



The supply of ceramics to Portuguese North African strongholds in the 15th and 16th centuries: New archaeometric data from Ksar Seghir and Ceuta

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ABSTRACT

The present study aims to present new archaeometric data from a wide typological rank of ceramics collected in Ksar Seghir (Morocco) and Ceuta (Spain), two different archaeological sites in the south bank of the Strait of Gibraltar occupied by the Portuguese from the 15th to the middle of the 16th centuries. We characterise and illustrate the most common ceramic fabrics and shapes found in these settlements, only possible by the intensive excavation of these two sites. Its mineralogical and chemical analyses confirm the idea that Seville and Lisbon were the most significant production centres in the Iberian Peninsula to supply the two Portuguese North African strongholds. These results reinforce the idea that Seville and Lisbon, besides being two great pottery workshops, played a complementary role as key cities in the logistics of the Iberian overseas expansion since its early beginning. The combination of the typological and archaeometrical studies will allow to better identify the centres of production of these ceramics, which were widely disseminated in the Mediterranean Sea and the Atlantic Ocean during this period.

1. Introduction

This study addresses the chemical and mineralogical characterization of the fabrics of 44 ceramics from Ksar Seghir (Morocco) and 42 from the Murallas Reales [Royal Walls] of Ceuta (Spain). These archaeological sites are located on the south bank of the Strait of Gibraltar, some 20 km apart (Fig. 1). Both were important settlements during the Middle Ages under the Almoravid, Almohad and Marinid dynasties, conquered by the Portuguese in 1415 and 1458 respectively. The difference is that Ceuta was, and still is, a great city since classical times, while Ksar Seghir was founded in the 12th century and was left in ruins after the Portuguese abandoned it in 1550, being a visitable archaeological site today.

The first relevant archaeometrical studies on the Ksar Seghir site were conducted by Charles L. Redman, heading an American-Moroccan team, between 1974 and 1981. These works, which involved the excavation of about 18% of the site, resulted in the identification of two major

occupation phases, a medieval Islamic phase and a Portuguese phase (synthesis in Redman, 1986). As far as the study of archaeological materials from the Portuguese levels is concerned, Redman's team performed some general approaches to the ceramic materials. The description of the most significant types combines information about the history of the site and the evolution of the excavations (Redman et al., 1978, 1979, 1980; Redman and Boone, 1979). At the same time, more specific studies on ceramics and their spatial setting were carried out, originating a number of monographs (Boone, 1980; Sinopoli, 1980; Redman: 190-201, 1986) and articles (Redman, 1980a, 1980b, 1982; Boone, 1984). A theoretical reflection on methodological approaches to medieval ceramics was also undertaken (Redman, 1979).

The first archaeometrical analyses, focused on Islamic productions, were conducted at this stage, involving the analysis of 43 conical plates dated to between the 13th and 15th centuries, from consuming contexts, and five trivets from a kiln found at Ksar Seghir (Myers and Blackman, 1986: 55). This ceramic kiln would have been used during the Marinid

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Fig. 1. Location of the main sites discussed.

occupation, but was dismantled before the Portuguese arrival (Redman, 1986: 71). Using petrography and chemical analysis by neutron activation, three large chemical groups were recognized, revealing one probable local production, directly related to Ksar Seghir, and other two from unknown sites, but possibly related to the Strait of Gibraltar region (Myers and Blackman, 1986: 61-64). A second archaeometrical study, conducted in the wider context of the worldwide diffusion of Sevillian pottery during late 15th and 16th centuries, included the analysis of 11 shards from Ksar Seghir and established a connection between the findings of tin lead-glazed earthenware painted in blue or in blue and purple and the pottery workshops of Triana (Myers et al., 1992: 136-137).

Since 2012, a Portuguese-Moroccan mission, coordinated by Abdelatif El-Boudjaj and André Teixeira, has resumed research in Ksar Seghir, with the excavation of new areas and the review of data from the previous interventions. Regarding ceramic materials, one of the objectives of this project is to broaden the knowledge of the typological series, narrowing down their chronological periods, detailing areas of provenance and commercial routes, as well as establishing a clearer connection between objects and archaeological contexts (Teixeira et al., 2013, 2016; Teixeira and Torres, 2018). This research also includes new studies on the archaeometry of ceramics, benefiting from the contributions of the last decades in this subject area, concerning the regions of Seville and Lisbon (Iñáñez, 2007; Iñáñez et al., 2008, 2009; Fernández de Marcos et al., 2017; Gómez Ferrer et al., 2013). The ceramic samples for this archaeometrical study were gathered between 2015 and 2016 from the site's reserves and are illustrative of the most important types documented archaeologically in Ksar Seghir. They all belong to the same excavation area (E17N10-E18N10-E18N9), inhabited over the centuries, with a clear stratigraphic sequence and whose materials have been studied in depth (Teixeira et al., 2016).

Concerning Ceuta, archaeological activity has been intense in the last decades, namely regarding the medieval period, with numerous discoveries revealing the city's extensive urban perimeter and the richness of its structures and material culture, largely destroyed by the Portuguese conquest. The archaeometrical study carried out in Ceuta is related to the Puerta Califal [Caliph Gate] site, part of the Murallas Reales of Ceuta, the city's main defensive curtain, dividing it from the

interior of the territory. The *cerca* [wall] was built by the middle of the 10th century on the initiative of the Caliphate of Córdoba, and its main gate remained in use throughout the Middle Ages, including the first century of Portuguese occupation (Villada Paredes and Gurriarán Daza, 2013). The medieval wall and gate were hidden in the 1540s, when the Portuguese built a bastioned curtain, which nevertheless kept the previous structure intact, as an internal structure of the new fortification. This new design involved the excavation of a large wet moat, where urban waste accumulated over the centuries, being eventually incorporated into the space between the wall facings. Thus, the excavation of these inner spaces resulted in the recovery of archaeological materials that constitute a good repository of the material culture of Ceuta prior to the middle of the 16th century, including of course a large amount of waste corresponding to the Portuguese occupation. Several interventions have been carried out near the Puerta Califal (Villada Paredes, 2012), the most recent and substantial one took place in 2013, covering an area of 115 m² and reaching a depth of about 10 m, from the wall-walk of Ceuta's Murallas Reales to the bedrock (Villada Paredes et al., 2016). The archaeological materials recovered during this excavation were studied by the authors in 2014 and 2015 and are about to be published, samples for archaeometrical analysis were selected among the most representative groups. To sum up, the 86 samples analysed belong to broadly coeval archaeological contexts. Thus, the ceramic materials of Ceuta are dated to between 1415 and the 1540s, in Ksar Seghir the datings range from 1458 to 1550. These two cities of the southern shore of the Strait of Gibraltar probably featured similar supply and economic activities, according to historical and archaeological data.

2. The ceramic assemblage

Our study focused on four ceramic types and their most significant subtypes: tin lead-glazed earthenware (without decoration, with decoration in blue, in blue and purple, and half-dipped white and green), lead-glazed earthenware (honey, green and honey-brown), unglazed red earthenware (without finishing, with red slip or also burnished) and unglazed beige earthenware. This differentiation is made according to their surface finishing characteristics, primarily separating the glazed from the unglazed ceramics. In the glazed group we are able to

Table 1
Ceramic sample description.

ANID	Site	Chemical Group	Type	Form	Chronology
CTA001	Ceuta	SEV-B	unglazed beige	big jar	1415–1540
CTA002	Ceuta	SEV-B	unglazed beige	small jar	1415–1540
CTA003	Ceuta	SEV-B	unglazed beige	basin (lebrillo)	1415–1540
CTA004	Ceuta	SEV-B	unglazed beige	small jar	1415–1540
CTA005	Ceuta	SEV-B	unglazed beige	small jar	1415–1540
CTA007	Ceuta	Lisboa2	unglazed red	jar	1415–1540
CTA008	Ceuta	Lisboa2	unglazed red	pot	1415–1540
CTA009	Ceuta	SEV-B	tin–lead glazed	plate	1415–1540
CTA010	Ceuta	SEV-B	tin–lead glazed	plate	1415–1540
CTA011	Ceuta	SEV-B	tin–lead glazed	plate	1415–1540
CTA013	Ceuta	SEV-B	honey lead-glazed	plate	1415–1540
CTA014	Ceuta	SEV-B	honey lead-glazed/ brown	plate	1415–1540
CTA016	Ceuta	SEV-B	honey lead-glazed/ brown	plate	1415–1540
CTA020	Ceuta	SEV-B	honey lead-glazed	plate	1415–1540
CTA028	Ceuta	Lisboa2	unglazed red	cup	1415–1540
CTA030	Ceuta	Lisboa2	unglazed red	jar	1415–1540
CTA032	Ceuta	Lisboa2	unglazed red	pot	1415–1540
CTA033	Ceuta	Lisboa2	unglazed red	pot lid	1415–1540
CTA035	Ceuta	Lisboa1	unglazed red	pan	1415–1540
CTA036	Ceuta	Lisboa2	unglazed red	pot lid	1415–1540
CTA040	Ceuta	SEV-B	tin–lead glazed	bowl	1415–1540
CTA042	Ceuta	SEV-B	tin–lead glazed/blue	basin (lebrillo)	1415–1540
CTA043	Ceuta	SEV-C	tin–lead glazed/blue/ purple	bowl	1415–1540
CTA044	Ceuta	SEV-C	tin–lead glazed	bowl	1415–1540
CTA045	Ceuta	SEV-C	tin–lead glazed/blue	plate	1415–1540
CTA046	Ceuta	SEV-C	tin–lead glazed/blue	plate	1415–1540
CTA047	Ceuta	SEV-C	tin–lead glazed/blue/ purple	plate	1415–1540
CTA050	Ceuta	SEV-C	green lead-glazed	basin (lebrillo)	1415–1540
CTA051	Ceuta	SEV-C	green lead-glazed	basin (lebrillo)	1415–1540
CTA052	Ceuta	SEV-C	green lead-glazed	basin (lebrillo)	1415–1540
CTA053	Ceuta	SEV-C	green lead-glazed	basin (lebrillo)	1415–1540
CTA054	Ceuta	SEV-C	green lead-glazed	chamber pot	1415–1540
CTA055	Ceuta	SEV-C	tin–lead glazed/green	bowl	1415–1540
CTA056	Ceuta	SEV-C	green lead-glazed	mortar	1415–1540
CTA057	Ceuta	SEV-C	tin–lead glazed/green	bowl	1415–1540
CTA058	Ceuta	SEV-C	tin–lead glazed/green	plate	1415–1540
CTA059	Ceuta	SEV-C	honey lead-glazed	bowl	1415–1540
CTA061	Ceuta	SEV-C	honey lead-glazed	chamber pot	1415–1540
CTA062	Ceuta	SEV-C	honey lead-glazed	bowl	1415–1540

Table 1 (continued)

ANID	Site	Chemical Group	Type	Form	Chronology
CTA063	Ceuta	SEV-C	honey lead-glazed	picther	1415–1540
CTA064	Ceuta	SEV-C	honey lead-glazed	picther	1415–1540
CTA065	Ceuta	SEV-C	honey lead-glazed	bowl	1415–1540
KSG001	Ksar Seghir	SEV-B	green lead-glazed	basin (lebrillo)	1458–1550
KSG002	Ksar Seghir	SEV-B	green lead-glazed	basin (lebrillo)	1458–1550
KSG003	Ksar Seghir	SEV-B	green lead-glazed	basin (lebrillo)	1458–1550
KSG004	Ksar Seghir	SEV-B	honey lead-glazed	basin (lebrillo)	1458–1550
KSG005	Ksar Seghir	SEV-B	honey lead-glazed	plate	1458–1550
KSG006	Ksar Seghir	SEV-B	honey lead-glazed/ brown	plate	1458–1550
KSG007	Ksar Seghir	SEV-B	honey lead-glazed/ brown	plate	1458–1550
KSG008	Ksar Seghir	Lisboa2	unglazed red	basin (lebrillo)	1458–1550
KSG010	Ksar Seghir	Lisboa1	unglazed red	pot	1458–1550
KSG012	Ksar Seghir	SEV-B	tin–lead glazed	bowl	1458–1550
KSG013	Ksar Seghir	SEV-B	honey lead-glazed	chamber pot	1458–1550
KSG015	Ksar Seghir	Lisboa2	unglazed red	pot	1458–1550
KSG016	Ksar Seghir	SEV-B	tin–lead glazed	bowl	1458–1550
KSG017	Ksar Seghir	SEV-B	unglazed beige	big jar	1458–1550
KSG019	Ksar Seghir	SEV-B	tin–lead glazed/green	plate	1458–1550
KSG020	Ksar Seghir	SEV-B	tin–lead glazed	plate	1458–1550
KSG021	Ksar Seghir	SEV-B	unglazed beige	mortar	1458–1550
KSG022	Ksar Seghir	SEV-B	unglazed beige	basin (lebrillo)	1458–1550
KSG023	Ksar Seghir	SEV-B	unglazed beige	small jar	1458–1550
KSG024	Ksar Seghir	SEV-B	unglazed beige	big jar	1458–1550
KSG025	Ksar Seghir	SEV-B	unglazed beige	big jar	1458–1550
KSG026	Ksar Seghir	SEV-B	unglazed beige	big jar	1458–1550
KSG027	Ksar Seghir	Lisboa2	honey-brown lead-glazed	pan	1458–1550
KSG028	Ksar Seghir	Lisboa2	honey-brown lead-glazed	pan	1458–1550
KSG030	Ksar Seghir	Lisboa2	honey-brown lead-glazed	pan	1458–1550
KSG031	Ksar Seghir	Lisboa2	honey-brown lead-glazed	pan	1458–1550
KSG032	Ksar Seghir	Lisboa2	lead-glazed unglazed red	pot	1458–1550
KSG033	Ksar Seghir	Lisboa2	honey-brown lead-glazed	pan	1458–1550
KSG034	Ksar Seghir	Lisboa2	honey-brown lead-glazed	pan	1458–1550
KSG037	Ksar Seghir	Lisboa2	unglazed red	pot	1458–1550
KSG039	Ksar Seghir	Lisboa2	unglazed red	pot lid	1458–1550
KSG040	Ksar Seghir	Lisboa2	unglazed red	pan	1458–1550
KSG042		SEV-B		bowl	1458–1550

(continued on next page)

Table 1 (continued)

ANID	Site	Chemical Group	Type	Form	Chronology
	Ksar Seghir		tin-lead glazed/blue/purple unglazed red	basin (lebrillo)	1458–1550
KSG043	Ksar Seghir	Lisboa2	tin-lead glazed	plate	1458–1550
KSG044	Ksar Seghir	SEV-B	tin-lead glazed	plate	1458–1550
KSG045	Ksar Seghir	SEV-B	tin-lead glazed/blue/purple	plate	1458–1550
KSG046	Ksar Seghir	SEV-B	tin-lead glazed/green	plate	1458–1550
KSG048	Ksar Seghir	SEV-B	honey lead-glazed	bowl	1458–1550
KSG049	Ksar Seghir	SEV-B	honey lead-glazed	bowl	1458–1550
KSG050	Ksar Seghir	SEV-B	honey lead-glazed/brown	plate	1458–1550
KSG051	Ksar Seghir	SEV-B	honey lead-glazed	pitcher	1458–1550
KSG052	Ksar Seghir	Lisboa1	unglazed red	pan	1458–1550
KSG053	Ksar Seghir	Lisboa1	unglazed red	pan	1458–1550
KSG054	Ksar Seghir	SEV-B	honey lead-glazed	pitcher	1458–1550

macroscopically distinguish the opaque glaze, that commonly mix lead oxide, silica and a portion of tin in its chemical composition, and the transparent glaze, which in this case use basically lead oxide and silica (Lister and Lister: 80-87, 1982; Picon et al.: 42-45, 1995). In fact, although we did not chemically test these samples' glazes, past research on similar ceramic typologies has shown their chemical composition (Iñáñez, 2007; Tite et al., 1998; Molera et al., 1999). For this type of ceramics, the analytics made until today show the common use of: cobalt for blue, manganese for purple and brown, iron for honey, and copper for green (Molera et al., 1997; Molera et al., 2013; Iñáñez et al., 2013; Pérez-Rodríguez et al., 2020; Sanchez-Garmendia et al., 2020)

The more frequent forms at both sites were analysed within these types, in this paper, the samples corresponding to each subtype are identified and drawings of the equivalent complete forms found in archaeological contexts from Ksar Seghir or Ceuta's Murallas Reales are provided (Table 1).

2.1. Tin lead-glazed earthenware

Thus, the tin lead-glazed earthenware (Fig. 2), with its white opaque surface, includes undecorated exemplars (KSG012, KSG016, KSG020, KSG044, CTA009, CTA010, CTA040), but also decorated in blue (CTA042, CTA045, CTA046), in blue and purple (KSG042, KSG045, CTA043, CTA044, CTA047), or half-dipped white and green (KSG019, KSG046, CTA055, CTA057, CTA058). This ceramic technological type is usually associated with tableware, such as plates and bowls. There are two types of plates: one is truncated conical, with an upright rim and omphalos base (KSG019, KSG020, KSG044, KSG045, CTA009, CTA010, CTA045, CTA047, CTA058), and the other is hemispherical, with a broad rim and an omphalos base (CTA046). Bowls are hemispherical, carinated, with a slightly everted rim and a foot-ring (KSG012, KSG016, KSG042, CTA040, CTA043, CTA044, CTA055, CTA057). The fabrics of these fragments have light colours, from beige (KSG016, KSG019, KSG020, KSG042, KSG044, CTA009, CTA010, CTA043, CTA044, CTA045, CTA046, CTA047, CTA055, CTA057, CTA058), to light pink (KSG012, CTA040) and light orange (KSG045, KSG046, CTA042), with a small amount of very fine to medium temper. The less frequent *lebrillos* [basins], truncated conical shaped, have an everted and thickened rim,

and flat base (CTA042).

2.2. Lead-glazed earthenware

Lead-glazed ceramics show honey (Fig. 3) and green colours (Fig. 4). Honey glazed types include plates, bowls, pitchers, *lebrillos* and chamber pots. The plates are truncated conical, with an upright rim and omphalos base (KSG005, KSG006, KSG007, KSG050, CTA013, CTA014, CTA020), while bowls are hemispherical, carinated, with an upright rim and omphalos base (KSG048, KSG049, CTA059, CTA062, CTA065). Both forms are integrally glazed and their fabrics are usually beige (KSG005, KSG048, CTA013, CTA059, CTA065), light pink (KSG049) or orange (KSG006, KSG007, KSG050, CTA014, CTA020, CTA062). Pitchers are elliptical, with an upright rim and a long neck, more or less grooved, with a vertical handle and flat base (KSG051, KSG054, CTA063, CTA064). The *lebrillos* are truncated conical, have a flat base and an everted and thickened rim (KSG004). Chamber pots are cylindrical or truncated conical, with a flat base and a broad rim, with two vertical handles (KSG013, CTA061). In these three cases, the internal surfaces are fully coated, as opposed to the external surfaces, which are only partially covered, leaving the lower part of the piece unglazed. The surfaces are light coloured, but the fabrics range from beige (KSG051, CTA063) to orange (CTA061), light pink (KSG004, KSG013, CTA064) and light orange (KSG054), with few fine to small temper. The inner surface of plates, bowls and *lebrillos* is sometimes decorated in underglaze manganese (KSG006, KSG007, KSG050, CTA014). This underglaze technique has been proven by the appearance of several of ceramic production wastes in archaeological sites in Seville and also in Jerez de la Frontera (Spain) (Pleguezuelo, 1997: 356), where one can observe the biscuit fragments painted with manganese decoration, previous to the application of the lead-glaze. The use of the technique is attested in the Islamic Mediterranean since the 9th century (Salinas and Pradell, 2018: 9), and in the Iberian Peninsula since the 10th century (Salinas, in press).

Green lead-glaze can be found on plates, bowls, cups, mortars and *lebrillos*. The plates and bowls seem to be part of the half-dipped white and green pieces and were therefore previously mentioned. The cups have a flat base, with a globular body, and a long and flaring neck, they show a tin-glaze on the inner surface, only reaching the middle of the body on the outer surface and overlaid by the green glaze (CTA011). The *lebrillos* feature the same formal characteristics as the honey lead-glazed (KSG001, KSG002, KSG003, CTA050, CTA051, CTA052, CTA053, CTA054), even if some exemplars show one or two excised diagonal lines in the rim (KSG001). The mortars are truncated conical, with a flat base and an inverted and thickened rim (CTA056). These fragments feature light coloured surfaces, but the fabrics range from beige (KSG003, CTA011, CTA051) and light pink (KSG001, CTA052, CTA054), to light orange (CTA056) and orange (KSG002, CTA050, CTA053), with few fine to small temper.

The assemblage of lead-glazed ceramics used for cooking foodstuffs, such as pots (KSG031, KSG034) and pans (KSG027, KSG028, KSG030, KSG033) (Fig. 5) was also analysed. The glaze is honey-brown, a designation used to include the different colour shades these pieces generally have, from dark honey, to orange, brown or even greenish (Pleguezuelo: 49, 1993; Gutiérrez: 53, 2000). These variations are macroscopically justified by the colour of each object fabric, although further research could clarify or propose other explanations. This effect is probably due to small changes resulting from processes of reduction and oxidation of Fe during firing in the kiln (for an in-depth discussion, see Molera et al., 1997). The use of these objects over the fire also contributes to the alterations in their appearance. Unlike the previous ones, these ceramics feature less purified and grainier fabrics, with a medium amount of small-sized temper, and dark colours, between orange (KSG027, KSG028, KSG030, KSG033, KSG034) and red (KSG031). Pots have a globular body, with an upright rim framed by incised lines on the exterior, with two vertical handles, starting directly at the rim.

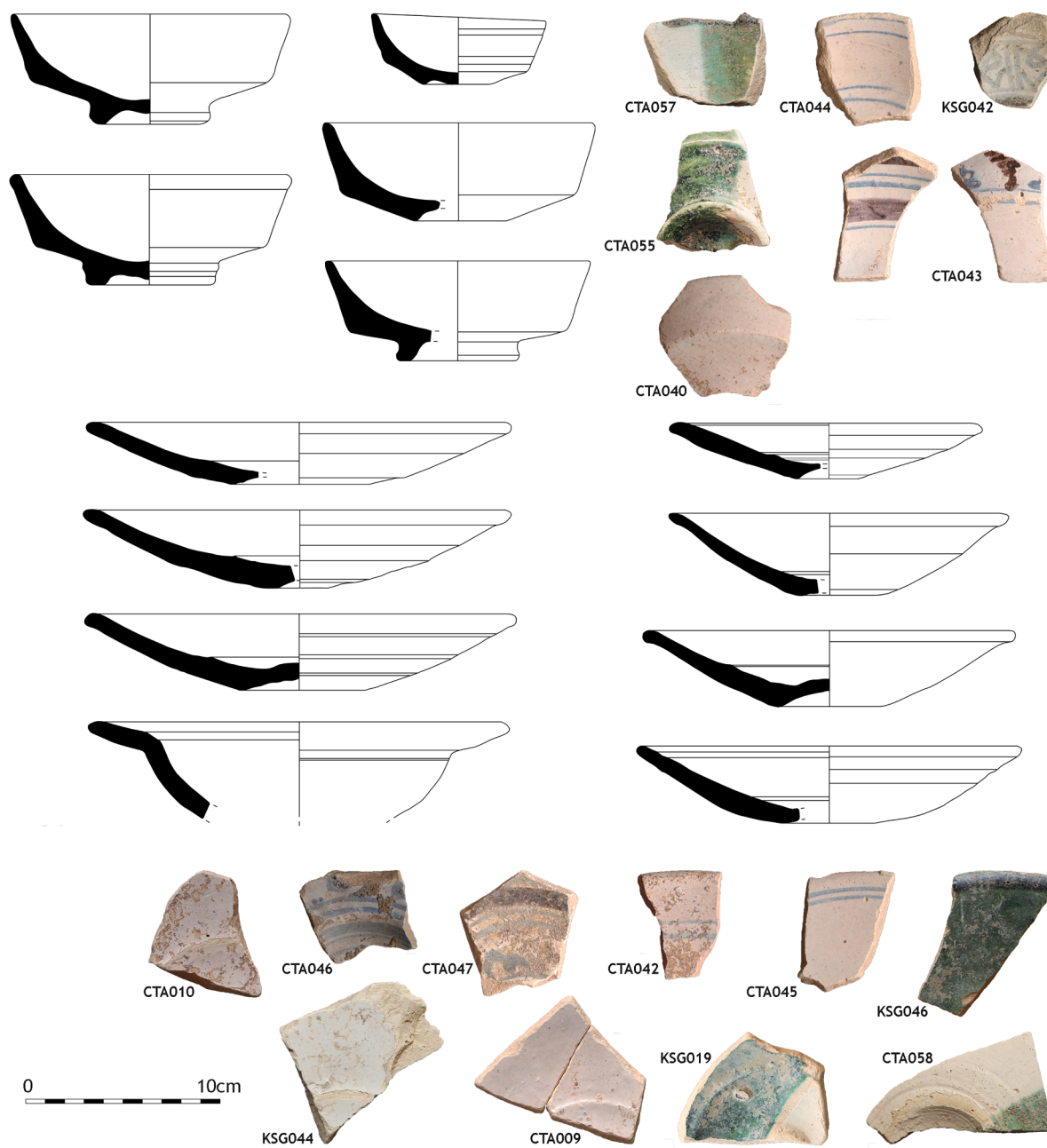


Fig. 2. Main types of tin-lead glazed ceramics.

Pans are truncated conical, with an everted and thickened or bifid rim. Some exemplars have very small vertical handles, basically decorative (KSG027).

2.3. Unglazed red earthenware

The unglazed earthenware is represented by two different groups, one with darker, reddish-coloured fabrics and another with lighter, beige-coloured fabrics, in the first group, we selected forms used for cooking foodstuffs, such as pans (KSG040, KSG052, KSG053, CTA035), pots (KSG010, KSG015, KSG032, KSG037, CTA032) and lids (KSG039, CTA033, CTA036), but also multi-functional objects, such as *lebrillos* (KSG008, KSG043, CTA008), or objects related to water storage and consumption, such as jars (CTA007, CTA030) and cups (CTA028) (Fig. 6). These pieces, like the glazed earthenware to be used over the fire, feature orange-coloured, grainy textured fabrics, with an medium

amount of small temper (KSG008, KSG010, KSG039, KSG040, KSG052, KSG053, CTA035), but also slightly finer, light orange to orange fabrics, a small amount of small-sized temper (KSG015, KSG032, KSG037, KSG043, CTA007, CTA008, CTA028, CTA030, CTA032, CTA033, CTA036). Despite the fact that these pieces are unglazed, in some cases a red slip was applied (KSG008, KSG015, KSG032, KSG037, KSG039, KSG040, KSG043, KSG052, KSG053, CTA007, CTA008, CTA030, CTA033, CTA036), sometimes burnished over the more visible surfaces (KSG008, KSG043, CTA008, CTA033).

Pots have a globular body, but two major types can be distinguished: one with an upright rim, framed by incised lines on the exterior, with vertical handles starting directly at the rim (KSG010), and another with a thickened rim, forming a square section, with two vertical handles (KSG015, KSG030, KSG032, KSG037, CTA032). Pans are hemispherical with an inverted rim (KSG040, CTA035) and, sometimes, with horizontal triangular handles. The lids are truncated conical and are

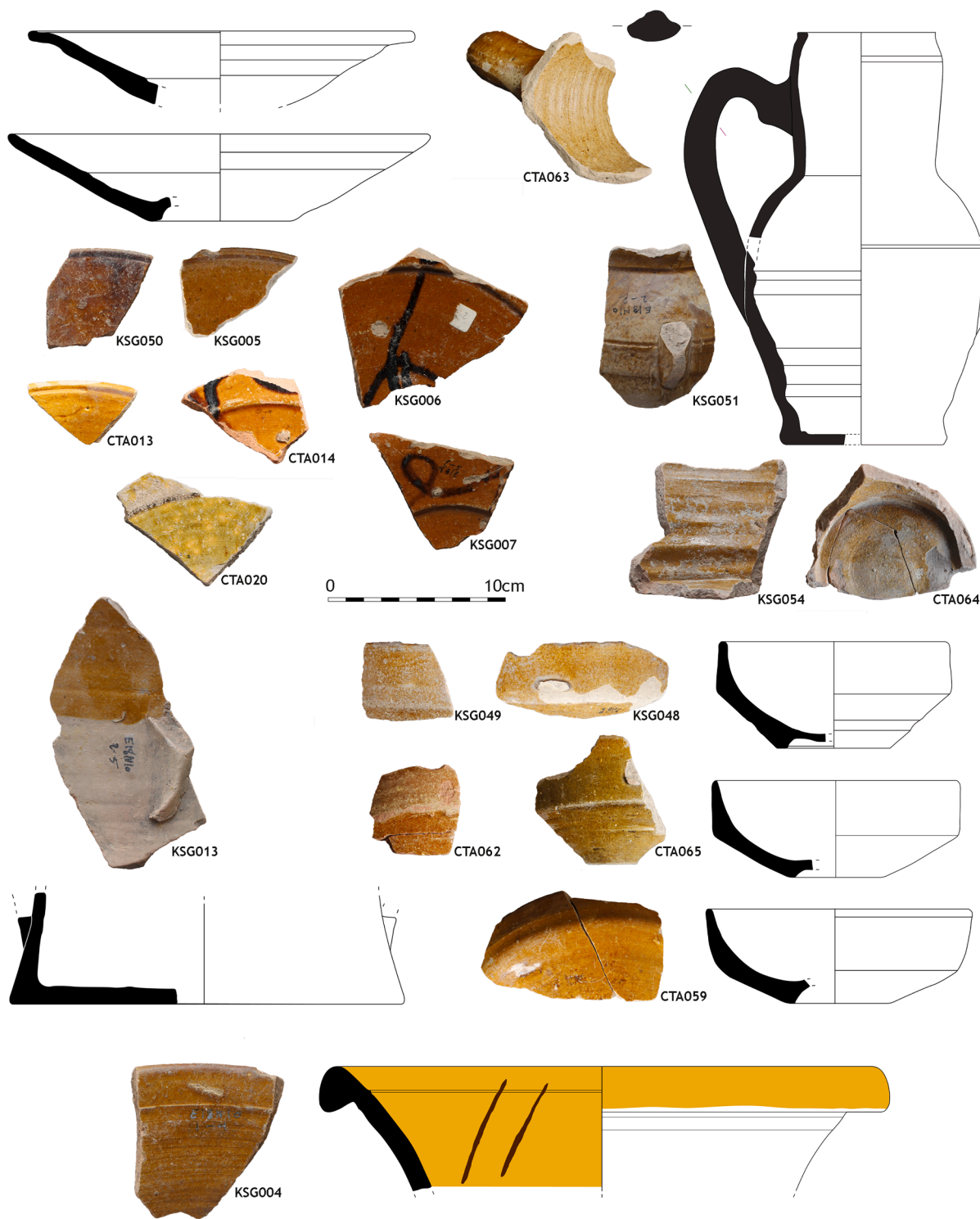


Fig. 3. Main types of honey lead-glazed ceramics.

represented by three different types: one with a flat base, an everted rim and a knob in the middle (KSG039), another with the same base and an inverted rim (CTA036), and a third, with a foot-ring and everted and thickened rim (CTA033).

2.4. Unglazed beige earthenware

The unglazed beige earthenware exemplars include pieces used for storage and transportation, such as the big jars (KSG017, KSG024, KSG025, KSG026, CTA001) and smaller jars (KSG023, CTA002,

CTA004, CTA005), along with mortars (KSG021) and the *lebrillos* (KSG022, CTA003) (Fig. 7). The big jars are large pieces, pyriform or oval, with a concave base, a straight and flaring neck, with an upright, or slightly everted, rim. The fabrics are beige (KSG024, CTA001) or light orange (KSG025, KSG026, CTA003), with visible small to medium temper, in large amounts, resulting in their grainy texture (KSG017). Mortars are truncated conical, with flat bases and inverted rims. The *lebrillos* documented herein are truncated conical with a broad rim, decorated with a wavy groove. The small jars are characterized by their long necks and upright rims. These pieces feature more purified fabrics,

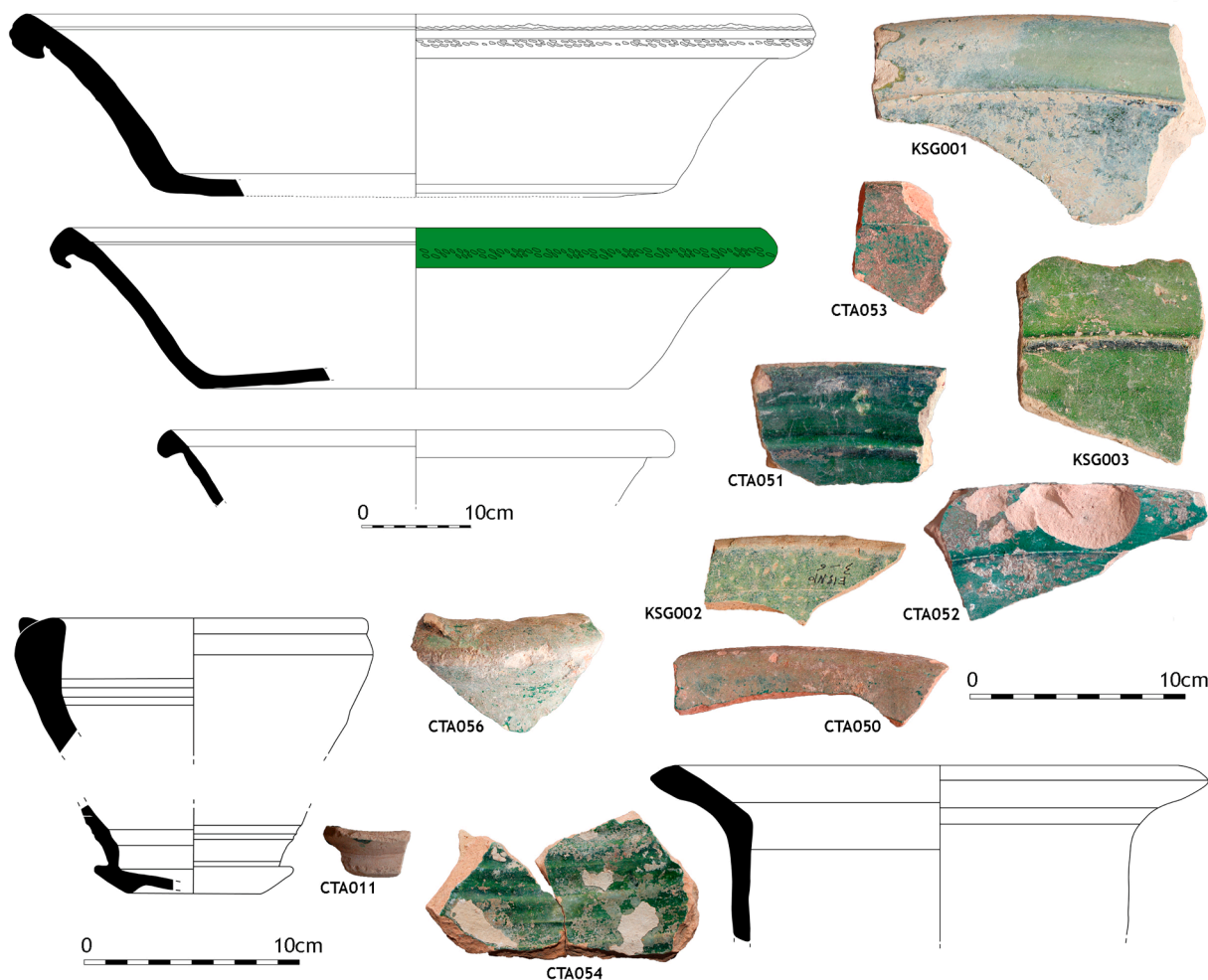


Fig. 4. Main types of green lead-glazed ceramics.

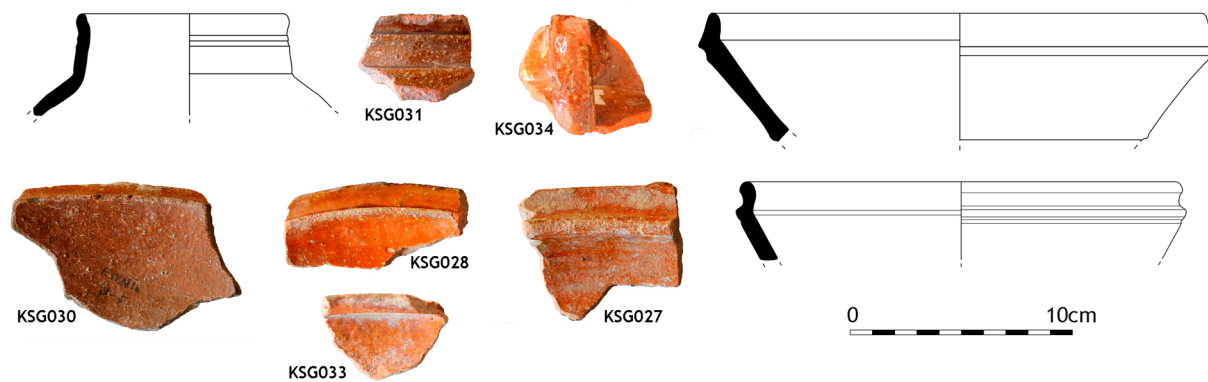


Fig. 5. Main types of honey-brown transparent lead-glazed ceramics.

with a medium amount of small temper, their colour ranges from beige (KSG022), to light pink (KSG021, CTA002, CTA005) and light orange (KSG023, CTA004).

3. Analytical routine and methodology

3.1. Inductively coupled plasma mass spectrometry (ICP-MS)

The chemical study of the 86 shards' ceramic fabrics (see Table S1) was carried out by means of inductively coupled plasma mass

spectrometry (ICP-MS).

In the current study, 10 g of each ceramic shard was powdered using a Fritsch pulverisette 6 (Fritsch GmbH) planetary milling machine equipped with tungsten carbide cells and balls for 4 min at 300 rpm. Glazes and exterior surfaces were mechanically removed prior to grinding by means of a tungsten carbide burr bit, leaving only the central core of the shard for chemical and mineralogical analysis. The removal of the glaze coats and outer surfaces minimizes the effect of contamination of the ceramic matrix (e.g. Iñáñez et al., 2008). Powdered specimens were stored in polyethylene vials for transport to the

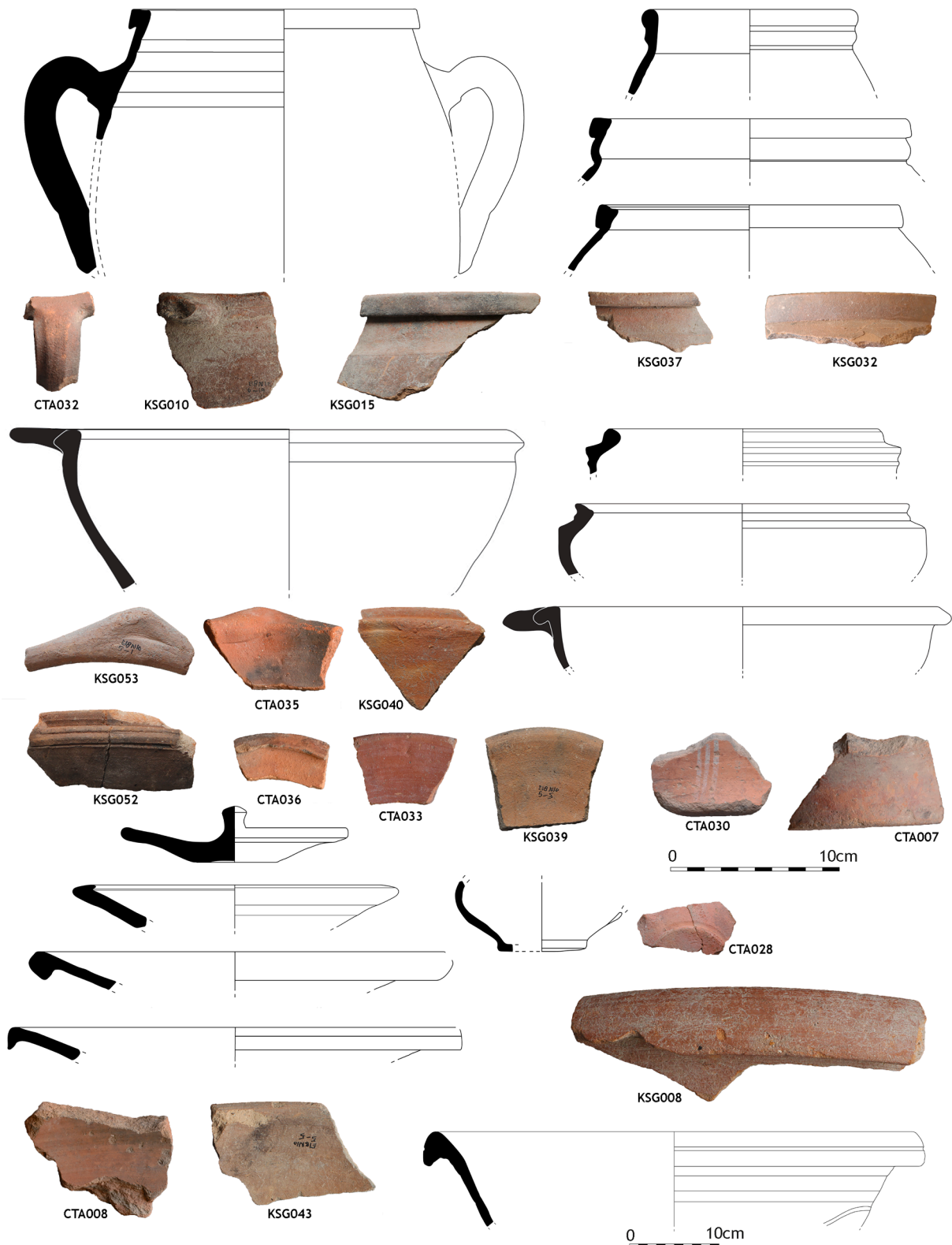


Fig. 6. Main types of red fabric wares.

laboratory.

Sample manipulation, reagents purification and ICP-MS analysis were carried out in a 100 clean laboratory. Both 250 mg of each powdered unknown sample and certified reference materials used for external calibration (CRM from the Geological Survey of Japan: andesite JA2, granodiorite JG-1,

granite JG-2, and basalt JB-3) were prepared for ICP-MS measurements by alkaline fusion using 500 mg LiBO_2 as flux agent in Pt-Au crucibles, followed by acid dissolution of the melt. ICP-MS chemical measurements were carried out using a NexION 300 ICP/MS (PerkinElmer, Ontario, Canada), provided with Rytan cross-flow nebulizer, Scott-type double pass spray chamber and

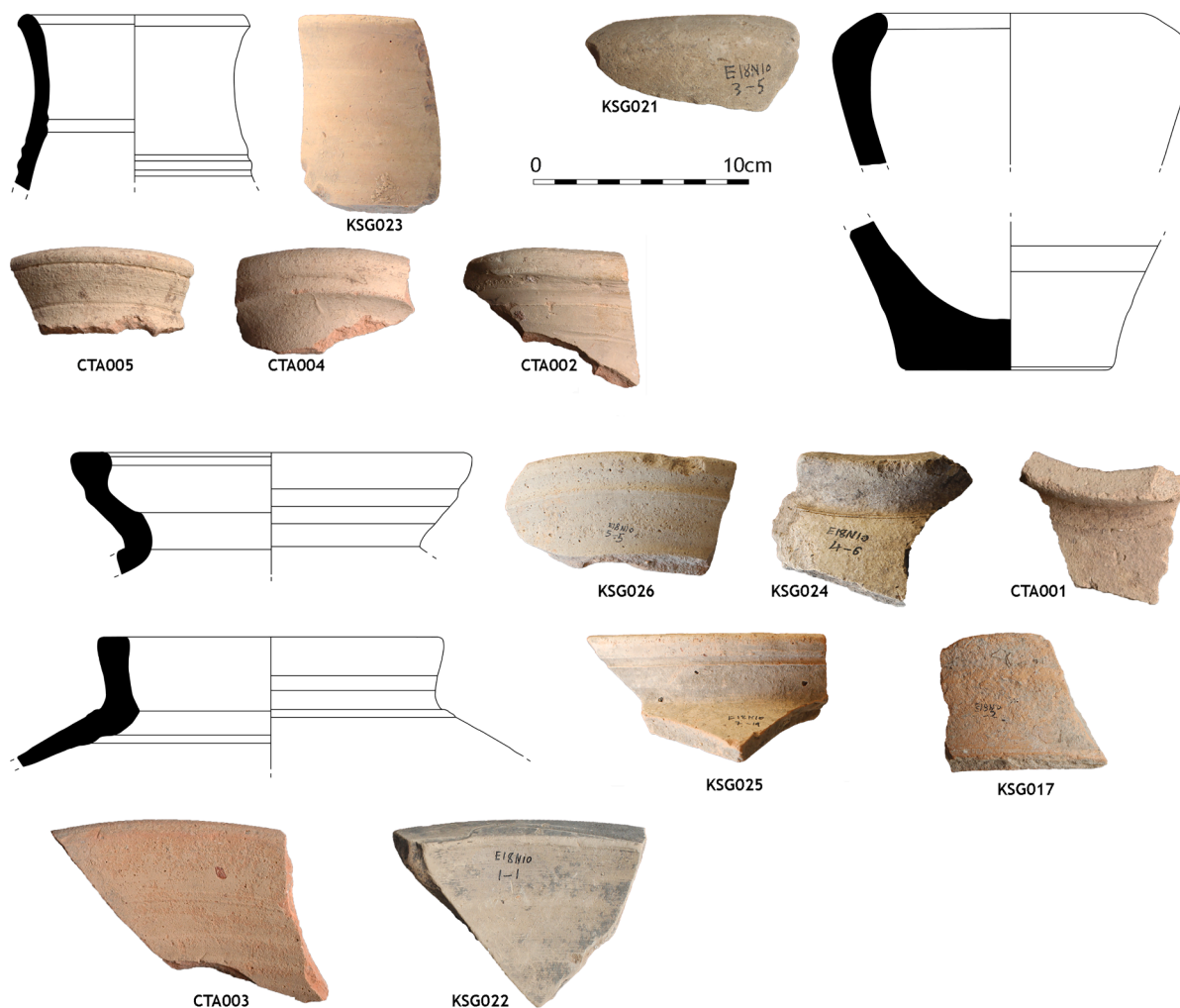


Fig. 7. Main types of beige earthenware.

standard nickel cones., using Argon as carrier gas, and equipment optimization was conducted every day before any batch of analyses with a 10 ng/mL standard solution of Mg, Rh, In, Ba, Pb and U. The concentrations of a wide range of analytes ^{27}Al , ^{31}P , ^{88}Sr , ^{120}Sn , ^{90}Zr , ^{93}Nb , ^{133}Cs , ^{137}Ba , ^{139}La , ^{140}Ce , ^{141}Pr , ^{142}Nd , ^{147}Sm , ^{153}Eu , ^{158}Gd , ^{159}Tb , ^{164}Dy , ^{165}Ho , ^{166}Er , ^{169}Tm , ^{174}Tb , ^{175}Lu , ^{180}Hf , ^{181}Ta , $^{206+207+208}\text{Pb}$, ^{232}Th and ^{238}U (Internal standards: In and Bi) were analysed in standard mode, while ^{23}Na , ^{24}Mg , ^{28}Si , ^{39}K , ^{44}Ca , ^{47}Ti , ^{51}V , ^{52}Cr , ^{55}Mn , ^{56}Fe , ^{59}Co , ^{60}Ni , ^{63}Cu and ^{66}Zn (Internal standard: In) were analysed in collision mode with He as cell gas. In the interest of space, quantitative results and analytical conditions of the ICP-MS chemical analysis are reported in Table S1 and S2, respectively (for a thorough description of the analytical procedure, see García de Madina-beitia et al., 2008; and Calparsoro et al., 2019).

3.2. X-ray diffraction (XRD)

Mineralogical phases of the 86 powdered ceramics were characterized by X-ray diffraction (XRD), employing a powder diffractometer PANalytical Xpert PRO, with a vertical goniometer with Cu-K α radiation ($\lambda = 1.5418 \text{ \AA}$) at 40 kV of voltage and 40 mA, with an angular range (2θ) scanned between 5 and 70° . Mineral phases present in the samples were identified using X'pert HighScore (PANalytical) software and ICDD mineral files from database PDF2 (International Centre for Diffraction Data, Pennsylvania, USA). The mineralogical identifications obtained by XRD have provided important information about the main firing phases, cryptocrystalline, from which it has been possible to calculate the

Estimated Firing Temperatures (EFT) and alteration and/or contamination phases.

3.3. Methodology

The statistical approach is based on Aitchison's and Buxeda's studies on compositional data (Aitchison, 2008, 1986; Aitchison et al., 2000; Buxeda and Kilikoglou, 2003; Buxeda, 1999). In this case, the statistical routine consisted on using ratios of logarithms obtained by the division of all the components of the data set (e.g. chemical elements here), by the component that introduces the lowest chemical variability to the entire set of ceramics (see Martín-Fernández et al., 2015) for a thorough discussion on the use of log-ratio principles).

The chemical data were explored by means of different multivariate statistical procedures using R software (R Core Tema, 2014) and Calparsoro scripts based on Buxeda's routines and observations (Calparsoro, 2020). The application of multivariate statistical techniques to multielemental chemical data easily enables identification of compositional groups. Therefore, chemical similarity of individuals, and subsequently their suggested provenance in agreement with the provenance postulate (Weigand et al., 1977), was tested using Principal Component Analysis (PCA). In order to assess the provenance of unknown samples from Ceuta and Ksar Seghir sites, they were compared against well-known archaeometrical reference groups from the main production centres of the Iberian Peninsula (Buxeda et al., 2015; Iñáñez et al., 2008, 2009).

Table 2

Calculated average, standard deviation, maximum and minimum values for the ICP-MS concentrations according to the groups identified. Oxides are expressed in wt % and the rest in ng/g.

	Lisboa-1 (n = 4)				Lisboa-2 (n = 20)				Sev-B (n = 42)				Sev-C (n = 20)			
	average	st.dev	max.	min.	average	st.dev	max.	min.	average	st.dev	max.	min.	average	st.dev	max.	min.
Al2O3	15.4	1.8	17.6	13.6	18.1	2.7	21.5	13.5	14.2	1.4	17.4	11.0	13.5	1.0	16.5	11.9
Ba	627	279	836	235	455	135	748	260	447	95	765	85	467	64	573	337
CaO	1.5	0.5	2.3	1.0	0.7	0.3	1.6	0.4	16.1	4.9	29.3	5.8	17.1	4.7	23.3	6.1
Ce	93.5	34.2	140.5	66.8	93.6	17.2	128.3	57.6	82.1	9.5	109.4	52.9	80.2	5.0	93.9	71.7
Co	25.9	7.9	37.4	19.8	22.3	8.3	37.8	10.9	18.5	5.9	40.5	3.4	19.8	4.5	32.7	14.5
Cr	96.6	10.5	106.8	83.3	88.0	16.7	119.3	59.0	88.8	24.7	173.5	6.3	84.0	8.7	109.2	69.7
Cs	9.3	2.8	13.3	6.7	13.2	4.5	18.2	5.9	6.7	1.5	10.6	3.8	5.2	0.8	6.7	3.5
Dy	3.97	0.43	4.55	3.63	5.03	0.83	6.98	3.54	4.98	1.16	11.55	4.03	5.55	0.34	6.25	5.15
Er	2.32	0.19	2.54	2.07	2.68	0.32	3.40	2.03	2.71	0.68	6.71	2.24	2.81	0.15	3.10	2.55
Eu	1.13	0.13	1.23	0.95	1.43	0.29	2.02	0.88	1.26	0.22	1.73	0.09	1.50	0.11	1.75	1.30
Fe2O3	10.87	3.92	15.32	7.34	6.13	2.46	10.33	1.94	8.54	2.77	14.54	3.02	5.55	0.45	6.64	4.85
Gd	5.17	0.59	6.00	4.59	6.64	1.23	9.17	4.04	6.01	0.74	9.01	5.01	6.20	0.32	6.94	5.66
Hf	5.86	1.11	7.16	4.71	4.75	1.26	7.96	3.26	5.63	2.25	11.32	2.65	5.57	0.52	6.37	4.45
Ho	0.73	0.04	0.78	0.70	0.83	0.09	1.08	0.67	0.85	0.16	1.81	0.74	0.80	0.04	0.87	0.75
K2O	1.80	0.17	2.01	1.60	3.17	1.17	4.43	0.90	2.48	0.49	3.54	1.36	2.51	0.62	3.49	1.48
La	37.24	5.87	45.32	32.82	44.52	7.10	59.68	31.54	39.02	4.51	51.43	20.45	40.43	2.36	45.76	36.42
Lu	0.38	0.04	0.41	0.32	0.42	0.05	0.52	0.33	0.44	0.15	1.34	0.34	0.47	0.03	0.52	0.41
MgO	1.12	0.28	1.34	0.71	1.38	0.40	2.32	1.00	4.60	3.57	19.30	1.52	3.48	0.66	4.55	1.89
MnO	0.0635	0.0916	0.2000	0.0047	0.0887	0.0821	0.2012	0.0038	0.1777	0.8995	5.8675	0.0155	0.0840	0.0154	0.1000	0.0500
Na2O	0.818	0.215	1.110	0.644	1.076	0.295	1.580	0.650	1.036	0.353	2.360	0.309	1.047	0.159	1.410	0.820
Nb	14.2	4.1	20.0	11.3	16.5	3.6	26.8	11.7	15.5	1.5	20.3	11.9	14.9	0.8	16.2	13.3
Nd	36.2	5.3	40.9	29.9	47.9	9.4	62.4	28.5	40.3	4.6	55.3	30.9	36.2	2.1	41.3	33.5
Ni	45.3	6.8	51.3	37.0	41.5	14.7	73.2	18.8	56.1	40.3	250.4	-1.0	35.9	6.8	52.8	26.9
P2O5	0.3	0.1	0.4	0.2	0.2	0.1	0.4	0.1	0.2	0.1	0.7	0.1	0.3	0.1	0.4	0.1
Pb	44.57	18.11	71.09	31.14	311.77	-	1584.48	25.27	3031.96	-	13266.41	24.27	4413.86	-	18284.28	345.32
Pr	9.43	1.35	11.00	7.83	12.13	2.26	15.85	7.65	10.40	1.20	14.47	7.34	9.37	0.52	10.65	8.54
Rb	105.0	11.5	118.2	90.4	174.8	59.5	232.6	61.5	115.4	39.0	322.1	70.8	96.8	18.2	138.9	73.5
SiO2	66.5	6.0	74.7	60.4	62.6	6.9	79.4	53.5	51.3	8.0	70.3	40.1	61.3	4.5	72.2	54.4
Sm	6.03	0.65	6.46	5.06	7.62	1.46	10.30	4.74	6.80	0.78	9.02	5.16	6.99	0.47	7.94	6.24
Sn	11.3	-	26.4	1.4	14.8	-	46.0	3.9	22.1	-	67.5	2.1	20.0	-	118.3	3.0
Sr	109.4	21.8	137.3	88.6	81.0	13.7	106.9	56.4	387.3	101.8	623.3	25.3	428.7	95.5	523.6	199.2
Ta	1.60	0.39	2.11	1.23	2.20	0.47	2.92	1.27	1.86	0.30	3.08	1.42	1.32	0.08	1.44	1.12
Tb	0.71	0.04	0.76	0.67	0.93	0.17	1.29	0.58	0.88	0.16	1.74	0.71	0.89	0.05	1.01	0.80
Th	8.7	1.7	10.4	6.9	8.7	2.5	14.0	3.9	8.4	7.3	52.1	1.8	11.0	0.8	13.4	9.9
TiO2	0.658	0.127	0.820	0.530	0.828	0.123	1.080	0.610	0.755	0.083	0.930	0.610	0.755	0.049	0.850	0.630
Tm	0.36	0.01	0.38	0.35	0.41	0.05	0.55	0.32	0.44	0.15	1.31	0.32	0.45	0.03	0.51	0.40
U	2.63	0.24	2.83	2.31	2.59	0.51	3.69	1.54	2.97	1.74	13.33	2.08	3.21	0.35	4.02	2.77
V	102.6	15.7	124.7	88.4	99.0	14.6	132.3	72.6	89.0	18.5	111.4	7.7	93.0	12.7	125.5	68.7
Yb	2.39	0.09	2.50	2.28	2.65	0.27	3.24	2.09	2.77	0.81	7.65	2.30	2.83	0.15	3.08	2.56
Zn	54	30	95	26	141	164	622	16	211	377	2049	8	128	52	254	64
Zr	124	14	135	105	92	53	228	17	110	82	395	7	209	21	244	166

Ksar Seghir - Ceuta (n=86)

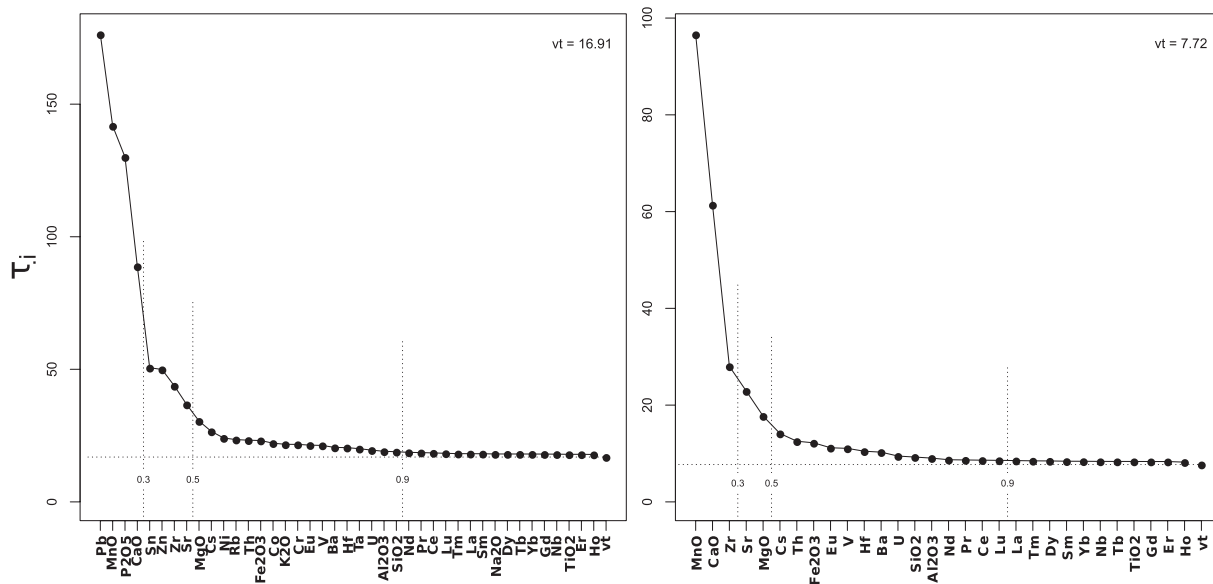


Fig. 8. Left: Graphical representation of the evenness of the compositional variability of 86 analysed ceramics by ICP-MS. Right: Graphical representation of the evenness of the compositional variability of 86 analysed samples by ICP-MS, after excluding some elements for the statistical analysis. (vt = Total variability. H2% = percentage of information entropy over the maximum possible).

While analytical error has been minimized by using very strict quality protocols during sample preparation, the potential for contamination exists nonetheless and a conservative approach to data interpretation is enforced to prevent spurious interpretations. Thus, Co was removed from consideration during the statistical assessment because cobalt is a well-known binder in the planetary milling tungsten carbide cell. Moreover, Pb and Sn were not used in the statistical treatment since these elements are major components of the glaze composition for tin lead-glazed and lead-glazed earthenwares. Furthermore, Ni and Zn showed values under the quantification limit and high variability in the results obtained by ICP-MS and were not used. In addition, P_2O_5 was also neglected in statistical routines due to its high variability and potential as a key-role element in alteration processes (Lemoine and Picon, 1982; Pradell et al., 1996). At the same time, Rb, Na_2O and K_2O values have been used with certain restrictions due to the existence of a relevant number of ceramics that showed the presence of analcime or wairakite. The presence of such minerals is related to a double alteration and contamination process, which occurs with the leaching of potassium and, oftentimes, rubidium, from the ceramic body, with a subsequent increase of sodium because of analcime-wairakite crystallization from circulating waters (Buxeda and Mommseni Tsolakidou, 2002; Iñáñez et al., 2007; Schwedt et al., 2020). Therefore, this alteration and contamination process is very difficult to quantify and correct, giving values on these chemical elements that are impossible to correct at the moment (Buxeda et al., 2002; Iñáñez, 2007; Schwedt et al., 2020).

4. Chemical characterization results

In order to process the chemical composition results (Table 2 and Table S1), the studied individuals were compared with the database of late-medieval and modern ceramics generated in recent years (for more information see Iñáñez, 2007; Iñáñez et al., 2008, 2009; Buxeda and Iñáñez, 2010; Iñáñez et al., 2007b (Buxeda et al., 2015; Buxeda et al., 2011)). This database consists of more than 1000 individuals analysed using multiple analytical techniques: ICP-MS, XRF, AAN, SEM, XRD. Most of the ceramics contained here correspond to the main production centres of the Iberian Peninsula (Seville, Lisbon, Aveiro, Talavera de la Reina, Puente del Arzobispo, Paterna, Manises, Barcelona, Reus, Vilafranca del Penedès, Lleida, Teruel, Muel, Villafeliche, Logroño, Orduña

and Elosu), as well as to a multitude of peninsular consumer centres. There is also a number of individuals from the Canary and Azores Islands, as well as a large number of American colonial ceramic materials, especially from Panama, Peru, Colombia, Argentina, the Dominican Republic and the United States of America, among others. In this work, only ICP-MS chemical data was used to compared unknown ceramics with the above-mentioned known reference groups previously analysed.

Regarding the chemical nature of the samples under study, the calculated total variation (tv) of the chemical dataset is very high (tv = 16.91) (Fig. 8), denoting the polygenic characteristics of the different fabricreference groups and the unknown ceramic shards (Buxeda and Kilikoglou, 2003). The study of the compositional variation matrix has shown that, in addition to the elements already known as problematic due to their relationship with phases of alteration and/or contamination (As, Na_2O , K_2O and Rb), there is a relatively high variability of other chemical elements used in the first statistical approach. These elements are mainly MgO and CaO, in addition to Sr. However, CaO and MgO can be directly related to technology and behavioural action by the potters, who left their cultural and technological imprint on it (for a deeper discussion on this topic, see Buxeda et al., 2008; Skibo and Schiffer, 2008; Fowler et al., 2019). The elements that can be considered mainly responsible for this variation are MnO, CaO, Pb, Sr, MgO, P_2O_5 , all of them showing a $tv/\tau_i < 0.5$, in contrast, the variable that introduces the least variability into the data set is Nb ($tv/\tau_i = 0.929$). However, when considering the contribution of the elements CaO, Pb, Sr, MgO, P_2O_5 , possibly due to alterations that are difficult to identify by simple chemical analysis, the variable that introduces the least variability to the set of data consists of Ho ($tv/\tau_i = 0.889$), while the total variability is significantly reduced (tv = 7.72) (Fig. 8). For these reasons, Ho has been chosen as a divisor in the subsequent transformation into logarithms of ratios.

Concerning the statistical analysis of the chemical data set, in order to assign a provenance, we have performed a principal component analysis taken into account the reference groups that are historically coherent with the possible main sources of ceramic supply at that time of the North African strongholds: Lisbon and Seville, as well as Manises, Muel and Talavera to a lesser extent. Hence, a statistical analysis of principal components has been carried out on the sub-composition

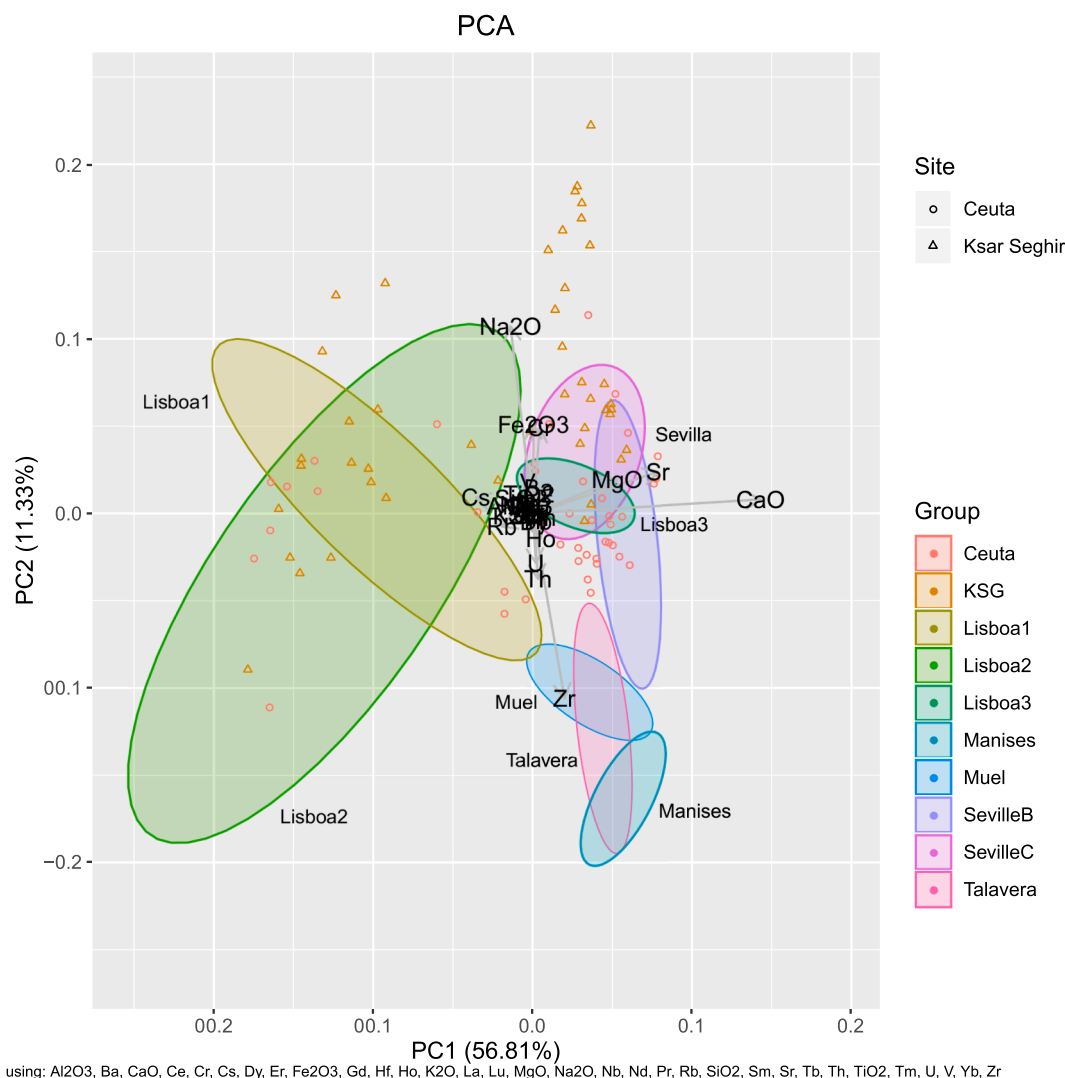


Fig. 9. Principal Component Analysis of unknown samples from Ceuta and Ksar-Seghir and relevant reference groups of Talavera, Manises, Muel, Lisbon and Seville.

Al₂O₃, BaO, CaO, Ce, Cr, Cs, Dy, Er, Eu, Fe₂O₃, Gd, Hf, K₂O, La, Lu, MgO, Na₂O, Nb, Nd, Pr, Rb, Si₂O, Sm, Sr, Tb, Th, Ti₂O, Tm, U, V, Yb, Zr with the additive log-ratio transformation using Ho as divisor (Fig. 9). The study of the principal components indicates that the first seven principal components account for 95% of the variance of the data set, with the first two principal components explaining a variance of 83.55% of the data set

Delving deeper into the statistical exploratory study, and as a result of the above, following the statistical study summarized in Fig. 10, the existence of four large groups has been established: two groups formed by ceramics compatible with the Seville reference groups (SEV-B and SEV-C) and two groups formed by ceramics compatible with a Lisbon origin (Lisbon-1 and Lisbon-2). In the case of the Seville ceramics, we have established that the clusters are composed of calcareous ceramics, whereas the Lisbon ceramics, which include red fabric ceramics, have little calcareous content, in order to further research the different origins suggested above, a detailed study of the different productions has been carried out with respect to the reference groups identified for each production centre.

As can be seen in Fig. 11, the group labelled as SEV-B is the most numerous (42 shards) and is mainly composed of honey lead-glazed ceramics (CTA013, CTA016, CTA020, KSG004, KSG005, KSG013, KSG048-KSG051, KSG054), some with underglaze painting in brown (CTA014, KSG006, KSG007), and tin lead-glazed ceramics, both

undecorated (CTA009-011, CTA040, KSG012, KSG016, KSG017, KSG020, KSG044) and painted in blue and purple (CTA042, KSG042, KSG045), or half-dipped white and green (KSG019, KSG046). Fragments of unglazed beige earthenware (CTA001-CTA005, KSG021-KSG026), as well as exemplars of *lebrillos* glazed in green (KSG001-KSG003) are also included in this group. This SEV-B compositional group can be related, in terms of composition, with the Sev-03 group recently identified by Fernández de Marcos and collaborators (2017). The group named SEV-C includes only glazed ceramics (20 shards), either tin or lead-glazed: the tin lead-glazed ceramics include fragments decorated in blue and purple (CTA043-CTA047), or half-dipped white and green (CTA055, CTA057, CTA058), the lead-glazed is either honey (CTA059, CTA061-CTA065) or green coloured (CTA050-CTA054, CTA056).

Moreover, it is interesting to note that none of the studied pieces from Ceuta and Ksar Seghir show chemical compatibility with the third reference group from Seville, SEV-A, composed mainly of storage and transport ceramics (e.g. *botijas* [olive jars] and jars), with beige fabrics and a clearly calcareous composition (CaO 14.6%) (Table 2). Actually, most of the studied North African ceramic assemblages feature table and kitchenware and consumption items, rather than transportation vessels.

On the other hand, Fig. 12 summarizes the chemical differentiation between the ceramic groups identified as originating from Lisbon. Thus, the Lisbon-1 group is composed of fragments of unglazed red earthenware (CTA035, KSG010, KSG052, KSG053), as well as the Lisbon-2

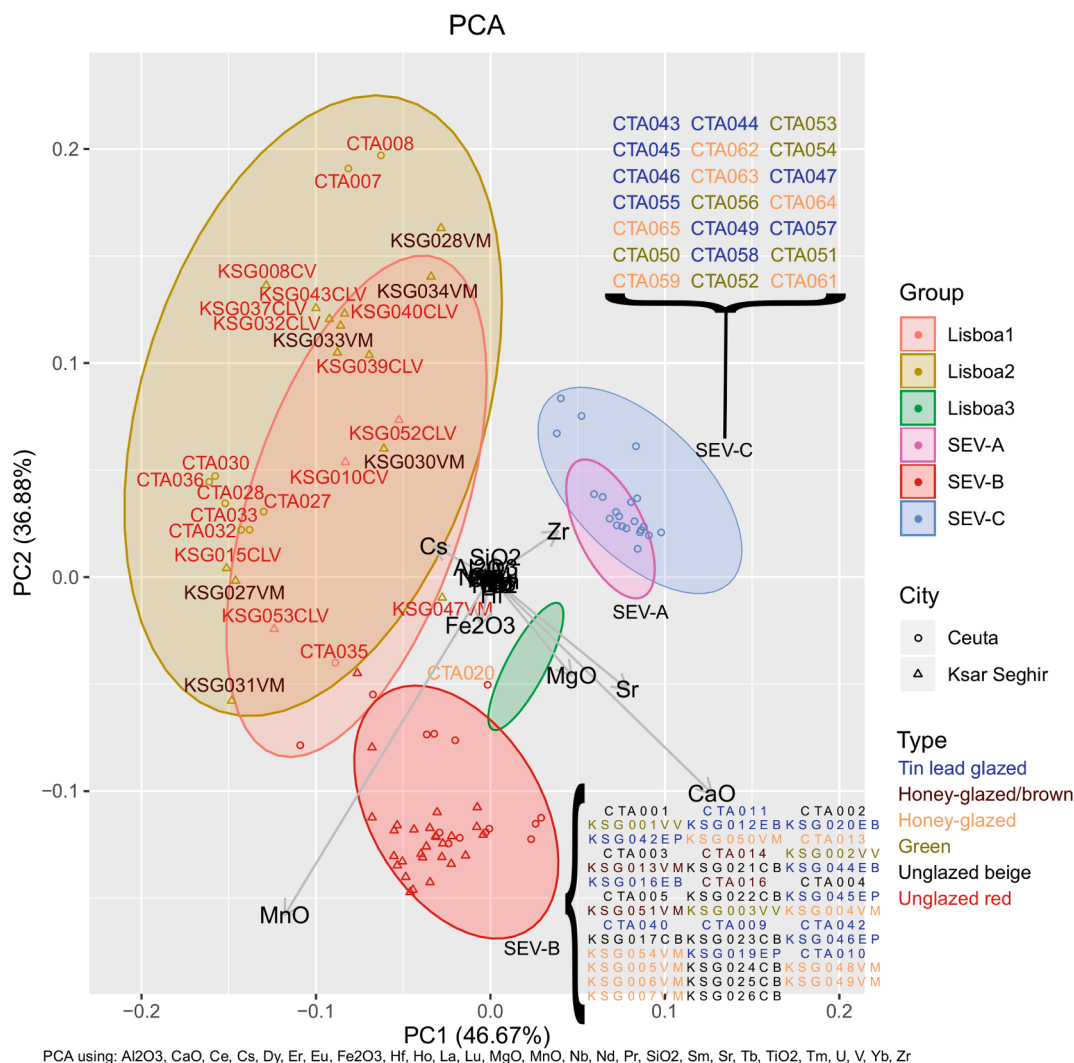


Fig. 10. Principal Component Analysis of unknown samples from Ceuta and Ksar-Seghir and relevant reference groups of Lisbon 1, 2 and 3 and Seville A, B and C.

group (CTA007, CTA008, CTA028, CTA030, CTA032, CTA033, CTA036, KSG008, KSG015, KSG032, KSG037, KSG039, KSG040, KSG043). Honey-brown lead-glazed ceramics (KSG027, KSG028, KSG030, KSG031, KSG033, KSG034) can also be chemically associated with the Lisbon-2 group, although further sampling would be required to confirm the hypothesis of provenance, since the archaeological data points to other directions.

5. Mineralogical characterization results

The positions of the characterized samples from the Ksar Seghir and Murallas Reales of Ceuta sites can be seen in the system SiO₂-Al₂O₃-CaO + Fe₂O₃ + MgO (Fig. 13). This triangular diagram shows how most of the individuals are located in the quartz-anorthite-wollastonite thermodynamic equilibrium triangle and in the wollastonite-anorthite-mullite triangle. Therefore, and according to the temperature reached during the firing, it can be theoretically assumed that the phases that crystallize during the firing will be those that form its thermodynamic equilibrium triangle (Heimann, 1989, Maggetti, 1982). Likewise, it can be seen that most of red fabric ceramics show a tendency to be non-calcareous (with CaO below 5%), while the rest of productions show a tendency to be calcareous (with CaO above 5%, and above 15% in many cases).

The identified crystalline phases were mostly: quartz, plagioclases, gehlenite, pyroxene, calcite, illite, hematite, goethite, wairakite. Quartz,

SiO₂ hexagonal, is considered a constitutive mineral of the ceramic raw material, as well as potassium feldspar, KAlSi₃O₈. Plagioclases, solid solution NaAlSi₃O₈ - CaAl₂Si₂O₈, can be part of the raw material, but can also be formed during firing as a result of the reaction between silicates and carbonates at high temperatures, in most samples it can be found in variable quantities, corresponding to an albite. However, in some cases of samples with calcium silicates, it could be of a more calcic plagioclase (closer to an anorthite). Gehlenite, Ca₂Al(AlSi)O₇ tetragonal, is considered a result from the reaction between silicates and carbonates at high temperatures. The calcium pyroxene (diopside) Ca(Mg,Al)(Si,Al)²O₆ monoclinic, is considered a result of the reaction between silicates and carbonates at high temperatures. Calcite, CaCO₃ trigonal, could be part of the raw material, but this would indicate that the temperatures at which this carbonate decomposes have not been reached. Furthermore, it is also frequent as a secondary phase in many ceramics, product of alteration, recarbonation, adhesion to the fabric after burial, et cetera. Phyllosilicates of the mica group, mainly illites, are a component of the raw material and are identified mainly by their basal reflections (001) at 10 Å (approx. 9° 2theta) and 5 Å (approx. 17,6° 2theta), and by a non-basal reflection at approx. 19,8° 2theta. Finally, other mineral phases have also been identified. For example, hematite, Fe₂O₃, has been identified in several individuals, especially those with a more reddish colour. This phase is generally the product of iron oxidation during the firing process, although it may be part of the raw material. The presence of goethite, FeO(OH) orthorhombic has also been identified, even

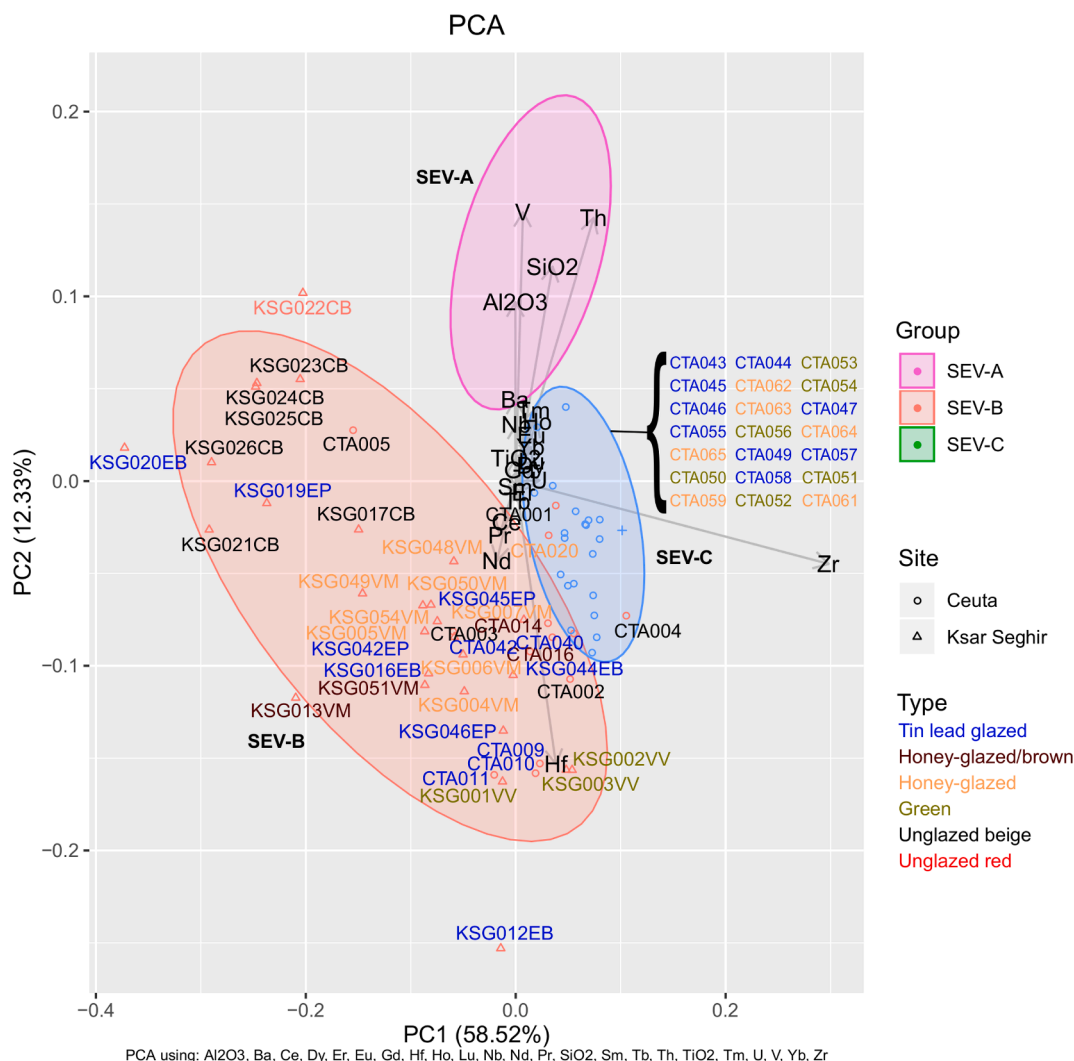


Fig. 11. Principal Component Analysis of unknown samples from Ceuta and Ksar-Seghir and relevant reference groups of Seville.

though it may be part of the raw material, its presence in ceramics is considered a product of iron oxide alteration, since this phase decomposes at temperatures below 300 °C. Moreover, the presence of wairakite, $\text{Ca}(\text{Al}_2\text{Si}_4\text{O}_{12}\cdot 2\text{H}_2\text{O})$, the calcium analogue of analcime, $\text{NaAlSi}_2\text{O}_6\cdot \text{H}_2\text{O}$, was detected, i.e. a hydrated calcium zeolite whose presence in archaeological ceramics, mainly as a secondary phase, can be explained by the alteration of the vitreous phase of calcareous ceramics fired at high temperatures. The crystallization of wairakite or analcime implies the fixation of calcium or sodium, respectively, from the circulating water (Buxeda et al., 2002; Schwedt et al., 2020; Iñáñez et al., 2007b). Table S3 summarizes the mineral associations for each studied ceramic individual, including an indication of its chemical group and identified fabric.

From the observation of the diffractograms results it can be deduced that, basically, there are two types of ceramic associations: the calcareous ceramics, in which calcium silicates (gehlenite and/or diopside) were identified, and which include calcite (in almost all cases), and occasionally wairakite as secondary phases, and the non-calcareous ceramics, which feature low or no CaO content, favouring the formation of iron oxides (mainly hematite), resulting in reddish coloured fabrics.

With regard to calcareous ceramics, this group basically includes ceramics with tin-glaze or honey or green lead-glaze, with beige, orange or pink fabrics. The colour is a result of the use of calcareous fabrics and oxidation firing, the decomposition of carbonates and the formation of

pyroxenes that incorporate Fe in their crystalline structures inhibit the formation of hematite (Molera et al., 1998a, 1998b). These pieces correspond to the SEV-B and SEV-C groups described above.

Moreover, up to four fabrics have been identified in the SEV-B group, corresponding to their mineral phases (Table S3, Fig. 14). Thus, fabric F-I, corresponding to three unglazed beige earthenware pieces (CTA002, CTA003 and KSG017), shows clear illite-muscovite peaks at the 10 Å peak of the diffractogram, plagioclase and diopside, there is also some evidence of calcite, possibly secondary. EFT is estimated in the range of 850–900 °C. Two honey lead-glazed individuals with brown decoration (CTA014 and KSG006) show traces of hematite, as well as goethite alteration in the latter, and were assigned to a variant of the first fabric, F-Ib (Table S3, Fig. 14). Fabric F-II, which includes a wide variety of ceramics, shows a mineral association, which, in addition to the beginning of the decomposition of illite-muscovite being evident, shows peaks of gehlenite and diopside (F-IIa), as well as calcite (F-IIb), with an estimated EFT in the range of 850–950 °C. Likewise, fabric F-IIc, composed only of the melted honey lead-glazed piece KSG050, shows the presence of alkaline feldspar and hematite, with an EFT of 850–900 °C. On the other hand, fabric F-IIId, which also includes a variety of ceramics, features mainly quartz, plagioclase, gehlenite and diopside, as well as calcite in almost all the included ceramics. Furthermore, this fabric shows no traces of illite-muscovite, while some of the pieces show crystallization of wairakite (labelled as F-IIId). Therefore, its EFT would be in the upper range of 850–950 °C. Fabric F-

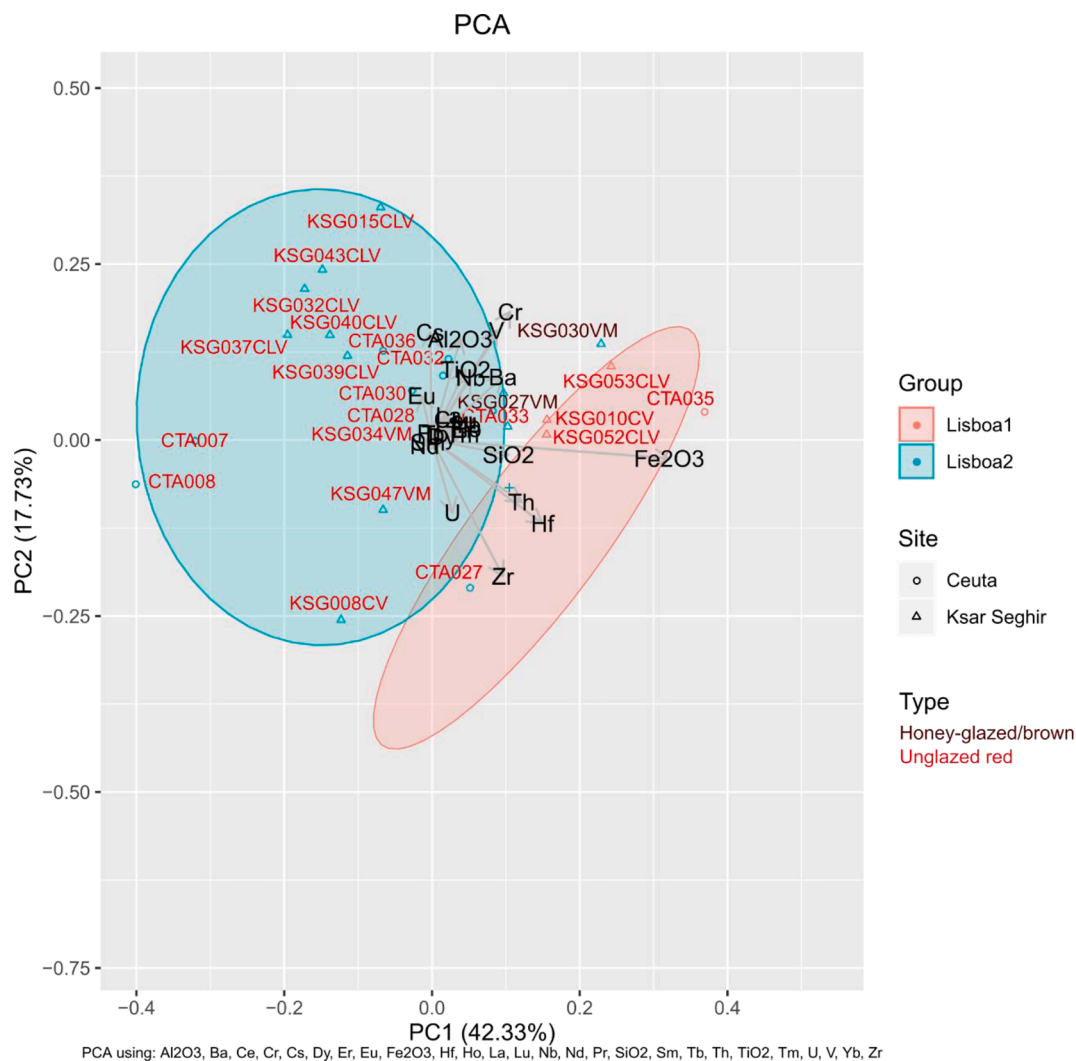


Fig. 12. Principal Component Analysis of unknown samples from Ceuta and Ksar-Seghir and relevant reference groups of Lisbon.

III shows no traces of illite-muscovite, and along with the presence of hematite, gehlenite and diopside, an EFT in the range of 900–950 °C for the first sub-fabric and 950–1000 °C for the second one can be estimated, since a higher temperature is required to form a more extensive vitreous phase facilitating the crystallization of zeolites according to the process described above. Finally, fabric F-IV shows similar characteristics to fabric F-IIIa, but with hematite reflections. Thus, an EFT in the range of 950–1000 °C is estimated (Table S3, Fig. 14).

The SEV-C chemical group includes five mineralogical fabrics. Fabric F-I, consisting of a single piece with honey lead-glaze (CTA061, Table S3, Fig. 15), shows a mineral association with clear illite-muscovite peaks, as well as alkali feldspar, plagioclase and traces of hematite, with an estimated EFT around 850–900 °C. Fabric F-II, which includes glazed and tin lead-glazed ceramics of all types, also shows illite-muscovite reflections, as well as plagioclase, gehlenite and diopside, in addition, calcite contents, possibly secondary, are also present. Therefore, an EFT in the upper range of 850–950 °C is estimated. It is interesting to note that the variant of this fabric, F-IIb, shows signs of crystallization of wairakite in two pieces (CTA043 and CTA057, both tin lead-glazed decorated in blue and purple, and in half-dipped white and green, respectively). The estimated temperature is in the upper range of 900–950 °C. Fabric F-III, with only one honey lead-glazed fragment (CTA062), is the upper evolution of fabrics F-I and F-II, since it no longer shows signs of illite-muscovite but does have hematite, its EFT is estimated at 950–1000 °C. Fabric F-IV, including a single individual with

the same characteristics (CTA065), shows an association similar to F-III, but without hematite, although with crystallization of wairakite, so its EFT has been determined in the range of 1000–1050 °C. Almost all the individuals that compose the calcareous groups show the presence of calcite, as well as quartz and plagioclase in their diffractograms (Table S3, Fig. 15).

In contrast, non-calcareous ceramics are represented by the chemical groups Lisbon-1 and Lisbon-2, featuring fabrics with darker orange and red colours, with low CaO contents, which does not enable the incorporation of Fe in silicates of neoformation (mainly pyroxenes and gehlenite), hematite is thus formed, resulting in these colours. The reddest and most hematite-rich samples do not (or hardly) retain illite reflections. The Lisbon-1 chemical group, composed only of unglazed red earthenware pots and pans, includes three identified mineralogical fabrics, in this respect, fabric F-I shows an association of mineral phases composed mainly of quartz, plagioclase and clear illite-muscovite peaks. Thus, an EFT in the range of 850–950 °C has been determined for this fabric. Fabric F-II shows a similar EFT and mineralogical association to the previous fabric but with the presence of hematite. On the other hand, fabric F-III does not show any signs of illite-muscovite, although hematite is present, EFT is estimated at 900–950 °C (Table S3, Fig. 16).

Finally, the Lisbon-2 group is composed mainly of ceramics with very little or no calcareous material, without calcium silicates, in which potassium feldspar is often identified along with plagioclases (mainly albite), usually retaining illite reflections (Table S3, Fig. 17). This is the

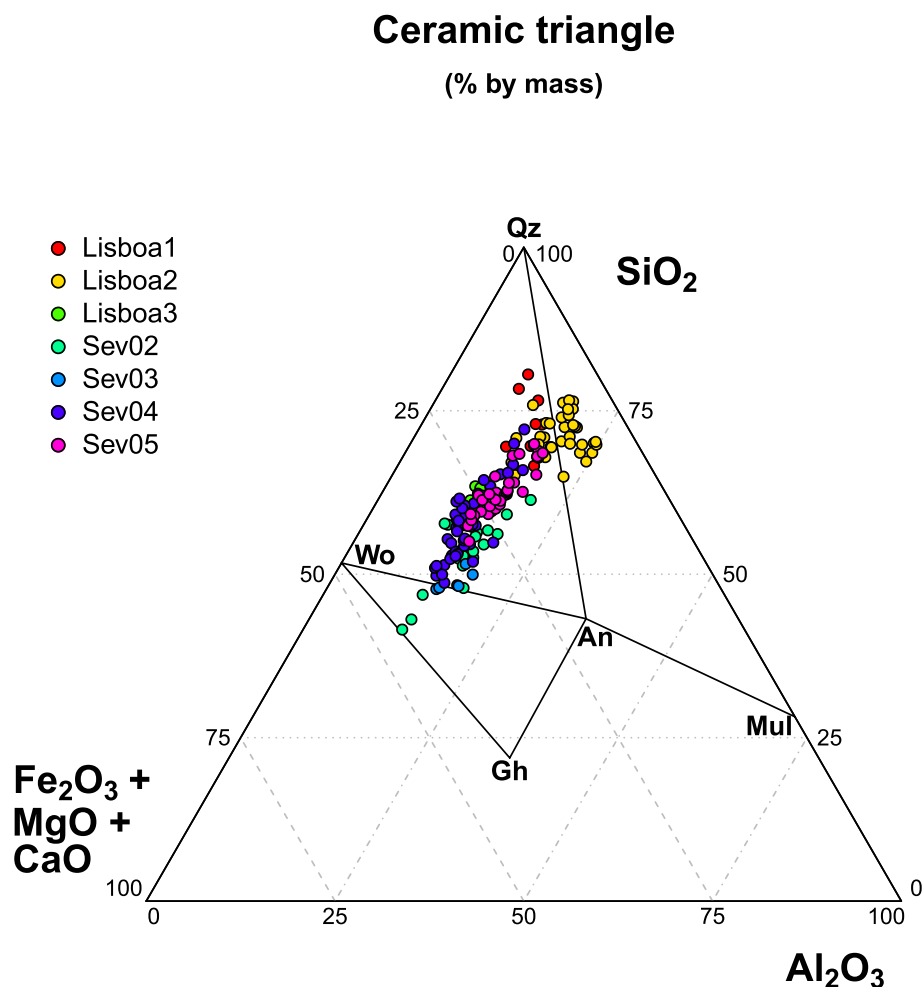


Fig. 13. Ternary diagram showing the compositions of SiO₂, CaO and Al₂O₃ of the 86 shards classified by their corresponding typology. An Anorthite, Gh Gehlenite, Mul mullite, Qz quartz and Wo Wollastonite (abbreviations after [Whitney and Evans, 2010](#)).

case of fabrics F-I and F-II, which are composed of unglazed red fabric ceramics. These fabrics can be differentiated by the absence and presence, respectively, of plagioclase in their fabrics, with an estimated EFT between 800 and 850 °C. Fabric F-III, with an estimated EFT of about 850 °C, is very similar to the previous one, but with more evident reflections of hematite in its diffractograms. Fabric F-IV, identified at higher temperature, includes two pieces with honey-brown lead-glaze (KSG031 and KSG034), this fabric shows no traces of illite-muscovite, but features potassium feldspar and hematite. Therefore, EFT has been estimated in the range of 900–950 °C. Finally, the honey-brown lead-glazed piece KSG027 is the only one included in fabric F-V, it features only quartz and hematite, in terms of the major mineral phases identified. Thus, a high temperature can be estimated, with an important vitreous phase evidenced in the diffractogram and in the range of 950–1000 °C (Table S3, Fig. 17).

6. Discussion

The detailed analysis and crossing of the data from the macroscopic and archaeometrical analyses allowed us to confirm the generic assignation of provenances to a good part of the ceramic types based on archaeological parallels for Ksar Seghir and Ceuta ([Boone: 120-129, 1980](#); [Redman: 190-198, 1986](#); [Teixeira et al., 2013](#); [Teixeira and Torres, 2018](#)). Particularly in the case of the Sevillian examples, it reinforced the analysis made in the 1990's for the eleven shards of tin lead-glazed earthenware from Ksar Seghir ([Myers et al., 1992: 136-137](#)). For other ceramic types, doubts remain unsolved until further research,

especially one that includes characterizing new production centres from Portugal and South Spain. Our research revealed the following evidence.

The tin lead-glazed ceramic, with light fabric colours, between beige, light pink and light orange, is present in the two well documented production groups (SEV-B and SEV-C), both in the undecorated version, and in the versions painted in blue, blue and purple, and half-dipped white and green. Indeed, the starting hypothesis for these productions is that they originate from the region of Seville, throughout the 15th and 16th centuries, as indicated by the archaeological findings recorded by American teams ([Deagan: 56-57, 1987](#); [Marken: 94-139, 1994](#)), confirmed by studies carried out at this Andalusian city ([Pleguezuelo and Lafuente: 228, 1995](#); [Pleguezuelo: 356, 1997](#); [Iñáñez: 307-329, 2007](#); [Fernández de Marcos et al., 2017](#)). The archaeometrical analyses previously performed at Ksar Seghir linked these productions to the pottery workshops of Triana ([Myers et al., 1992: 136-137](#)).

Regarding the honey lead-glazed ceramics, its fabrics are usually beige, light pink, light orange and orange, and belong to the SEV-B and SEV-C groups, some shards showing underglaze decoration in brown originate from SEV-B. Concerning the green glaze, both its beige, light pink and orange fabrics were produced in the SEV-B and SEV-C pottery centres. These lead-glaze productions were connected with the Seville or lower Guadalquivir manufactures from the 15th to early 16th centuries, considering the parallels recovered in the *bóvedas* [vaults] of this city ([Amores and Chisvert, 1993: 288-289, 291-292, 314-315, 321, 323, Pleguezuelo et al., 1999: 274, 276, 288, 291](#)), or in Jerez de la Frontera ([Barrionuevo Contreras, 2008-2009: 271, 274](#)). Some honey lead-glazed ceramics have also been identified as being produced in the workshops

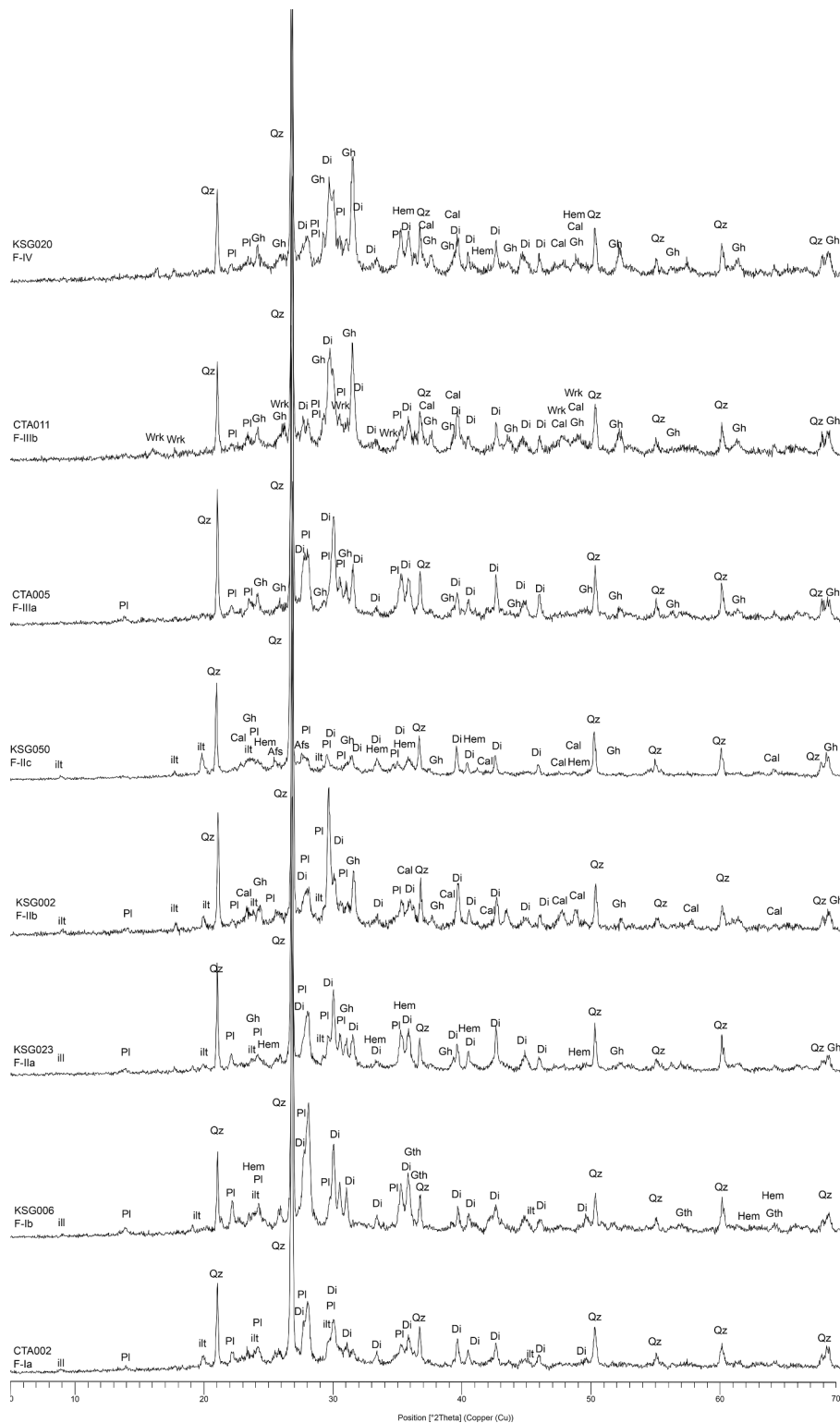


Fig. 14. Representative diffractograms of each identified fabric in Seville-B chemical group (abbreviations after [Whitney and Evans, 2010](#)).

of Palos de la Frontera (Huelva, Spain) ([Campos Carrasco et al., 2018: 260–261](#)).

Another analysed assemblage was the honey-brown lead-glaze pottery used for cooking foodstuffs, with less purified and grainier fabrics, dark-coloured, between orange and red. This group is chemically close to the Lisbon-2 productions, but the results should be taken with some caution, since these pieces do not have many equivalents in the archaeological parallels from the Portuguese capital. Parallels for the

use of these pieces can be found in the lower medieval levels of Seville ([Huarte Cambra et al., 1999: 150](#)), in the 15th and 16th centuries at Los Guájares (Granada) ([García Porras, 1995: 249–250, 254–255](#)), although their production origin has never been clearly indicated. The production of these items is attested in the 16th century in the vaults of the cloisters of Santo Domingo, in Jerez de la Frontera ([Barrionuevo Contreras, 2008–2009: 275–276](#)), and also in the workshops of Palos de la Frontera (Huelva) ([Campos Carrasco et al., 2018: 260–261](#)). For this reason, we

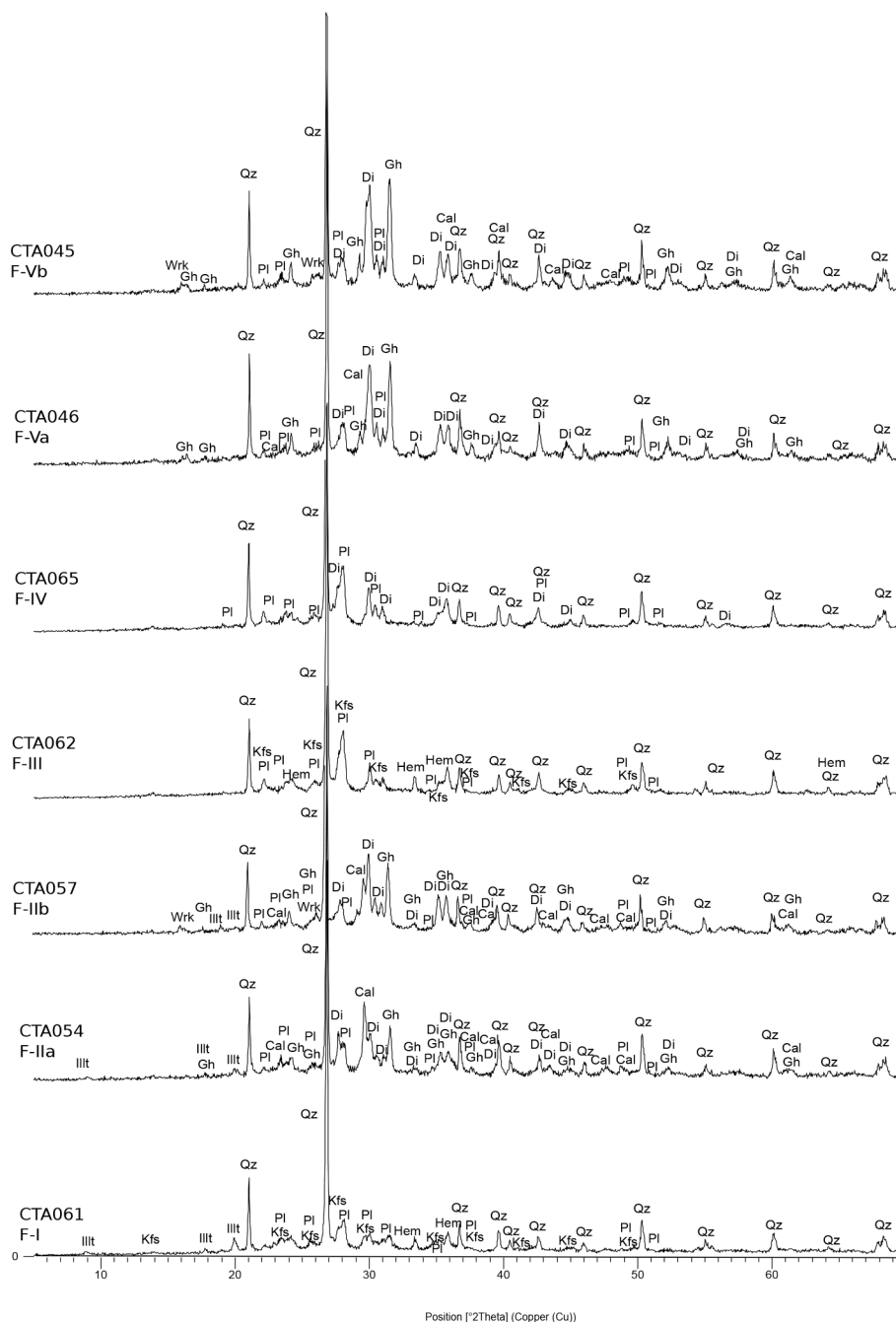


Fig. 15. Representative diffractograms of each identified fabric in Seville-C chemical group (abbreviations after Whitney and Evans, 2010).

underline again the need for a deeper study on this type of ceramics.

On the contrary, the unglazed red and orange earthenware, ascribed to Lisbon-1 and Lisbon-2, have a clear archaeological association to the two production centres of this city. The whole assemblage resembles Portuguese productions, particularly from the *Mouraria* neighbourhood, the part of the city where the Muslim community settled after the Christian conquest, having a strong pottery production tradition from Middle Ages till Contemporary times (Marques et al., 2012; Nunes and Filipe, 2012; Teixeira et al., 2000).

Finally, and concerning the unglazed beige earthenware, we have noticed that, for the fabrics with grainy textured (used for example in jars), with beige and pale orange colours, the corresponding ceramic workshop is SEV-B, the same applies to the finer textured fabrics, with tones between beige and light pink. Parallels for these ceramics can be found in the pottery workshops of the Sevillian region (Amores and

Chisvert, 1993: 280, 281, 288, 293, 305, Pleguezuelo et al., 1999: 275, 281, Barrionuevo Contreras, 2008–2009: 265, 268, 271, 275–277).

Following the results of this archaeometric study, it can be confirmed that a considerable import of ceramics from Seville and/or the Guadalquivir area was taking place in the Portuguese North African strongholds, as already indicated by historical data. Most of Seville's production was used for food consumption at the table, as well as for serving water or wine, in addition to the transportation and storage of products. The Portuguese pieces were mostly used in the preparation of food, as well as water storage and individual consumption of water. In fact, according to Robert Ricard, “the maritime centre of Andalusian commerce with the Portuguese-Moroccan cities and the source of their supply in Spain was the Cadiz - Puerto de Santa Maria - Jerez area”, in addition to the cities of Malaga, Gibraltar and Tarifa (Ricard, 1955: 168–169). The published documentation attests to the supply of these North

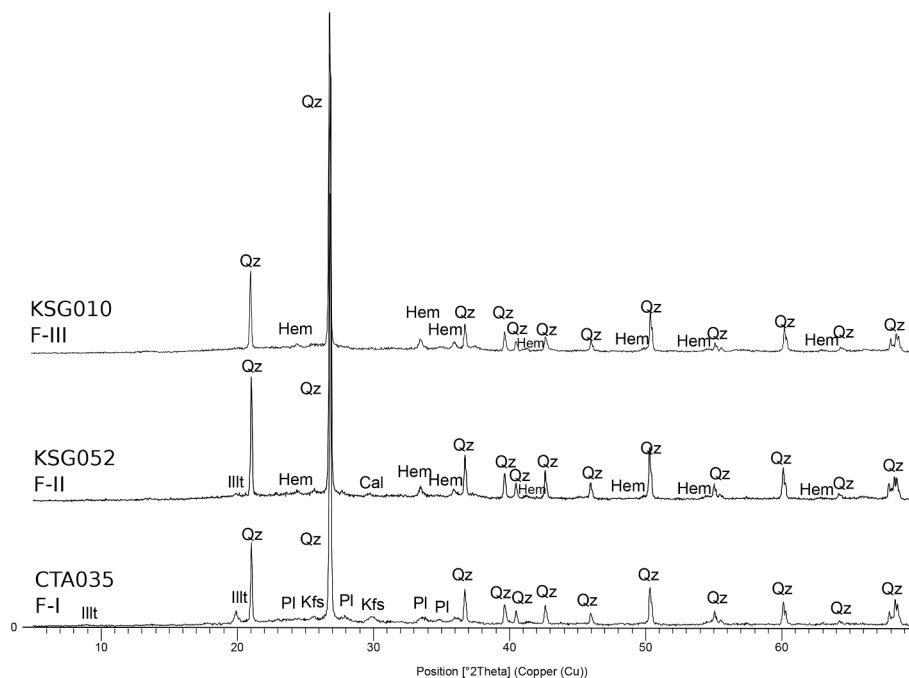


Fig. 16. Representative diffractograms of each identified fabric in Lisbon-1 chemical group (abbreviations after Whitney and Evans, 2010).

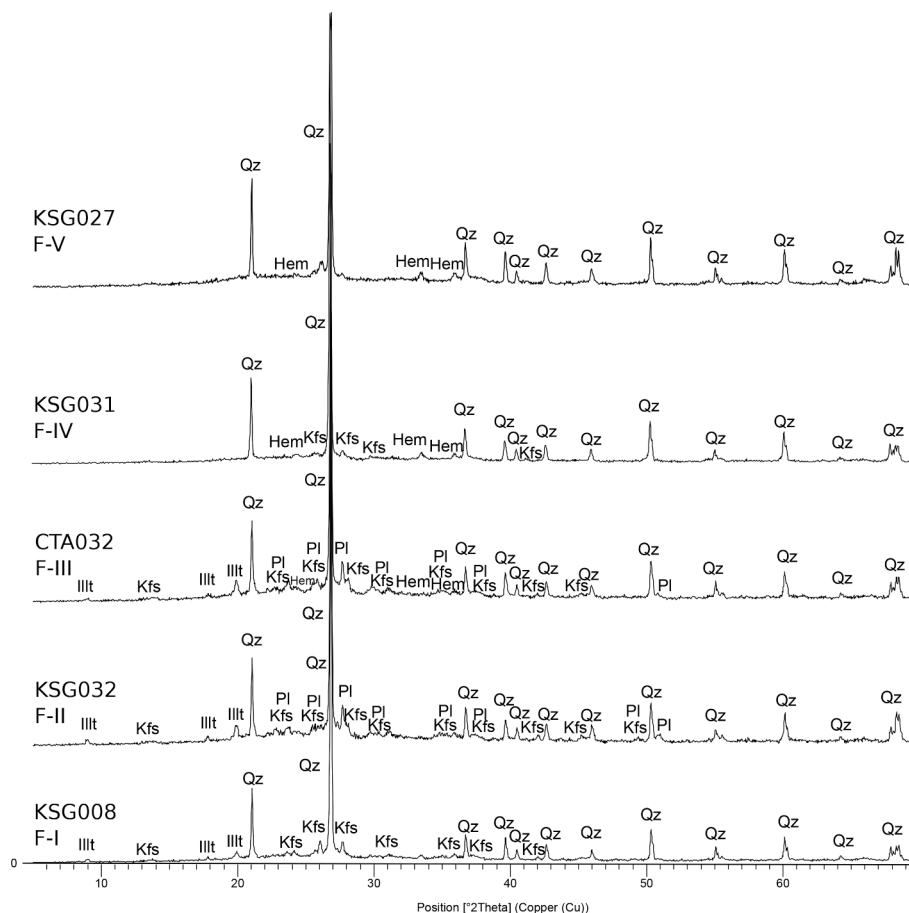


Fig. 17. Representative diffractograms of each identified fabric in Lisbon-2 chemical group (abbreviations after Whitney and Evans, 2010).

African strongholds by the cities of southern Portugal or Lisbon, or by the ports of Andalusia, especially concerning wheat, wine, olive oil, fruits and other foodstuffs, as well as materials for war, construction,

clothing and other aspects of daily life, like ceramic. If in the 15th century the origin of supplies seems to have been concentrated in the areas of Lisbon, the Algarve and the islands of Madeira and Azores, in

the 16th century the ports of Andalusia gained importance, especially through a factory established by the Portuguese Crown in this region, which travelled between several cities of southern Spain (Ricard: 143-164, 1955; Godinho, 1982, vol. III: 269; Braga and Braga, 1998: 85).

These results raise also interesting questions about the Andalusian ceramic production of the 15th and 16th centuries and the supply of the Portuguese North African strongholds. The different manufacture groups identified in the city of the Guadalquivir are quite interesting. The presence of the SEV-C pottery centre stands out, which, in the scope of our research and up to now, seem to have focused its production on glazed wares for domestic use. In archaeological terms, this group is composed of shards from the ceramic waste dumps found in the Triana quarter (Seville), partly related to the famous potters Pisano and Valladares, both great promoters of decorated tin lead-glazed pottery in Seville during the Modern period (Sánchez Cortegana, 1994; 2007). On the other hand, SEV-B is the centre of origin of about 50% of the samples and therefore includes all the Sevillian ceramic types identified here. The wide range of its production could possibly be explained by its larger size, perhaps with a more massive production for export, or with a preferential commercial connection with those two North African cities.

7. Conclusion

This paper involved the archaeometrical characterization of 86 shards from the archaeological sites of Ksar Seghir and the Murallas Reales of Ceuta. Thanks to this study it has been possible to determine the existence of two large clusters identified as chemically compatible with a Seville and/or Guadalquivir Valley provenance. Moreover, it has been identified a group chemically compatible with the Lisbon reference group. In broad terms, the Sevillian origin of the tin lead-glazed, honey lead-glazed, green lead-glazed and unglazed beige earthenware series is clear, while the unglazed red and a few honey-brown lead-glazed earthenware can be ascribed to Lisbon pottery workshops, though with serious doubts in the latter. In technological terms, the majority of Sevillian pieces are calcareous, mainly produced in an oxidation atmosphere at a temperature of around 900 °C, resulting in light-coloured ceramics (beige, light pink and light orange). On the contrary, the red ceramics feature little or no calcareous components, and the formation of mineral phases originates red fabrics (red or orange), also at temperatures of about 900 °C. Furthermore, post-depositional contaminations, such as crystallization of wairakite and goethite, have been detected, possibly due to the interaction of the ceramics' porous matrices with the atmosphere and the circulating waters. On one hand, the presence of the wairakite-analcite solid solution can be explained as the alteration of the vitreous phase of calcareous ceramics fired at high temperature, in which calcium or sodium from the circulating waters is fixed and precipitates these cryptocrystalline minerals, in this case. On the other hand, a fraction of the Fe minerals existing in the red fabric ceramics, mainly hematite, were probably transformed into goethite by hydroxylation reactions.

The key role of the Sevillian region and western Andalusia in the maintenance of the Portuguese possessions in North Africa is clearly emphasized in this study. The Portuguese seem to give preference to neighbouring markets, in a completely economic logic, instead of supplying its settlements according to political criteria. A topic that could benefit from these archaeological studies is precisely the complementarity and mutual help between Portugal and Spain at this early stage of their overseas expansions, many decades before the union of the respective crowns. Let us also stress here that all production centres, whether from Seville or Lisbon, are represented both at Ceuta and at Ksar Seghir. However, we would like to point out that the SEV-C group was only found in the city of Ceuta. These indications suggest the existence of possible preferences in the commercial and supply connections of these Portuguese North African cities, along with a trend towards a certain productive specialization of the pottery workshops and a choice of clays, according to the type of ceramics and their functionality. It

would be quite interesting to address these aspects in more detail, as a further development of our research.

Despite these results, we must be cautious when generalizing, especially in the case of the honey-brown lead-glaze and unglazed red earthenware, for which we need more samples of other production centres, including local and regional ones, to ensure the origin(s) of these ceramics. Although it was not possible to identify ceramic kilns or ceramic production wastes at Ksar Seghir and Ceuta for the Portuguese period, we must be aware of pre-existent Islamic local productions. Llano de las Damas pottery production centre in Ceuta, located outside the city walls, was deactivated between the end of the 14th and the beginning of the 15th century, maybe with the arrival of Portuguese (Hita Ruiz and Villada Paredes, 2000: 292-293). Further research will analyse ceramic wastes from this site, which can determine their chemical and technological characteristics. Despite its chronological limitation, the characterisation of these ceramics composition may help to clarify the idea of a disruption in local pottery production during the Portuguese period. The archaeological and archaeometrical results thus support this idea, although further studies with MNV counts might clarify the weight of imports versus other productions. For the moment, published works have noted the significance of imported ceramics on the daily life of these strongholds inhabitants regarding possible local productions (Boone, 1980; Redman, 1986; Teixeira et al, 2013).

Also, the role played by other relevant production centres in the area, such as Malaga, among others, is still not very clear. Furthermore, the Islamic technological disruption or adaptation during the Portuguese occupation is yet another interesting issue that must be raised and addressed in future studies. In any case, it is to be hoped that the path taken on the basis of this work can be followed again in the future with more archaeometric analyses, combined with sound archaeological studies, which will allow us to delve deeper into this and other discussions and extend our knowledge of the commercial links and the logistics of the first European empires of the Modern Age, between the Mediterranean and the Atlantic.

CRedit authorship contribution statement

Javier G. Iñáñez: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Visualization, Project administration. **Joana Bento Torres:** Conceptualization, Investigation, Resources, Writing - original draft, Visualization. **André Teixeira:** Conceptualization, Investigation, Resources, Writing - review & editing, Visualization, Supervision, Project administration, Funding acquisition. **Uxue Sánchez-Garmendia:** Conceptualization, Investigation, Resources, Writing - review & editing, Visualization. **Estefania Calparsoro-Forcada:** Conceptualization, Investigation, Resources, Writing - review & editing, Visualization. **Gorka Arana:** Methodology, Validation, Investigation, Resources, Data curation, Writing - review & editing, Supervision.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jasrep.2021.102908>.

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