the spectral interference from uranium fission products in the determination of barium, rare earths and zirconium were also taken into consideration (Gouveia et al., 1987; Martinho et al., 1991).

The concentrations of trace and major elements were used as variables for a multivariate statistical study and for a detailed analysis of the element distributions, including elements not very susceptible to mobilizing processes (Trindade et al., 2011). This problem must have special attention, as it can occur not only during the ceramics manufacture processes, but also during its use and burial (post-depositional processes) (Golitko et al., 2012). Multivariate statistical methods were employed by using the Statistica program (Dell Inc., 2016), like the joining tree-clustering method, by using the absolute concentration of the chemical elements as variables, with diverse amalgamation methods and similarity/correlation coefficients. The abundance of data obtained by the chemical characterization and thus the high amount of variables and cases studied in the current work needs the use of combined data analysis and statistical methods. It is commonly a challenge to examine and interpret the significance of many elements, in terms of processes, and even more difficult to observe or understand the relationships between a large group of elements. The application of multivariate data analysis and statistical techniques combined with elemental ratios, provide support for data interpretation and subsequent archaeological interpretations. Exploratory multi-variate methods include: plots of all relevant pairs of data, assessment of the individual distributions, adjusting for missing data, detecting atypical observations, computing robust means, correlations, principal components analysis, cluster analyses, etc., specially aiming to identify geological processes, and so geochemical signatures for clays and ceramics.

The mineralogical composition of both whole samples and clay-sized fractions (< 2 μm) was obtained by XRD using a Philips diffractometer, Pro Analytical, with Cu Kα radiation at 45 kV and 40 mA. Spectra were gathered in the case of the whole sample at a step size of 1° 2θ/min from 3° to 70° 2θ and in the case of the < 2 μm fraction at a step size of 1° 2θ/min from 3° to 35° 2θ. For the whole rock sample preparation, samples were powdered in an agate mortar; the powder was then side loaded into a glass sample holder to obtain randomly oriented specimen, thus minimizing preferred orientations. The clay-size fraction was obtained by sedimentation according to Stokes law, and then piped and dried at room temperature on glass slides to produce a thin oriented layer. Ethylene glycol-solvated and 500 °C heated slides were also prepared. Especially attention was done to eventual neoformed minerals obtained by firing processes (Trindade et al., 2009, 2010).

In some cases, thin section petrography technique was applied to ceramic sherds, to better observe textural features by conventional optical microscopy.

3. Materials and contexts

The Fraga da Pena (FP) archaeological site is a ceremonial walled enclosure in Beira Alta region, Central Portugal, located in a huge granitic tor, impressively conspicuous in the surrounding country, occupied at the transition Chalcolithic - early Bronze Age (last quarter of the third millennium BC). The FP archaeological remains revealed a significant quantity of rare goods, mainly bell beakers, and a relative absence of objects related to daily life, reinforcing, together with the general architecture of the site and its location in the landscape, the exceptionality of the context (Valera, 2016, 2007, 1997). Among the several typological groups identified, four were selected for compositional analyzes, comprising a total of seventy ceramic samples: (i) bell beakers (Fig. S1 in the Supplementary materials); (ii) decorated vessels with combed incisions; (iii) vessels with morphological and decoration patterns of Chalcolithic tradition; and (iv) Bronze Age new morphologies (Dias et al., 2005a, 2003, 2002, 2000b). The surrounding geological context of the settlements is granites with quartz, aplite-pegmatite and dolerite veins and in restrict area the schist-greywacke complex. Thirty samples of residual clay materials were collected essentially derived by weathering of granites and their veins, specially the dolerite ones, which are the most clayey materials of the area. Clay samples correspond to different stages of weathering of these geological materials.

The Porto Torrão archaeological site is located in Ferreira do Alentejo municipality, Beja district, Alentejo, South Portugal, attributed to the Late Neolithic and Chalcolithic (Arnaud, 1993; Valera, 2010; Valera and Filipe, 2004;). Porto Torrão is a known large Chalcolithic site since the eighties (Arnaud, 1993), but only in 2002 it became clear it corresponded to a complex set of ditched enclosures, especially due to the emergency excavations that detected the presence of ditches for the first time (Valera and Filipe, 2004), related to the building of a high voltage electricity line. Those ditches were from different chronologies, being the inner one filled with Late Neolithic materials and the outer one with Chalcolithic materials, with the presence of bell beakers. More recently, again in a context of emergency excavation works, other ditches, several necropolis of tholoi and hypogea were identified around the enclosures (Santos et al., 2014; Valera, 2010; Valera et al., 2014a), pointing to a large complex and one of the biggest of Iberia. Bell beakers from the Porto Torrão archaeological site, as well as Pre-Beaker ceramics, modern ceramics, together with potential raw materials from site surrounds were the aim of a previous study (Cabral et al., 1988). More recently, additional samples of bell beakers and other pottery were studied aiming a better differentiation from this site and other Portuguese and Spanish sites along the Guadiana basin (Odriozola et al., 2009a). The geological context of the site comprises gabbro-diorites, quartz, porphyries, Silurian and Devonian schists, greywackes, and Paleogene-Miocene and Pliocene sediments. This diversity and environmental factors have certainly affected the chemical composition of soils and clays in that area. The archaeological settlement is located on Paleogene-Miocene sediments. Nowadays working potteries at nearby Beringel village are also located in those Tertiary sediments. However, they prefer to use raw materials derived by weathering from gabbros and diorites, the so-called “Barros de Beja”. From Porto Torrão site, we have analyzed ninety-seven archaeological ceramic sherds, comprising sixty bell beakers:

a) Fifty-five sherds from two different archaeological contexts - the pre-beaker (non-decorated; decorated; loom-weights) and the beaker levels (non-decorated; decorated; loom-weights).
b) Thirty-one bell beakers decorated and non-decorated from early excavations works and eleven sherds of common pottery.
c) Eleven clay samples derived by weathering from available geological background in the area, including some explored at the time by local potters (soils derived by weathering from gabbros and diorites; from schists and greywackes; Paleogene-Miocene and Pliocene sediments).
d) Five samples of modern ceramics made by local potters and abandoned potteries.

The Perdigões site is another large complex of ditched enclosures in South Portugal, occupied during the second half of the 4th–3rd millennium B.C in the Reguengos de Monsaraz region, in Évora district. The site spreads over an area of 16 ha, and presents a sequence of 13 roughly concentric ditches. Work developed so far suggests that the enclosures might have functioned as a place of social aggregation and negotiation, where ceremonial activities, namely involving funerary practices and human body manipulations would have particular importance (Valera et al., 2014b). The topographic and landscape relations, allowed the development of interpretative discourses of the spatial organization of the site, also by means of astronomical and cosmological aspects (Valera, 2008). In addition, this site dovetail structures that are distributed differently over time, so that we may speak of “several” Perdigões lifelong the site occupation (Valera et al., 2014c). The on-going research is attentive to the fact that we are dealing with a reality that was dynamic, diverse, and complex, obtained from a register that provides fossilized palimpsest as a static, homogeneous, partial known (Valera et al. 2014b, 2007). The burial remains are diversified, and mainly include pottery, lithic artefacts, limestone and bone idols, pecten shells, ivory items, adorn artefacts, etc. On a global perspective the pottery includes all the typical morphologies of the Late Neolithic and Chalcolithic of the Southwest Iberia, and differences occur between some of the Chalcolithic funerary votive pottery and recipients from other contexts. Previous works on provenance and production technologies of the funerary ritual pottery, when compared with ceramics from other contexts (Dias et al., 2007, 2005b), on a diachronic approach (Neolithic to Chalcolithic) (Dias et al., 2012), reveal a ceramic production strategy, and a special role of bell beakers in the framework of the later enclosures and some of the funerary contexts. In this work ceramics attributed to the Chalcolithic (seventy sherds), and to the Neolithic contexts (thirty sherds) were studied. The Chalcolithic ones include ceramic recipients associated to funerary rituals (Tomb 1), to domestic typologies integrated in Tomb 1, to domestic typologies from other contexts and to funerary recipient ceramics from Tomb 2. Neolithic pottery is mainly from sector Q, ditch 6. Nineteen bell beakers were analyzed (Fig. S2 in the Supplementary materials). Potential raw materials were also collected, comprising clay samples from the Estremoz-Barrancos sector (SEB) of the Ossa Morena Zone. Field-work and sampling included residual clays derived by weathering from diorites and gabbros, granodiorites and tonalites from the Reguengos de Monsaraz massif (RM), from schists, dolerite veins, vulcanites and schists with metabasites, and sedimentary clays (Tertiary) in the Reguengos de Monsaraz – Mourão – Póvoa region (Dias, 2013; Dias et al., 2012, 2007).

The Monte do Tosco (MT) is a walled Chalcolithic enclosure, reoccupied during the early Bronze Age, where a late bell beaker context has been excavated (Valera, 2013, 2006, 2000). During the Chalcolithic (first half of the 3rd millennium BC) occupation, at least two hut stone structures were identified. The construction of a circular stone hut, where several beaker shards and metal tools were collected, are associated to a later reoccupation (Late Chalcolithic/Early Bronze Age). This site is located in the top of a hill, over the Alcarrache River, in the Mourão municipality (Évora district, Alentejo, South Portugal).

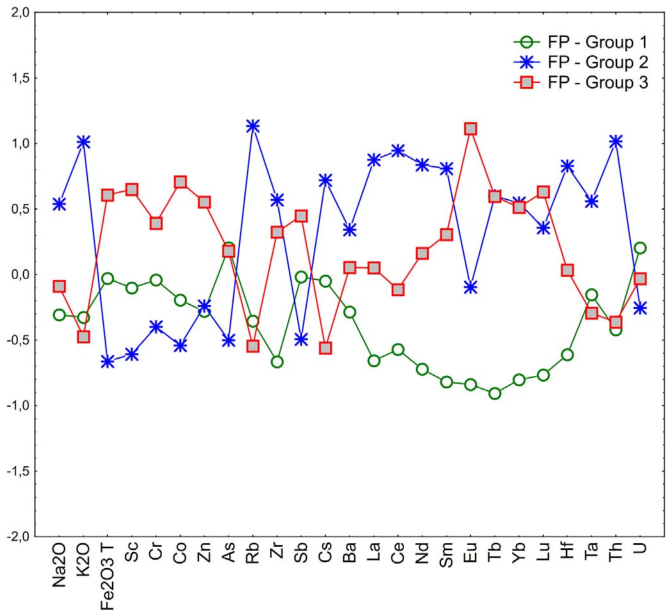

Fig. 2. Plot of means for each cluster (K-means method) using chemical results of Fraga da Pena ceramics as variables.

Materials analyzed in this work include Chalcolithic ceramics, Bronze Age ceramics from the bell beakers context, common ware and big recipients, as well crucibles, comprising thirty-five samples. For provenance issues and raw materials exploitation strategies, the same clay materials previously mentioned for the Perdigões case study (weathered schists, greywackes, diorites and gabbros and Tertiary clays) were used, as they belong to the same region.

4. Results and discussion

4.1. The bell beakers from Fraga da Pena, Beira Alta, Central Portugal

Compositional analyzes of all studied ceramic typologies from FP (Table S1 in the Supplementary materials), reveal that bell beakers present a more homogeneous chemical composition, though some nail printed bell beakers present a different behavior. The main geochemical features observed and statistical approaches, like the k-means clustering method, heighten three main groups (Fig. 2): Group 1 (G1) comprises mostly bell beakers; Group 2 (G2) is essentially composed by Chalcolithic and Bronze Age typologies, as well as combed incisions ceramics, and only a few bell beakers; Group 3 (G3) includes, like the first group, essentially bell beakers, and also Chalcolithic and Bronze Age typologies.

Regarding the potential raw materials of the region, dolerites provide the most clayey samples, though the aplite-pegmatite veins also weathered to argillaceous materials. The mineralogical associations obtained for clay samples are shown in Table 1. Plagioclase reaches the higher levels in granites. Iron oxides and pyroxenes reach the higher levels in dolerites. Micas are present in all cases. The better differentiation within these samples is the phyllosilicates, plagioclase, iron oxides and pyroxenes proportion. Regarding the clay fraction (< 2 μm) (Table 2), smectite and kaolin minerals are good fingerprints, as they predominate respectively in the mafic and in the felsic materials (Dias et al., 2003, 2000b).

Chemical composition of potential raw materials clearly differentiates the clays derived by weathering of granites and those derived by weathering of dolerites, respectively by their Na and K contents (related to plagioclase and alkali feldspar), and Cr, Co, Sc and Fe (elements with geochemical affinity with ferromagnesian minerals). Although it is possible to clearly differentiate between these different geological materials, in some cases also important chemical differences occur within each type, such as in the rare earth elements (REE) contents, as they result from different grades of weathering.

A comparison study between chemical composition of ceramics and clays enables to correlate the three defined ceramic groups with regional geological materials. The application of multivariate data analysis and statistical techniques combined with elemental ratios, support that the chemical composition of the groups including mostly bell beakers (G1 and G3) has more affinity with basic rocks, like the dolerite samples, enriched in elements associated to ferromagnesian minerals. The few dissimilarities found are due to variations among dolerite veins due to different stages of weathering, and they are specially revealed by REE fractionating. Still, internal composition variation of the source is lower than intersource variation.

The chemical composition of Chalcolithic and some Bronze Age typologies (G2) points to the use of more felsic materials, like local/regional granites, with higher contents of chemical elements (Na, K, Rb, Cs) related with the presence of feldspars. In addition higher amounts of REE, especially light rare earth elements (LREE), as well of Zr, Hf, Ta and Th, and a depletion in elements of the first transition series occur.

Textural features obtained by petrography, together with mineralogy and chemistry, reveal that when in presence of fine pottery, a more positive correlation exists with dolerites, and in coarser pastes with granites, even in both cases non-plastic grains of granite origin were always detected. In the case of bell beakers, specially the "nail printed" ones, which are in general more fine and have less temper, it is more obvious the correlation with dolerites as raw materials. In this later case, a careful production technology occurred, with the use of well-selected materials of dolerite origin. There are also ceramics with no specific typology presenting fine textures, but with material of granite origin, also well selected. For beyond the bell beakers case, in most of the ceramics, non-plastic grains occur in great amounts added by the potter in the process of assembling the paste, with temper grains with various sizes. In this case, the paste shows a high irregularity of grain size distribution, with lumps of clay, giving a very irregular texture and a very nebulous and irregular aspect of the orientation of the clays. The clay-temper grain mixture material used as paste shows that the clays were certainly not ground sieved and mixed as well as the temper grains. On the contrary, bell beakers, especially the "nail printed" ones, present a thinner paste with well selected non-plastic grains, which even distribution and grain size gives a more regular texture, indicating a careful mixing and working of the clay resource and temper grains, thus a different process of making the pot.

Therefore, the Fraga da Pena bell beakers, especially the "nail printed" ones, present chemical, textural and mineralogical particularities, which point to a more careful local production technology, even there are also bell beakers with compositional similarities to other typologies. Outliers were defined by using statistical techniques, like the hierarchical tree clustering method using the Euclidean distances as the similarity coefficient. In the bell beakers from Fraga da Pena outliers defined comprise a few ceramics (seven = 10%) presenting a chemical composition that differs significantly from the local

Table 1
Mineralogical associations of the bulk sample of geological materials derived by weathering of different rocks in FP surrounding area.

Granites	Quartz ± Alkali Feldspars ± Plagioclases ± Micas ± Phyllosilicates ± (Goethite ± Pyroxenes)
Schist-Greywacke Complex	Quartz ± Alkali Feldspars ± Phyllosilicates ± Plagioclases ± Micas ± Gibsite ± Goethite
Dolerite Veins	Quartz ± Phyllosilicates ± Alkali Feldspars ± Micas ± Plagioclases ± Pyroxenes ± Goethite ± Ilmenite ± Hematite
Aplite-pegmatite Veins	Alkali Feldspars ± Phyllosilicates ± Quartz ± Plagioclases ± Micas ± Goethite ± Pyroxenes

Table 2
Mineralogical associations of the clay (< 2 μm) fraction of geological materials derived by weathering of different rocks in FP surrounding area.

Granites	Kaolinite ± Smectite ± Illite (± Interstratified)
Schist-Greywacke Complex	Kaolinite ± Illite ± Gibsite ± Iron Oxides
Dolerite Veins	Smectite ± Kaolinite ± Illite ± Chlorite ± Interstratified ± Iron Oxides
Aplite-pegmatite Veins	Kaolinite ± Smectite ± Illite

productions, indicating the use of different raw materials (importations?). It should be noted that four of these outliers are bell beakers, mainly of the "International complex" and imitations.

4.2. The Bell Beakers from Porto Torrão, Alentejo, Southern Portugal

The approach used in this work for the interpretation of chemical data (Table S2 in the Supplementary materials) enables to differentiate three compositional groups for the Porto Torrão ceramics, which main differences are clearly shown in the plot of means (k-means clustering method, Fig. 3). Pre-beaker and beaker vessels exist in all of those compositional groups together with local clays and modern ceramics produced at local potters, which indicates that the clays employed for manufacturing the ceramics of those groups lasted a long period of time, they use local resources, and no special differentiation was taking into account to produce the bell beakers. Geochemical composition of ceramics and the available raw materials point toward the production of pre-beaker and beaker pottery mainly with clays from the same nature, specifically the clays derived by weathering of gabbros and diorites ("Barros de Beja"). Nevertheless, some ceramics also point to the resource of clays derived by weathering of schists and greywackes, and only a few ceramics present chemical similarity with the Tertiary clays, occurring at the site.

The best geochemical fingerprints for ceramics and clays of Porto Torrão are the REE distribution. Tertiary clays and the smaller group of ceramics (Group 1 comprising 16 ceramic samples of all the typologies) do not have Eu anomaly (Eu/Eu*), or have negative Eu anomaly (0,56 ≤ Eu/Eu* ≤ 0,98). They also have the higher values of REE sum (sum REE average = 122,06), especially the LREE, with different degrees of REE fractionation, and the higher ratio light REE and heavy REE (LREE/HREE). On the other hand, clays derived by weathered gabbros and diorites, and related ceramics (group 2) have a positive Eu anomaly (1,28 ≤ Eu/Eu* ≤ 2,90) (Eu/Eu* refers to Eu anomaly = $EuN / \sqrt{(SmN)\cdot(GdN)}$) and lower values of REE (sum REE average = 26). In schists and greywackes, the positive Eu anomaly also exists but it is not so strong. Therefore, the two main groups are chemically differentiated mainly due to Eu/Eu*, REE contents and REE fractionating. Nevertheless, other elements also contribute to the differentiation of group 1, namely the lower Na, Fe, Sc, Cr and Co contents, and the higher K, Rb, Cs, Hf and Th contents, indicating a more felsic nature of the raw materials. A more detailed geochemical study enables to detach a group of ceramics - group 2, including 26 ceramic samples of all the typologies. These samples have higher Fe, Sc, Co, Eu and HREE contents (Fig. 3).

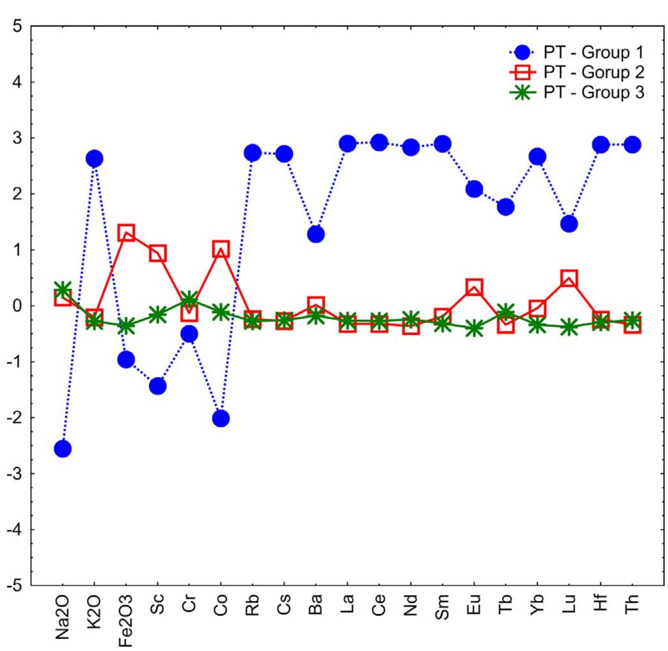

Fig. 3. Plot of means for each cluster (K-means method) using chemical results of Porto Torrão ceramics and of local/regional clays, as variables.

Mineralogical composition doesn't allow to clear differentiate ceramics and clays, as most of them has as predominant mineralogical association Quartz + Feldspars (Plagioclase) + Amphiboles (Hornblende), and in a few samples Calcite + Dolomite + Micas + iron hydroxides (goethite). It is important to enhance the constant presence of Plagioclase and Hornblende in ceramics, which is a characteristic of the materials around the site, supporting the idea of local production of pottery, as was also indicated by chemical analysis.

The positive europium anomaly found for most of the samples is certainly related with their high plagioclase content, as if a magma crystallizes stable plagioclase, most of the Eu will be incorporated into this mineral causing a higher than expected concentration of Eu in the mineral versus other REE in that mineral (a positive anomaly).

Chemical and mineralogical composition obtained for ceramics (bell beakers and other typologies) from Porto Torrão Chalcolithic site and related clay materials, have had an important contribution to the bell beakers circulation problematic, as they clearly indicate that bell beakers were not introduced in the area as a result of some trade or "prestige good" distribution network, but were produced locally.

4.3. The Bell Beakers from Perdigões, Alentejo, Southern Portugal

The mineralogical associations found for both the bulk and < 2 μm fraction of local and regional clay materials contribute to the distinction of studied clay samples. Nevertheless, the chemical analyses of trace elements played the most important role in obtaining regional geochemical fingerprinting of both residual and sedimentary clay materials from the Paleozoic and Cenozoic of the studied region. The residual clays derived from the weathering of the dolerite veins were generally enriched in all elements, especially REE. On the opposite end, the sedimentary Tertiary clays, due to the diluting effect made by the high amounts of carbonates, have the lower amounts of all chemical elements, especially REE, with the exception of As and Sb, whose ions are commonly fixed by clays and calcium. The clays from weathered diorites and gabbros, granodiorites and tonalites of Reguengos de Monsaraz have similar geochemical behavior as clays from weathered schists with metabasites, with high amounts of elements related with ferromagnesian minerals and HREE; LREE depletion is explained by the presence of heavy minerals like allanite. Clays derived by the weathering of schists have geochemical similarities with vulcanite clays, especially due to the high amounts of REE, and similar contents of Fe, Sc, Cr, even vulcanites are slightly enriched in Zn, As and Sb (Dias, 2013).

In a geochemical point of view, a good differentiation was achieved within and between residual clay materials and sedimentary clays of

Reguengos de Monsaraz – Mourão – Póvoa region. The clays derived by weathering of the lithologies of the Reguengos de Monsaraz massif (granodiorites & tonalites, and gabbro-diorites) have an intense LREE depletion, due to the presence of heavy minerals like allanite. They also have high contents of elements related to ferromagnesian minerals, and an increase of albite in plagioclase and of microcline and quartz, and a decrease of mafic minerals like amphibolite from the gabbro-diorite term to the granodiorites and tonalites term. Clays derived by weathering of schists have high amounts of K, Fe, Sc, Cr, Co, Rb, REE, Ta, Th and U. The clear enrichment in K, Rb, Ga may be explained as a consequence of the importance of phyllosilicates in parent rocks and of its terrigenous origin; high contents of REE may be explained by their richness in Ordovician parent rock and interstratified quartzites in this schists, and not the schists itself. The clay rich material resulting of the weathered dolerite vein has in general high contents of all elements, related to their preference to be fixed in clays, iron oxides and heavy minerals. Tertiary clays have a general depletion of all elements, especially REE, as a diluting effect of the presence of high amounts of carbonates in these samples, and are enriched especially in As, as arsenate ions tend to be readily fixed by calcium.

The comparative study of both potential raw materials and ceramics performed by using multivariate data analysis combined with elemental ratios, enables to determine three chemical groups of Chalcolithic pottery that were produced in general with raw materials from the geological contexts close to Perdigões, i.e. clays derived from the diorite and gabbros alteration. Nevertheless, the third group consisting almost exclusively of "funerary" containers from tomb 1, points to the resource of more diversified raw materials. This group, being the smallest and the more chemically heterogeneous, points to the use of weathered schists far from Perdigões > 5 km to the north, south and west. It includes only funerary ceramics, which is also consistent with the hypothesis of the necropolis of Perdigões was in use by distant communities. Regarding Neolithic samples, a certain chemical homogeneity exists, but two samples are detachable from the others due to higher amounts of chemical elements from the first transition row, more related with ferromagnesian minerals.

In a mineralogical point of view, we have the same type of mineralogical association for both chronologies (Quartz ≥ Phyllosilicates > Ca Plagioclase > K Feldspar > Amphibole. Still amphibole is more consistently present in Neolithic samples (especially in the former mentioned loners) and in a few samples, biotite can reach 25%. No high temperature mineral phases were identified.

In a diachronic approach, chemical results (Table S3 in the Supplementary materials) enable to establish compositional patterns of Neolithic, Chalcolithic and correlated clays. The Fe, Sc, Ba, As, Zn, Ta, U and LREE contents have a higher range in the Chalcolithic funerary pottery. A few of these funerary ceramics point to a complete different source, especially in the REE. One bigger group reflects the existence of more chemical homogeneity (majority of Neolithic and Chalcolithic domestic and funerary ceramics); another one is clearly explained by REE, especially LREE (majority of Chalcolithic domestic ceramics and some Neolithic ones). Quartzodiorite derived clays were probably the most used raw material for those groups, as well as diorites and associated gabbros, and in some cases Tertiary clays. On the other hand, a third group is explained specially by Fe, Sc, Cr and Co (mainly funerary ceramics of tomb 1 and one Neolithic sample). This group, which is the smallest, is the only one, which points to the use of the weathered schists far from Perdigões, as mentioned above. In addition, there are some outliers in tomb 1 ceramics. The diachronic approach of compositional features of ceramics from the Neolithic to the Chalcolithic reinforces the resource to local clay raw materials.

The bell beakers samples when analyzed separately have a chemical heterogeneity shown by a dispersion of values of chemical elements contents. Nevertheless, two main tendencies can be observed, some ceramics have high contents of REE, K, Rb, Cs, Th and U, others high contents of Fe, Sc, Cr, Co and Zn, and 3 samples are completely different

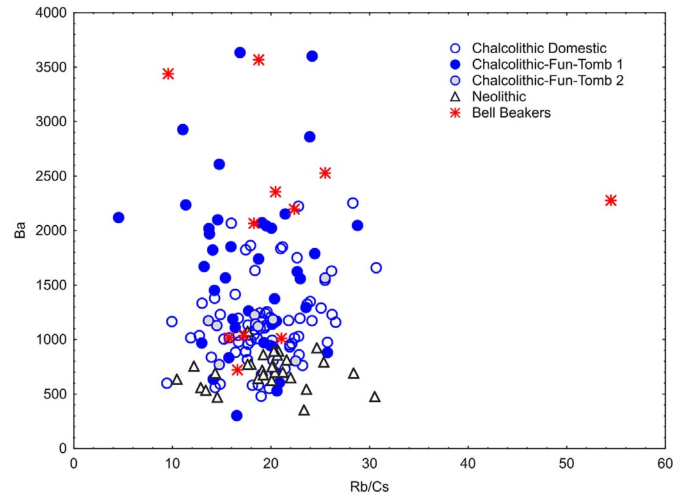

Fig. 4. Bivariate plot of Ba vs. Rb/Cs in ceramic samples from the Chalcolithic and Neolithic contexts, the domestic and funerary ceramics (Fun) and the bell beakers from Perdigões site.

with high values of As, Rb and Cs, and low values of REE. This chemical heterogeneity observed in the bell beakers from Perdigões reinforces the hypothesis of diverse provenances with the resource to different kinds of raw materials.

A comparison of the chemical composition of the bell beakers with all the other typologies of the diverse archaeological contexts studied in Perdigões, highlights the higher dispersion of chemical contents observed in bell beakers (Fig. 4), like also observed for ceramics of the Chalcolithic funerary contexts, particularly of tomb 1. This plot can be used as a reference for further comparative studies of other bell beakers, and/or other typologies from the same site or other contemporaneous archaeological sites. Some bell beakers have chemical composition similar to the other typologies, associated with local raw materials close to the archaeological site, such as the clays resulting from the weathering of diorites and gabbros, quartzodiorites, and Tertiary clays. Others bell beakers are associated with the tomb ceramics that used clays resulting from weathered schists, far from Perdigões (these ceramics are the ones with higher phyllosilicate contents, with finer textures). Only a minority of bell beakers have distinct chemical composition, moreover with no correlation to studied raw materials.

4.4. The Bell Beakers from Monte do Tosco, Alentejo, Southern Portugal

Ceramics from Monte do Tosco are divided in three groups in relation to its chemical composition (Fig. 5) (Table S4 in the Supplementary materials). The importance of this subdivision lies on the coincidence of each of those groups with a certain chronology. Group one (1) embraces 80% of Chalcolithic ceramics; the second group (2) comprises all the Bronze bell beakers (60%) and other Bronze and Chalcolithic ceramics; the third group (3) is mainly composed of Bronze Age ceramics (80%).

Chalcolithic ceramics can be differentiated from Bronze Age ceramics by a chemical composition more correlated with the basic rocks of the region, namely with clays derived by weathering from quartzodiorites, diorites and gabbros.

Chemical composition of bell beakers is more homogeneous than other Bronze Age ceramics, presenting higher amounts of Hf and less of Fe, Sc, Cr, Co and REE. Bell beakers and some other Bronze Age ceramics present a similar chemical composition, with a correspondence with clays derived by weathering from schists and in a few cases with Tertiary clays.

Hence, at Monte do Tosco site, a local production for all ceramic typologies can be delineated, including bell beakers. Still a few outliers are defined comprising the analyzed crucibles and three bell beakers,

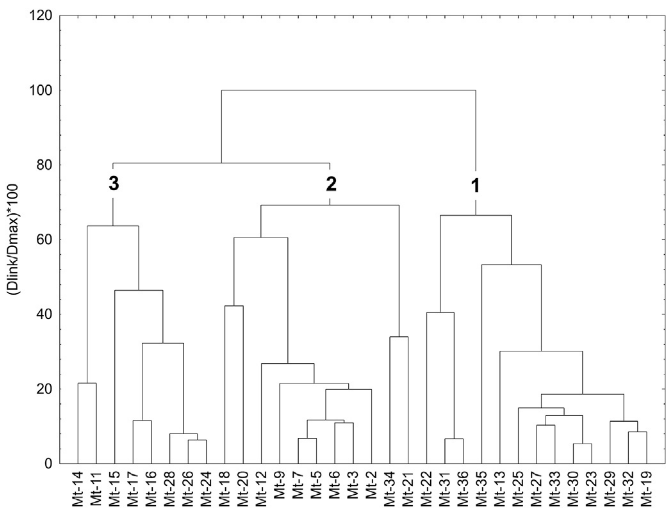

Fig. 5. Tree diagram for Monte do Tosco ceramics by using the UPGMA (Unweighted pair-group average) as amalgamation (linkage) rule and the Pearson coefficient as distance measure. Group 2 includes the Bell Beakers.

with a diverse chemical composition, with much higher REE, Hf and Zr contents, pointing to the use of different raw materials from the local/regional resources, probably a different provenance.

5. Conclusions

The archaeometric study by means of geochemistry and mineralogy of bell beakers and other typologies from four archaeological sites of central and southern Portugal (Beira-Alta: FP; Alentejo: PT, MT and PRD) yields groups of ceramic sherds comprising various typologies with similar composition at each site, indicating mostly a local provenance. Potential raw materials were also analyzed, compared with those ceramics and attribution of provenance was achieved in many cases. Trace elements composition together with clay minerals association obtained in both ceramics and raw clays have had a crucial role when discussing the provenance and circulation of bell beakers.

Undoubtedly, in the studied cases, the methodological approach by using interdisciplinary sciences combined to solve the bell beakers mechanisms of circulation problem became very useful and of greatest efficiency, as it contributes to establish which ceramics were produced locally, and which ones were imported. In the studied cases, local production was ascertained for the majority of the cases, nevertheless some exogenous ceramics exist. In general, results point to the circulation of the "*Idea*" *and* "*prototypes*", as local productions are clearly identified (even some with a careful production technology). Nevertheless, importations of Beakers occur, but local productions seem to prevail. In three archaeological sites, most of the defined outliers are bell beakers, pointing to an exogenous provenance. Considering that the transport of these fragile ceramic objects over great distances would be demanding, it enhances the importance of these typologies over a certain period.

This approach helps to conceptualize the diffusion models of this typology, for it recognizes bell beaker local assimilations through copy or stylistic recreation. In hinterland areas, such as the ones of the analyzed sites, the results suggest that beakers are an addition to local social processes and were integrated and reproduced locally. Although they maintain the main original stylistic patterns in the South (no clear local styles have been identified for beakers in that region), while in Beira Alta some combed decorations patterns may represent local reinterpretations.

For each studied site, significant compositional variations of pottery sherds, divided by groups were identified, that appear to reflect differences in the geographical locations and the geology of clay sources, thus the pottery found in each specific site possesses distinct geochemical signatures.

A relatively large number of sherds having a similar composition indicate either the use of raw materials from a single source, or the production of a consistently uniform ceramic paste over time. On the other hand, in some cases, multiple regional sources were defined. This is relevant, especially because there is a tendency for the small sites to present just one specific beaker style (i.e. Monte do Tosco, Valera, 2007) while the large enclosures (i.e. Porto Torrão or Perdigões) show influences of several beaker styles. At Perdigões beakers are kept out of funerary contexts, that also do not present any purpose of individualization, contrasting to what is known in many European regions (Czebreszuk, 2014; Fitzpatrick, 2011; Heyd, 2007; Hofman and Bickle, 2011). Indeed decorated beakers also rarely appear in funerary contexts (Valera et al., 2014b, 2014c) and the same general situation has been identified in the neighboring areas of Spanish Extremadura and Western Andalusia (García Rivero, 2006; Hurtado, 2005; Lazarich González, 2005). Indeed, the fact that in Alentejo decorated beakers rarely appear in funerary contexts, seems to indicate that they have their main social use outside graves, contrasting with what happens in Portuguese Estremadura and in the central – north Portugal, that reveal situations closer to what happens in continental Europe (Prieto Martínez and Salanova, 2015; Salanova, 2011, 1998). In this sense, it is essential to place variability at the core of the bell beaker phenomenon, as we have found multiple regional facets of material culture for the studied sites. This period must be set in terms of extended sociability and sharing of common traditions and values (Linden, 2007, 2004).

Thus, the obtained results are in accordance with other contextual observations, indicating that Bell Beakers, more than an homogeneous phenomena, correspond to a material expression (recursively active) of complex and asymmetric social relations, generating considerable regional and contextual variances, assuming diversified social roles in different areas. In this scenario, local productions are in relation with interregional systems of circulation of ideas and materials that reached their pick of development in the middle/third quarter of the 3rd millennium BC, especially in southern Iberia, before a generalized collapse.

The potential role of bell beakers in the spread of new ideas and technologies in parallel with their local productions reflects a juxtaposition of knowledge systems existing in the 3rd millennium BC in Europe.

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