Assessing the territorial influence of an Iberian worship site. The chemical characterisation of the terracotta from the Iron Age sanctuary of La Serreta

Abstract
This paper presents the study of the prestigious terracotta votive figurines from the Iberian Iron Age sanctuary of La Serreta (Alicante province, Spain) composed of 174 items. Portable X-ray fluorescence (PXRF) was used to identify elemental markers that permit us to observe the differences between local and non-local terracotta figurines and furthermore to evaluate the geographical influence of the La Serreta sanctuary using Principal Component Analysis (PCA). The Partial Least Squares Discriminant Analysis (PLSDA) statistical method was also used to classify the figurines of uncertain geographical origin. The resulting groups were related to typological and stylistic groups of figurines and the distribution in different sites in the region.

1. Introduction
The Iron Age in the Mediterranean area of the Iberian Peninsula (8th to 1st centuries BC) is characterised by the development of complexity with patron-client forms of social relationship, the emergence of urbanism, and economic development with interregional exchange. The introduction of local Iberian economies into the Mediterranean trade system has been implicated in the development of complex socio-political systems (Ruiz 2008). The Iberian elites enacted several strategies, including patron-client relationships, prestige goods exchange systems, and commensality or competitive feasting (Ruiz and Molinos 1998; Grau Mira 2011). In this social environment the role of sanctuaries was crucial to the aristocratic control of rituals as part of power strategies, mainly during the 4th and 3rd centuries BC (Grau Mira 2016).

The main ritual practice attested in the Iron Age sanctuaries of Iberia is offerings made by worshippers in the presence of the divinity. At the majority of the sanctuaries, especially those in the south-east of the Iberian Peninsula, many objects have been found. They take on a specific form in each place: in the sanctuaries of the Jaén area (Andalusia, southern Iberia) ex-votos in the form bronze figurines were deposited; at El Cigarralejo (Murcia, south-eastern Iberia) the offerings were mainly in the form of small carvings of horses; while at El Cerro de los Santos (Albacete, Spanish Meseta), the main type of ex-voto was a carving of a person representing the offerors (Moneo, 2000). The sanctuary of La Serreta, linked to one of the most important Iberian towns in the Valencia region (eastern Iberia), stands out among these worship sites because the worshippers' offerings were in the form of terracotta statuettes in different styles representing human figures. With approximately 430 pieces, it is one of the largest assemblages of terracotta ex-votos from the Iberian world.

This collection was found extremely fragmented and dispersed in the archaeological strata of the sanctuary excavated in the 1920s on the summit of La Serreta Mountain, near the large Iberian town at the same site (Visedo, 1922a, 1922b, Olcina et al., 1998; Grau Mira, 2002; 2016). The mountain is 1040 m above sea level; it stands out from the surrounding territory and visually dominates a large number of the villages in the area (Figure 1). Its visual presence was doubtless a decisive factor in choosing the site of the town and the Iberian sanctuary, which would have played a fundamental role in the social integration of the Iberian populations through religious ties, as previous archaeological studies have proposed (Grau Mira, 2016).
The excavation of the sanctuary revealed a very diverse assemblage of finds, among which of particular note were the aforementioned terracotta figurines, as well as other objects including Roman lamps, Samian ware, coins, black-glazed ware, Iberian ware, cookware, spindles and metal objects. Some of these assemblages have been studied in detail (Juan 1987-88; Garrigós and Mellado 2004; Lara 2005; Poveda 2005; Horn 2011).

The votive deposit is made up of different offering objects that were buried in pits on the mountaintop (Visedo, 1922a, 1922b). The offerings included the Iberian votive figures analysed here (Figure 2A), Gallic and Hispanic terra sigillata, mainly plates, including forms Drag. 15/17, 18, 24/25 (Poveda 2005), Roman lamps dated to the 2nd and 3rd centuries AD (Lara 2005) and coins dated mainly to the 3rd and 4th centuries AD (Garrigós and Mellado 2004) (Figure 2B).

The finds tell us of a long sequence of occupation from the 3rd century BC to the 5th century AD. In those eight centuries of religious use, the practices carried out at the site would have changed, as evidenced by the wide diversity of finds. Thus, the offerings ranged from terracotta figures between the 3rd and 1st centuries BC, Roman tableware and coins from the 1st to the 3rd centuries AD, and lamps from the 2nd to the 5th century AD. In this paper, we will focus on the terracotta ex-votos and their relationship with the Iron Age rituals.

Typological and stylistic analyses of the terracotta figurines have identified different formal groups that can be linked to a ritual diversity or to changes in the offerings over the three centuries of use during the Iberian period. The variations in the style and manufacture of the figurines may also be due to their different origins. Based on formal analyses, it has been proposed that there were different groups that included schematic figures, realistic masks, realistic female figures, realistic male figures, representations of groups of persons and incense burners (Juan 1987-88; Horn, 2011, 153). It has traditionally been assumed that the majority of these figurines are from the same period—the 3rd and 2nd centuries—and of local origin. Unfortunately, the fieldwork carried out during the 1920s did not provide a detailed enough stratigraphy to be able to resolve the question (Visedo, 1922a).

In our opinion, doubt should be cast on this local origin in the case of certain figurines made with fabrics that, from a macroscopic point of view, are very different to the majority of those considered to be part of the local assemblage.

With the aim of investigating the possible variation in the origin of the pieces and to attest the territorial influence of the worship site in the Iberian period, a compositional analysis was carried out and the results were compared to those from an archaeological analysis focusing on the typological and stylistic approach and possible parallels for the pieces. The studied assemblage consisted of 174 terracotta pieces. They included different types that we have initially grouped according to formal and stylistic criteria and the characteristics of the ceramic pastes as seen in visual macroscopic observations. The groups will be defined below.

This study was carried out with a portable X-ray Fluorescence (PXRF) device, thus avoiding the transportation and destructive sampling of the prestigious figurines of the La Serreta collection. Portable X-ray fluorescence results were used to identify elemental markers that allowed us to observe differences between local and non-local terracotta figurines and furthermore to evaluate the geographical influence of La Serreta sanctuary using Principal Component Analysis (PCA). Finally, the Partial Least Squares Discriminant Analysis (PLS-DA) statistical method was used to classify figurines of uncertain origins.

2. Materials and Methods

2.1. Samples
Samples were directly measured by PXRF without undergoing any pre-treatment process. Each sample consisted of one piece of pottery and three surface readings were taken. Any possible dirt or dust on the surface was cleaned and contamination problems related to the measured surface were avoided. Furthermore, chemical elements that suffered interference related to the analytical method and the matrix analysed were not taken into consideration.

The studied assemblage consisted of 174 terracotta pieces. They included different types that we have initially grouped according to formal and stylistic criteria and the characteristics of the pottery fabric as seen in visual macroscopic observations. The groups are as follows (Table 1):

Local 1: Tableware produced in local potteries. Although they are not actually offering figurines, we have taken as examples of the clays used locally an assemblage of tableware and storage and transportation vessels manufactured in the territory and used in the town of La Serreta (Grau Mira, 1998-99). The pieces are made with a very refined, fine clay with barely any visible temper and a floury feel that stains when touched. They have mainly orangey colourings, ranging from a pinkish chestnut to a reddish orange. Specifically, there are 20 pieces that account for 10% of the assemblage.

Non-Local 2: To characterise the pieces of non-local origin, we selected a set of figurines whose ceramic pastes are completely different to those produced locally. This is a rough, hard fabric with small visible non-plastic inclusions, sometimes with particles of limestone. The colourings are mainly yellowish chestnut. All the pieces in this non-local group are Guardamar-type incense burners (Abad Casal, 1992), so-called because they are frequently found at the archaeological site of that name in the south of Alicante province, some 70 km away. We were very restrictive in the selection of the pieces of non-local origin, selecting those with fabrics that differed most from the local ones, limiting this assemblage to 8 pieces.

Probably-Local 3: This group consists of a large assemblage of figurines made with very similar fabrics to those of the local group, although there are some variations in the fabrics between the pieces. The main characteristic is that the human faces of these figurines were mass produced using only eight different moulds that were all very similar (Horn 2011: 164). From this feature, we deduce that the series was probably local. It is a large assemblage of 72 pieces.

Unknown 4: This large group was put together with pieces that do not belong to any of the above categories and that could be of local or foreign origin. The colourings and textures of the fabric are heterogeneous and range from yellowish to orange. Among the stylistic types, we find some Guardamar-type incense burners that are probably non-local and other Hellenistic-style incense burners le shape of a female head that also do not appear to be of local origin, judging by their fabrics. Alongside these, there are schematic pieces manufactured with a clay very similar to the local one, but that have completely different schematic features to those made with moulds. There are 74 pieces in this group.

2.2. PXRF analysis

X-ray fluorescence spectra were obtained directly using a portable S1 model Titan energy dispersive X-ray fluorescence spectrometer from Bruker (Kennewick, Washington, USA) equipped with a Rhodium X-ray tube and an X-Flash® SDD detector. For instrument control, the S1RemoteCtrl (Geochem-trace programme) and S1Sync software from Bruker was used to measure percentages of Al₂O₃, SiO₂, K₂O, CaO, Ti, Fe, Mn, Sr, Ni, Zn, Y, Zr. For spectra treatment, ARTAX software from Bruker was also used. The certified material, soil NIM GBW07408 (see Table 2) was used as a standard reference for evaluating the accuracy of the data obtained with the method used (Gallello et al., 2015).
2.3. Data analysis

For statistical analysis, twelve variables (Al₂O₃, SiO₂, K₂O, CaO, Ti, Fe, Mn, Sr, Ni, Zn, Y, Zr) were used for modelling.

Principal Component Analysis (PCA) was used to explore the spectra dataset, reducing the number of variables and providing a deeper insight into the structure of the variance of the dataset. Mean centring of spectra and autoscale were used as a pre-processing step prior to modelling, and cross validation (CV) was carried out using leave and out. PXRF elemental results of Al₂O₃, SiO₂, K₂O, CaO, Ti, Fe, Mn, Sr, Ni, Zn, Y and Zr were used to obtain the model.

Partial Least Squares Discriminant Analysis (PLSDA) is a frequently used classification method (Gallello et al. 2013). Here, it was applied to differentiate between local (LOCAL1) and non-local (NON-LOCAL3) pottery. To build the calibration model, fifteen samples classified as local pottery and six non-local samples were selected, resulting in a calibration set with twenty-one samples and twelve variables. The remaining local (five) and non-local (two) pottery samples were used as an external validation set with a total of seven samples and twelve variables for evaluating the model performance. Seventy-two probably local (PROBABLY LOCAL2) and seventy-four unknown (UNKNOWN) samples were included in the test set to predict their class, resulting in two matrices made up of seventy-two and seventy-four samples and twelve variables. Pre-processing (mean centre and autoscale) and cross validation (CV) with “leave one out” were used.

Data analysis was carried out using the PLS Toolbox 6.5 from Eigenvector Research Inc. (Wenatchee, WA, USA) running in Matlab R2014b from Mathworks Inc. (Natick, MA, USA).

3. Results and Discussion

3.1. XRF analysis of the La Serreta terracottas

As described above, all 174 samples were measured by PXRF (see supplementary material table). Table 3 summarises the mean concentration values expressed in percentages of Al₂O₃, SiO₂, K₂O, CaO, Ti, Mn, Fe, Sr, Ni, Zn, Y, Zr. SiO₂ is the main component of all classes with mean values that vary from 32% to 51%, followed by Al₂O₃ at levels from 12% to 15%. CaO levels range from 5.2% to 9.8%. K₂O is present at percentage levels from 2.1% to 2.3% also with low levels of Ti, Mn, Fe, Sr, Ni, Zn, Y, Zr. It would be possible to partially complete the remaining percentage if the CO₂ contents were analysed. Unfortunately, destructive analysis cannot be carried out on the studied objects.

Mean concentrations of data obtained from each class with standard deviation of average values confirm that the elemental profile results make it possible to differentiate local samples from non-local samples, especially by their SiO₂ elemental composition. Therefore, in case of this compound, the obtained values for the LOCAL1 and NON-LOCAL 3 categories are not statistically comparable, at a probability level of 95%.

3.2. Revealing pottery distribution applying PCA

Figure 3 shows the results obtained from PCA, which was used to gain a deeper insight into the complex dataset. The first principal component (PC1) contains the main part of the variance in the data (33.68%), followed by PC2 (15.56) and PC3 (14.25). Scores plots represent data points (i.e. samples) projected into the calculated PC space. They are
常常用于数据探索，距离样本之间的距离可以与它们的相似性相关。从图3a的得分图中，可以明显地看到本地瓷器（LOCAL1）与非本地（NON-LOCAL3）的区别。一个更为显著的发现是，空间顺序在分数图上清晰可见。可以观察到在PC1中元素对的直接贡献及其方向。正如上面解释的那样，PC1包含有用的信息，用于区分每个变量（即元素）对PC1的贡献，属于该组的样本。

**3.3. Classification of Probably-Local and Unknown groups**

对古代陶器的研究可以提供关于原材料来源的有趣数据。此外，存在一个关于陶器元素组成和对象来源之间的相关性。然而，由于历史因素，无法将某一特定组别与某特定对象精确对应。为解决这一问题，使用了主成分分析（PCA）来分类本地（manufactured nearby the sanctuary）和非本地陶器。分类集的建立，并不包括未知组别（except for one sample that is more similar to the NON-LOCAL3 class）。大型本地 ceramics是由于在样本分数图中变量之间的差异所导致的。在图3b的荷载图中，每个变量的荷载值可以通过绝对强度来衡量。同样，PCA也对PLSDA模型的统计意义进行了验证。如有一个样本更接近于本地瓷器的分布，本地瓷器的CaO水平明显低于本地瓷器，而非本地瓷器。此外，CaO水平在本地瓷器中高于非本地瓷器。同样，Fe，Y和Zr浓度在本地瓷器中更高。

**3.4. Evaluation of the geographical influence of La Serreta sanctuary**

LOCAL1和NON-LOCAL3在它们的元素组成方面有明显的差异。LOCAL1组的样品具有SiO2，Ti和其他 trace elements的更高含量，这与本地瓷器的黏土起源有关，这与以前的假设基于风格研究一致。
The results obtained show that the PROBABLY-LOCAL2 group is linked to productions manufactured in the area. Some of the variations in composition could be due to the use of clays that, although they come from the same region, are not from exactly the same quarry, which would incorporate a margin of variation into the results from the samples (Figure 5).

From a strictly archaeological point of view, it is possible to hypothesise that these pieces were produced and used exclusively in La Serreta and its territory. First, it should be pointed out that at none of the other sites in Alicante province where terracottas have been found are there any that correspond typologically to the figurines made with moulds that make up this group (Horn, 2011). Secondly, this same type of figurine is found in the 3rd-century-BC levels of the Iberian town. For example, one of the terracotta pieces identical to those of this group was found in the entrance fortification (Llobregat et al., 1995, Plate 7). In our opinion, the fact that this type of piece is only found at La Serreta and not in other towns is the archaeological confirmation that they are probably of local origin and coincide with the most active period of the shrine in the 3rd century BC.

The NON-LOCAL3 group can be linked to some of the pieces from the unknown group, such as 744, 747, 752, 754, 759, 777, 781, 783, 923, 948 and 977. These pieces correspond typologically to the Guardamar-type incense burners and therefore have the same shapes as those of the non-local group (Figure 6). Initially they were not included in the NON-LOCAL3 group, as their pastes were not clearly different. However, during this study a similarity has been observed both in the style and elemental composition of the fabrics that allows their inclusion in this group.

From the point of view of the archaeological contexts of the finds, it is important to indicate that this type of incense burner, unlike the other types of terracotta figurines, is often found in other parts of south-eastern Iberia (Figure 7), suggesting they may have been produced in places other than La Serreta. For example, they are frequently found at El Castillo de Guardamar (Alicante province), together with other finds from the 3rd-1st centuries BC, which suggests there was a place of worship at that site (Abad, 1992, 233-234). Some very fragmented examples have been found in the Late Iberian levels of L’Alcúdia d’Elx (Alicante province) (Moratalla and Verdú 2007: 344, Fig. 2.2). A mould for this type of piece was found in the excavations of El Tossal de les Basses in L’Albufereta (Alicante province) (Rosser 2007). Examples were also found during the excavations at the Iberian sanctuary of La Malladeta in La Vila Joiosa (Alicante province) (Rouillard et al. 2014: 51-53 and 164). Finally, some examples have been found at El Tossal de la Cala (Finestrat, Alicante province) (Sala and Verdú, 2014). All these sites, most of which are on the coast, suggest a possible regional production that would have reached La Serreta from the neighbouring territories.

Regarding the UNKNOWN4 group, we have been able to distinguish a subgroup consisting of incense burners (Figure 8) with a different style to the aforementioned Guardamar-type. They are very similar to the previous group and, looking at the analytical results, they should be considered non-local. These pieces, whose prototypes tend to come from the Greek and Punic world, were quickly imitated in the Iberian territory, where they were very widely distributed to numerous settlements (Horn, 2011).

Finally, we would like to point out that most of the pieces in the UNKNOWN4 group are figurines with schematic-type representations (Figure 9). They are very simply-made with manual fashioning of the fresh clay and the application of details such eyes or embellishments. The pastes of these pieces are very similar to those of the probably-local group and we believe they are local productions. The fact that pieces of this type are found in the ancient town’s dwellings —such as the example from Sector F (Grau Mira, 1996, Fig. 19)— or in the entrance fortification (Llobregat et al., 1995, Plate 7) would appear to support the hypothesis that these pieces were manufactured and used locally.
In summary, we have pieces from diverse origins that open up an interesting panorama for interpreting the territorial influence of the Iberian sanctuaries. In this way, we can indicate that the terracotta pieces left as offerings at La Serreta correspond to two different circuits.

The first circuit would be represented by the terracotta pieces depicting the worshippers, the so-called "PROBABLY-LOCAL2" group and an "UNKNOWN4" subgroup of figurines with schematic representations of people that correspond to offerings from the territory of the Iberian town of La Serreta and the Alcoi Valley (Grau Mira, 2002). The link between the towns and villages and the worship site is thus demonstrated and can be related to the need to create kinship ties between the local populations in order to build a sense of community (Grau Mira, 2016).

The second circuit would be represented by the pieces that can be clearly associated with the NON-LOCAL3 group, which includes some from the Unknown subgroup with the same features. In general, they are incense burners, mainly of the so-called Guardamar-type, together with others of different characteristics. These pieces are frequently found in other settlements in the region from where they possibly came, although we have yet to discover the exact place of production. We can currently indicate two possibilities. The first is the sanctuary of Guardamar, from which we have the largest assemblage of these pieces, more than fifty examples, that give the name to the specific type. The second option is La Albufereta in Alicante from where we have a mould used to manufacture this type of piece, although unfortunately it was found out of context and not associated with a production area.

Moreover, the data presented allow us to corroborate the circulation and patterns of trade between Iberian territories, a line of research that has barely begun to be developed (Mata et al., 2000).

4. Conclusions

The extensive group of terracotta figurines from La Serreta is a reference collection for the study of the artistic manifestations in clay from the Iberian world. Due to this importance, it has attracted the attention of researchers who have studied it from the stylistic and typological perspectives. We can now add to this with the possibilities offered by archaeometric analysis.

The chronology attributed to the collection is between the 3rd and the 1st centuries BC; in other words, a time span of approximately 250 years, which suggests that the assemblage is made up of pieces manufactured in different periods and at different places. That supposition has been confirmed by analysing the mineralogical composition and comparing the results to the archaeological information based on the formal types and the contexts of the finds of similar pieces, both in the settlement of La Serreta itself and at other Iberian sites in the region.

The existence of two clearly established groups —one local and another of foreign origin— is evidence of the influence of the sanctuary of La Serreta in its own territory (Grau Mira, 2016), but it also expands the scale of interaction to a larger regional area from which the pieces would have come. This opens up opportunities for interpreting new possibilities that need to be explored, such as whether we are dealing only with the circulation of terracotta pieces or whether we are looking at offerors who arrived from other parts, bringing the pieces with them.

PXRF analysis has been confirmed as a suitable technique for determining the elemental composition of the La Serreta figurines collection and also as a safe method to ensure the preservation of the objects. PCA has shown that the elemental profile of local
and non-local pottery is different, due to the higher SiO₂, Al₂O₃, K₂O, Ti, Fe, Y and Zr contents in the local ones.

The PLSDA results confirmed a major enrichment of SiO₂ and Ti and other trace elements in local pottery. Furthermore, unknown and probably local figurines were classified as local or non-local. Therefore, our proposed classification method proved to be very useful in identifying the regional origins of an uncertain pottery assemblage.

This study confirms that the reconstruction of a regional circuit for these objects is feasible using stylistic, morphological and elemental analyses. The data show that there is clear evidence of ritual practices shared within the communities of different territories in south-eastern Iberia. The offering of identical objects that circulated and were deposited in diverse sanctuaries opens new prospects for the interpretation of the worship sites and their role in building regional identity.

Acknowledgements

The authors would like to thank the Camil Visedo Archaeological Museum of Alcoi, particularly its director J. M. Segura Martí, for providing access to the La Serreta terracotta collection and for the permission to use the photographs that illustrate this paper. Ignasi Grau acknowledges the financial support of the Ministerio de Economía y Competitividad (Project HAR2015-64601-C3-2-R). Gianni Gallello acknowledges the financial support of the Generalitat Valenciana (PROMETEO project II/2014/077), the Ministerio de Economía y Competitividad-Feder (Project CTQ 2014-52841-P and Project CTQ 2012-38635) and the European Commission (Project H2020-MSCA-IF-2015 704709-MATRIX).

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### Table 1

Description of the samples from La Serreta.

<table>
<thead>
<tr>
<th>Group</th>
<th>Individuals</th>
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<td>Local</td>
<td>1431, 2344, 2601, 2613, 15,648, 16,510, 16,723, A1, A2, A3, A4, A5, A6, A8, A9, A10, A11, A12, A13, A14</td>
</tr>
<tr>
<td>Non-local</td>
<td>757, 781, 783, 787, 790, 791, 987, 995</td>
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</tbody>
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Table 1. Description of the samples from La Serreta.

### Table 2

XRF analysis accuracy check.

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<tr>
<th>Element</th>
<th>Obt. value</th>
<th>Cert. value</th>
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<tr>
<td>AI2O3</td>
<td>11.86 ± 0.49</td>
<td>11.92 ± 0.15</td>
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<tr>
<td>SiO2</td>
<td>58.2 ± 1.7</td>
<td>58.61 ± 0.13</td>
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<td>K2O</td>
<td>2.284 ± 0.014</td>
<td>2.42 ± 0.04</td>
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<td>CaO</td>
<td>7.83 ± 0.09</td>
<td>8.27 ± 0.12</td>
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<td>Ti</td>
<td>0.3829 ± 0.0010</td>
<td>0.390 ± 0.012</td>
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<tr>
<td>Fe</td>
<td>4.07 ± 0.002</td>
<td>4.08 ± 0.07</td>
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<tr>
<td>Mn</td>
<td>0.0692 ± 0.00007</td>
<td>0.065 ± 0.002</td>
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<tr>
<td>Sr</td>
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<tr>
<td>Zn</td>
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<tr>
<td>Y</td>
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<tr>
<td>Zr</td>
<td>0.0220 ± 0.0006</td>
<td>0.0229 ± 0.0012</td>
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</tbody>
</table>

Values expressed as percentages (%) and ppm for trace elements. CRM (GBW07408) obtained values (Obt. value) and certified values (Cert. value) of the analysed elements.

Table 2. XRF analysis accuracy check

Note: Values expressed as percentages (%) and ppm for trace elements. CRM (GBW07408) obtained values (Obt. Value) and certified values (Cert. Value) of the analysed elements.
Table 3. Chemical composition of samples from La Serreta by XRF (ID)

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<th>Class</th>
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<th>Mn</th>
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<td>0.0015</td>
<td>0.001</td>
<td>0.002</td>
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<td>Mean</td>
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Note: Concentration of element mean values in the studied classes and standard deviation (SD) expressed as a percentage (%) and ppm for trace elements.

Figure 1. Map of the La Serreta area showing the locations of the Iberian Iron Age sanctuary (marked by the star) and the Iberian settlements: dark points are hillfort sites; light points are rural sites. The dark square marks the modern town of Alcoi.
Figure 2. Archaeological items recorded at La Serreta sanctuary. A: The Iberian terracotta figurines studied here; B) Roman items: oil lamps, coins and Samian ware.
Figure 3. PCA results. Study of the La Serreta samples XRF elemental scores (a) and loadings (b) of PC1, PC2 and PC3.
Figure 4. PLSDA results. Predicted class membership of samples contained in the calibration “Cal. set” and validation “Val. set” set (a) and VIP scores of the PLSDA model (b). Predicted class membership of samples contained in the calibration test “Cal. test” and test set “UNKNOWN4” (c). Predicted class membership of samples contained in the calibration test and test set “PROBABLY LOCAL2” (d).

Figure 5. Examples of probably-local figurines.
Figure 6. Examples of non-local and unknown group figurines.
Figure 7. Map of south-eastern Iberia with the sites at which Guardamar-type burners are found (the non-local group of figurines in our analysis)

Figure 8. Examples of analysed unknown group figurines

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