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Analytical and multivariate study of roman age architectural terracotta from northeast of Spain

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Abstract

Roman culture employed architectural terracotta made from baked clay as original material to manufacture ceramic pieces. It was often used as a basis for construction of functional and/or decorative elements in roofs, such as plane and curve tiles as well as antefixes with their corresponding "imbrexes". Some of them are conserved nowadays. They were collected in Roman quarries discovered in old cities and villages sited in the *Hispania Citerior* (northeast of Spain in Roman age). A study of the origin and manufacturing process (moulding, baking, touching up and painting) of these terracotta pieces has been made on the basis of the data obtained from a physicochemical characterization of samples. The used techniques were mainly flame absorption and emission spectrometry for the elemental analysis (major and minor elements), dilatometry for the study of thermal behaviour, scanning electron microscopy (SEM) for observation of thin layers and X-ray diffraction spectrometry (XRD) for mineralogical composition. In addition, a supervised pattern recognition programme was applied to the results for a selected group of 85 samples and five variables (chromium, copper, lead, nickel and zinc contents). Dilatometry and SEM results showed baking temperatures of these materials below 900 °C and the existence of zones with very different porosity in the same ceramic piece. Results obtained from multielemental analysis and multivariate statistical study by linear discriminant analysis lead us to the following conclusions: (i) the high content of lead found in a large number of antefixes demonstrates the use of lead oxide as an additive in the lime grout treatment, (ii) different contents of Cu, Zn, Cr, and Ni were indicative of the use of varied clay types in the manufacture process (even in the same production centre) as well as of the existence of a pigmentation process, although this last affirmation is not corroborated by the presence of remains of evident painting in the ceramic pieces, (iii) samples can be classified according to the places where these pieces came from and (iv) more variety in their composition was found in Roman age terracotta production centres.
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1. Introduction

Ancient architectural terracotta comprises all of those pieces made from baked clay and used as functional and/or decorative elements in construction. There is not much infor-mation about the production, employment and evolution of ancient architectural terracotta but it seems that Roman culture acquired from Etruscan the habit of employing terracotta elements for covering and decorating their house's roofs [1]. Among these ceramic pieces, plane and curve tiles as well as antefixes were the most often used in roof construction [2]. In particular, antefixes were ornamental pieces located at the end of a tile-line, placed in vertical position and fixed to the

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corresponding curve tile with the aid of an intermediate piece called "imbrex". They could be made of diverse forms (circular, square, triangular, lobed, etc.) and could represent varied figures (head, palm, etc.) [1]. In particular, Fig. 1 shows an example of roof construction indicating the locations of the different elements.

Terracotta was made in most cases of clays from sedimentary type, which had a variety of colours and paste qualities depending on the origin zone. Potters used to wash the clay with water and purify it trough a decantation process. Then, they usually mixed it with sand or other substances (chamotte, fragments of volcanic rocks, etc.) in order to decrease the plasticity of the ceramic pastes or to prevent cracks during baking. They left to dry all pieces into their moulds before the baking process took place in kilns at 600–1000 °C during several days under an oxidant atmosphere [3]. The process was continued by filling the pores of the ceramic pieces using the "engobium", a lime grout (sometimes with addition of lead oxide) or fine clay suspended in water in order to obtain a homogeneous and very smooth surface. Finally, the ceramic material was subject to a second baking process at 600–900 °C [4]. After this, some of the ornamental pieces, like antefixes, were touched up and painted [5] using natural substances such as ochre, cinnabar, minium and several iron hydroxides (red and yellow colours), carbon (black colour), copper compounds (blue and green colours), kaolin (white colour), chromium compounds (yellow shades), nickel substances (greenish-blue shades), etc. However, these colours have seldom withstood the passing of time, because of this pigmentation usually took place after the baking process.

Traditionally, archaeological methods used for characterization of ancient terracotta [6,7] did not enable us to obtain enough information about several aspects regarding to their nature and origin. On the other hand, information about archaeological sites [8] where fragments of ancient architectural terracotta were collected is usually vague, imprecise and sometimes many of the archaeological pieces were located far from their original source. Taking into account all these aspects, chemical and mineralogical data obtained from them help us to establish relations between the composition of the ceramic materials and the raw materials employed, the technological aspects of manufacture process and the origin zone [9–11].

The aim of this work was to obtain mineralogical, chemical and dilatometric information about ancient terracotta samples collected in Roman quarries discovered in old cities and villages sited in the *Hispania Citerior* (northeast of Spain in Roman age). These results allowed us to compare the original materials used with the clay from the surrounding areas where archaeological samples were collected, with the purpose of knowing whether the analysed pieces were made of raw materials from the surrounding areas of the corresponding ancient village or were imported from other areas. In the same way, the presence of large amounts of not typical substances, such as chamotte and fragments of volcanic rock, was indicative of a deliberated addition of these components in the manufacturing process in order to improve the physical properties of the ceramic paste. In addition, the presence of relative high amount of other elements such as zinc, cooper, chromium, nickel, etc. in antefixes could prove the use of different clay types in the fabrication process and the existence of a pigmentation process, in spite of the absence of remains of painting in the pieces.

Due to the high number of variables involved in the manufacturing process, it is often very difficult to establish a connection between the ceramic object and the raw materials [12,13]. This circumstance, joined to the large number of samples analysed, drove us to use statistical procedures that provide a way to find possible connections among a high number of variables and classify samples into compositional groups [11,14–17]. In this paper a multivariate statistical study by linear discriminant analysis using the SPSS program was applied to 85 architectural terracotta samples with the purpose of classifying these samples into groups sharing similar chemical composition in order to establish how the manufacturing process of these architectural terracotta pieces took place and when it was possible to establish their origin. It introduces us to the importance of chemical data and chemometrics in classification of terracotta pieces within architectural realm, where the traditional archaeological methods used for characterization of ancient terracotta did not enable us to obtain enough information.

2. Experimental

2.1. Description of the samples

The ceramic analysed were fragments of Roman age terracotta coming from old cities and villages sited in the *Hispania. Citerior* [18] and located, at present, in different National Archaeological Museums of Spain and catalogued by
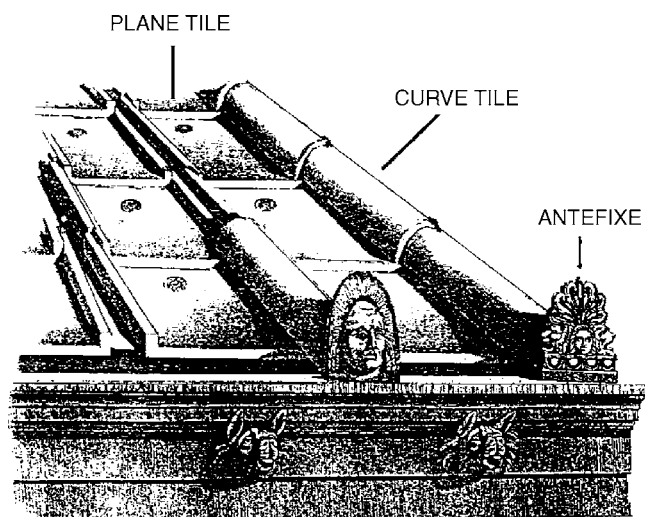

Fig. 1. Terracotta elements in a Roman roof construction.

Ramos [1]. The terracotta production centres and their diffusion areas are represented in the map of Fig. 2, being indicated in parenthesis the number of samples collected from each zone, with the exception of two samples of unknown origin. A total of 85 samples were analysed probably corresponding to the period included between the IV century B.C. and the II century A.D. They were classified in 14 tiles, 42 antefixes, 18 imbrexes, 3 corresponding to rest of "engobium" and 8 raw materials (clays) from different zones.

Samples were taken from the inner part of fragments in the case of tiles or from the back in the case of antefixes, with the aid of a scalpel. In all cases, a minimum part of sample was taken to minimize damage to archaeological objects. Each sample was ground in an agate mortar with a pestle before analysis, in order to secure homogeneity and to reduce the particle size. Due to the small amount of sample in most cases (sometimes less than 100 mg), it was not always possible to carry out replicates of analyses.

2.2. Physical analyses

Thermal behaviour of terracotta samples coming from tiles and antefixes was carried out using a Adamel–Lhomargy dilatometer in the following conditions: a 7 mm of length cylinder was used in an air stream in a range of temperatures from 25 to 1000 °C, with a heating rate of $5\,°C\,min^{-1}$, using an alumina standard.

In addition, observations of 1 μm layers corresponding to some ceramic samples, polished with diamond, were made using a Philips-XL 30 scanning electron microscope (SEM).

2.3. Chemical and mineralogical analyses

Chemical analyses of major and minor elements were performed by flame absorption and emission spectrometry (FAAS) in a Perkin Elmer 503 spectrometer [19]. Previous dissolution of samples was carried out in the following way:

Fig. 2. Map of the terracotta Production Centres and their distribution.

a minimum amount of sample was treated with hydrofluoric acid in an open vessel, heating in a hot plate until dryness. This treatment was followed by addition of aqua regia, heating again until dryness. The residue was dissolved with 1 ml of concentrated hydrochloric acid and diluted with water to the mark in Teflon volumetric flasks. Al, Fe, Ca, Mg, Cu, Ni, Cr, Zn and Pb were determined by atomic absorption spectrometry, and Na and K by emission mode in the same instrument. Silica was determined using an alkaline fusion when there were enough sample amounts.

Mineralogical data were obtained from X-ray diffraction analyses using a Philips PW 1035 Diffractometer. This technique allowed us to identify and semi-quantify minerals in the samples.

Care was taken to keep the contamination to a minimum. Ultrapure water was used throughout and all reagents used were of analytical grade.

2.4. Chemometric study

A statistical processing of the data was carried out with the SPSS 11.5 Programme using the concentration of Pb, Cu, Zn, Cr and Ni as variables of the 85 analysed samples. Supervised pattern recognition was applied in this study. Linear discriminant analysis was used for hard classification purposes, trying to establish possible connections among groups of samples and variables. This procedure is useful for classifying the terracotta dataset into groups according to the places where the samples were found. It generates a small number of functions of quantitative measurements, which are linear combinations of the standardized pattern variables with weight coefficients. These functions are called canonical discriminant functions and help discriminate among groups of terracotta samples with different origin. The procedure assumes that the variables are drawn from population with multivariate normal distributions and that variables have equal variances.

3. Results and discussion

3.1. Physical analyses

Fig. 3 shows an example of the typical dilation-contraction curve obtained for a large number of these samples. The dilation curve versus temperature ($d\Delta l/l_0/dT$) shows two variations in the ranges of 550–650 and 650–700 °C. The expansive effect of about 600 °C can be attributed to the transformation of quartz (α–β transformation). The material contracts slightly until a temperature of 750 °C is reached, in this moment the sinterisation step starts. The presence of calcium carbonate causes a second expansive effect about 700 °C due to the formation of CO_2. In the same way, the thermal effect around 800 °C is due to the carbonate coming from calcite and changes were not observed at higher temperatures. The total dilation observed in the temperature range from 25 to 1000 °C was lower than 2%. These typ-
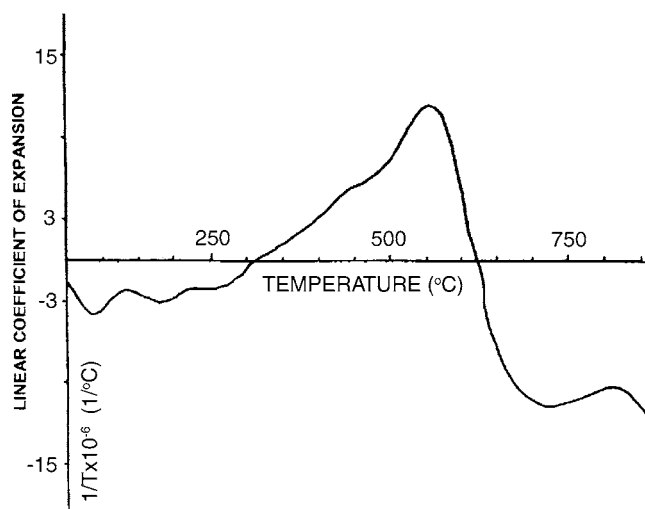

Fig. 3. Dilatometric curve for a tile from Tarraco (Spain).

ical linear contraction–expansion effects were comparable than those previously obtained in similar Roman materials [14]. In this sense, the presence of gehlenite has been previously described at temperatures under 800 °C in kaolinitic clays [20], which contained calcium carbonate; and also is recognised the formation of gehlenite from lime, starting at 850 °C. Likewise, thermal effects were not observed in previously constant heating rate dilatometric studies of tiles and antefixes, containing calcium carbonate in 900–950 °C range.

In our case, there are several evidences of the use of temperatures not higher than 900 °C in the baked process in the oven: (i) the total dilation in the studied range lower than 2%, without thermal effects at temperatures higher than 850 °C, (ii) alteration of gehlenite and decarbonation of calcium carbonate from a secondary origin observed in several samples by SEM, and (iii) the presence of quartz, calcite, relic clay materials, gehlenite, pseudowollastonite and anorthite obtained by XRD analyses.

Additionally, SEM analysis and thermal treatment of tiles at temperatures close to 900 °C, showed a stable microstructure containing amorphous, crystalline and porous phases. SEM analyses were also indicative of the existence of zones with very different density in the same ceramic sample. As an example, it can be observed a microphotograph (Fig. 4) corresponding to a tile from Tarraco where the different sinterisation led to an inner zone with an elevate porosity and another outer zone more dense and compact. This fact could be explained by: (i) certain variables involved in the baked process, as time and temperature, did not provide adequate conditions to obtain a homogeneous material, (ii) the recovering process of the outer face of some of the pieces ("engobium").

3.2. Chemical and mineralogical analyses

Major elements such as Si, Al, Mg, Ca, Na, K and Fe were determined in the samples. Obtained data showed SiO_2 content between 45 and 70% mainly present as quartz. Al_2O_3
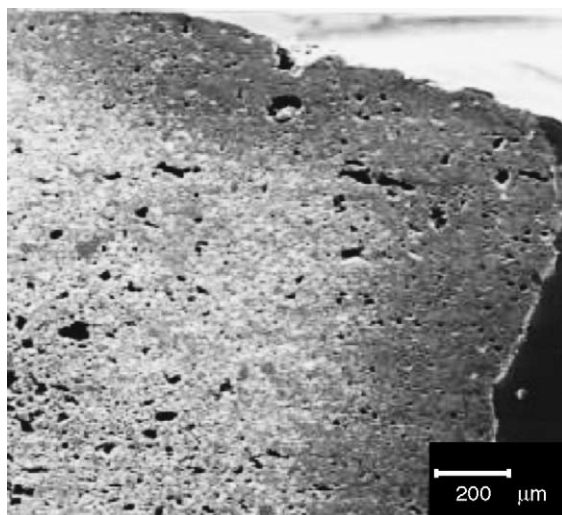
Fig. 4. Photomicrograph of the ceramic body showing two different zones. The core is characterized by its high porosity.

average content was of 15%, with a minimum value of 10%. K_2O showed contents between 2 and 3% and Na_2O lower than 1%. The contents of MgO were very uniform (1–3% in all samples) whereas CaO showed a great variability caused mainly by additional treatments such as the lime grout.

High fluctuations were observed for Fe_2O_3 (0.2–10.8 %). Samples coming from ALB, BAE and COM (see Fig. 2) showed the highest concentration of Fe, with average values of 6.1, 5.2 and 5.5%, respectively. The lowest average value corresponded to samples from MAS (2.7%). This element is an indicator of the raw material nature, as well as of the manufacture process. By comparison between pairs of antefix with their corresponding imbrex it can be seen that they had generally similar Fe content. However in some particular cases it was considerably higher in the antefixes (9COM with 9.3% in the antefix and 4.5% in the imbrex, and 3TER with 3.7% in the antefix and 1.3% in the imbrex) or in the imbrexes (8ALB with 2.2% in the antefix and 5.8% in the imbrex, and 7ARC with 0.9% in the antefix and 2.8% in the imbrex). These examples can be considered as a signal of different clays used in their manufacture, or even of the different production centres.

Minor elements were also determined. They were chosen taking into account that their unusual high concentrations could show the existence of a pigmentation process. In any case, differences in concentration could be indicative of the use of different clays deliberately mixed or added coming from the same or other areas. All these cases represent an evidence of the attempt by the ancient potters of improving the terracotta properties. Table 1 shows the average values and

Table 1
Average values ± standard deviations for minor chemical elements

Place	Number of samples	Cu ($\mu g\,g^{-1}$)	Zn ($\mu g\,g^{-1}$)	Pb ($\mu g\,g^{-1}$)	Cr ($\mu g\,g^{-1}$)	Ni ($\mu g\,g^{-1}$)
Tarraco (TAR)	15	51 ± 26	85 ± 25	170 ± 172	48 ± 20	55 ± 16
Emporium (EMP)	11	39 ± 17	93 ± 31	82 ± 35	81 ± 27	61 ± 26
Termes (TER)	9	48 ± 8	103 ± 28	123 ± 46	85 ± 36	77 ± 19
Complutum (COM)	7	37 ± 10	102 ± 33	76 ± 30	56 ± 17	49 ± 16
Mas de Gomandi (MAS)	6	26 ± 11	71 ± 24	61 ± 69	72 ± 54	42 ± 13
Clunia (CLU)	6	27 ±7	58 ± 19	129 ± 70	61 ± 29	56 ± 9
Baetulo (BAE)	5	35 ± 9	116 ± 24	62 ± 21	49 ± 29	39 ± 8
Martyrium de la Alberca (ALB)	4	24 ± 3	76 ± 11	32 ± 20	41 ± 12	50 ± 16
Illici (ILLI)	3	27 ± 4	58 ± 21	222 ± 172	27 ± 6	41 ± 2
Arcobriga (ARC)	2	26 ± 2	85 ± 12	125 ± 28	83 ± 6	35 ± 39
Saguntum (SAG)	2	29 ± 7	139 ±103	70 ± 29	50 ± 1	48 ± 12
Els Villarens (VIL)	2	25 ± 1	95 ± 15	99 ± 3	120 ± 3	81 ± 40
Unknown samples (NAM[a])	2	44 ± 5	97 ± 16	56 ± 24	70 ± 69	47 ± 11
Els Antigons (ANT)	1	21	66	33	50	32
Bouada /BOU)	1	11	77	59	73	35
Dianium (DIA)	1	67	160	37	30	35
Iluro (ILU)	1	40	114	116	120	60
Els Munts (MUN)	1	14	117	137	35	90
Palentia (PAL)	1	46	88	63	74	58
Pintia (PIN)	1	27	130	50	64	56
Salou (SAL)	1	23	80	53	28	37
Toletum (TOL)	1	35	78	54	195	41
Uxama (UXA)	1	24	78	107	53	59
Vic (VIC)	1	26	85	102	123	109
Extreme values						
Minimum value		14	30	10	18	8
Maximum value		108	212	698	195	117
Mean for all samples	85	37 ± 17	90 ± 31	103 ± 93	65 ± 35	54 ± 20
R.S.D.[b] for all samples		50	35	90	54	37

[a] National Archaeological Museum of Madrid.
[b] Relative standard deviations expressed in percentage.

the standard deviations for all these elements considering the origin place; as well as the extreme values, the mean and the R.S.D. for all samples. Data obtained showed global average contents of these elements that decrease in the following order: Pb > Zn > Cr > Ni > Cu.

Fig. 5 represents Box and Whisker plots for minor chemical components in all samples. In these plots, each box encloses the middle 50%, where the median is represented as a horizontal line inside the box. Vertical lines extended from each end of the box (called whiskers) enclose data within 1.5 interquartile ranges. Values than fall beyond whiskers, but within 3 interquartile ranges (*suspect outliers*), are plotted as individuals points (O). Far outside points (*outliers*) are distinguished by a special character (✳).

Taking into account graphic representation for Cu, Zn, Cr and Ni, box and whisker plot for Cu and Ni shows a higher symmetry distribution of data than Zn and Cr, where a slight positive asymmetry is observed. *Outliers* and *suspect outliers* above the upper quartile are observed in all cases except for Ni where only *suspect outliers* are observed. In the case of Zn, *suspect outliers* are also observed below the lower quartile. Distribution dispersion increases in the following order Cu < Ni < Zn < Cr.

Pb representation has a different scale. This element shows a slight positive asymmetry with *suspect outliers* and *outliers* above the upper quartile. Pb concentration is in close relation with the existence of certain lime grout treatments that included lead oxides as additives ("engobium"). So, there are three samples of covering remains, two of them of "engobium remains" with Pb concentrations of 194 and 326 $\mu g\,g^{-1}$ (23TAR and 1ILI), and another covering with about 37 $\mu g\,g^{-1}$ (3ALB) corresponding to other lime grout treatments without lead oxides addition. These data are very interesting since samples from ALB and ILI should have been exported from the production centre of Cartagonova. Consequently, in this centre should have used engobium from two kinds. According to the XRD results, one of them (sample from ALB) is constituted mainly by quartz in a very high proportion. It means that the covering was made with finely powdered sand with a small amount of calcite and plagioclases to allow the adherence, and with a very low level of Pb similar to that found in clays (39 $\mu g\,g^{-1}$). This type of covering was used as a degreasing. By contrast, the other engobium (ILI) seems to be about 50% sand mixed with other materials composed by plagioclases, calcite and dolomite with a remarkable high level of Pb.

Only samples found in places with three or more samples have been taken into account for comparison among different average values in the following comments. Samples coming from CLU, ILI, TAR and TER had a lead content above the average value (103 $\mu g\,g^{-1}$), corresponding the outliers (Fig. 5) to two samples from TAR (one of the production centres). In the same way, antefixes showed in most cases higher Pb contents than tiles and than their respective imbrexes, because tiles were decorative elements and they were subjected to a lime grout treatment containing lead oxide.

The average value of Cu content was 37 $\mu g\,g^{-1}$. Samples coming from TAR and TER showed contents above this value (51 and 48 $\mu g\,g^{-1}$, respectively), and those coming from ALB and MAS below this value (24 and 26 $\mu g\,g^{-1}$, respectively). Analysed clays showed Cu contents about 30 $\mu g\,g^{-1}$. These copper contents were slightly higher in antefixes than in tiles whereas they were very similar in antefixes and their corresponding imbrexes. An exception was the case of two antefixes (35EMP and 59TAR) with Cu concentrations of 86 and 108 $\mu g\,g^{-1}$, probably due to a pigmentation process.

Samples coming from BAE showed the highest Zn average contents (116 $\mu g\,g^{-1}$) in contrast to samples from ILI and CLU with the lowest values (both 58 $\mu g\,g^{-1}$). Samples from BAE showed certain addition of Zn compared to those found in the analysed clays (with an average content of 77 $\mu g\,g^{-1}$), probably due to their use to obtain a white pigmentation. In some cases antefixes showed higher Zn contents than tiles and imbrexes and on the contrary there are tiles and imbrexes with higher Zn contents than antefixes. The latter results can be explained by the use of different clays to make pieces with different function.

Tiles, most antefixes and their corresponding imbrexes and analysed clays, showed similar composition with respect to Cr and Ni (average values of 65 and 54 $\mu g\,g^{-1}$, respectively). However, the samples of the production centres of EMP and TER showed contents of Cr and Ni above these average values (81 and 85 $\mu g\,g^{-1}$, respectively, for Cr and 61 and 77 $\mu g\,g^{-1}$, respectively, for Ni). Samples from ILI showed the lowest average content in Cr and Ni (27 and 41 $\mu g\,g^{-1}$, respectively). Concerning to antefix–imbrex pairs, concentrations of both elements were comparable but in some exceptional cases Cr content in antefix was higher than in its imbrex (sample 1BAE 99 $\mu g\,g^{-1}$ (A) and 40 $\mu g\,g^{-1}$ (I) and sample 1CLU 115 $\mu g\,g^{-1}$ (A) and 50 $\mu g\,g^{-1}$ (I)). Similar behaviour was observed in antefix–imbrex pairs respect to Ni concentrations.

3.3. Chemometric study

Several chemical parameters (concentrations of Cu, Cr, Ni, Pb and Zn) were selected for the multivariate statistical study. They were chosen as indicators of the manufacturing process and of the original materials as well as for showing the largest dispersions of the values among samples. The accumulation of certain elements analysed in the ceramic paste can be explained taking into account two considerations: the original material and the manufacturing process. In this way, there are differences in composition among clays coming from different areas. In addition, certain elements such as Zn, Cu, Cr and Ni were added as mineral components during the manufacture process in order to obtain, for example, an appropriate pigmentation, and Pb was a melting element added to the lime grout.

Linear discriminant analysis was applied to the results of the chemical concentrations of these five elements in the 85 considered samples. This procedure is designed to develop a
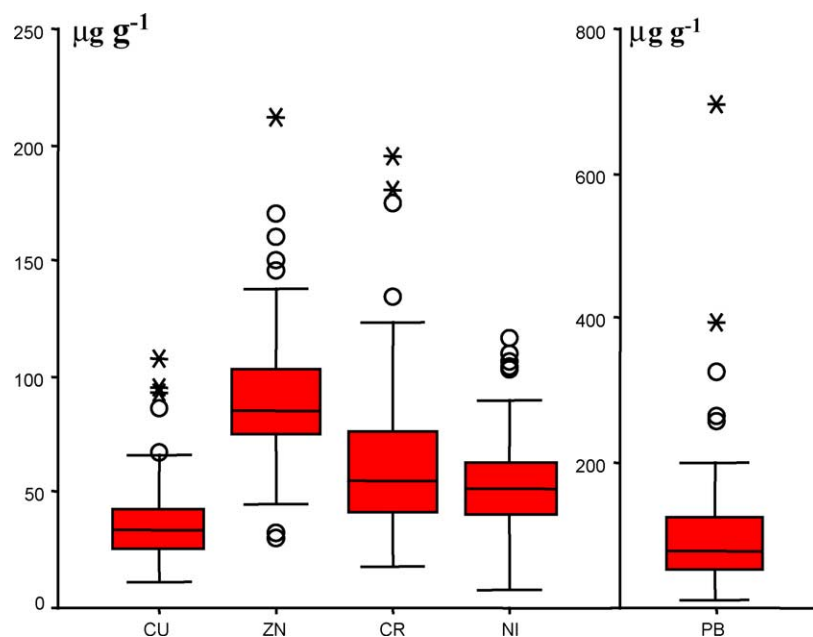
Fig. 5. Box and Whisker plots for minor chemical elements.

set of discriminating functions, which can help classify samples and predict the origin of an unidentified terracotta sample based on the values of other quantitative variables. Eightyfive cases were applied to this study and five predictor variables were entered. Eight different categories were selected according to the places where more than three samples were found, in such a way that each category is well represented. In this sense, 63 cases were used to develop a model in order to discriminate among the eight different categories (locations).

The results of this study are plotted in Fig. 6. This is a representation of the terracotta pieces as a function of two most outstanding canonical discriminant functions, which explain

Fig. 6. Linear discriminant analysis of the archaeological terracotta pieces.

73.3% of the total variance and try to establish a classification of the samples according to the places where they came from. These functions with P-values less than 0.05 are statistically significant at the 95% confidence level. Function 1 is a linear combination of the different variables and the most significant standardized coefficients are -0.643 for Pb and 0.589 for Cr. In case of function 2, they are -0.630 for Zn, 0.613 for Ni and 0.563 for Cu.

The 22 samples not used to develop the model are represented as small simple dots and are considered as non-grouped cases. The samples found in each location are grouped inside an enclosure and are characterized by a centroid, represented by ⬦. This point is the average for each group (unique values in the classification factor field) that uses the discriminant functions.

Some considerations should be taken into account in this point. Two terracotta samples from Mas de Gomandi (MAS) and one from Complutum (COM) were estimated as aberrant patterns (or outliers) and they were not included in their corresponding group. The sample from COM was collected from a Basilica and the two samples from MAS were clays collected in this location, one of them is a high roast clay taken from the inner part of a current kiln and the other is also a local clay but with a different composition from the terracotta pieces.

The 85 samples considered in this study have a widespread provenance (more than 20 different locations), a high variety of sample type (tiles, antefixes, imbrexes, engobium remains and raw materials) and even they are dated in a large period of time (six centuries). This fact makes difficult to establish a good classification and distribution of the samples in well-defined clusters, even when there are not enough samples from several places to develop consistent model and groups.
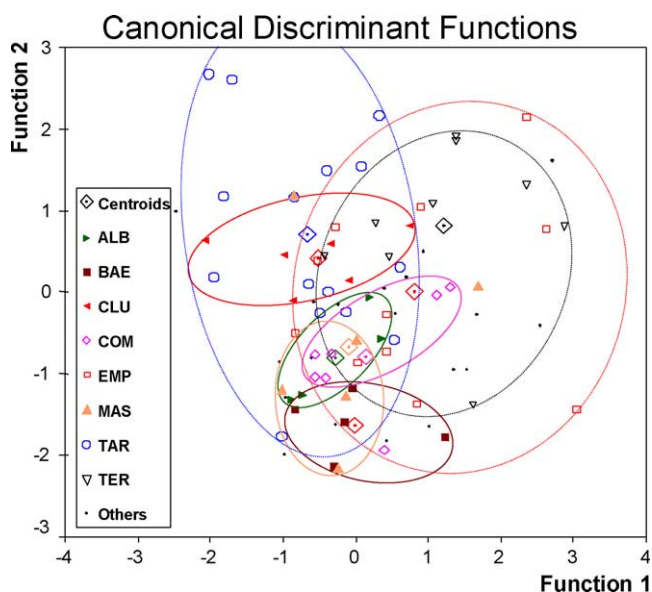
Fig. 6 is the best model. It is not very inter-group discriminant, but some interesting conclusions can be deduced. The groups formed by less than seven samples (ALB, BAE, CLU, COM, MAS) are constituted by pieces with a relatively similar intra-group composition (small circles in the figure). Some of these groups are located in the same area with their centroids very close together and there are some samples in the intersection of several clusters. This indicates that there are not significant evidences of their differences in their composition in spite of they are located in very distant settlements.

There are also three groups, marked with dotted lines, from Emporium (EMP), Termes (TER) and Tarraco (TAR) with a wide dispersion of their samples, TER and EMP in the same area and TAR in a different one, mainly as a consequence of the high Pb content in the TAR samples. These Roman age sites are known as terracotta production centres that exported pieces to other places for roof constructions. Their large variability and dispersion among samples can be attributed to the different treatment and manufacture processes, to the use of different muddy places where raw materials were obtained and also to the fact that the different samples belong to a large period of time that included several centuries.

4. Conclusions

Dilatometry, XRD and SEM results showed baking temperatures of these materials below 900 °C and the existence of zones with very different porosity in the same ceramic piece. Results obtained from multielemental analysis and linear discriminant analysis lead us to the following conclusions: (i) samples can be classified according to the places where these pieces came from, (ii) the high content of Pb found in a large number of antefixes demonstrates the use of lead oxide as an additive in the lime grout treatments, (iii) different contents of Cu, Zn, Cr and Ni were indicative of the use of different clay types in the manufacture process (even in the same production centre) as well as of the existence of a pigmentation process, although this last affirmation is not corroborated by the presence of remains of evident painting in the ceramic pieces and (iv) more dispersion in sample composition was found in Roman age terracotta production centres (Emporium, Termes and Tarraco).

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