CHEMICAL CHARACTERIZATION OF TIN-LEAD GLAZED POTTERY FROM THE IBERIAN PENINSULA AND THE CANARY ISLANDS: INITIAL STEPS TOWARD A BETTER UNDERSTANDING OF SPANISH COLONIAL POTTERY IN THE AMERICAS*

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Majolica pottery was the most characteristic tableware produced in Europe during the Medieval and Renaissance periods. Because of the prestige and importance attributed to this ware, Spanish majolica was imported in vast quantities into the Americas during the Spanish Colonial period. A study of Spanish majolica was conducted on a set of 186 samples from the 10 primary majolica production centres on the Iberian Peninsula and 22 sherds from two early colonial archaeological sites on the Canary Islands. The samples were analysed by neutron activation analysis (NAA), and the resulting data were interpreted using an array of multivariate statistical approaches. Our results show a clear discrimination between different production centres, allowing a reliable provenance attribution of the sherds from the Canary Islands.

KEYWORDS: MAJOLICA, NEUTRON ACTIVATION ANALYSIS (NAA), PROVENANCE, MULTIVARIATE STATISTICS, IBERIAN PENINSULA, CANARY ISLANDS

INTRODUCTION

Tin-lead glazed pottery, also know as majolica, is an earthenware pottery characterized by a creamy light-buff coloured ceramic body and an opaque white tin-lead glaze that covers the entire outer surface of the vessel. The most characteristic feature of majolica pottery lies in the metallic-oxide decorations that are applied on top of the opaque white glaze coat. The opaque white glaze is composed of sand (e.g., quartz) and lead, that serves as a flux to decrease the temperature needed for melting SiO_2. The glaze is opacified with particles of tin oxide (SnO_2) and also by the action of extant quartz and feldspar inclusions. These inclusions, and the bubbles that result from the firing process, absorb, scatter, and/or reflect incident light, thereby giving the transparent glaze a white appearance. Due to this opacity, decoration is normally applied to the outer surfaces of the glaze coat (Kingery and Aronson 1990; Tite *et al*. 1998; Molera *et al*. 1999; Iñañez 2007).

During the 15th, but especially the 16th, century, Spanish majolica production flourished as Italian-influenced decorative styles diffused into the Iberian Peninsula. Consequently, black and especially green motifs—colours associated with Islamic ceramic traditions—were progressively replaced by blue patterns, sometimes mixed with other colours, especially yellow.
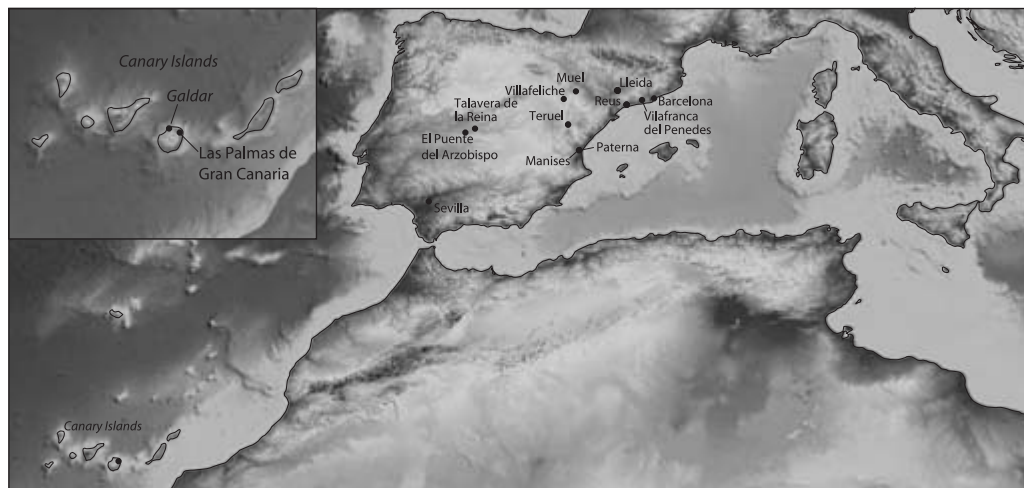
Figure 1 *Map of the main sites cited in the text.*

By the end of the 16th and 17th centuries, majolica polychrome was produced in large quantities throughout the Iberian Peninsula. At the same time the production of lustreware declined, both in quantity and quality. Some of the primary production centres during this period include Barcelona, Reus, Vilafranca del Penedès and Lleida in the Catalan area; Manises in the Valencian area; Muel and Villafeliche in Aragon; and Talavera de la Reina, Puente del Arzobispo and Seville in Central and South Spain, respectively (Fig. 1).

As commercial trade between the Americas and Europe increased during the 16th and 17th centuries, the port of Seville became the primary port of trade for Spain. The increased importance of Seville occurred because this city's inland fluvial port protected it from pirate attacks and served as both the departure point and the final destination for most of the Spanish galleons that traded with the Americas in the so-called 'Carrera de Indias'. In order to supervise the traffic of goods, a bureau of trade was established in 1503, called Casa de la Contratación, only 11 years after the discovery of the Americas. The Casa de la Contratación had, for more than 200 years (1503–1717), its primary office in the city of Seville.

The Canary Islands were a key stopping point for Spanish vessels travelling to the Americas, because of the need to take advantage of the ocean currents and Alisios (Trade) winds that blow directly to the Caribbean from the eastern Atlantic. Moreover, the Canary Islands signified, for all the Spanish ships, the last geographical region controlled by the Spaniards before starting their long open-sea voyage. The Canary Islands were also a required stop for legal reasons, given that the Castilian Kingdom established a rigid control over the trade of items to the New World. Because of this organization of trade, Seville became the primary production centre for majolica exported outside of Spain, whereas Talavera became the most important production centre for majolica consumed within the Castillian Kingdom and emerged as the official supplier of royal tableware.

From an archaeometric point of view, and despite the fact that many significant works concerning majolica pottery technology have been published, most of which are focused on lustreware (for instance, see Padeletti and Fermo 2003; Pradell *et al*. 2005; Polvorinos *et al*. 2006; Roqué *et al*. 2007), archaeometric knowledge about tin-lead glazed pottery produced in the Iberian Peninsula is uneven. Consequently, there is an overall lack of chemically defined
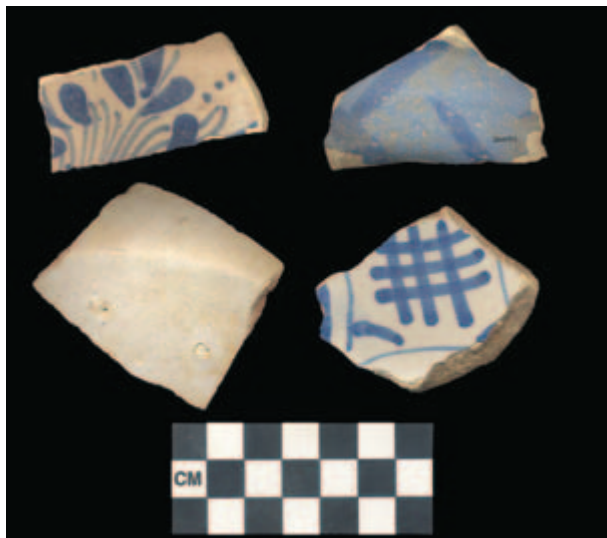
Figure 2   *Examples of majolica pottery from Gran Canaria island. From left to right and top to bottom: MJ0280, MJ0282, MJ0264 and MJ0274.*

reference groups that characterize the primary production centres. Nonetheless, there have been a few studies that have focused on production sites in the Iberian Peninsula, such as Paterna and Manises (Jornet *et al*. 1985; Hughes and Vince 1986; Hughes 1991, 1995; Molera *et al*. 1996, 2001; Iñañez 2007), Barcelona, Lleida, Reus and Vilafranca del Penedès (Buxeda *et al*. 2001; Iñañez 2007; Iñañez and Buxeda 2007; Iñañez *et al*. 2007a,c) and Talavera, Puente and Seville (Criado *et al*. 2002; Iñañez *et al*. in press). In addition, a few studies have examined the occurrence of Spanish majolica at overseas sites (e.g., the Americas), most of which refer to production in Seville (Olin *et al*. 1978; Maggeti *et al*. 1984; Olin and Blackman 1989; Myers *et al*. 1992; Olin and Myers 1992; Vaz *et al*. 1997; Jamieson and Hancock 2004). However, when many of these provenance studies were conducted on majolica found in the Americas, there was a significant lack of knowledge regarding actual kiln sites at Seville. During the past several years archaeological excavations have revealed a substantial number of such kiln sites, which can be used to establish the reference groups (Lorenzo Morilla *et al*. 1990; Mercado Hervás *et al*. 2001; Mesa Romero and Castañeda de la Paz 2001; Iñañez *et al*. 2007b).

   In this paper, we summarize the results from compositional analysis of 186 majolica sherds obtained from the 10 primary production centres located on the Iberian Peninsula and their comparison with a sample of 22 majolica sherds obtained from two early colonial sites on the Canary Islands: la Cueva Pintada (Gáldar) and the ancient convent of San Francisco (Las Palmas), both on the island of Gran Canaria (Fig. 2 and Table 1). The production sites, which date from the 14th to the 18th centuries, have been involved in the manufacture of tin-lead glazed pottery since the Middle Ages, and some continue to produce majolica even today. Consequently, majolica production at a few of these places achieved high prestige due to their high quality and aesthetic value. The exportation of this pottery to the Americas, which became the case for the Seville and Talavera production centres, eventually resulted in the establishment of autochthonous workshops, such as Puebla or Mexico City, both in Mexico (Castro 1988; Gámez Martínez 2003; LaBrecque *et al*. 2003; Rodríguez-Alegría *et al*. 2003).

Table 1   *Summary of the studied shards according to their decoration type and chronology and with regard to their production and reception archaeological sites*

Sites	Centuries	Green and black	Green	Blue	Blue and green	Lusterware	White plain	Polychrome	Sgraffito	Non-glazed	Total
Barcelona		–	–	4	–	16	–	–	–	–	20
Drassanes	16th to 17th	–	–	4	–	9	–	–	–	–	13
H. Santa Creu	16th to 17th	–	–	–	–	7	–	–	–	–	7
Lleida		–	–	11	–	–	–	1	–	3	15
Obradors	16th to 17th	–	–	2	–	–	–	1	–	2	5
St. Anastasi	16th to 17th	–	–	5	–	–	–	–	–	–	5
Remolins	16th to 17th	–	–	4	–	–	–	–	–	1	5
Manises	15th	–	–	–	–	15	–	–	–	–	15
Muel	16th to 17th	–	–	16	10	–	–	–	–	–	26
Puente	16th to 18th	–	–	13	–	–	–	1	–	1	15
Reus	16th to 17th	–	–	–	–	3	12	–	–	–	15
Sevilla		–	–	21	–	1	9	5	–	–	36
Pureza	16th to 17th	–	–	3	–	1	5	3	–	–	12
Valladares	16th to 17th	–	–	8	–	–	2	–	–	–	10
Plaza Armas	16th to 17th	–	–	8	–	–	2	–	–	–	10
Museu Ceràmica	15th to 16th	–	–	2	–	–	–	2	–	–	4
Talavera		–	–	13	–	–	–	1	–	–	14
Mirasol	16th to 17th	–	–	12	–	–	–	–	–	–	12
Museu Ceràmica	16th to 17th	–	–	1	–	–	–	1	–	–	2
Vilafranca	16th to 17th	–	–	1	–	1	13	–	–	–	15
Villafeliche	17th to 18th	–	–	15	–	–		–	–	–	15
Canary Islands		1	1	9	–	3	6	1	1	–	22
Cueva Pintada	15th to 16th	–	1	–	–	1	2	–	–	–	4
San Francisco	16th to 17th	1	–	9	–	2	4	1	1	–	18
Total		1	1	103	10	39	40	9	1	4	208

Our goal is to obtain a more precise understanding of majolica pottery exports to the Americas through the study of cultural material found at the Canary Islands reception centres. To accomplish this goal, we require a reliable compositional database, based on pottery from the primary production sites of the Iberian Peninsula dating from the 14th to the 18th centuries or, in other words, from the appearance of majolica until the introduction of porcelain production in Spain during the second half of the 18th century. By identifying compositional reference groups for majolica production within the Iberian Peninsula, we can then begin to identify the provenance of majolica pottery that was exported from Spain to the Canary Islands and the Americas. The identification of these production centres has important implications for understanding changing sociopolitical and economic relationships between Spain and the New World—relationships that may be at odds with historical documents of the era.

METHODS

Table 1 lists the 208 majolica sherds that were analysed in this study (for a more detailed description, pictures and drawings, see Iñañez 2007—available online at http://www.tesisenxarxa. net/TDX-0205107-115739/). All of the specimens were obtained from either extant museum collections or from contemporary archaeological excavations. Our sampling strategy was strictly focused on kiln-related materials, to maximize the probability that the materials included in this study were a product of their respective workshops and production centres. Consequently, we focused on ceramics from archaeologically and historically documented majolica kiln dumps; although in some cases we also sampled sherds from other types of archaeological deposits, such as the roof vaults at the old Hospital de la Santa Creu in Barcelona. Most of the reference samples included in this study (97) were obtained from the Museu de la Ceràmica de Barcelona, a repository that has large reference collections for most of the primary majolica production sites in Spain, and Mr J. A. Cerdà from the Associació Catalana de Ceràmica. We also obtained 15 sherds from the Museu Comarcal Salvador Vilaseca de Reus and 15 samples from the Museu de Vilafranca del Penedès. An additional 32 majolica fragments were selected from three different archaeological excavations within the city of Seville (Pureza, that is linked to the famous artist Niculoso Pisano's workshop, Valladares and Plaza de Armas). The samples were generously provided by the Museo Arqueológico de Sevilla, where the material from these excavations is curated. Finally, we obtained 15 specimens from the Servei Arqueològic de la ciutat de Lleida from three different archaeological sites (Obradors, St. Anastasi and Remolins) and 12 samples from Talavera de la Reina that were kindly provided by Mr A. Sánchez Cabezudo. Colonial sherds from the Canary Islands were generously provided by the Parque Arqueológico Cueva Pintada de Gáldar (Gran Canaria) and the Museo Canario for the archaeological site of Ancient Convent of San Francisco (Las Palmas de Gran Canaria) (Table 2).

Located in the modern-day city of Gáldar, La Cueva Pintada represents one of the most significant prehispanic settlements on Gran Canaria Island. Known as Agaldar between the sixth and 16th centuries, the site, which was the capital of one of the indigenous Guanche chiefdoms, was abandoned shortly after the Spanish conquest. Agaldar was resurrected after the Spanish conquest as the Spanish settlement of Gáldar. La Cueva Pintada (the painted cave) is a large prehispanic artificial cave that was discovered in 1862. The cave is significant, not only because of its size, but also because it houses examples of indigenous Canarian artwork that depicts the household environment. The area where this site is located was used for growing crops from the 18th century until the 1970s, when archaeological and conservation work was

Table 2   *Classification and description of samples from the Canary Islands*

ANID	Description	Form	Date(s)
MJ0236	Lusterware	Plate	15th–16th
MJ0237	Sevillan white	Plate	15th–16th
MJ0241	Sevillan white	Plate	15th–16th
MJ0253	Green	Tile	15th–16th
MJ0258	Columbia simple	Bowl	16th–17th
MJ0262	Sevillan white	Bowl	16th–17th
MJ0268	Isabela polychrome	Porringer	16th–17th
MJ0269	Delft series	Plate	16th–17th
MJ0270	Ligurian blue on white	Plate	16th–17th
MJ0272	Ligurian blue on white	Plate	16th–17th
MJ0275	Portuguese blue on white	Plate	16th–17th
MJ0284	*Sgraffito*	Plate	16th–17th
MJ0285	Lusterware	Plate	16th–17th
MJ0286	Lusterware	Plate	16th–17th
MJ0287	Columbia simple	Plate	16th–17th
MJ0288	Sevillan white	Plate	16th–17th
MJ0289	Delft series	Plate	16th–17th
MJ0290	Ligurian blue on white	Plate	16th–17th
MJ0291	Ligurian blue on white	Plate	16th–17th
MJ0292	Portuguese blue on white	Plate	16th–17th
MJ0293	Catalan blue on white	Plate	16th–17th
MJ0294	Green and manganese	Plate	16th

initiated to protect the cave and ultimately to open it to the public (Onrubia *et al*. 2004, and references therein).

In contrast to La Cueva Pintada, the convent of San Francisco at Las Palmas de Gran Canaria (Gran Canaria Island) was built at the end of the 15th century. It is one of the most important historical sites, because it represents the earliest foundation of the Franciscan order on the island during the conquest/acculturation process of the Canary Islands. The convent remained active until 1835, when the Franciscans were forced to leave as a result of a general confiscation ordered by the Spanish government, known as Mendizabal's confiscation. After being used by the army for more than 100 years, the buildings were finally abandoned in the mid-20th century. In 1992 the Museo Canario de Las Palmas de Gran Canaria initiated archaeological excavations at the site, where a large quantity of majolica pottery, not only Spanish, but also Italian and Portuguese, was recovered. The occurrence of non-Spanish majolica at San Francisco de Las Palmas suggests a more complex scenario regarding the role of the Canary Islands in the trade network between Europe and the Americas (Sosa Suárez 2002).

In the present study, 10 g of each collected sample was powdered using a Spex Mixer (mod. 8000) tungsten carbide cell for 12 min. Prior to grinding, glazes and exterior surfaces were removed mechanically by means of a tungsten carbide abrading tool, leaving only the inner part of the ceramic for analysis. This step served to minimize contamination of the ceramic matrix by glaze and soil. Powdered specimens were stored in polyethylene vials for transport to the laboratory.

Chemical analyses were conducted by neutron activation analysis (NAA) at the University of Missouri Research Reactor's Archaeometry Laboratory (MURR). Prior to weighing, the

powdered pottery samples were oven-dried at 100°C for at least 24 h. Approximately 150 mg of each sample was weighed in small polyvials used for short irradiations. At the same time, 200 mg of each sample was weighed into high-purity quartz vials used for long irradiations. Along with the majolica samples, reference standards of SRM-1633a (coal fly) and SRM-688 (basalt rock) were prepared, as well as quality control samples of SRM-278 (obsidian rock) and Ohio Red Clay (for analytical conditions, see Glascock *et al.* 2007).

At MURR, NAA of pottery consists of two irradiations and a total of three gamma counts. Short irradiations involve a pair of samples being transported through a pneumatic tube system into the reactor core for a 5 s neutron irradiation using a thermal flux of 8×10^{13} n cm^{-2} s^{-1}. After 25 min of decay, the samples are counted for 720 s using a high-resolution germanium detector. This count yields data for nine short-life elements: Al, Ba, Ca, Dy, K, Mn, Na, Ti and V. For the long irradiation, bundles of 50 or 100 of the encapsulated quartz vials are irradiated for 24 h at a flux of 5×10^{13} n cm^{-2} s^{-1}. Following the long irradiation, samples decay for 7 days, and then are counted for 1800 s (known as 'middle count') on a high-resolution germanium detector coupled to an automatic sample changer. This middle count yields determination of seven medium half-life elements: As, La, Lu, Nd, Sm, U and Yb. After an additional two-week decay, a second count for 9000 s is carried out on each sample. This final measurement allows quantification of 17 long-life elements: Ce, Co, Cr, Cs, Eu, Fe, Hf, Ni, Rb, Sb, Sc, Sr, Ta, Tb, Th, Zn and Zr (Glascock *et al.* 2007).

Statistical analysis of the data followed Aitchison's approach and Buxeda's observations on compositional data (Aitchison 1986, 1996; Buxeda 1999; Aitchison *et al.* 2000, 2002; Buxeda and Kilikoglou 2003). The statistical procedure consists of the use of base-10 logarithms of ratios obtained by dividing all the components, in this case the elements, by the element that introduces the lowest chemical variability to the entire set of specimens. The use of logarithms compensates for differences in magnitudes between major elements, such as Al and Fe, and trace elements, such as the lanthanide and rare earth elements (e.g., La, Ce, Sm etc.). Additionally, log-transformed data can sometimes highlight possible perturbations in the chemical data as a result of diagenesis, contamination or other alteration processes (Buxeda 1999).

Data were examined using an array of multivariate statistical procedures. The application of multivariate statistical techniques to NAA data facilitates identification of compositional groups. The similarity between specimens, and subsequently to their hypothetical provenance according to the provenance postulate (Weigand *et al.* 1977), was examined using scatter plots, whereas stepwise discriminant analysis (DA) was performed to assess the archaeological classifications and the chemical groups proposed by scatter plots. In addition, Mahalanobis distance was used to describe the statistical probability, when group sizes permitted, of the separation between defined groups and those individuals that remained unclassified. Mahalanobis distance takes into account variances and covariances in the multivariate group and is analogous to expressing the distance from a univariate mean in standard deviation units (Baxter 1999; De Maesschalck *et al.* 2000). In that sense, Mahalanobis distance can also be converted into probabilities of group membership for each individual (Glascock 1992; Neff *et al.* 2003).

Although sample preparation was conducted taking great care to minimize the analytical error, the potential for contamination exists nonetheless, and a conservative approach to data interpretation is warranted. For example, cobalt had to be removed from consideration during the statistical treatment because the tungsten carbide cell used to grind the samples exhibits traces of Co in its chemical composition (cobalt is a known binder in tungsten alloys). Additionally, Ni concentrations were below detection limits for many of the samples and subsequently were removed from consideration.
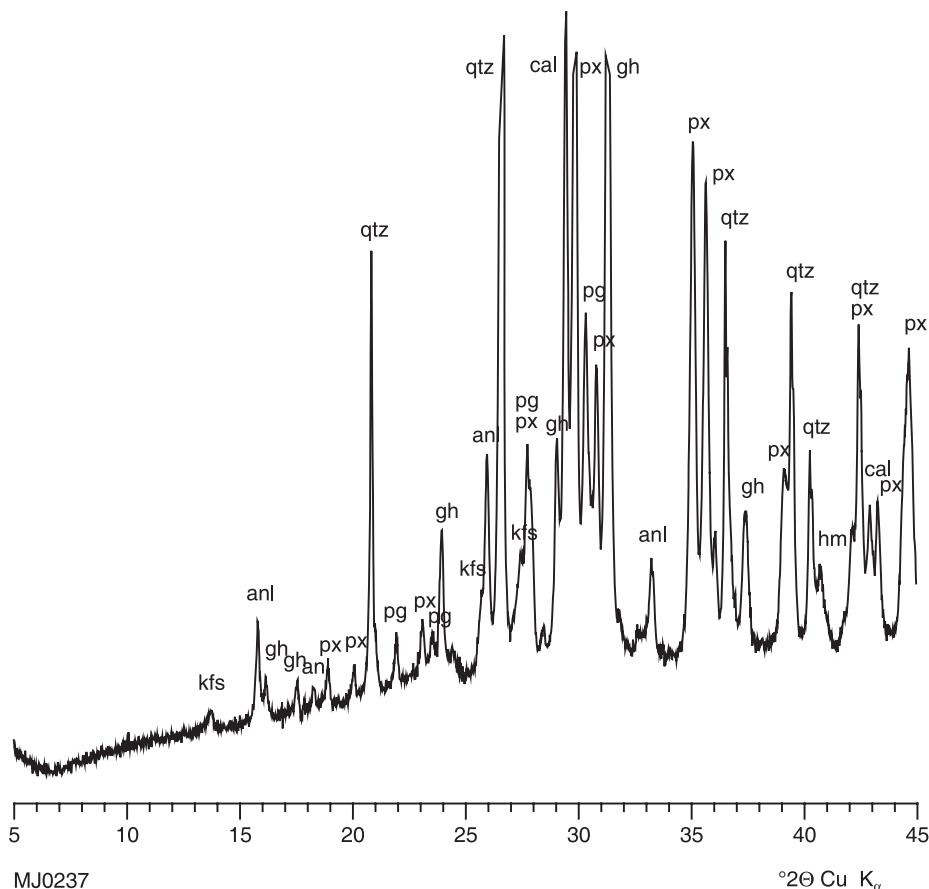
Figure 3   *XRD diffractogram of the sample MJ0237, representative of the affected pottery. anl, Analcime; cal, calcite; gh, gehlenite; hm, hematite; ill, illite–muscovite; kfs, alkaline feldspar; pg, plagioclase; px, pyroxene; qtz, quartz.*

Conversely, given that most of the specimens had also been analysed by X-ray diffraction, it has been possible to observe that a relevant number of the analysed majolica sherds exhibited a double process of alteration and contamination (Fig. 3), also documented in previous studies (Iñañez 2007; Iñañez and Buxeda 2007). This process occurs with the leaching of potassium and, sometimes, rubidium, from the matrix, with a subsequent enrichment of sodium because of analcime crystallization (Buxeda *et al.* 2002; Schwedt *et al.* 2006; Iñañez 2007, and references therein). Because these alteration and contamination processes affect those components in the matrix composition, without any possibility of calculating a satisfactory correction, Na, K, and Rb were removed from consideration during the statistical analysis.

RESULTS AND DISCUSSION

The variability of each chemical component was first taken into account in this study and assessed by calculating the variation matrix using the S-plus program (MathSoft 1999), which provides information about those components that introduce higher variability to the data set (Table 3).

Table 3 *Compositional variation matrix from the majolica production centres from the Iberian Peninsula and the reception centres from the Canary Islands. In each column i (I = 1, . . . ,S) are the variances after a logratio transformation using the component x_i as divisor. vt = Total variation; τ_i = total sum of variances in column i; vt/τ_i = percentage of variance in the logratio covariance matrix using the component x_i as divisor due to the total variation; $r_{v,\tau}$ = correlation between the values τ_{ij} (i ≠ j) and the corresponding values τ_i (j = 1, . . . ,i − 1,i + 1, . . . ,S).*

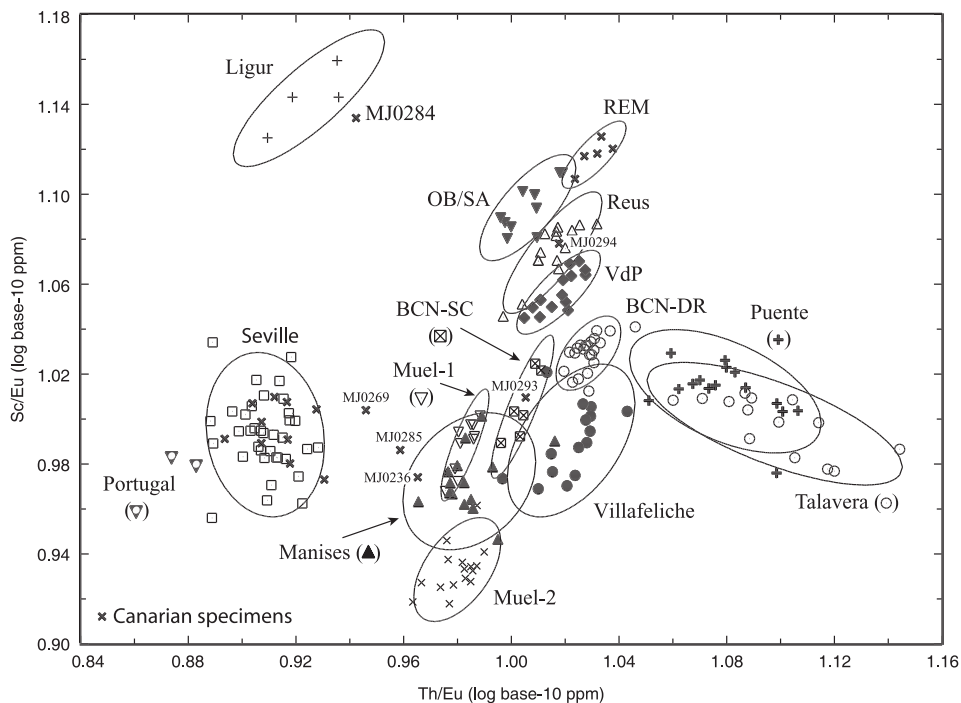| Var.Matrix	As	La	Lu	Nd	Sm	U	Yb	Ce	Cr	Cs	Eu	Fe	Hf	Rb	Sb	Sc	Sr	Ta	Tb	Th	Zn	Zr	Al	Ba	Ca	Dy	K	Mn	Na	Ti	V
As	0.00	0.25	0.26	0.25	0.25	0.35	0.26	0.25	0.31	0.42	0.25	0.24	0.30	0.40	0.36	0.25	0.53	0.25	0.25	0.26	0.30	0.30	0.24	0.37	0.36	0.25	0.37	0.30	0.55	0.25	0.26
La	0.25	0.00	0.01	0.00	0.00	0.07	0.01	0.00	0.09	0.18	0.00	0.02	0.04	0.11	0.26	0.01	0.29	0.02	0.01	0.01	0.07	0.04	0.01	0.09	0.16	0.01	0.07	0.10	0.37	0.01	0.05
Lu	0.26	0.01	0.00	0.01	0.01	0.05	0.01	0.01	0.09	0.21	0.01	0.03	0.04	0.13	0.28	0.02	0.28	0.02	0.01	0.01	0.07	0.03	0.02	0.11	0.14	0.01	0.09	0.10	0.34	0.02	0.06
Nd	0.25	0.00	0.01	0.00	0.00	0.07	0.01	0.00	0.09	0.19	0.00	0.02	0.04	0.11	0.28	0.02	0.30	0.02	0.01	0.01	0.07	0.04	0.01	0.10	0.16	0.01	0.08	0.10	0.37	0.02	0.06
Sm	0.25	0.00	0.01	0.00	0.00	0.06	0.00	0.00	0.09	0.19	0.00	0.02	0.03	0.12	0.28	0.02	0.29	0.01	0.01	0.01	0.07	0.03	0.01	0.10	0.15	0.00	0.08	0.10	0.36	0.01	0.06
U	0.35	0.07	0.05	0.07	0.06	0.00	0.07	0.06	0.18	0.24	0.08	0.11	0.12	0.13	0.34	0.09	0.29	0.07	0.07	0.05	0.15	0.10	0.07	0.13	0.20	0.08	0.11	0.22	0.48	0.08	0.14
Yb	0.26	0.01	0.01	0.01	0.00	0.07	0.00	0.00	0.09	0.20	0.01	0.02	0.03	0.12	0.28	0.02	0.31	0.02	0.01	0.01	0.07	0.03	0.02	0.10	0.16	0.01	0.09	0.10	0.35	0.02	0.06
Ce	0.25	0.00	0.01	0.00	0.00	0.06	0.00	0.00	0.09	0.19	0.00	0.02	0.04	0.11	0.27	0.01	0.29	0.02	0.01	0.00	0.07	0.04	0.01	0.09	0.15	0.01	0.07	0.10	0.37	0.01	0.05
Cr	0.31	0.09	0.09	0.09	0.09	0.18	0.09	0.09	0.00	0.33	0.07	0.04	0.13	0.28	0.30	0.04	0.22	0.11	0.09	0.11	0.08	0.13	0.07	0.20	0.17	0.09	0.21	0.12	0.27	0.07	0.05
Cs	0.42	0.18	0.21	0.19	0.19	0.24	0.20	0.19	0.33	0.00	0.22	0.22	0.29	0.07	0.51	0.20	0.70	0.16	0.20	0.16	0.25	0.30	0.16	0.23	0.46	0.20	0.16	0.40	0.70	0.22	0.24
Eu	0.25	0.00	0.01	0.01	0.00	0.08	0.01	0.00	0.07	0.22	0.00	0.01	0.03	0.14	0.26	0.01	0.26	0.02	0.01	0.02	0.06	0.03	0.01	0.10	0.14	0.01	0.09	0.08	0.34	0.01	0.04
Fe	0.24	0.02	0.03	0.02	0.02	0.11	0.02	0.02	0.04	0.22	0.01	0.00	0.07	0.16	0.25	0.00	0.26	0.04	0.02	0.03	0.04	0.07	0.01	0.11	0.16	0.02	0.11	0.07	0.30	0.02	0.02
Hf	0.30	0.04	0.04	0.04	0.03	0.12	0.03	0.04	0.13	0.29	0.03	0.07	0.00	0.20	0.32	0.07	0.32	0.05	0.04	0.05	0.14	0.02	0.07	0.16	0.18	0.03	0.14	0.13	0.39	0.04	0.12
Rb	0.40	0.11	0.13	0.11	0.12	0.13	0.12	0.11	0.28	0.07	0.14	0.16	0.20	0.00	0.42	0.14	0.61	0.10	0.12	0.09	0.19	0.21	0.11	0.16	0.40	0.13	0.06	0.32	0.68	0.15	0.20
Sb	0.36	0.26	0.28	0.28	0.28	0.34	0.28	0.27	0.30	0.51	0.26	0.25	0.32	0.42	0.00	0.25	0.49	0.34	0.28	0.28	0.27	0.34	0.28	0.29	0.41	0.28	0.39	0.25	0.53	0.27	0.25
Sc	0.25	0.01	0.02	0.02	0.02	0.09	0.02	0.01	0.04	0.20	0.01	0.00	0.07	0.14	0.25	0.00	0.26	0.03	0.02	0.02	0.04	0.07	0.01	0.10	0.15	0.02	0.10	0.08	0.31	0.02	0.02
Sr	0.53	0.29	0.28	0.30	0.29	0.29	0.31	0.29	0.22	0.70	0.26	0.26	0.32	0.61	0.49	0.26	0.00	0.33	0.30	0.32	0.34	0.29	0.28	0.39	0.17	0.29	0.46	0.31	0.42	0.26	0.25
Ta	0.25	0.02	0.02	0.02	0.01	0.07	0.02	0.02	0.11	0.16	0.02	0.04	0.05	0.10	0.34	0.03	0.33	0.00	0.02	0.01	0.10	0.05	0.02	0.13	0.18	0.02	0.08	0.16	0.41	0.03	0.08
Tb	0.25	0.01	0.01	0.01	0.01	0.07	0.01	0.01	0.09	0.20	0.01	0.02	0.04	0.12	0.28	0.02	0.30	0.02	0.00	0.01	0.07	0.04	0.02	0.11	0.16	0.01	0.09	0.10	0.35	0.02	0.06
Th	0.26	0.01	0.01	0.01	0.01	0.05	0.01	0.01	0.11	0.16	0.02	0.03	0.05	0.09	0.28	0.02	0.32	0.01	0.01	0.00	0.07	0.05	0.01	0.09	0.18	0.01	0.06	0.12	0.40	0.03	0.06
Zn	0.30	0.07	0.07	0.07	0.07	0.15	0.07	0.07	0.08	0.25	0.06	0.04	0.14	0.19	0.27	0.04	0.34	0.10	0.07	0.07	0.00	0.14	0.06	0.15	0.18	0.07	0.17	0.09	0.25	0.07	0.05
Zr	0.30	0.04	0.03	0.04	0.03	0.10	0.03	0.04	0.13	0.30	0.03	0.07	0.02	0.21	0.34	0.07	0.29	0.05	0.04	0.05	0.14	0.00	0.07	0.16	0.17	0.03	0.14	0.13	0.38	0.04	0.12
Al	0.24	0.01	0.02	0.01	0.01	0.07	0.02	0.01	0.07	0.16	0.01	0.01	0.07	0.11	0.28	0.01	0.28	0.02	0.02	0.01	0.06	0.07	0.00	0.10	0.17	0.02	0.07	0.12	0.38	0.02	0.04
Ba	0.37	0.09	0.11	0.10	0.10	0.13	0.10	0.09	0.20	0.23	0.10	0.11	0.16	0.16	0.29	0.10	0.39	0.13	0.11	0.09	0.15	0.16	0.10	0.00	0.26	0.11	0.12	0.18	0.50	0.11	0.13
Ca	0.36	0.16	0.14	0.16	0.15	0.20	0.16	0.15	0.17	0.46	0.14	0.16	0.18	0.40	0.41	0.15	0.17	0.18	0.16	0.18	0.18	0.17	0.17	0.26	0.00	0.15	0.31	0.17	0.26	0.14	0.16
Dy	0.25	0.01	0.01	0.01	0.00	0.08	0.01	0.01	0.09	0.20	0.01	0.02	0.03	0.13	0.28	0.02	0.29	0.02	0.01	0.01	0.07	0.03	0.02	0.11	0.15	0.00	0.09	0.10	0.35	0.02	0.06
K	0.37	0.07	0.09	0.08	0.08	0.11	0.09	0.07	0.21	0.16	0.09	0.11	0.14	0.06	0.39	0.10	0.46	0.08	0.09	0.06	0.17	0.14	0.07	0.12	0.31	0.09	0.00	0.25	0.67	0.10	0.15
Mn	0.30	0.10	0.10	0.10	0.10	0.22	0.10	0.10	0.12	0.40	0.08	0.07	0.13	0.32	0.25	0.08	0.31	0.16	0.10	0.12	0.09	0.13	0.12	0.18	0.17	0.10	0.25	0.00	0.23	0.10	0.09
Na	0.55	0.37	0.34	0.37	0.36	0.48	0.35	0.37	0.27	0.70	0.34	0.30	0.39	0.68	0.53	0.31	0.42	0.41	0.35	0.40	0.25	0.38	0.38	0.50	0.26	0.35	0.67	0.23	0.00	0.33	0.29
Ti	0.25	0.01	0.02	0.02	0.01	0.08	0.02	0.01	0.07	0.22	0.01	0.02	0.04	0.15	0.27	0.02	0.26	0.03	0.02	0.03	0.07	0.04	0.02	0.11	0.14	0.02	0.10	0.10	0.33	0.00	0.05
V	0.26	0.05	0.06	0.06	0.06	0.14	0.06	0.05	0.05	0.24	0.04	0.02	0.12	0.20	0.25	0.02	0.25	0.08	0.06	0.06	0.05	0.12	0.04	0.13	0.16	0.06	0.15	0.09	0.29	0.05	0.00
τ_i	9.25	2.35	2.51	2.44	2.36	4.26	2.48	2.34	4.20	8.19	2.35	2.52	3.62	6.16	9.60	2.41	10.11	2.90	2.53	2.53	3.73	3.60	2.49	4.95	6.22	2.46	4.98	4.70	11.96	2.56	3.28
vt/τ_i	0.24	0.94	0.88	0.90	0.93	0.52	0.89	0.94	0.52	0.27	0.93	0.87	0.61	0.36	0.23	0.91	0.22	0.76	0.87	0.87	0.59	0.61	0.88	0.44	0.35	0.89	0.44	0.47	0.18	0.86	0.67
$r_{v,\tau}$	0.94	0.99	0.99	0.99	0.99	0.95	0.99	0.99	0.88	0.85	1.00	0.99	0.98	0.83	0.88	0.99	0.66	0.97	0.99	0.97	0.94	0.97	0.98	0.95	0.70	0.99	0.89	0.82	0.55	1.00	0.95
vt	2.19																														

Figure 4 *Bivariate plot using log base 10 Th/Eu and Sc/Eu as variables, showing the 15 reference compositional paste groups and the Canary individuals. Ellipses represent a confidence interval of 90%.*

Consequently, the elements As and Sb were removed due to their high variability ($vt/\tau_i < 0.25$), which is presumably provided by possible contamination processes during burial, such is the case of As. Also, Sb is known as a contaminant of Sn, an important component of majolica glazes, and many majolica exhibit yellow decoration that is essentially made of Sb. Therefore, higher Sb concentrations have likely diffused into the clay matrix. As mentioned above, Rb, K and Na were removed because they are involved in the previously cited alteration, consequently exhibiting high chemical variability too ($vt/\tau_i < 0.45$). In addition, U and Tb were also removed from consideration due to their poor analytical precision. Although Ba also exhibits a high variation, this is mainly reported by few samples (MJ0104, MJ0141, MJ0341 and DIA317) with three to seven times the normal Ba amounts of their respective groups, probably as a result of contamination (Table 4). Therefore, Ba was retained as a variable in the statistical treatment once this latter problem was taken into account. Following the exclusion of these elements, a base-10 log ratio transformation was applied to the following subcomposition: La, Lu, Nd, Sm, Yb, Ce, Cr, Cs, Fe, Hf, Sc, Sr, Ta, Th, Zn, Zr, Al, Ba, Ca, Dy, Mn, Ti and V, using Eu as divisor because it introduced the lowest variability to the data set ($vt/\tau_i = 0.93$).

The results are summarized in Figure 4, Figure 5 and Table 4. An examination of various projections of the data facilitated the identification of 13 discrete chemical reference groups attributed to the primary production centres on the Iberian Peninsula: Barcelona Drassanes (BCN-DR), Barcelona Santa Creu (BCN-SC), Reus, Muel-1 and -2, Villafeliche, Manises, Puente del Arzobispo, Vilafranca del Penedès (VdP), Lleida Obradors/Sant Anastasi (OB/SA),

Table 4 *Mean and standard deviation of the 208 majolica sherds from the main production centres of the Iberian Peninsula and the reception centres of the Canary Islands according to the different chemical groups identified by NAA. All values are expressed as ppm (μg/g) except those expressed as weight % in brackets*

Groups Elements	BCN-DR (n = 14) Mean sd	BCN-SC (n = 7) Mean sd	Ligur (n = 4) Mean sd	Manises (n = 17) Mean sd	Muel-1 (n = 10) Mean sd	Muel-2 (n = 15) Mean sd	OB/SA (n = 10) Mean sd
As (ppm)	23 ± 13	34 ± 15	16 ± 7	19 ± 1	30 ± 9	16 ± 2	26 ± 11
La (ppm)	38 ± 1	36 ± 2	27 ± 1	36 ± 1	40 ± 1	38 ± 1	35 ± 3
Lu (ppm)	0.4 ± 0.0	0.4 ± 0.0	0.3 ± 0.0	0.4 ± 0.0	0.4 ± 0.0	0.4 ± 0.0	0.4 ± 0.0
Nd (ppm)	33 ± 2	31 ± 3	24 ± 1	31 ± 1	35 ± 2	32 ± 2	28 ± 2
Sm (ppm)	7 ± 0	6 ± 0	5 ± 0	6 ± 0	7 ± 0	7 ± 0	6 ± 0
U (ppm)	3 ± 0	3 ± 0	2 ± 0	3 ± 0	3 ± 0	3 ± 0	3 ± 0
Yb (ppm)	3 ± 0	3 ± 0	2 ± 0	3 ± 0	3 ± 0	3 ± 0	2 ± 0
Ce (ppm)	76 ± 3	72 ± 4	55 ± 3	70 ± 2	80 ± 2	78 ± 3	70 ± 6
Co (ppm)	17 ± 6	17 ± 3	31 ± 11	15 ± 2	20 ± 1	20 ± 2	24 ± 4
Cr (ppm)	67 ± 4	62 ± 6	301 ± 28	64 ± 3	75 ± 3	57 ± 4	90 ± 7
Cs (ppm)	18 ± 2	6 ± 2	4 ± 1	9 ± 1	8 ± 0	7 ± 0	8 ± 1
Eu (ppm)	1.2 ± 0.0	1.2 ± 0.1	1.0 ± 0.0	1.2 ± 0.0	1.4 ± 0.0	1.3 ± 0.0	1.2 ± 0.1
Fe (%)	3.41 ± 0.14	3.13 ± 0.22	4.04 ± 0.24	3.20 ± 0.13	3.83 ± 0.23	2.99 ± 0.08	3.99 ± 0.35
Hf (ppm)	5 ± 0	5 ± 0	4 ± 1	5 ± 0	5 ± 0	7 ± 0	4 ± 0
Ni (ppm)	32 ± 26	16 ± 26	204 ± 13	37 ± 13	34 ± 21	14 ± 20	42 ± 23
Rb (ppm)	222 ± 24	111 ± 20	60 ± 33	129 ± 10	133 ± 6	116 ± 5	118 ± 17
Rb (ppm)*	227 ± 21					114 ± 2	
Sb (ppm)	4 ± 3	5 ± 5	2 ± 1	1 ± 0	4 ± 0	3 ± 0	5 ± 2
Sc (ppm)	13 ± 1	12 ± 1	14 ± 1	11 ± 0	14 ± 0	11 ± 0	14 ± 1
Sr (ppm)	159 ± 43	215 ± 27	676 ± 180	357 ± 41	373 ± 45	315 ± 33	606 ± 51
Ta (ppm)	1.2 ± 0.1	1.0 ± 0.1	0.9 ± 0.1	1.2 ± 0.1	1.2 ± 0.0	1.1 ± 0.0	1.2 ± 0.1
Tb (ppm)	0.9 ± 0.1	0.8 ± 0.1	0.8 ± 0.0	0.8 ± 0.1	0.9 ± 0.1	0.9 ± 0.1	0.8 ± 0.0
Th (ppm)	13 ± 0	12 ± 1	9 ± 1	11 ± 0	13 ± 0	12 ± 0	12 ± 1
Zn (ppm)	120 ± 24	94 ± 11	108 ± 6	70 ± 11	85 ± 16	63 ± 4	103 ± 12
Zr (ppm)	122 ± 17	129 ± 22	120 ± 10	135 ± 15	144 ± 22	170 ± 12	112 ± 12
Al (%)	7.43 ± 0.32	6.73 ± 0.45	6.66 ± 0.29	6.86 ± 0.34	7.96 ± 0.17	6.55 ± 0.25	8.23 ± 0.78
Ba (ppm)	516 ± 164	374 ± 28	239 ± 63	342 ± 26	596 ± 63	530 ± 46	362 ± 68
Ba (ppm)†	474 ± 51						
Ca (%)	11.05 ± 0.93	16.77 ± 1.35	12.50 ± 3.17	15.88 ± 0.97	11.21 ± 0.57	10.74 ± 0.35	14.37 ± 1.61
Dy (ppm)	5 ± 0	4 ± 0	4 ± 0	4 ± 0	5 ± 0	5 ± 0	4 ± 0
K (%)	2.45 ± 0.82	1.89 ± 0.30	1.32 ± 0.49	2.43 ± 0.17	2.65 ± 0.26	2.51 ± 0.18	2.18 ± 0.31
K (%)*	3.18 ± 0.35					2.59 ± 0.17	
Mn (ppm)	553 ± 38	501 ± 23	486 ± 92	481 ± 55	654 ± 77	571 ± 38	651 ± 22
Na (%)	0.64 ± 0.41	0.33 ± 0.06	0.94 ± 0.14	0.25 ± 0.04	0.31 ± 0.09	0.26 ± 0.04	0.39 ± 0.08
Na (%)*	0.33 ± 0.13					0.25 ± 0.04	
Ti (%)	0.36 ± 0.04	0.35 ± 0.02	0.35 ± 0.02	0.33 ± 0.02	0.39 ± 0.04	0.38 ± 0.04	0.39 ± 0.05
V (ppm)	93 ± 7	87 ± 15	104 ± 7	85 ± 16	95 ± 8	67 ± 6	127 ± 10

Table 4   *(Continued)*

Groups Elements	Portugal (n = 2) Mean sd	Puente (n = 15) Mean sd	REM (n = 5) Mean sd	Reus (n = 15) Mean sd	Seville (n = 36) Mean sd	Talavera (n = 14) Mean sd	VdP (n = 15) Mean sd
As (ppm)	33 ± 10	22 ± 2	39 ± 8	16 ± 2	18 ± 11	22 ± 10	19 ± 4
La (ppm)	27 ± 2	38 ± 1	42 ± 0	30 ± 1	31 ± 2	39 ± 2	42 ± 1
Lu (ppm)	0.3 ± 0.0	0.5 ± 0.0	0.4 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.4 ± 0.0	0.4 ± 0.0
Nd (ppm)	25 ± 2	35 ± 1	33 ± 1	26 ± 2	26 ± 2	36 ± 3	37 ± 2
Sm (ppm)	5 ± 0	7 ± 0	7 ± 0	5 ± 0	6 ± 0	7 ± 0	7 ± 0
U (ppm)	3 ± 0	5 ± 0	4 ± 1	3 ± 1	2 ± 0	5 ± 0	4 ± 0
Yb (ppm)	2 ± 0	3 ± 0	3 ± 0	2 ± 0	2 ± 0	3 ± 0	3 ± 0
Ce (ppm)	56 ± 4	80 ± 2	84 ± 1	60 ± 3	62 ± 3	80 ± 4	84 ± 2
Co (ppm)	18 ± 8	14 ± 1	31 ± 7	15 ± 2	15 ± 4	15 ± 3	18 ± 2
Cr (ppm)	80 ± 4	58 ± 3	106 ± 3	72 ± 5	72 ± 8	53 ± 5	78 ± 2
Cs (ppm)	3 ± 0	12 ± 0	11 ± 1	15 ± 3	5 ± 1	10 ± 1	9 ± 0
Eu (ppm)	1.1 ± 0.1	1.2 ± 0.0	1.4 ± 0.0	1.0 ± 0.0	1.1 ± 0.1	1.2 ± 0.1	1.4 ± 0.0
Fe (%)	2.77 ± 0.08	3.55 ± 0.19	5.01 ± 0.08	2.98 ± 0.11	3.21 ± 0.24	3.27 ± 0.29	4.25 ± 0.09
Hf (ppm)	5 ± 1	5 ± 0	4 ± 0	3 ± 0	5 ± 0	6 ± 1	5 ± 0
Ni (ppm)	0 ± 0	17 ± 20	58 ± 35	33 ± 23	20 ± 19	15 ± 32	35 ± 26
Rb (ppm)	50 ± 13	165 ± 9	169 ± 8	113 ± 12	73 ± 13	157 ± 19	149 ± 8
Rb (ppm)*				116 ± 12	69 ± 14	154 ± 10	148 ± 8
Sb (ppm)	1 ± 0	2 ± 1	5 ± 0	2 ± 1	3 ± 3	2 ± 1	3 ± 0
Sc (ppm)	10 ± 1	13 ± 1	19 ± 0	12 ± 1	11 ± 1	12 ± 1	16 ± 0
Sr (ppm)	359 ± 16	354 ± 46	750 ± 213	496 ± 70	455 ± 62	329 ± 40	229 ± 33
Ta (ppm)	1.4 ± 0.1	1.5 ± 0.1	1.3 ± 0.0	0.9 ± 0.0	0.9 ± 0.1	1.5 ± 0.1	1.3 ± 0.1
Tb (ppm)	0.6 ± 0.1	1.0 ± 0.2	1.0 ± 0.1	0.7 ± 0.1	0.8 ± 0.1	1.0 ± 0.1	1.0 ± 0.1
Th (ppm)	8 ± 1	15 ± 0	15 ± 0	10 ± 0	9 ± 1	15 ± 1	15 ± 0
Zn (ppm)	66 ± 6	81 ± 5	138 ± 13	79 ± 12	73 ± 9	80 ± 8	127 ± 5
Zr (ppm)	146 ± 11	161 ± 13	113 ± 12	92 ± 19	134 ± 16	154 ± 21	147 ± 20
Al (%)	5.76 ± 0.42	8.21 ± 0.43	9.34 ± 0.92	6.82 ± 0.33	5.94 ± 0.57	7.77 ± 0.56	8.84 ± 0.27
Ba (ppm)	160 ± 0	407 ± 39	513 ± 28	602 ± 633	343 ± 126	377 ± 35	641 ± 62
Ba (ppm)†				439 ± 43	323 ± 38		
Ca (%)	19.93 ± 0.80	12.63 ± 0.74	8.89 ± 0.42	17.82 ± 0.76	14.73 ± 1.29	13.02 ± 0.98	8.61 ± 0.61
Dy (ppm)	3 ± 0	5 ± 0	5 ± 0	4 ± 0	4 ± 0	5 ± 0	5 ± 0
K (%)	0.96 ± 0.35	2.37 ± 0.19	2.92 ± 0.53	1.95 ± 0.28	1.43 ± 0.35	2.27 ± 0.34	2.79 ± 0.29
K (%)*				2.14 ± 0.22	1.50 ± 0.31	2.33 ± 0.23	2.84 ± 0.25
Mn (ppm)	246 ± 49	520 ± 25	657 ± 16	420 ± 51	652 ± 137	495 ± 29	741 ± 67
Na (%)	0.74 ± 0.24	0.37 ± 0.04	0.77 ± 0.25	0.42 ± 0.09	0.65 ± 0.19	0.48 ± 0.16	0.38 ± 0.14
Na (%)*				0.37 ± 0.07	0.62 ± 0.19	0.43 ± 0.05	0.34 ± 0.04
Ti (%)	0.45 ± 0.02	0.38 ± 0.04	0.37 ± 0.04	0.32 ± 0.02	0.34 ± 0.03	0.35 ± 0.03	0.41 ± 0.02
V (ppm)	58 ± 1	80 ± 8	167 ± 12	98 ± 9	82 ± 12	68 ± 5	119 ± 10

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Table 4   (Continued)

Groups Elements	Villafeliche (n = 13)		MJ0269 (Delft)	MJ0284 (Sgraffito)	MJ0287	MJ0288	MJ0293	MJ0294
	Mean	sd	Composition					
As (ppm)	11 ± 6		12	6	11	12	25	6
La (ppm)	39 ± 2		24	40	31	30	35	35
Lu (ppm)	0.4 ± 0.0		0.3	0.4	0.4	0.3	0.4	0.4
Nd (ppm)	32 ± 2		21	34	27	27	31	28
Sm (ppm)	7 ± 0		4	6	6	5	6	6
U (ppm)	7 ± 1		2	2	3	3	2	3
Yb (ppm)	3 ± 0		2	2	3	2	3	2
Ce (ppm)	81 ± 2		49	84	63	59	71	66
Co (ppm)	20 ± 3		24	100	21	22	25	24
Cr (ppm)	69 ± 3		74	159	90	73	64	82
Cs (ppm)	7 ± 1		6	8	5	4	6	10
Eu (ppm)	1.3 ± 0.0		0.9	1.4	1.2	1.1	1.2	1.2
Fe (%)	2.91 ± 0.14		2.31	5.65	3.13	2.97	3.17	3.73
Hf (ppm)	6 ± 0		4	4	7	6	5	5
Ni (ppm)	28 ± 26		42	102	25	0	0	45
Rb (ppm)	162 ± 12		39	152	83	59	67	141
Rb (ppm)*	166 ± 13							
Sb (ppm)	2 ± 1		1	1	1	1	3	1
Sc (ppm)	13 ± 1		9	20	11	10	12	14
Sr (ppm)	628 ± 34		531	180	420	569	255	685
Ta (ppm)	1.2 ± 0.0		0.8	1.6	1.1	0.9	1.0	1.3
Tb (ppm)	0.8 ± 0.1		0.5	0.8	0.7	0.7	0.8	0.8
Th (ppm)	14 ± 0		8	13	9	9	12	12
Zn (ppm)	68 ± 11		62	141	78	210	92	112
Zr (ppm)	165 ± 15		137	116	208	156	136	117
Al (%)	7.39 ± 0.47		5.02	9.15	6.03	5.30	6.56	8.53
Ba (ppm)	544 ± 45		148	293	253	280	285	305
Ba (ppm)†								
Ca (%)	12.05 ± 0.71		16.23	5.19	11.23	16.43	16.83	14.54
Dy (ppm)	5 ± 0		3	4	4	4	4	5
K (%)	2.76 ± 0.23		0.91	2.18	1.60	1.17	1.28	2.21
K (%)*	2.87 ± 0.18							
Mn (ppm)	336 ± 23		320	1105	380	547	542	225
Na (%)	0.21 ± 0.05		0.72	0.52	0.62	0.65	0.61	0.29
Na (%)*	0.19 ± 0.03							
Ti (%)	0.40 ± 0.03		0.23	0.47	0.32	0.32	0.34	0.37
V (ppm)	79 ± 7		76	132	88	85	88	90

*Values for those groups without taking into account samples showing analcime in their diffractograms.
†Ba values for those groups which include samples with aberrant Ba contents.
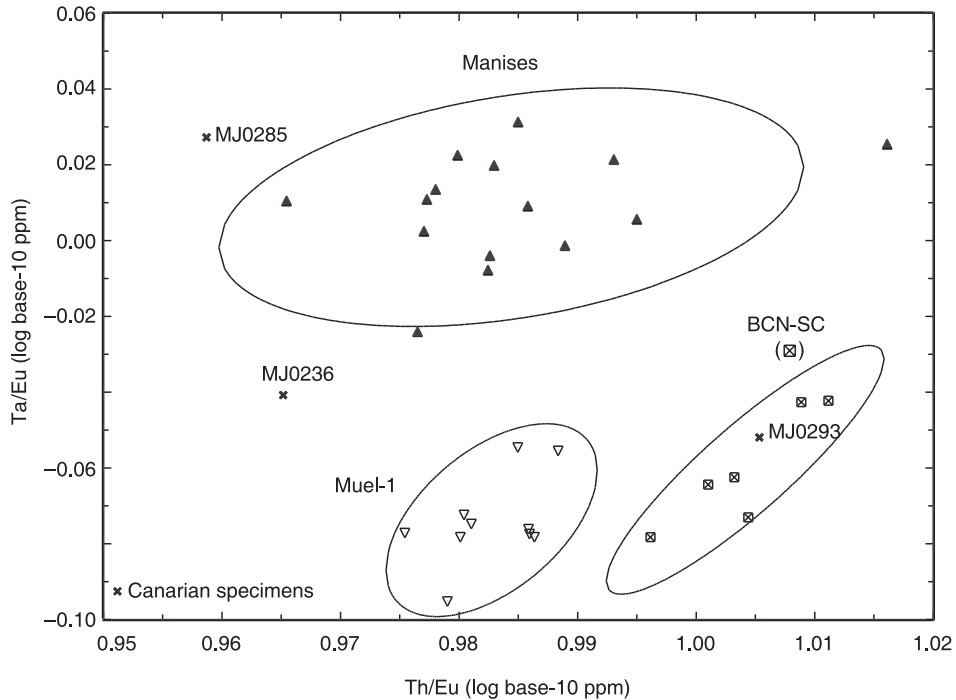
Figure 5   *Bivariate plot using log base 10 Th/Eu and Ta/Eu as variables showing the BCN-SC, Manises and Muel-1 group separation.*

Lleida Remolins (REM), Talavera and Seville. We also identified two additional paste reference groups that we attribute to unidentified Ligurian and Portuguese productions. Despite the low number of specimens that comprise the Ligurian ($n = 4$) and the Portuguese ($n = 2$) groups, there is archaeological and chemical evidence that corroborate their existence. The suggested Ligurian group exhibits relatively high chromium and nickel concentrations that may indicate an ophiolitic origin for these sherds, such as the Ligurian and northern Italy regions (Maggetti 2005), reinforcing their typological/stylistic classification as either Ligurian or northern Italian productions. Moreover, and possibly as a consequence of the high standardization and chemical similarity of the materials, some of the groups that can be separated show a slight amount of overlap. Consequently, and for better visual group separation purposes, separation of the BCN-SC, Manises and Muel-1 groups are depicted in a separate plot (Fig. 5) using Th/Eu and Ta/Eu base-10 logarithm values as variables. Unambiguous separation of the Talavera and Puente groups presents further problems due to their chemical similarity. These cities are geographically very close to each other and have had a very similar majolica production history during the past four centuries. Many problems hinder discrimination between these production centres, especially on the basis of their decorative styles and typologies. Additionally, tableware production styles from Puente have traditionally been considered to be an imitation of the Talavera style (Sánchez-Pacheco 1997). Therefore, and related to their similar geological environment, chemical data generated for several sherds analysed from both sites exhibit similar compositions (Iñañez *et al.* in press), resulting in an overlapping group structure that requires additional study.

By projecting the Canary Islands samples against the Iberian Peninsula reference groups (Figs 4 and 5), we assert that most of the pottery sampled from the Canary Islands can be attributed to Seville. This conclusion is in agreement with the historical and archaeological data. In addition, two lustreware specimens, one from the Cueva Pintada (MJ0236) and one from the Ancient Convent of San Francisco (MJ0285), are an excellent match with the Manises reference group. One blue on white sherd (MJ0293) also presents a high chemical similarity with the Catalan group of BCN-SC. Despite the ability to attribute many of the Canary Island samples to specific production locales, there are a few samples that do not correspond with any of our reference groups (MJ0262, MJ0269, MJ0284 and MJ0294). According to the archaeological data, MJ0284 is a *sgraffito* ware, which is technologically different and also exhibits higher Ni and Cr amounts, albeit not as high as the Ligurian majolica. Sample MJ0269 has been classified archaeologically as Delftware, a plausible interpretation given that chemical composition of this sample does not seem to match any reference group from the Iberian Peninsula or Italy. Conversely, sample MJ0289 also has been classified by typology and decoration also as Delftware, but its chemical composition clearly indicates a Sevillian origin. Finally, as discussed above, there are six samples that we attribute to the Ligurian region and to Portugal.

In order to achieve a better discrimination among the groups discussed above, a stepwise discriminant analysis was performed on the chemical data set, using all of the analysed samples. The most suitable components for running the discriminant analysis were the same as those used for the scatter plots following the log ratio transformation using Eu as divisor (Table 3). As discussed above, elements presumed to be problematic because of alteration or contamination processes, such as Co, Na, Rb or K, and those components also dismissed from the scatter plots because of their high variability or analytical problems (e.g., As, U, Ni, Tb and Sb), were not considered in the stepwise discriminant analysis.

Stepwise discriminant analysis (DA) is a powerful tool to assess the groups identified by other approaches. Using a stepwise selection algorithm, it was determined that 17 variables (the log ratio transformed components: Cr, Th, Sc, Cs, Sm, Sr, Ta, Ca, Hf, Fe, Ce, La, Al, Mn, Ti, V and Yb) are significant predictors of majolica groups. The 12 discriminating functions with *P* values less than 0.05 are statistically significant at the 95% confidence level. The evaluation of all the sherds classified according to the previous plots has shown a very high success score: 98.54%, in which 203 out of 206 sherds match their suggested group according to the scatter plots and the archaeological information. Results are summarized in Figure 6, using the two first discriminant functions. As a result, DA has operated in a successful manner for those sherds from different archaeological contexts that result in a single chemical group in the spatial distribution, such as the cases of Talavera and Puente. Thus, unique groups for the producing towns of Talavera and Puente were identified according to their multivariate chemical differences, although one specimen from Puente and two from Talavera still indicate an ambiguous origin. Regarding the Canary specimens, 11 out of 22 of the samples are assigned to a Sevillian origin by DA, as expected by the scatter plots and the historical documentation. The three lustreware majolicas are attributed to Seville (MJ0286) and to Manises (MJ0236 and MJ0285), whereas the blue on white specimen (MJ0293) clearly is attributed to the BCN-SC group, supporting the previous classification made by examination of scatter plots. Additionally, the proposed Ligurian and Portuguese groups are supported by DA, forming their own respective groups with no attribution to any other group. Nevertheless, individual MJ0269, archaeologically classified as Delft series, is classified by similarity as a Sevillian product, as are MJ0284 and MJ0294 with OB/SA.
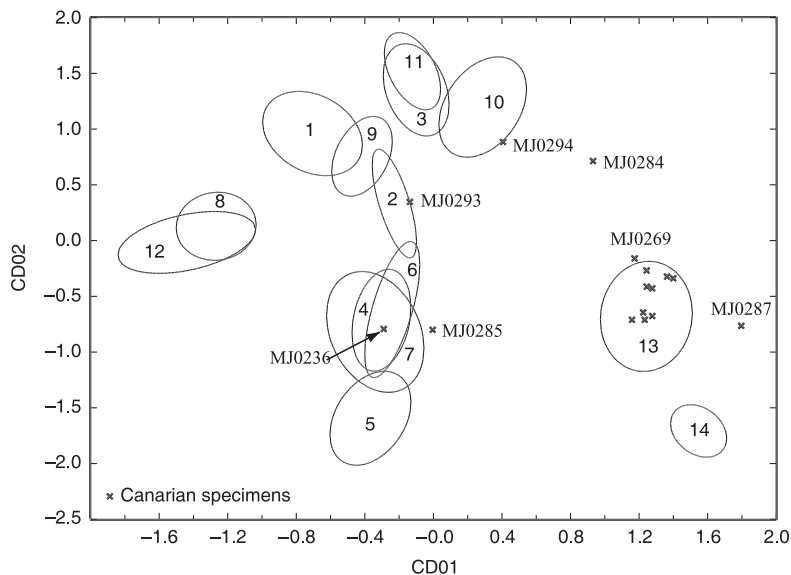
Figure 6 *Bivariate plot using the first two discriminant functions as variables, showing the group separations suggested by canonical discriminant analysis: 1, BCN-DR; 2, BCN-SC; 3, Reus; 4, Muel-1; 5, Muel-2; 6, Villafeliche; 7, Manises; 8, Puente del Arzobispo; 9, VdP; 10, OB/SA; 11, REM; 12, Talavera; 13, Seville; 14, Portugal; and individuals from the Canary Islands. The Ligurian group is not represented because its high chemical differences compress the rest of the groups in the graphic representation. Ellipses represent a confidence interval of 90%.*

In order to assess the statistical strength of the groups suggested by the scatter plots and confirmed by the stepwise DA, a probabilistic group assignment was obtained using Mahalanobis distance (MD). However, the limited number of samples for some of the reference groups precluded the possibility of employing all the chemical variables measured by NAA or the 18 most discriminating transformed variables for this data set, because most of the suggested groups are comprised of about 15 specimens. Thus, another scale-reducing statistical technique is needed, such as principal components analysis (PCA). PCA on the variance–covariance matrix was performed in the same way as the stepwise DA and the scatter plots (see above). Likewise, As, K, Na, Rb U, Tb and Sb were not included for the reasons discussed above. The PCA indicated that 95% of the cumulative variance was accounted for in the nine first principal components, resulting in a good estimation of the overall composition of the majolica sherds. Given that the majolica production centre groups identified by scatter plots and DA are usually smaller in number than 18 and normally larger than 11, it was possible to calculate MD probabilities using the first nine principal components to assess the membership probabilities for Canarian samples assigned to each group and those that form their own groups, such as the Ligurian and Portuguese sherds. It must be highlighted that the two groups that do not agree with the number-restriction rule were the chemical groups proposed for REM and BCN-SC, each having fewer than 11 sherds.

The MD calculations (Table 5), confirm that the groups previously identified by scatter plots and DA are statistically robust, and are also in agreement with a previous study performed by X-ray fluorescence that includes more than 600 specimens (Iñañez 2007). Consequently, most of the sherds are assigned with a higher probability, greater than 5%, to their respective

Table 5 *Membership probabilities in percentage and suggested provenance according to Mahalanobis distance calculations, discriminant analysis and scatter plots*

ANID	Membership probabilities (%)				
	Manises	*Talavera*	*Seville*	*Best group*	*Suggested provenance*
MJ0236	5.10	0.27	0.03	Manises	Manises
MJ0237	0.01	0.06	64.56	Sevilla	Seville
MJ0241	0.04	0.12	60.34	Sevilla	Seville
MJ0253	0.01	0.02	58.79	Sevilla	Seville
MJ0258	0.03	0.26	98.07	Sevilla	Seville
MJ0262	0.13	0.10	7.06	Sevilla	Seville
MJ0268	0.02	0.08	97.83	Sevilla	Seville
MJ0269	0.00	0.04	0.30	–	Delft (?)
MJ0270	0.00	0.00	0.00	–	Ligur
MJ0272	0.00	0.00	0.00	–	Ligur
MJ0275	0.18	0.00	0.00	–	Portugal
MJ0284	0.01	0.00	0.00	–	Italy (?)
MJ0285	10.86	1.26	0.02	Manises	Manises
MJ0286	0.01	0.04	14.21	Sevilla	Seville
MJ0287	0.03	0.04	0.11	–	Unassigned
MJ0288	0.00	0.01	0.00	–	Unassigned
MJ0289	0.03	0.09	3.81	Sevilla	Seville (?)
MJ0290	0.00	0.00	0.00	–	Ligur
MJ0291	0.00	0.00	0.00	–	Ligur
MJ0292	0.05	0.00	0.00	–	Portugal
MJ0293	39.65	0.60	0.01	–	BCN-SC*
MJ0294	0.12	0.00	0.00	–	Unassigned

*Although MJ0293 has a high membership probability of belonging to the Manises group, we suggest a BCN-SC origin (see Figs 4 and 5 and the Discussion).

chemical groups. Furthermore, the provenance of the specimens that showed a discrepancy among the classification by scatter plots or by DA is now in agreement with MD probabilities. Therefore, eight of the samples from the Canary Islands sites can be assigned to the Seville reference group, including MJ0262, which exhibited certain differences in the scatter plots. In the same way, MJ0236 and MJ0285 exhibit high membership probabilities of belonging to the Manises group, as suggested by scatter plots and DA. Although the probability of MJ0289 is slightly below 5%, it is more than 30 times higher for the Sevillian than for the other groups. Therefore, a Sevillian origin is suggested, which is also in agreement with the archaeological data and the study of the scatter plots and DA.

As is common with MD probabilities, some samples could not be assigned to any group— but given that we have not analysed the entire universe of possibilities, unassigned samples are to be expected. For example, none of the pottery archaeologically classified as Ligurian and confirmed to be distinct by scatter plots and DA shows any probability of belonging to a reference group from the Iberian Peninsula. The low MD probabilities and the high Ni and Cr concentration of these sherds reinforces the hypothesis that these sherds were produced in the Ligurian region or northern Italy. In addition, the two samples classified as Portuguese by archaeologists, and which based on scatter plots and DA also could not be linked to any

reference group from Spain, have low probabilities of membership in any of the references groups. Sample MJ0284, which is typologically classified as a *sgraffito* ware, exhibits a very low probability of membership in any of the reference groups, suggesting that this sample should be classified as unassigned. Moreover, and as a result of the number restrictions for MD calculations, MJ0293 could not be compared with the BCN-SC group due to the low number of samples assigned to this reference group. Although the sample shows a high membership probability with the Manises group, a BCN-SC provenance can be suggested in agreement with the DA attribution and scatter analysis (see Figs 4, 5 and 6). Future studies, which will increase the number of samples for the reference group allowing the MD calculations, may corroborate the final provenance suggested for this sample. Samples MJ0287 and MJ0288, which were assigned to Seville on the basis of scatter plots and DA, were not assigned to any of the reference groups, due to their low MD membership probabilities. Therefore, we classified these as unassigned. Finally, sample MJ0294, the only black and green majolica, cannot be assigned to any reference group by scatter plots or MD. Consequently, it must remain as unassigned.

The results confirm the existence of differences among the two reception sites from Gran Canaria Island in terms of pottery provenance. Pottery from La Cueva Pintada shows two different origins, Seville and Manises, whereas pottery from the Ancient Convent of San Francisco in Las Palmas was obtained from at least five different areas: Seville, Liguria, Portugal, Barcelona and Delft. The variation in pottery consumed at both sites likely results from the different roles and chronologies that occurred at each site. Whereas La Cueva Pintada samples are dated to the late 15th and early 16th centuries, pottery from the Ancient Convent of San Francisco date to the 16th and 17th centuries. Moreover, La Cueva Pintada was a secondary place in terms of importance, since the capital of the island was located at Las Palmas. In contrast, the Convent of San Francisco was an important religious community in the capital and the entire island during the 16th and 17th centuries. Presumably, it had greater access to trade goods as a result of its location and socio-economic/religious role.

In terms of types of pottery, the results show that lustreware recovered at Gran Canaria originated from Seville and Manises. This is reasonable, given that the most important production from Manises is lustreware, and that this was the most luxurious ceramic ware until the beginning of the 17th century (Soler 1997).

Surprisingly, the blue on white (MJ0293) sample can be attributed, according to the scatter plots and DA analyses, to the Catalan BCN-SC group—a group so far comprised exclusively of lustreware pottery. Therefore, the results from Gran Canaria are not only the first identification of BCN-SC ceramics outside of Barcelona, but also this sample provides compelling evidence for the first example of a non-lustreware ceramic that has been identified as a product of the BCN-SC group.

Sevillian-produced majolica is by far the most represented ceramic in Gran Canaria. Moreover, it also is very heterogeneous in terms of kinds of decorative type: lusterware ($n = 1$), Sevillian white ($n = 4$), Columbia simple ($n = 2$) and Isabela polychrome ($n = 1$). In addition, one green tile also has a Sevillian origin (MJ0253). A special case is raised by one sample, MJ0289, archaeologically classified as Delft. In fact, as has been addressed above, sample MJ0269 was archaeologically classified as a Delft product. The fact that chemically this sample does not cluster to any defined group, and in the absence of other data, would seem to support an actual Dutch origin. In contrast, sample MJ0289, which chemically corresponds to the Seville reference group, might be a Delft imitation. The existence of such imitations has been hypothesized on the basis of historical and archaeological evidence (Pleguezuelo and Sánchez Cortegana 1997) and has now been confirmed archaeometrically.

Blue on white ceramics are represented by a single sample attributed to the BCN-SC group, two Portuguese samples and four Ligurian samples. Based on the current sample, this decorative type exhibits the widest geographic dispersion concerning its origin among the pottery from Gran Canaria.

Finally, a single *sgrafitto* specimen could not be assigned to any of the compositional groups. However, it is noteworthy that its Fe content was the highest among the studied materials. This feature, together with a relatively medium Ca content, provides a characteristic red colour for the clay body. This colour, which would be undesirable in majolica, is a desired effect for *sgraffito* pottery (Lazzarini *et al*. 1980).

CONCLUSION

The origin of the Spanish Colonial trade monopoly with the Americas can be attributed to the establishment of the Casa de la Contratación in Seville in 1503. This organization directly controlled all trade of goods that were shipped to the Americas from the Castilian kingdom. Consequently, between the 15th and the 18th centuries Sevillian potters produced the majority of majolica exported to the Americas (Sánchez Cortegana 1994). Therefore, Sevillian pottery has a high occurrence in the archaeological records of the Canary Islands and in sites in the Americas. The present study confirms that most of the majolica from the sites of La Cueva Pintada and the Convent of San Francisco are in agreement with a Sevillian provenance, but that Seville was not the only production centre that was exporting pottery shipped to the Americas.

Historical sources, especially numerous notarial and commercial manuscripts generated by the Casa de la Contratación and preserved in the Archivo de Indias, inform us about the strong controls in this trade network. Furthermore, the repetitive pleas by the Canary Islands merchants to increase their market share with the Americas also point to such a situation. The occurrence of pottery of different origins might then be explained by several possibilities that do not exclude each other. On the one hand, despite the very tight control of the Castilian monopoly, vigorous smuggling activity could be plausible, based on the archaeological and archaeometric studies. It has to be highlighted that, from the Iberian Peninsula (except Portugal and its colonies) only Castilian merchants had licence to the trade with America, therefore other entities could not legally provide pottery from other sources. As a result of this monopoly, territories belonging to the Aragon kingdom, such as Barcelona or Valencia, would not have been allowed to trade with America until 1778. On the other hand, objects of non-Sevillian origin could have been included in cargos by Castilian merchants themselves, when the potters from Seville were unable to provide a comparable product (Pleguezuelo and Sánchez Cortegana 1997; Pleguezuelo 2003). Surprisingly, no majolica ware from Talavera de la Reina was identified within the analysed sample, despite the relevant role that this production centre played during the 16th and 17th centuries in Spain.

The present study suggests the arrival of ceramics, and possibly other goods, from different places around the Mediterranean, such as Italy, Manises or Barcelona, Portugal and possibly the Netherlands. The extent of the participation of all these different production centres, and their impact and the variety of products involved in this process, remains uncertain. However, this study shows that the trade network, the most important one at that time, is far more complex than previously assumed. The exact mechanisms by which the different pottery entered these flows, whether licit or illicit, might not be easily understood, but the work must be extended in order to obtain a more comprehensive picture. The role of the Canary Islands in this network becomes, then, a central subject of research for achieving a deeper understanding of the colonial artefacts found in the Americas.

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