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Pottery making tradition in Logroño: an archaeometric approach to the Late Medieval workshops

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Abstract

This paper deals with the findings of Hospital Viejo site of Logroño (La Rioja, Spain), which yielded the largest evidence of local pottery production, comprising three kilns and a potters' dump (13^{th}-15^{th}centuries). The study of pottery production in inland Iberia provides valuable information on the material conditions in which the Iberian medieval and post-medieval society occurred. Yet, the territory of La Rioja has been largely eclipsed by studies at coastal and southern areas of Iberia, due to their role in maritime exchange. With the aim of understanding the specific incidence and evolution of medieval pottery at regional scale, 77 sherds (MNI 637) including glazed and unglazed ware and kiln utensils were archaeometrically examined by combining the use of ICP-MS, XRD and SEM-EDS. Likewise, NAA was applied for provenance analysis, including the collation with a large majolica NAA database. The results provided the first chemical fingerprint of the production from Logroño consisting on three compositional groups within the same workshop (LOG-

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A, LOG-B and LOG-C). Moreover, pottery trading from Teruel and Valencia was detected, sustaining the historical records of these trade-networks. The imports showed tin lead glazes obtained by known recipes, contrasting with simpler manufactures (mostly unglazed or only lead glazed) of contemporary Logroño.

Keywords pottery · ICP-MS · XRD · compositional groups · Logroño · majolica

1 Introduction

Logroño is the current capital city of La Rioja and is located in the middle Ebro valley in the northern Iberian Peninsula (see Figure 1). The Riojan territory went from being a peripheral area of Al-Andalus (ca. 8^{th} century) to the nerve center of life in the Kingdom of Pamplona (ca. 9^{th} - 10^{th} centuries). Logroño functioned as a strong communication node, becoming one of the most important urban settlements of medieval Navarre and Castile, both culturally and economically (Moya, 1994). The study of pottery production in inland Iberia provides valuable information on the complexity of inter-cultural contacts at a local scale and in general, the life conditions in which the Iberian medieval and post-medieval society existed. The reactivation of long-distance routes such as the Way of Saint James is the result of the interconnection of those local scenarios as well as the development of an internal regional commercial network, with ramifications in other areas of the peninsula and the continent. Yet, this territory has been frequently eclipsed by studies at coastal and southern areas of Iberia, due to their role in maritime exchange.

The present study deals with pottery production from the late medieval workshops of Logroño as well as its trading networks. Likewise, the diachronic evolution from the medieval to the post-medieval pottery workshops is revisited based on the results of the latest archaeological interventions (Gil and Luezas, 2018).

1.1 Historical Background

The medieval city's importance has its precedents during the 11^{th} century, when the king Sancho Garcés III of Pamplona incorporated the city in the Way of Saint James. Logroño's inclusion into the most important medieval route of Europe led it to flourish economically. Later, the prominence of the city was increased when Alfonso VI decreed that only two bridges could cross the Ebro river in that region, one at Logroño and the other at Miranda de Ebro, thus, re-enforcing the city's relevance as part of the compulsory trading routes. These events, along with the transfer of the frontier with Islamic territory to the peninsular interior, allowed Logroño to gain status and to gradually displace Nájera and Viguera (the only cities of the territory

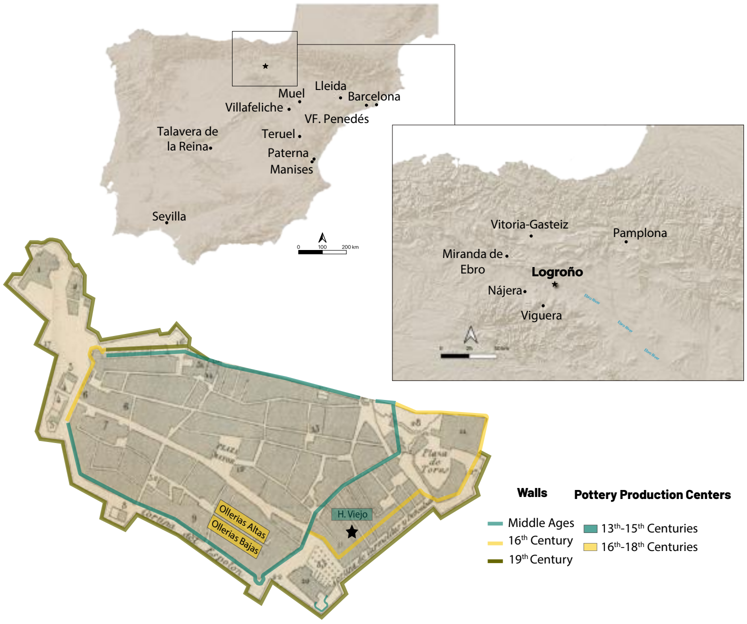

Fig. 1: Above: Main majolica production centers of Iberian Peninsula during 14^{th}-18^{th} centuries (Iñañez et al., 2008). Center: Location of Logroño and its neighboring cities. Below: Historical map of Logroño showing the location of pottery workshops and different phases of walls.

documented in the 10^{th} century) from the geopolitical order (Martínez, 2013). By the 12^{th} century, the territory of La Rioja became a fluctuating frontier between Castilians, Navarre and Aragonese. The area prospered and was enriched by the interchange of people and merchandise with a heterogeneous population where former neighbors of Logroño, those from nearby villages, as well as Franks, Jews and Mudejars in a minor extent, coexisted.

However, several events, including the modification of the Saint James Way at the end of the 13^{th} century impacted negatively on its economy. In addition, the city experienced a severe economic decline traversed by episodes of the plague that struck the city during the 14^{th} century. In contrast, due to its strategic location as a military plaza, Logroño obtained several benefits and it was granted with two annual fairs by Alfonso XI (1314). Likewise, it obtained the titles of “city” (1431) and of “very noble and very loyal” (1444). Later on, the city was granted with the right to vote in the Courts (1494), as well as to have a free market (Martínez, 2013).

At the beginning of the 16^{th} century, the reconfiguration of the city brought to an end the functioning of the medieval pottery kilns, whose activity was strongly influenced by the socio-political circumstances described above. The post-medieval pottery workshops would be placed in a different location, and would mark the beginning of a new period characterized by new forms of ceramic production. In addition, merchant companies were created in order to fulfill the requirements of more ambitious commercial activities. Thus, the trade (mainly related to wool) and their routes extended to the north of Europe (Goicolea, 2007).

Concerning the ceramic production of Logroño, two streets are documented as being dedicated to pottery activity during the 16^{th} century, namely, *Ollerías altas* and *Ollerías Bajas* (Glera, 1994). These correspond to current *San Juan* and *Ollerías* streets, respectively, and are located *extramuros* (see Figure 1). The toponymy of *Ollerías* —meaning the place of the pottery workshops— already implied the presence of a ceramic workshop, which was confirmed by the unearthing of a ceramic kiln in 2018 (Gil and Luezas, 2018). The kiln, ascribed to the 16^{th} century, constitute the first ever material evidence of the local pottery production of post-medieval chronology in Logroño.

1.2 The archaeological context of Hospital Viejo site

In the present work, the materials unearthed in Hospital Viejo Street of Logroño are addressed (see Figure 1), that is, the first Riojan archaeological site in which direct evidence of late-medieval pottery production has been documented. The archaeological findings, which comprise three ceramic kilns and a potter's dump, are the most extensive local ceramic collection unearthed until now (Martínez, 2013; Angulo and Porres, 2015). The large ceramic assemblage (thousands of sherds) includes kiln utensils, ceramic wasters, pantry pottery, cookware and tableware, with glazed and unglazed coatings, as well as fired clayey pastes (see Section 2.1).

Up to three superimposed workplaces were identified, in operation during the 13^{th}, 14^{th} and 15^{th} centuries. The pottery kilns were built with the purpose of serving from two to three generations and had a life-cycle of ca. 50 years each (Martínez, 2013). Moreover, the study of the stratigraphic sequence (see Figure 2) allowed for the identification of two different occupation phases (A and B) in each of the three workshops. These differed in the type and volume of materials, as well as the structures associated with the workshops. The installation of the first workshop was expanded on by the second structure in the second, which would have functioned more productively, but at some point, may have been abruptly disrupted by the influence of the plague (reporting the use of the kiln for calcination of the households). Finally, production rate of the third workshop would have diminished both in volume and quality, being finally abandoned. According to the extensive archaeological investigation carried out in this site, the workshops would have been run by Islamic potters, being abandoned after the Pragmatic[1] against the Mudejar community of 1502.

[1] Pragmatics of forced conversion is a term that could be applied to several decrees or other legal texts issued at the beginning of the 16^{th} century during the reign of the Catholic Monarchs and Charles V. It is the name that historiography gives especially to the

The location of these workshops was *extramuros*[2] with regard to the first wall of this city, in the area called "Villanueva" (i.e. new village). Their operational period lasted until the 15^{th}century, when Ferdinand II of Aragón (The Catholic) ordered the erection of the second fortification, including the Wall of Revellín, part of which delimits the pottery workshops under this study (See Figure 1).

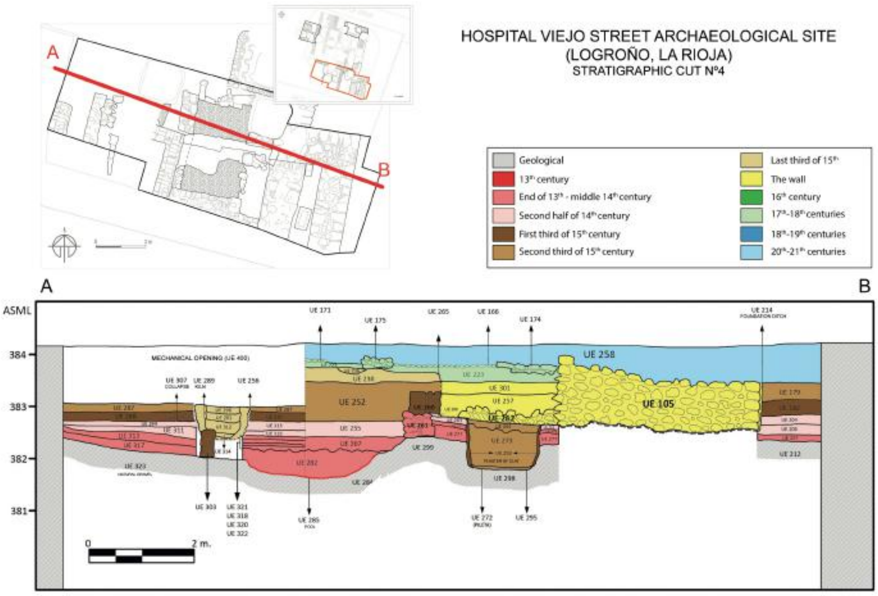

Fig. 2: Stratigraphic cut of the excavation performed in Hospital Viejo Street of Logroño. Source: Modified from ArqueoRioja

Pragmatics of February 14, 1502 Apparently the subject Muslims (Mudejars) were given the choice between exile or conversion to Christianity (Artola and Ledesma, 1991).

[2] In the past, pottery production was regarded as a nuisance activity. Therefore, it was usually carried out in the outskirts of the urban nuclei (e.g in Roman times *Lex Ursonensis*, Montelupo, Seville, etc). This situation changes over time. Whereas in the roman times it was carried out in the outskirts of the city according to the Lex Ursonensis, in the medieval Ages it was viewed differently. There are many examples of attesting the modification of the workshop localizations from *intramuros* to *extramuros*, which strongly depend on the regions and are provided in the late-medieval period and specially during the modern period.

1.3 Ceramologic Research Background

Regarding previous ceramologic regional studies, historical documentation evidenced pottery related activity in the Riojan territories during Roman[3] times (Luezas, 2014), Medieval Ages (Pérez, 1985; Martínez, 2013) and the post-Medieval period, where already differentiated pottery nuclei would have existed (Glera, 1994; Ceniceros Herreros, 2004, 2012). Nevertheless, none of those works deals with material evidences of pottery production such as kilns, and even less from the archaeometric perspective.

In addition, thanks to the Cadastre of Ensenada it is possible to grasp an idea of the pottery activity and craftsmen mobility during the post-medieval times. For the specific case of Logroño, the reception of potters from Zamora in the 16^{th} century is documented, where the origin of the potters extends to cities or villages such as Burgos, Viguera, Muel and Zaragoza, and regions such as Navarre and Alava during the 17^{th} century. Moreover, according to the registered news, there were familiar relationships that connected Logroño with Muel, a link that considerably increased during the 18^{th} century. Regarding the raw materials, the work of Glera mentions that Jose Bernardino del Busto (born in 1742) was the secretary of the Holy Inquisition in Logroño, as well as the administrator of the sale of lead and "lead alcohol" and in 1792 he granted to Manuel Blazo of Arnedo the administration and sale of both substances[4]. It can be said that in comparison with other sectors, pottery-related activity in the Riojan territory is still scarcely explored for late medieval and post-medieval periods. However, the finding of the workshops of Hospital Viejo Street has contributed significantly to changing this situation, since it constitutes the first unearthed structure associated with this type of economic activity during late medieval period (Martínez, 2013). Thus, its study plays a key role for the understanding of medieval and post-medieval pottery production in the northern Iberian Peninsula, as well as how the pottery activity evolved from the production of Hospital Viejo Street's workshops to the *Ollerías* street. That is to say, from the workshops located in the peripheral areas of the city and run by the Mudejar potters, to the workshops organized in guilds as can be inferred from the toponymy.

[3] Excavations performed in 2019 and 2020 are still evidencing the Roman pottery production activity nearby Logroño.

[4] "Lead alcohol" is a form in which galena or lead sulfide (PbS) was referred. Also known elsewhere as leaf alcohol, because of the laminar form in which the galena occurs (Glera, 1994). These substances were necessary to obtain lead-glazes used for the ceramic coatings.

2 Materials and Methods

2.1 Ceramic sample

The ceramic assemblage was unearthed from the thoroughly documented archaeological site of the Hospital Viejo Street from Logroño (Martínez, 2013). The sampling strategy focused on pottery production-related materials, such as kiln-utensils, to ensure the highest probability on matching their provenance with the local manufacture. Moreover, the most representative forms and styles were selected in order to provide a typological characterization of the pottery from the medieval workshops of Hospital Viejo Street (see Table 1).

Form and Style	N	MNI	Form and Style	N	MNI
Bowl	**13**	**24**	**Mortar**	**2**	**2**
Lead glazed (LG)	2	6	Lead glazed	1	1
LG-Honey + White Blue	1	1	LG + Stamped ”fleur-de-lis”	1	1
Tin lead glazed (TLG)	1	1	**Paintry Pottery**	**1**	**51**
TLG + White Black Green	2	4	Unglazed	1	51
TLG + White Blue	2	2	**Paste**	**4**	**49**
TLG + White Green	3	4	Unglazed	4	49
TLG + White Luster	1	2	**Pitcher**	**13**	**263**
LG + Mold Decoration	1	4	Lead glazed	3	13
Cooking Pot	**4**	**60**	LG + Incision	1	1
Unglazed	4	60	Unglazed	9	249
Cup	**1**	**1**	**Plate**	**3**	**4**
Lead glazed	1	1	Lead glazed	1	1
Jar	**5**	**54**	TLG + White Luster	1	1
Lead glazed	1	3	Honey + TLG White Black	1	2
LG + Grooved	1	5	**Porringer**	**1**	**4**
LG + Stamped	1	1	Lead glazed	1	4
Unglazed	1	7	**Roof Tile**	**9**	**48**
LG + Mold Decoration	1	38	Incision	1	2
Jug	**2**	**4**	Lead glazed	3	7
Unglazed	1	3	Unglazed	5	39
Unglazed + Black	1	1	**Undefined**	**9**	**49**
Kiln Utensil	**7**	**8**	Lead glazed	1	0
Unglazed	7	8	LG + Grooved	1	1
Lid	**3**	**16**	LG + Relieve	1	1
Incision	2	15	LG + Relieve + ”fleur-de-lis”	1	1
Tin lead glazed	1	1	Unglazed	5	46
			Total	**77**	**637**

Table 1: Quantification of analyzed ceramic sample (N) with respect to the whole assemblage (MNI) regarding ceramic forms (in bold) and styles.

The quantification of sherds was based on the minimum number of individuals (Shennan, 2014). The set of ceramics subjected to analysis is a sample (n = 77) of the whole assemblage (MNI = 637) and consists principally of kiln utensils, ceramic wastes, tableware, cookware and pantry pottery (see Table 1). These include unglazed, lead glazed and tin lead glazed coatings, some of which show blue, green, black, luster or a combination of these colors. Regarding the decorative techniques, among the lead glazed and unglazed pieces ribbed or stamped patterns can be found, as well as incisions or reliefs. Two of the analyzed ceramics (LOG037 and LOG054) showed a *fleur-de-lis* that was either stamped or as relief (see Figure 3).

According to the archaeologically characterized strata (see Figure 2), chronologically the selected ceramics dated from the 13^{th} to the 15^{th} centuries. In addition, ceramics corresponding to later chronologies (16^{th}-18^{th} centuries) were also selected in order to examine their compatibility with previously produced ceramic pastes and ascribe them, where appropriate, to exogenous provenances. Most of the analyzed ceramics correspond to the most active periods of the unearthed kilns, that is, during 1290-1350 and 1400-1500 (see Table 2).

An archaeological examination and documentation of a sub-sample of the materials was previously carried out (Angulo and Porres, 2015). Moreover, a Doctoral Thesis was conducted from an archaeological perspective based on the materials from the first of the two interventions (Martínez, 2013) and a summary focused on the main typologies and including the most representative ones (n = 35), was subsequently published (Martínez González, 2015). Despite there having been a large archaeological work documenting this site and its materials, until now none of the works have considered their archaeometric examination.

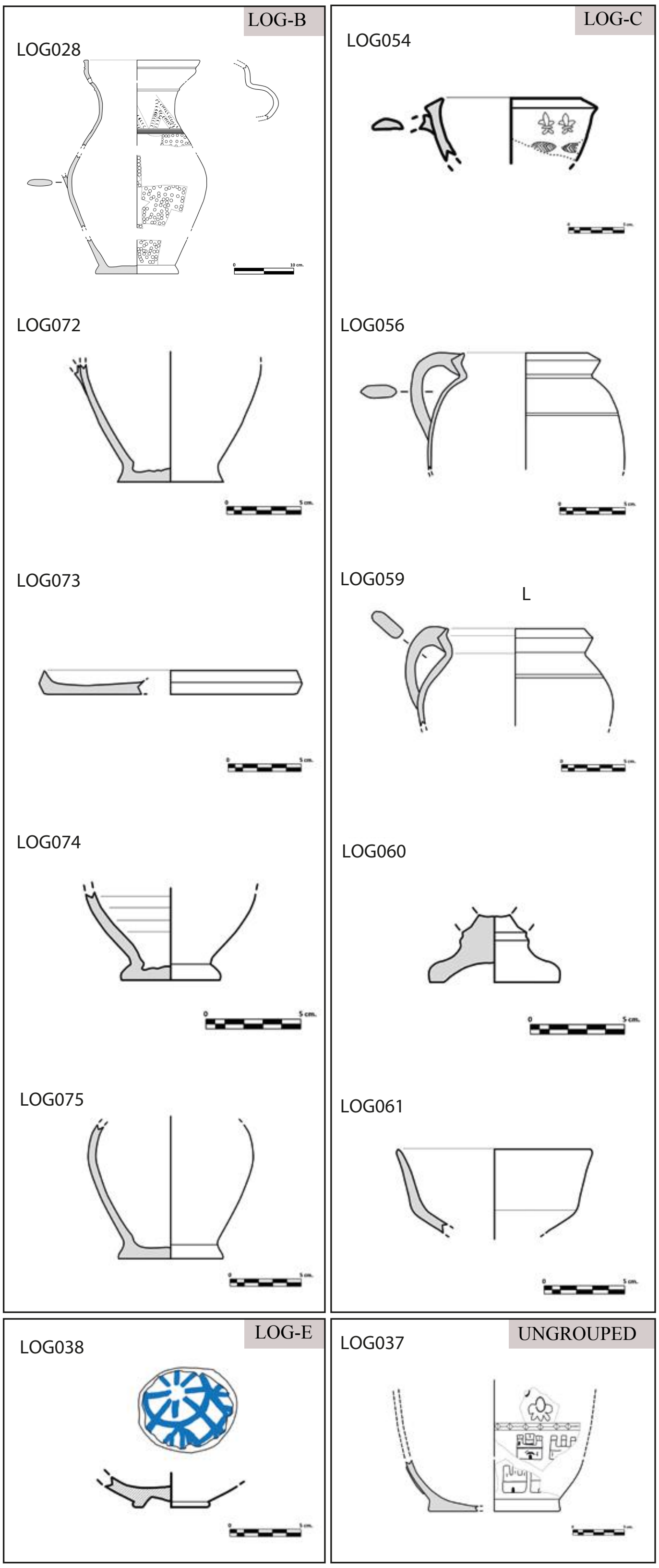

Fig. 3: Representative profiles of the ceramic assemblage from Hospital Viejo site in Logroño

Chronology	N	MNI
1200-1400	1	1
1200-1500	1	7
1290-1350	26	420
1350-1400	3	3
1350-1450	1	1
1400-1433	11	30
1433-1466	17	69
1466-1500	11	99
1500-1533	1	1
1600-1800	5	6
Total	**77**	**637**

Table 2: Approximate chronological frames of the ceramic sample (N) with respect to the whole assemblage (MNI).

2.2 Experimental

In order to carry out provenance analysis, the characterization of the paste compositions and mineralogical phases was carried out. Thus, the ceramic sherds were subjected to inductively coupled plasma mass spectrometry (ICP-MS) and X-ray diffraction (XRD) based analysis. Furthermore, a selection of 25 sherds was analyzed with NAA and the data compared to a database of majolica ceramic productions (Iñañez, 2005; Iñañez et al., 2008). In addition, a selection from these sherds (n = 16) was analyzed by means of scanning electron microscopy-energy dispersive spectrometry (SEM-EDS) in order to shed light on the understanding of the glazing technology used by the potters.

2.2.1 Provenance Analysis and Fabric Technology

For the provenance analysis and fabric technology characterization ICP-MS, NAA and XRD were used, under the following conditions:

Inductively Coupled Plasma Mass Spectrometry (ICP-MS): Each sherd was powdered (ca. 15 g) using wolfram carbide cells for 2-4 min at 300 rpm. Previously, surfaces were mechanically removed in order to minimize contamination from glaze and soil. The powdered samples were calcined (at 1000 °C), and along with the Certified Reference Materials (JB-3, JA-2, JG-1A, JG-2), were fused (250 g) in Pt-Au crucibles, mixing it with 500 mg of a flux ($LiBO_2$) at more than 1100°C (Madinabeitia et al., 2008). After dilution with nitric acid the solutions were analyzed by means of a Nexion 300 ICP-MS (Perkin Elmer). The internal standard solutions (Sc, Y, In, Be, Bi) were prepared from 1000 $\mu g/mL$ stock solutions of Alfa Aesar inside a class 100 clean room with an analytical balance with an uncertainty of ± 0.0001 g. The accuracy and reproducibility of the method was checked by repetitive analysis (n = 3) of the certified reference materials. For more details see the procedure followed elsewhere (Calparsoro, 2019; Calparsoro et al., 2019a,b,c).

Neutron activation analysis: NAA was conducted in the Archaeometry Laboratory of the University of Missouri Research Reactor (MURR). For more details see (Glascock, 1992). The powdered samples were dried at 100 °C for 24 h. Approximately 150 mg and 200 mg of sample were used for short and long irradiations, respectively. For short irradiations a thermal flux of $8{\cdot}10^{13} n/cm^{-2}s^{-1}$ was applied and γ rays were counted after 5 second of exposure and 25 minutes of decay, yielding data for: Al, Ba, Ca, Dy, K, Mn, Na, Ti and V. For the long irradiation (24 h) a flux of 5 x $10^{13} n/cm^{-2}s^{-1}$ was used. After seven days of decay the counting with a high-resolution germanium detector yielded data for As, La, Lu, Nd, Sm, U and Yb. After additional two-week decay, a second count was carried allowing quantification of 17 long-life elements: Ce, Co, Cr, Cs, Eu, Fe, Hf, Ni, Rb, Sb, Sc, Sr, Ta, Tb, Th, Zn and Zr. With the samples batch, SRM-1633a (coal fly) was prepared as standard, as well as quality control samples of SRM-278 (obsidian rock) and Ohio Red Clay (for analytical conditions, see (Glascock Michael D and Neff, 2007).

Statistical treatment: The statistical analysis applied in the current work is based on Aitchison's approach and Buxeda's observations on compositional data (Aitchison et al., 2000; Buxeda i Garrigós and Kilikoglou, 2003; Buxeda i Garrigós, 2008). Thus, similarity of sherds and subsequently their hypothetical provenance according to the provenance postulate (Weigand et al., 1977) was tested applying algorithms for compositional data written on R scripts. In order to compensate for the differences in absolute magnitude between major (%) and minor and trace elements (ppm), log-ratio transformations were employed dividing all chemical components by the element that introduces the lowest variability to the entire data set (alr) (Aitchison, 1986). This transformation was used for the principal component analysis (PCA). In contrast, for the hierarchical cluster analysis (HCA) centered log-ratios (clr) were preferred in order to identify better the clusters when plotting on a dendrogram the squared Euclidean distance using the centroid algorithm (Pawlowsky-Glahn and Egozcue, 2011). These transformations were chosen (alr and clr) in order to be coherent with other works in the same research line on pottery from the Iberian Peninsula (De Marcos García et al., 2017).

X-Ray Diffraction (XRD): XRD combined with SEM was used to characterize the fabrics and determine the Estimated Firing Temperatures (EFT). The EFT, which depends on several factors (redox, heating rate, maximum temperature, soaking times, etc.), can be linked to the mineral phases occurring in the fabrics (Tite, 1995) and the microstructure sintering level (Maniatis and Tite, 1981a).X-ray diffraction analyses were performed on the powdered fabric samples with a PANalytical X'pert PRO powder diffractometer equipped with a copper tube ($\lambda_{CuK\alpha_{mean}}$ = 1.5418 Å, $\lambda_{CuK_1\alpha}$ = 1.5,4060 Å, $\lambda_{CuK_2\alpha}$ = 1.54439 Å), vertical goniometer (Bragg-Brentano geometry), programmable divergence aperture, automatic sample changer, secondary graphite monochromator, and PixCel detector. The operating conditions for the Cu tube were 40 kV and 40 mA, and the angular range (2θ) was scanned between 5°and 70°. The treatment of the diffractogram data and the identification of the mineral phases present was carried out with the X'pert HighScore (PANalytical) software package in combination with the Powder Diffraction File (PDF-2) database (International Center for Diffraction Data (ICDD), Pennsylvania, USA).

For the micro-structural characterization of the pastes using SEM (the details are explained in the next section), fresh fractures were cut perpendicularly to Z axis of each piece in order to better assess the clayey inner structure, following the methodology used elsewhere (Maniatis and Tite, 1981b). For a better comparison of the microstructure, SEM images showing different vitrification levels were acquired at a x2000 magnification.

2.2.2 Glazing Technology

To evaluate the glazing technology, SEM-EDS was used in combination with XRD as described before.

Scanning Electron Microscopy-Energy Dispersive Spectroscopy (SEM-EDS): The glaze characterization was performed by EDS Quanta 200 from Fei environmental scanning electron microscopy (ESEM) in combination with the integrated energy dispersive XFlash 4010 X-ray analyzer from Bruker. Cross sections of the samples were previously prepared in epoxy molds and polished down to below 1 μm grain size. The elemental compositions of glazes were determined by EDS on areas of 50 μm^2 corresponding to each color. Data were collected at voltage of 15-20 KV and a current of 50 μA for the acquisition of images, and 100 μA for the analyses, at 11.5 mm working distance (WD) and low-vacuum. The detector was switched to secondary electrons (SE) for image acquisition and back scattered electrons (BSE) for analyses.

3 Results and Discussion

3.1 Evaluation by NAA and identification and provenance identification

A first approach based on NAA was performed on a sub-sample of 25 individuals. For provenance analysis, a collation with comparable NAA data of other majolica workshops was carried out (Iñañez, 2005; Calparsoro, 2019). The entire array of production centers tested was based on the contemporaneity and was the following: Barcelona, Lleida, Manises, Muel, Paterna, Sevilla, Talavera, Teruel, Villafranca de Penedes, Villafeliche and Nájera. As a result, the situation of the ceramics from Logroño was displayed within a broader perspective (see Figure 4). Whereas in all cases the matches were negative (except for Teruel and Valencia as discussed below), for the sake of clarity only the most significant sub-sample was included in the Figure 4 (for more details see the Supplementary Material).

Interestingly, two compositional groups were discriminated based on the sampling of wasters and unfinished wares of presumably local origin: LOG-B and LOG-C. These groups match the classification obtained by ICP-MS and are addressed in Sections 3.2.2 and 3.2.3, respectively. On the other hand, four sherds were clearly compatible with the provenance of Teruel (further discussed in Section 3.2.6). In addition, LOG037 and LOG038 showed differentiated composition within the dataset. The former (LOG037) was noted for its high Ba concentration, as well as minor and trace elements. This sherd was decorated with a *fleur-de-lis* (see Figure 3), which is often associated with a French origin (see further discussion in Section 3.2.7).

The latter (LOG038), was linked to a Valencian origin, based on the chemical compatibility observed by NAA. This hypothesis is supported by its

Valencian characteristic decoration of blue strokes on a tin lead glaze (see Section 3.2.5). Although in the Figure 4, this individual is clustered with Seville, previous studies demonstrated that the chemical discrimination of Manises from Seville is highly sensitive to Na and K (Iñañez et al., 2008). These elements were not included in the main statistical treatment for the potential alterations explained in Section 3.2). In contrast, including these elements (see Figure 4 of Supplementary material) allowed confirming that LOG038 is strongly compatible with the Valencian production center of Manises. Moreover, to exclude the possibility of a Sevillian origin, it was contrasted with the data from the extensive collection of Sevillian ceramics recently published (Fernández de Marcos, 2018) and no parallel was found.

Finally, it should be noted that none of the ceramics from Logroño matched the NAJ-A compositional group from Nájera characterized elsewhere (Calparsoro et al., 2019c). Therefore, from the present NAA evaluation it can be assumed that it is possible to discriminate between the pottery productions from these two neighboring cities (separated by 28 km), since the compositional groups from those production-centers revealed a sufficient level of resolution.

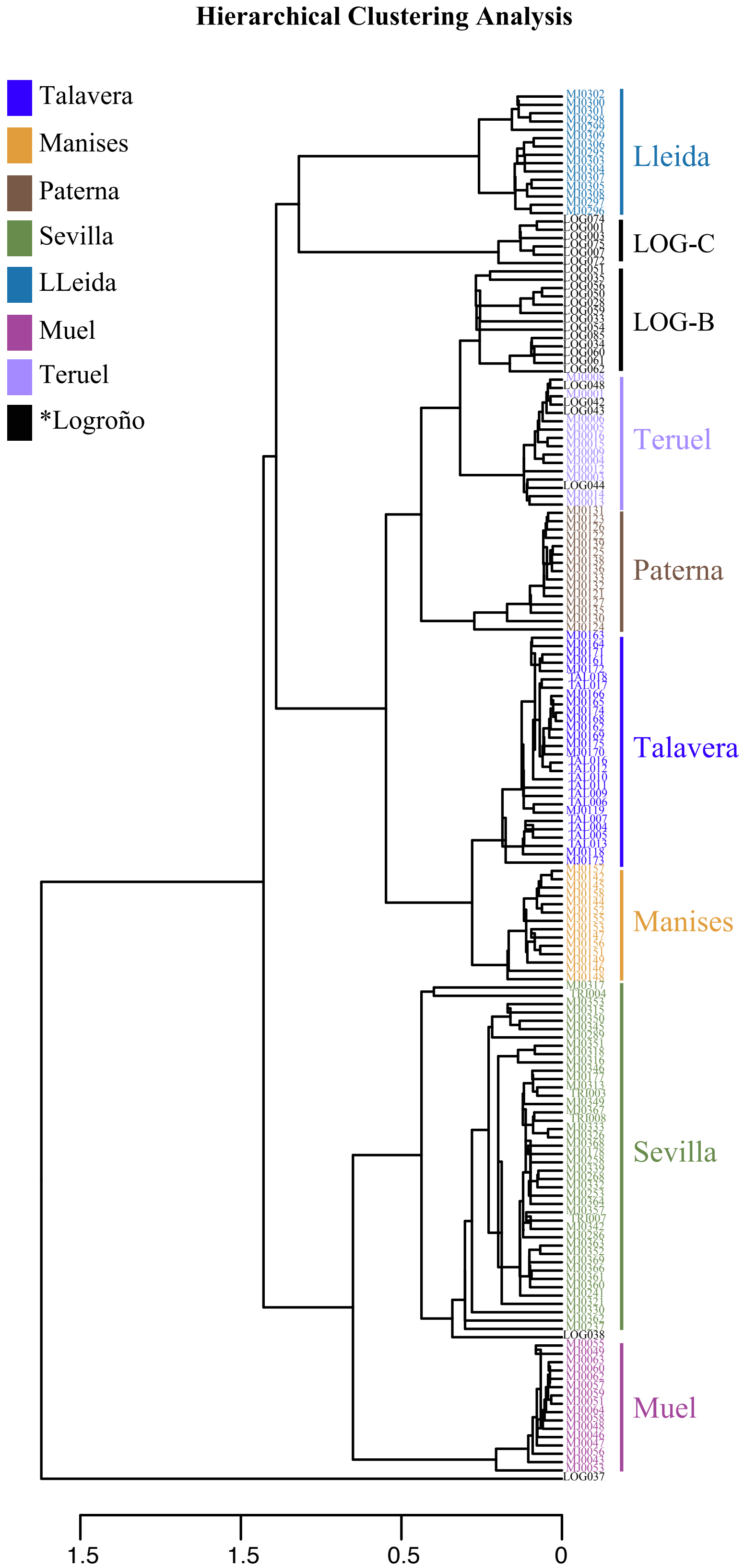

Fig. 4: HCA of 25 sherds from Logroño (in black), collated with compositional groups from main Iberian majolica production centers (Iñañez, 2005). LOG-B and LOG-C constitute isolated groups, four sherds are compatible with Teruel and LOG037 and LOG038 remain ungrouped. This sub-composition obtained by NAA is used: La, Lu, Nd, Sm, U, Yb, Ce, Cr, Cs, Eu, Fe, Hf, Sc, Tb, Th, Zr, Al, Ba, Dy, Mn, Ti and V.

3.2 Integrated discussion of the Compositional Groups (CG)

For the discrimination of the compositional groups (CG), the ICP-MS data of the 76 sherds were used (see Supplementary Material). Furthermore, NAA observations (addressed in the Section 3.1) were employed to endorse the results, as well as for the provenance identification.

Ideally, to obtain the CGs the compositional variance should be restricted to natural sources, excluding experimental errors and/or alterations arising from post-depositional processes. Accordingly, certain elements were not regarded for the statistical analysis: Pb and Sn due to the glaze-influence (Molera et al., 2001), P and Cu due to post-depositions (Freestone, 1984), Co and Ta due to sample-preparation process with wolfram carbide cell (Boulanger et al., 2013), and Na, K and Rb due to alteration by analcime (Schwedt et al., 2006). Moreover, Zn was not used because some of the values were very close to the limit of quantification (LOQ). Finally, in archaeological ceramics, the influence of the technological choices on the concentrations of CaO and Sr were taken into account. These can vary for a variety of reasons (e.g. carbonate types) (Buxeda i Garrigós, 1999; Schwedt et al., 2006; Fabbri et al., 2014).

The compositional heterogeneity was assessed by calculating the compositional variation matrix (CVM), which provides information about the variability introduced by each element into the dataset (see Figure 1 of Supplementary Material). A high value of the total variation (vt), as in this case (vt = 0.95), depicts a polygenic dataset (Buxeda i Garrigós and Kilikoglou, 2003). Thus, several compositional groups were expected within this ceramic assemblage.

Alternatively, the ternary diagram provides an overview of the mineralogical composition (see Figure 5), which is inferred from the chemical composition (Heinmann, 1989). Except for two individuals, a high mineralogical evenness was observed, since all sherds appeared within the wollastonite-quartz-anorthite triangle. Furthermore, two trends in CaO content were identified, depending on the final finish: lower calcareous for lead glazed and higher calcareous for tin lead glazed pottery. Historically, calcareous pastes have been preferred for tin lead glazed pottery, since they provide clearer pastes, and therefore, they require less opacifier, thus, reducing the production costs (Iñañez et al., 2008).

After the statistical analysis (see Figure 6), six compositional groups were identified (details of each sample are included in Supplementary material). The mean values with the standard deviations of each compositional group are summarized in the Table 3. The discussion of each compositional group combining the results yielded by ICP-MS, NAA, XRD and SEM-EDS is addressed in the following lines.

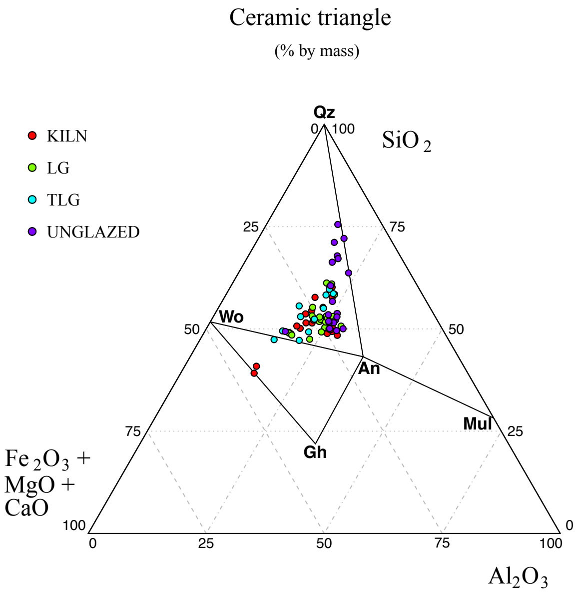

Fig. 5: Ternary diagram showing the compositions obtained by ICP-MS of SiO_2, Al_2O_3 and CaO+MgO+Fe_2O_3, corresponding to the 76 potsherds from Logroño. TLG: Tin lead glazed, LG: Lead glazed and KILN: kiln utensils. An:Anorthite, Gh:Gehlenite, Mul:Mullite, Qz:Quartz and Wo:Wollastonite. Aabbreviations after Whitney and Evans (2010).

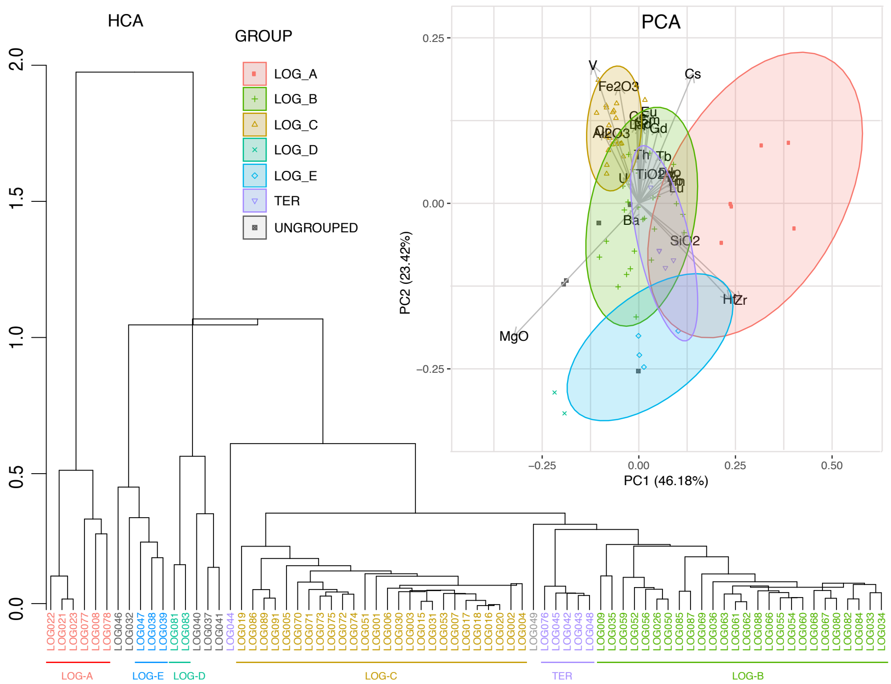

Fig. 6: Dendrogram of Euclidean squared distances using centroid algorithm and PCA of 76 individuals from Logroño on the sub-composition of Al_2O_3, Zr, Nb, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, U, MgO, SiO_2, TiO_2, V, Cr, Fe_2O_3. For the PCA Pr was used as a divisor in the alr transformation.

Element	LOG-A	SD	LOG-B	SD	LOG-C	SD	LOG-D	SD	LOG-E	SD	TER	SD
N of Individuals	(n = 6)		(n = 27)		(n = 25)		(n = 2)		(n = 3)		(n = 5)	
Al_2O_3	14.0	0.8	16.9	1.7	21.6	1.5	12.7	0.1	14.1	1.5	17.8	0.8
CaO	3.7	1.1	12.9	4.5	9.3	1.1	30.8	0.7	19.8	4.0	8.5	0.7
Fe_2O_3	5.1	0.7	5.8	0.7	7.7	0.4	3.4	0.1	4.9	0.5	6.0	1.2
K_2O	2.3	0.2	3.1	0.4	3.5	0.2	2.1	0.1	2.1	0.5	3.7	0.3
MgO	0.9	0.19	1.92	0.22	2.18	0.15	2.59	0.05	2.82	0.27	1.61	0.08
MnO	0.025	0.019	0.045	0.012	0.039	0.004	0.01	0.001	0.056	0.003	0.032	0.008
Na_2O	0.75	0.1	0.8	0.14	0.72	0.07	0.61	0.2	0.72	0.56	0.29	0.04
P_2O_5	0.18	0.07	0.25	0.07	0.35	0.19	0.19	0.01	0.47	0.18	0.24	0.06
SiO_2	55.5	4.9	43.9	4.1	43.1	2.7	33.1	1.3	41.6	3.2	48.3	1.6
TiO_2	0.75	0.05	0.69	0.08	0.82	0.03	0.46	0.01	0.72	0.1	0.71	0.02
Ba	474	53	537	71	572	44	322	38	562	132	566	25
Ce	92	6	95	12	119	5	53	1	88	9	89	3
Co	16	2	20	7	24	7	9	1	18	1	22	5
Cr	69	15	82	10	102	16	69	2	67	7	76	25
Cs	27	5	21	4	22	3	11	2	9	3	29	3
Cu	63	107	30	17	25	25	23	16	106	66	118	88
Dy	5.7	0.3	5.2	0.6	5.8	0.2	2.8	0.1	5.5	0.3	5.3	0.3
Er	3.1	0.2	2.7	0.3	3.0	0.1	1.5	0.1	2.9	0.1	2.7	0.1
Eu	1.5	0.1	1.5	0.2	1.8	0.1	0.7	0.1	1.4	0.2	1.4	0.1
Gd	8.3	0.3	7.6	0.9	9.1	0.4	3.8	0.1	7.6	0.4	7.3	0.3
Hf	5.1	0.6	3.3	0.6	2.9	0.4	2.0	0.1	4.0	0.3	3.4	0.2
Ho	1.1	0.07	0.93	0.1	1.05	0.05	0.5	0.01	1.05	0.05	0.94	0.04
La	45	3	46	6	58	3	27	1	44	4	43	1
Lu	0.47	0.02	0.41	0.06	0.44	0.02	0.22	0.02	0.44	0.04	0.42	0.02
Nb	17	1	18	2	20	1	13	0	17	1	19	1
Nd	38	1	39	5	48	2	21	1	37	2	37	2
Pb	48	17	(81-4001)*	-	(24-12077)*	-	1052	876	3534	893	934	464
Pr	10	0	10	1	12	0	6	0	9	1	9	0
Rb	120	11	143	14	156	10	104	14	89	9	186	10
Sm	8.0	0.3	7.8	1.0	9.6	0.4	4.0	0.1	7.5	0.5	7.5	0.4
Sn	5	0	(4-49)*	-	5	1	5	1	23	2	43	40
Sr	129	85	622	256	499	65	1930	175	527	123	235	55
Ta	1.1	0.1	1.1	0.1	1.2	0.1	0.9	0.1	1.0	0.1	1.3	0.1
Tb	0.99	0.05	0.90	0.11	1.03	0.04	0.48	0.10	0.90	0.04	0.88	0.03
Th	9	1	9	1	11	1	6	1	9	1	9	1
Tm	0.54	0.04	0.48	0.06	0.52	0.02	0.24	0.10	0.52	0.03	0.47	0.02
U	2.4	0.2	2.9	0.2	3.1	0.2	2.0	0.1	2.5	0.1	2.4	0.1
V	76	5	101	15	139	9	73	10	78	8	85	2
Yb	3.2	0.2	2.7	0.3	3.0	0.2	1.5	0.1	3.0	0.2	2.8	0.1
Zn	86	6	101	30	101	14	92	14	91	7	159	160
Zr	363	48	217	41	189	21	125	9	267	22	230	9

Table 3: Mean concentrations and standard deviation values (SD) of each compositional group from the Hospital Viejo archaeological site. TER: Teruel. Concentrations obtained by ICP-MS. Oxides are expressed in mass % and the rest in μg/g. *Pb and Sn ranges are given because they contain SD values higher than mean values due to their heterogeneous distribution.

3.2.1 LOG-A Compositional Group

The lowest concentrations of CaO and MgO (3.7 ± 1 and 0.9 ± 0.19 mass % respectively) characterize this group formed by six individuals (representing 75 MNI). In contrast, it shows the highest concentration of Zr and Hf, with values of 363 ± 48 μg/g and 5 ± 1 μg/g, respectively. According to the XRD analysis, all of the sherds have the same fabric type, F- I (see Table 4). The presence of illite-muscovite phases along with the K-feldspars (microcline) and plagioclases (albite), allowed estimating its EFT at 850-900°C (see Figure 13). The abundant quartz observed for this fabric is in accordance with the high concentrations of Zr and Hf, which are associated to the sandy phases in the ceramic body. As a result of all these factors, dark-reddish pastes were obtained (see Figure 7).

The forms include unglazed pots and lids used for cookware and pantry. Many of them were most probably wasters, since multiple cooking pots did not show firing traces from their use (they could have exploded during their own firing). Chronologically, the ceramics of this group correspond to two periods, 1290-1350 and the first third of the 15^{th} century, the former being all the cookware pieces and the storage from the latter. Therefore, it can be suggested that LOG-A underscores a local cookware production, which would have competed with Zamoran imports. The Zamoran ware was characterized by its refractory properties (with very micaceous pastes) and its trade has been documented since the 14^{th} century in the north of the peninsula (Solaun, 2005).

Fig. 7: A representative ceramic from the group LOG-A.

3.2.2 LOG-B Compositional Group

This group is characterized by the highest Al_2O_3 concentration of the whole dataset (21 ± 1 mass %). It shows calcareous pastes (CaO 13 ± 5 mass %). The 27 individuals that form it include a wide range of forms, which presumably would have been produced over the whole life-cycle of the three pottery workshops (13^{th}-15^{th} centuries). These are jars, porringers, small jugs and cups mostly unglazed but also coated by translucent greenish-honey glazes, as well as plenty of kiln utensils such as trivets or roof tiles.

Moreover, some of the sherds identified in this group correspond to strata of 17^{th}-18^{th} centuries (see Figure 2). Their compositional compatibility was taken as a basis to include them in the compositional group. Therefore, since their chronologies go beyond the functioning time of the unearthed kilns, it can be suggested that they were most probably manufactured in the post-medieval ceramic workshops of the *Ollerias* Streets introduced in Section 1.3 (see Figure 1).

After the mineralogical evaluation (see Table 4), two fabric types (F-I and F-II) were identified in LOG-B compositional group. Both showed the existence of quartz along with firing phases such as plagioclases (albite) and pyroxenes (diopside). Their coexistence with the lack of illite-muscovite phases and mullite, and the presence of K-feldspars (microcline), in the case of F-I, permits estimating the EFT at ca. 900°C. K-feldspars start decomposing at 850-900°C approximately. Therefore, their absence in the F-II accompanied by an enrichment of plagioclases and gehlenite (a sub-product of calcite) suggested a higher EFT than F-I, thus, ca. 950°C. Moreover, the detection of hematite in F-I and calcite in F-II are connected with the respectively higher Fe_2O_3 and lower CaO content of both fabrics.

Most of the pieces from this group are unglazed kiln utensils. Regarding the few pieces that are coated, they bear a lead glaze of varying PbO concentrations, as ascertained by SEM-EDS (see Table 5), whereas the only tin lead glazed sherd (LOG062) corresponds to the post-medieval chronologies.

Regarding LOG009, this piece shows manganese bearing decorations and dates to 1290-1350. Literature attests to the use of manganese based decoration since the mid-fourteenth century, first in jugs of common tableware and, later, from the beginning of the 15^{th} on pitchers and small jugs (Martínez, 2013). Indeed, similar forms have been documented in other findings of the city (Solaun, 2005, p. 233). Moreover, parallel ceramics from nearby locations, can be found in the medieval pottery workshop discovered in Calahorra (Luezas, 2014), whereas manganese decorations were ubiquitous after the 14^{th} century.

In some of the sherds the influence of the calcareous pastes could be observed regarding the clay-body and glaze interaction. During the firing, the molten glass reacts with the body and diffusion of the elements from the paste into the glaze and vice versa occurs (Molera et al., 2001). In this case, the formation of Ca-rich pyroxenes enhanced by the high concentration of CaO was identified. These are incorporated to the glaze, but they do not contribute to the interface formation because they appear floating on the glaze (see Figure 8). Likewise, K and Al rich crystallites growing from the clay-body to the glaze could be observed, especially in sherds rich in K-feldspars (see Figure 8). These interface crystallite formations play a role

GROUP	Fab.	EFT (°C)	Qz	Ilt-Ms	Mul	Afs	Cal	Hem	Gh	Pl	Px
LOG-A	F-I	850-900	xxxx	xx		x				x	
LOG-B	F-I	900	xxxx			x		x		xx	x
	F-II	950	xx				x		xx	xx	x
LOG-C	F-I	850-950	xx	x		x		x	x	xx	
LOG-D	F-I	1000-1050	x		x		xxx		xxx		x
LOG-E	F-I	900-1000	xxx				x	x	xx	x	x
TER	F-I	900-1000	xxxx			x		x		xx	x

Table 4: Fabrics identified in each compositional group and their main mineralogical phases and EFTs. Qz: Quartz; Ilt-Ms: Illite-Muscovite; Cal: Calcite; Gh: Gehlenite; Hem: Hematite; Pl: plagioclases; Px: Pyroxenes; Mul: Mullite; Spl: spinels. Abbreviations after (Whitney and Evans, 2010).

in the final stability of the ceramic coating (Molera et al., 2001). The nature of the interface is usually taken as indicative of single or double firing. According to this indicator, most of the ceramics subjected to analysis, experienced a single firing for the lead glazed pottery, while the tin lead glaze, would have been applied on bisque ceramics.

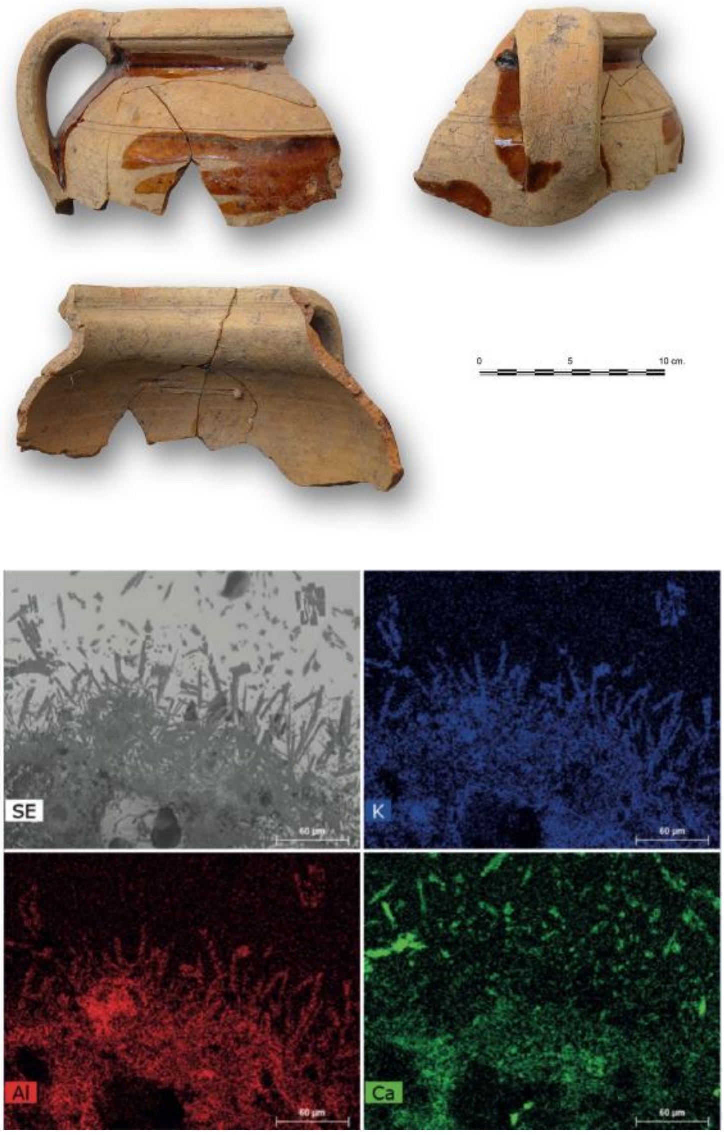

Fig. 8: A representative ceramic (LOG059) from the group LOG-B (Source: ArqueoRioja) and SEM-EDS images and Ca, Al and K elemental maps showing the growth of crystallites in ceramic body and diffusing into the glaze.

3.2.3 LOG-C Compositional Group

Based on its size (n= 25), LOG-C is the most chemically compact group of the assemblage (see Figure 6). While it is compositionally very similar to LOG-B (calcareous and iron-rich), this CG is featured by a high concentration of Al_2O_3 (21.6 ± 0.5 mass %), probably linked to the presence of mica. The pastes are finely decanted and orange-reddish (see Figure 9). After XRD analysis, one fabric type (F-I) was identified for this CG, characterized by the presence of quartz, with plagioclases and K-feldspars, together with hematite and gehlenite (see Figure 13). The coexistence of these phases and the weak peak of illite-muscovite indicated a low firing temperature (850-950°C). Consequently, the microstructure of the clay-body reached a low sintering level (see Figure 9).

The forms include mostly unglazed pitchers (MNI 263) and pantry pottery (MNI 51), (see Figure 3). Moreover, numerous kiln utensils and wasters constitute this group, including many potentially discarded pieces marked by firing-burns. On that basis, this CG could be linked to a local production, providing a clear chemical fingerprint. The scarce lead glazes present, were obtained by PbO-rich recipes (see Table 5).

Half of the ceramics from this group were manufactured in the period of 1290-1350 and the other half correspond to later chronologies (late 14^{th} and the 15^{th} centuries). Representative forms of this group such as the pitcher (e.g. LOG002) find parallels with micaceous grooved ceramics of Groups IV and V documented in nearby workshops (Solaun, 2005, p. 161) and (Pérez, 1985, p. 8). Seemingly, these pantry ware forms were abundant all over the northern Iberian peninsula, reporting specifically a certain cultural homogenization at regional level among the workshops of Logroño, Gasteiz and Valdegovía (Solaun, 2005, p. 371).

The diachronic evolution of ceramics from LOG-C reveals a continuity of an important local pantry and tableware ceramic production. Its chemical and mineralogical homogeneity reveals a manufacture by similar clays and/or using the same recipes during the entire operational period of the workshops from Hospital Viejo Street. This evenness coincides with the specialization of the craft which replaced a domestic and itinerant production that took place during ca. 12^{th} century (Solaun, 2005, p. 383)

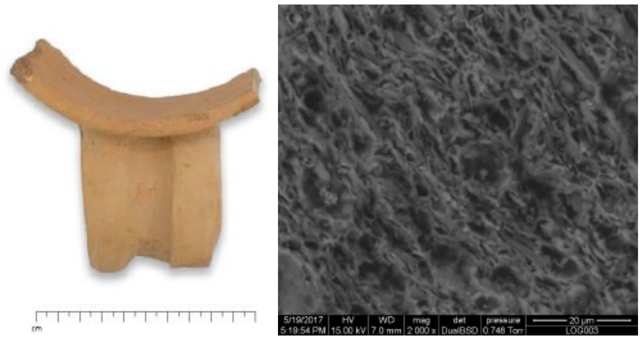

Fig. 9: A representative ceramic (LOG004) from the group LOG-C and SEM image showing the low sintering level of the paste.

3.2.4 LOG-D Compositional Group

LOG-D is the smallest group of the assemblage, comprising only two roof tiles (MNI 39). According to the statistical analysis (see Figure 6), the clear differentiation of this CG relies on its strikingly high CaO content (31 ± 1 mass %). Despite the omission of CaO and Sr, which cannot be exclusively regarded as provenance markers (see section 3.2), the results still permitted the clear discrimination of this group. The very different composition could also be observed in the ternary diagram (see Figure 5). According to the XRD analyses, both sherds showed F-I fabric type, that were characterized by very prominent peaks of calcite and gehlenite. Moreover, the coexistence of mullite and the secondary phases, such as gehlenite, suggests an EFT between 1000-1050°C. Roof tiles were extensively used as kiln tools, for separating the pieces during the firings. Therefore, this high EFT (the highest of the assemblage), could be linked to a recurrent firing, confirming their use as kiln utensils. Nonetheless, unlike trivets that are manufactured specifically as potters' tools, the possibility that the roof tiles were recycled from somewhere else, should be considered. Consequently, in the absence of other wasters and/or clues, this group could not unequivocally be linked to a local production, reason for which its provenance was considered undetermined.

Fig. 10: Roof tile from LOG-D, showing drops of glaze (LOG081).

3.2.5 LOG-E Compositional Group

This group includes three individuals that are characterized by bichrome tin lead glazes and buffer colored clay-bodies (see Figure 11). It is calcareous (16 $\pm$ 2 mass %) and could be discriminated primarily due to higher concentrations of MgO, Hf and Zr. The chemical differences matched those observed by NAA for LOG038. Moreover, this individual was ascribed to Manises as seen by NAA collation (see Section 3.1).

The XRD analysis pointed out one fabric type (see Table 4). The fabric (F-I) showed intense peaks of calcite (see Figure 13), in addition to the lack of illite and presence of gehlenite (a decomposition product of calcite). Thus, its EFT was estimated between 900-1000°C. This EFT is slightly higher than those observed for the local groups from Logroño, which can be linked to its exogenous provenance.

Two of the individuals correspond to 16-18th centuries (LOG038 and LOG039), while LOG047 dates to a previous chronology (15th century). The sherd ascribed to Manises (LOG038), shows geometric strokes painted with cobalt blue on white obtained by tin opacifier as ascertained by SEM-EDS (see Table 5). This type of decoration (see Figure 11) is considered as characteristic from certain Valencian workshops. See for instance the parallel ceramics from the local dump of Molí located in Paterna (Valencia) from the 14th century (Mesquida García, 2002) or the findings of Barri d'Obradors in Manises (Conesa Coll et al., 2013). Alternatively, a parallel piece ascribed to Valencia (inventory number N.441, S.LHV.003.54) was documented at the Hospital Viejo site in previous studies (Martínez, 2013). In addition, the luster decoration shown by LOG039 is usually associated with the Valencian area (Pérez-Arantegui et al., 2001; Conesa Coll, 2008). The closeness between Manises and Paterna workshops involves a known archeometric problem which hinders the proper chemical differentiation between the two Valencian provenances (Iñañez et al., 2008).

Fig. 11: Representative ceramics from LOG-E group (LOG038 and LOG039).

3.2.6 TER Compositional Group

The TER group is formed by tin lead glazed pottery showing characteristic green-black decorations from Teruel. According to the analysis performed by NAA, the origin of LOG042, LOG043, LOG044 and LOG048 is clearly associated to Teruel (see Section 3.1). The statistical analysis based on ICP-MS matches those results grouping these individuals together, and including LOG076 (see Figure 4).

This CG includes bowls from different periods: between the early 15^{th} to the early 16^{th} centuries (see Figure 12). The pastes of this group were iron-rich (6 ± 1.2 mass % of Fe_2O_3) and medium-high calcareous. Despite the high CaO content (CaO 9 ± 1 mass %), the reddish pastes contrasted to other majolica pottery identified in this study (LOG-E), which presented a more characteristic light buff colored clay-body of this ceramic typology (Iñañez et al., 2008). Regarding the XRD analysis, among the individuals of TER group only F-I type fabric was identified. The mineral phases detected were quartz, K-feldspars, plagioclases and pyroxenes (see Figure 13). The concurrence of all these phases permits estimating the EFT at 900-1000°C, revealing again higher firing temperatures in comparison to the unglazed productions from Logroño. Therefore, a higher firings-temperature could be expected from the bisque ceramics.

According to the archaeological and historical documentation, the green and black decorations, such as those shown by LOG042 and LOG043 are characteristic from Teruel (Conesa Coll, 2008). In this case, the greens were obtained by Cu, the blacks by Mn and the whites by Sn (see Table 5). These substances correspond to traditional recipes which have been used during centuries for majolica production (Iñañez et al., 2008).

Fig. 12: Representative ceramic from TER group ascribed to Teruel (LOG043)

3.2.7 Ungrouped Individuals

Apart from the discrimination of the local and exogenous groups, there were some sherds that could not be grouped and/or associated with any provenance. Firstly, LOG037, as observed by NAA was the most differentiated ceramic in terms of chemical composition, due especially to a striking Ba concentration (755 μg/g). According to ICP-MS this sherd was chemically similar to LOG041 (tin lead glazed decorated with blue) and LOG040, which shows a Mn-decoration applied on a whitish layer, being the only sherd from the entire assemblage bearing this kind of decoration. Although, they are chemically similar, their different typologies suggested that they most probably corresponded to different provenances.

In the case of LOG037, the decoration showing castles and a partial a *fleur-de-lis*, along with the completely different composition could potentially be linked to a French origin. Seemingly, in the late medieval Basque Country, the growing demand for ceramics surpassed local or regional markets, as the incomes of the residents were incremented due to the integrations into commercial circuits. In this context, imports from as far away as the French region of Saintes are documented (Solaun, 2005, p. 388). Therefore, it is not rare to think that these trades were extended to places like Logroño in the northern Rioja.

Secondly, LOG032 and LOG046 were not ascribed to LOG-E, since they exhibit differences in most of the trace and minor elements, as well as well as presenting a significantly higher concentration of Ba and MgO, among others. Moreover, they did not reveal any typological or chronological compatibility with the other sherds of LOG-E. For a proper archaeological interpretation more sherds with similar chemical features should be analyzed.

Additionally, LOG090 remained ungrouped due to its very low MnO concentration 0.003 mass %. This sherd showed a clear distinction within the ceramic assemblage even when MnO was not considered in the statistical

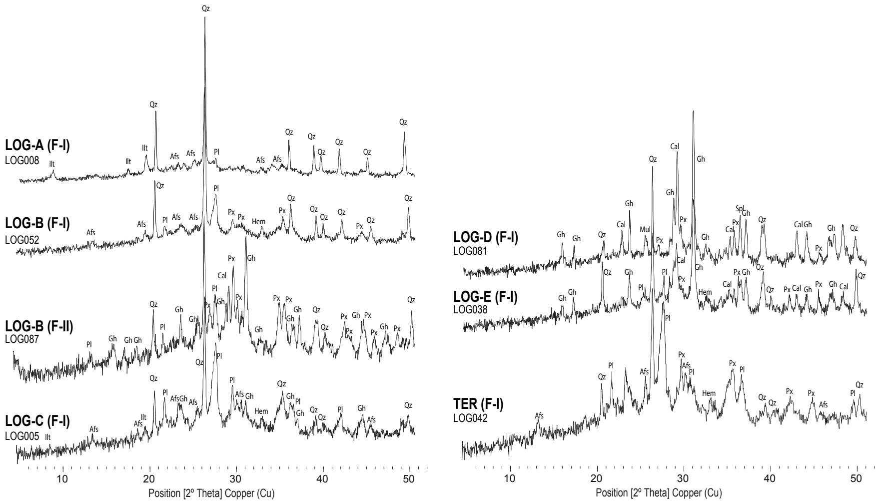

Fig. 13: XRD diffractograms of the fabrics identified in each compositional group of the ceramics from Logroño.

analyses. Finally, LOG049 showed a different chemical fingerprint and appearance in comparison to all the remaining ceramics studied.

Table 5: SEM-EDS Semi-quantitative concentrations of the glazes from a sub-sample of ceramics (n= 16) (concentrations expressed in mass %). Alk: Na_2O+ K_2O. Pb/Si*:PbO/ SiO_2. ND: Non detected.

Spectrum	GROUP	Color	K_2O	CaO	FeO	Na_2O	MgO	Al_2O_3	SiO_2	PbO	TiO_2	SnO_2	CuO	MnO	CoO	ZnO	P_2O_5	Pb/Si*	Alk.
LOG028	LOG-B	Honey	1.07	3.80	2.71	0.36	1.15	9.66	34.87	45.91	0.46	ND	ND	ND	ND	ND	ND	1.3	1.43
LOG034	LOG-B	Honey	1.60	6.32	4.09	0.19	0.44	6.04	30.87	50.09	0.19	ND	ND	ND	ND	0.06	ND	1.6	1.85
LOG050	LOG-B	Greenish	1.00	3.05	2.40	0.47	1.12	8.82	27.67	55.17	0.20	ND	ND	ND	ND	ND	ND	2.0	1.47
LOG054	LOG-B	Honey	0.95	3.49	3.96	0.49	0.78	7.30	31.05	51.33	0.19	0.01	ND	ND	ND	0.45	ND	1.7	1.44
LOG059	LOG-B	Honey	1.68	1.87	3.92	0.41	0.51	4.61	41.86	44.80	0.34	ND	ND	ND	ND	ND	ND	1.1	2.09
LOG060	LOG-B	Green	0.20	3.22	1.54	0.46	0.88	5.00	28.41	58.49	0.19	ND	1.55	ND	ND	ND	ND	2.1	0.74
LOG061	LOG-B	Honey	1.08	4.07	3.37	0.43	0.76	5.41	28.29	56.34	0.17	0.08	ND	ND	ND	ND	ND	2.0	1.50
LOG062	LOG-B	White	2.17	1.98	0.58	0.60	0.02	2.99	43.23	41.02	ND	7.41	ND	ND	ND	ND	ND	0.9	2.77
LOG085	LOG-B	Honey	0.55	3.32	5.39	0.64	0.65	3.31	30.06	55.61	0.47	ND	ND	ND	ND	ND	ND	1.9	1.19
LOG003	LOG-C	Honey	0.74	3.23	1.87	0.50	1.11	6.93	22.29	62.53	0.19	ND	0.59	ND	ND	ND	ND	2.8	1.24
LOG051	LOG-C	Brown	1.90	5.63	2.91	0.31	1.19	9.53	44.43	33.56	0.54	ND	ND	ND	ND	ND	ND	0.8	2.21
LOG075	LOG-C	Brown	1.16	2.42	1.49	0.88	0.71	4.71	39.69	48.27	0.19	ND	0.43	0.02	ND	ND	ND	1.2	2.04
LOG038b	LOG-E	Blue	7.44	3.92	1.34	0.47	0.70	3.40	52.88	26.15	0.12	3.26	ND	ND	0.15	ND	0.16	0.5	7.91
LOG038w	LOG-E	White	7.05	3.39	0.46	0.40	0.53	2.67	53.93	28.06	0.08	3.43	ND	ND	ND	ND	ND	0.5	7.45
LOG042b	TER	Black	3.35	6.05	2.53	0.11	0.20	7.85	46.38	16.98	0.78	6.83	0.72	3.94	ND	ND	4.24	0.4	3.46
LOG042g	TER	Green	1.61	1.26	0.42	1.65	0.20	4.77	47.57	37.11	ND	3.40	1.94	ND	ND	ND	ND	0.8	3.26
LOG042w	TER	White	2.28	2.12	0.79	1.92	0.47	7.22	56.49	23.11	0.03	4.87	ND	ND	ND	ND	0.70	0.4	4.20
LOG044w	TER	White	1.60	1.45	0.60	1.55	0.19	5.17	43.87	40.55	ND	5.02	ND	ND	ND	ND	ND	0.9	3.15
LOG037	UNGR	Honey	1.53	4.12	2.60	0.91	1.46	5.93	33.62	49.62	0.16	ND	ND	ND	ND	0.06	ND	1.5	2.44

4 Conclusions

This research constitutes the first archaeometric study of the pottery manufactured during the late medieval period in Logroño. The multi-analytical approach by means of ICP-MS, NAA, XRD and SEM-EDS applied on 77 individuals (MNI 637), yielded the identification of six compositional groups and permitted the assessment of their diachronic evolution (see Figure 14). Three of them could be linked to a local production (LOG-A, LOG-B and LOG-C), providing the first chemical fingerprint of the pottery from the late-medieval workshops of the current Riojan capital-city. In addition, after the collation with an extensive NAA majolica database, two groups of exogenous provenances were identified. Thus, pottery trading from Teruel (TER group) and Valencia (LOG-E group) was revealed, which supports the historical records about majolica trading from these more technologically advanced workshops. In the Valencian group, although NAA results pointed to a provenance from Manises, the discrimination between Manises and Paterna was not possible due to lack of chemical resolution. Additionally, another group was defined (LOG-D), characterized by a very distinct chemical composition (high calcareous). This group could neither be unequivocally linked to a local origin nor to a known exogenous production center. Nevertheless, it is clear that the pieces (roof tiles with drops of glazes from the firings) were used as kiln utensils by the potters of Logroño. Thus, their provenance, if is not local, is unlikely to be very far therefrom.

Regarding the local production of Logroño, three compositional groups were identified. The first, a low calcareous one, employed for cookware (1290-1350) and after a short interruption, for storage purposes, at the beginning of the 15thcentury (LOG-A). The characterization of this group permits the determination of a local production of cooking pots during the initial stages of

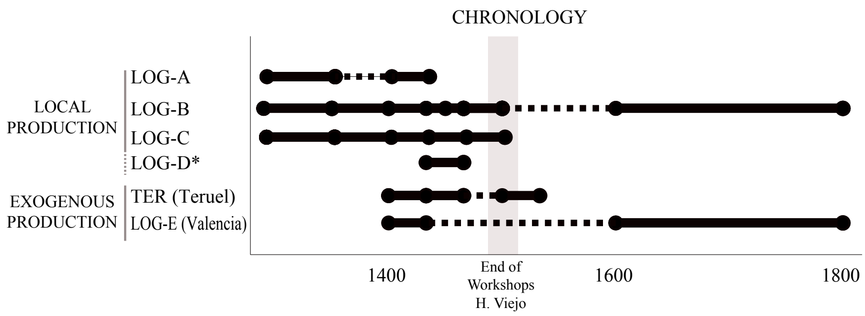

Fig. 14: Evolution of local and exogenous production identified in the site of Hospital Viejo from Logroño. *The provenance of LOG-D, could not be determined although is potentially local.

the workshop. This production volume may have declined by the 14^{th} century due to the influence of imports of the prestigious Zamoran pots during that century as previously documented (Solaun, 2005, p. 388). Nonetheless, the use of the same paste is observed later for the production of pantry ware such as lids. A total absence of material from this group is observed in the last stages of the workshop, coinciding with the construction of the Revellín Wall (directly linked with the abandonment of the Workshops of Hospital Viejo).

In addition, two calcareous groups were discriminated: LOG-B and LOG-C. Considering the diachronic evolution of LOG-B, the use of similar recipes and/or extraction sites could be inferred, producing ceramics from the 13^{th}-15^{th}centuries, but also from the 17^{th}-18^{th}centuries. Moreover, regarding the historical context (the relocation of the workshops after 15^{th} century), it can be suggested that the latter ceramics from this groups were most probably manufactured in the post-medieval ceramic workshops of one of the *Ollerias* streets (which is currently under archaeological investigation). While most of the ceramics were unglazed or lead glazed, the introduction of poorly applied tin lead glaze could be observed only on the post-medieval period for this group. In contrast, the LOG-C group showed the use of a paste which seems to have been present only during the operational period of the unearthed kilns (13^{th}-15^{th}centuries). This group was the most extensive and compositionally compact group. Thus, the it can be considered the group that best characterizes the production of the workshops from the Hospital Viejo site. All of the local groups showed pastes that were fired between 850 °C and 950 °C as the determined by XRD. These firing temperatures contrasted with those from the majolica pottery imports identified in this study, which were higher.

In general terms, it can be suggested that the late medieval workshops of Logroño manufactured mainly unglazed or lead glazed pottery during 13^{th}-15^{th}centuries. Those lead glazed pieces (with high Pb concentrations) contained a translucent glaze which, gave them a final honey-colored appearance. In contrast, most of the tin lead glazed pottery showing bichrome or polychrome decorations was traded from other production centers, such as Valencia or Teruel as determined by the present study.

The chromatic decorations include blue, black and green colors. All the opaque glazes were obtained through SnO_2 from concentrations ranging 3.26 to 7.41 mass % SnO_2. To obtain the final colors, traditional recipes based on different metallic oxides were used: the blue was acquired by CoO, while CuO was used to obtain the green color and MnO for the black.

The complexity of Riojan ceramic production in pre-industrial eras was highlighted in terms of archaeometric characterization, showing different chemical reference groups diachronically within the same production center (LOG-A, LOG-B and LOG-C). The under-representation of the locally produced tin lead glazed ceramics, coincides with the slow adoption of tin lead glazing technology which, was more widely extended in other parts of the Iberian Peninsula, such as the Valencian area.

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Declarations

Availability of Data

The data used for this work is included in the Supplementary Material.

Code Availability

The model and work flow based on R used for the statistical treatment is available and reproducible in the following repository:`https://github.com/esteful/arch_flow`

Conflict of interest

The authors declare that they have no conflict of interest.

Author's Contribution

E. Calparsoro and J.G. Iñañez designed the project, carried out the analyses and wrote the manuscript. G. Arana carried out the ICP-MS analyses and M.D. Glascock contributed with NAA analyses. All authors reviewed the manuscript before submission.

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