Applied Physics A

Combining chemical data with GIS and PCA to investigate Phoenician-Punic Cu-metallurgy

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Abstract:	This study focuses on the development and application of an innovative protocol which combines chemical data, GIS (Geographical Information System) and PCA (Principal Component Analysis), involving numeric (chemical composition) and categorical (typology of object, archaeological context, chronology and geographical areas) variables, as a simple tool to help in the visualization and interpretation of large multidisciplinary datasets on Cu-based alloy archaeological artefacts of the Phoenician-Punic metallurgy. The protocol is an useful tool for highlighting existing connections between specific alloy chemical compositions, the location of the original settlement where the artefact had been produced and the proximity to mining resources, waterways, and allochtonous presence such as, in the specific case of this study, the Phoenician and Punic influence in the Iberian bronze production during the Late Bronze Age- Iron Age. The protocol was tested successfully in a case study concerning the precise dating and provenance of bronze statuettes of unknown age and provenance from the Evora Museum collection in Southern Portugal where it confirmed and further refined earlier hypotheses based solely on archaeological and/or chemical studies. The results were interpreted with a unique perspective, to validate the GIS system in combination with experimental chemical-physical data to yield the identification of metallurgical sites of bronze production.
Response to Reviewers:	Ref.: Ms. No APYA –D-13 00940R1 - Combining chemical data with GIS and PCA to investigate Phoenician-Punic Cu-metallurgy List of changes according to the referees recommendations

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Figure

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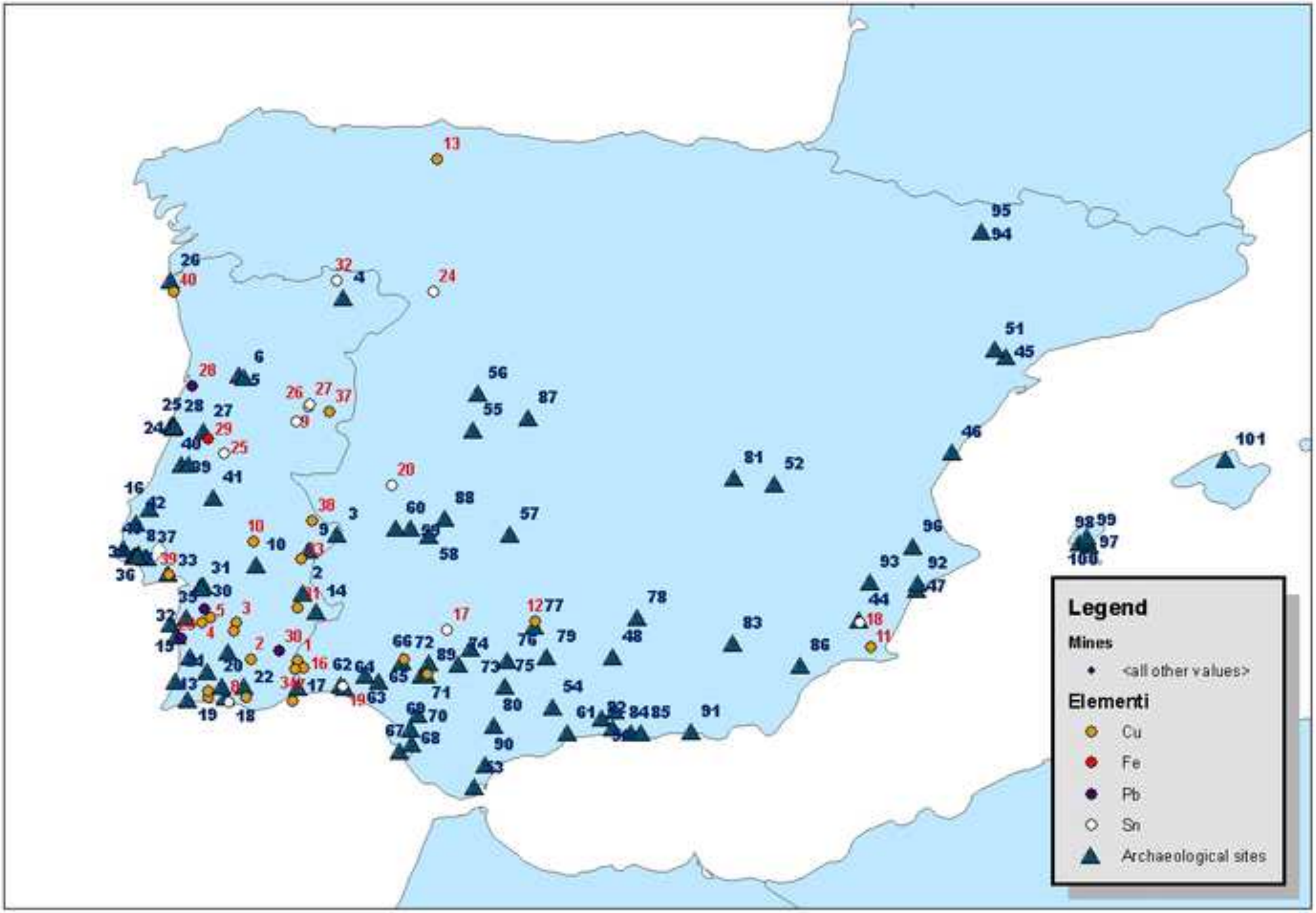

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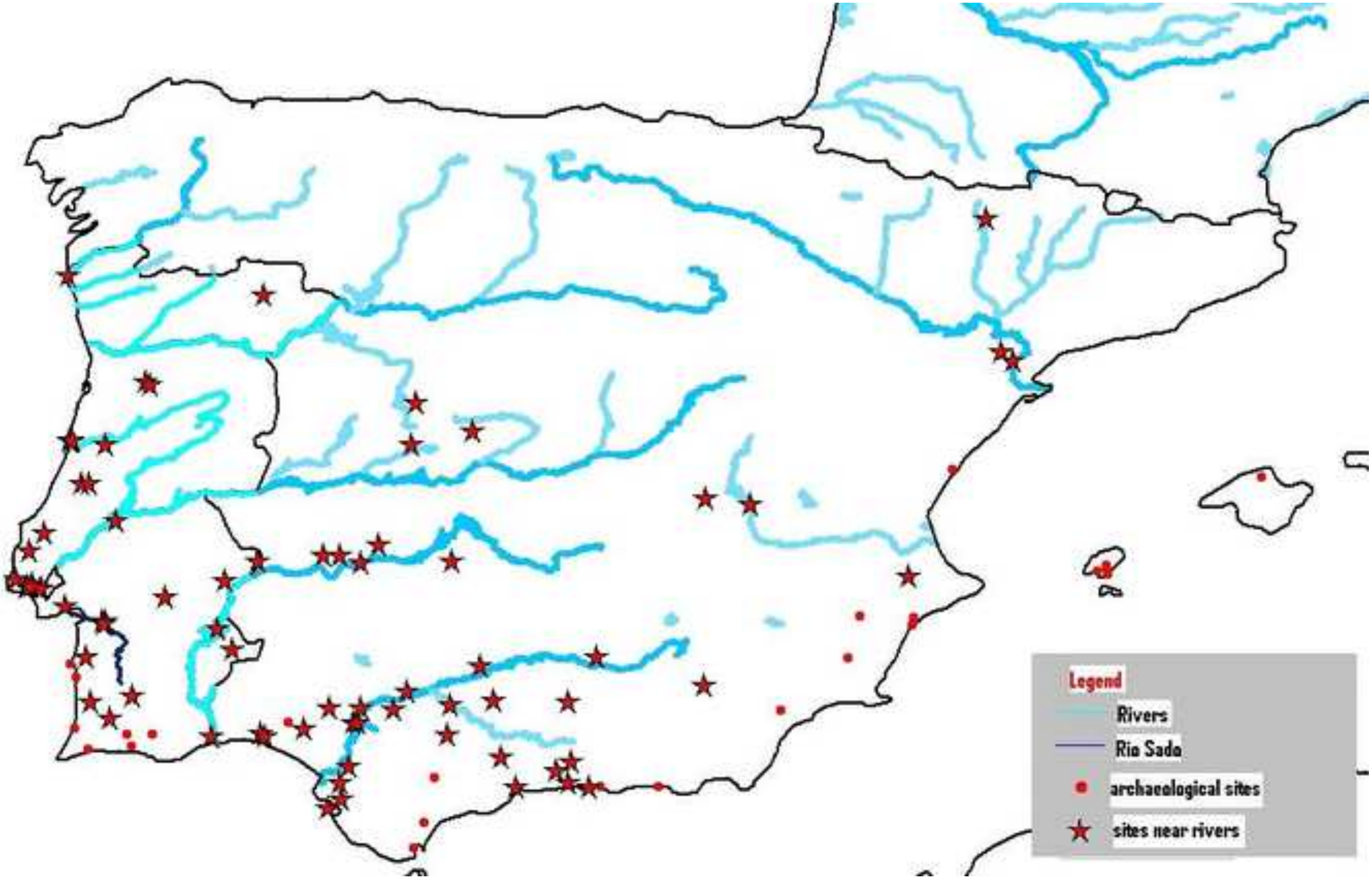

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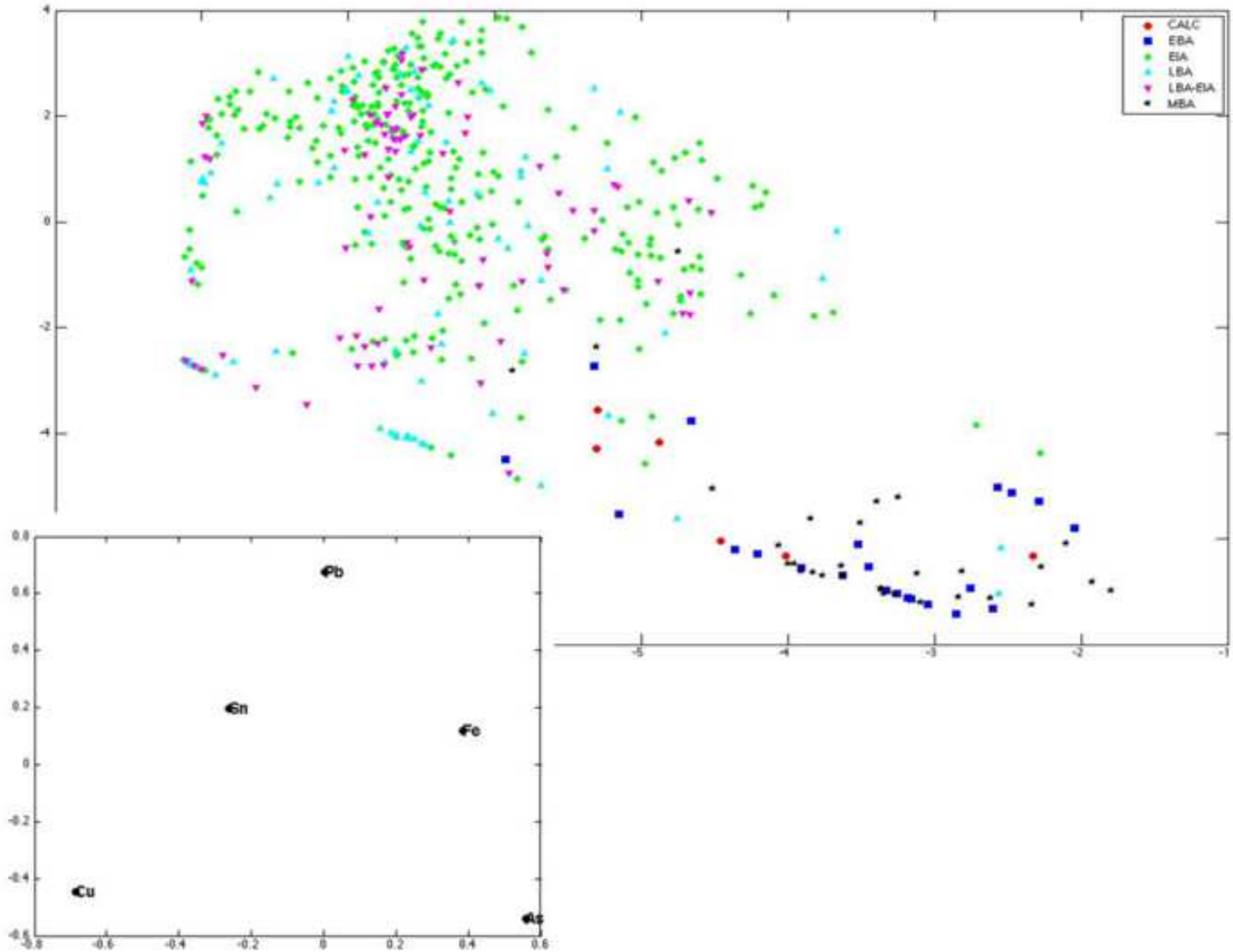

Figure 6

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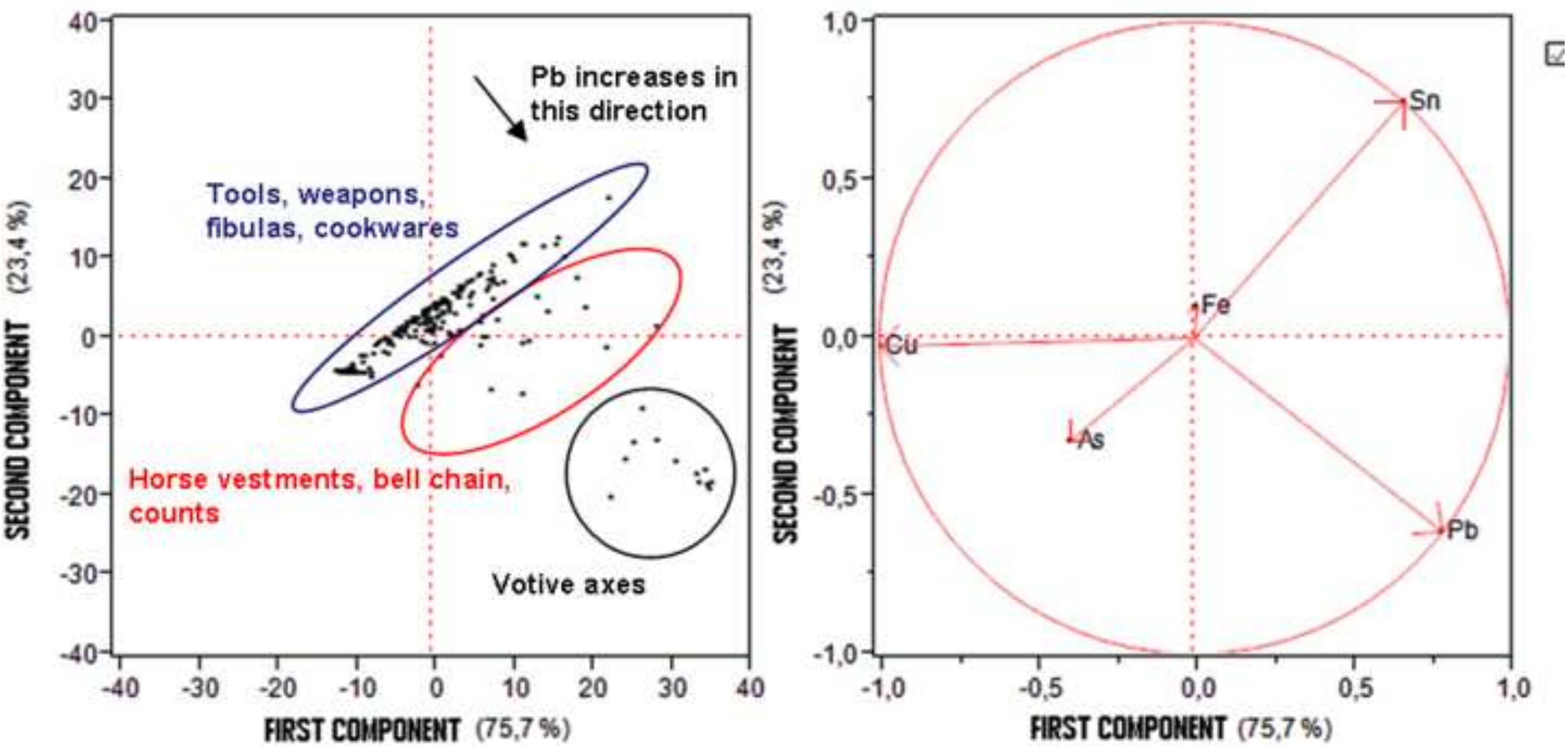

Figure 7

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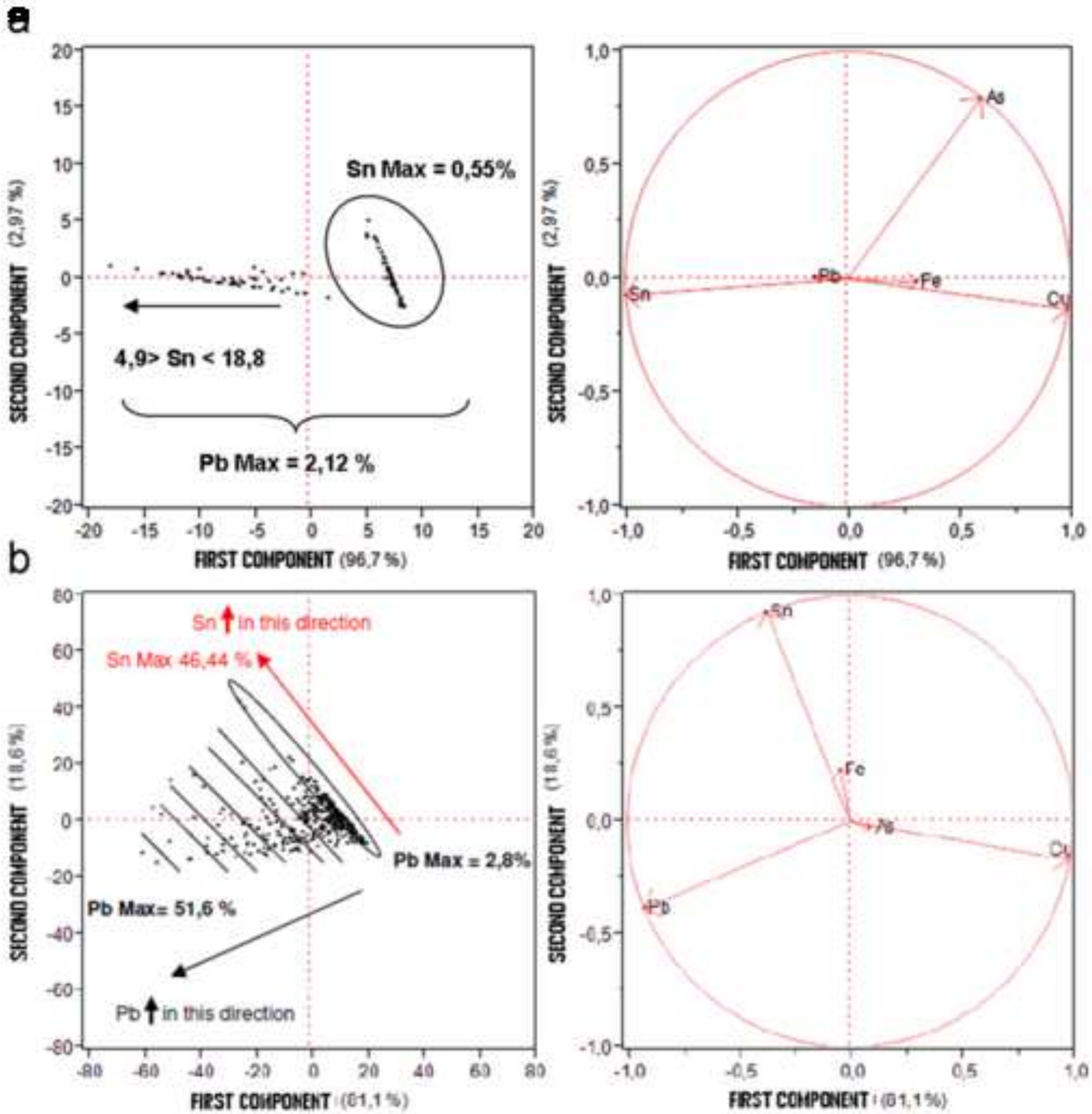

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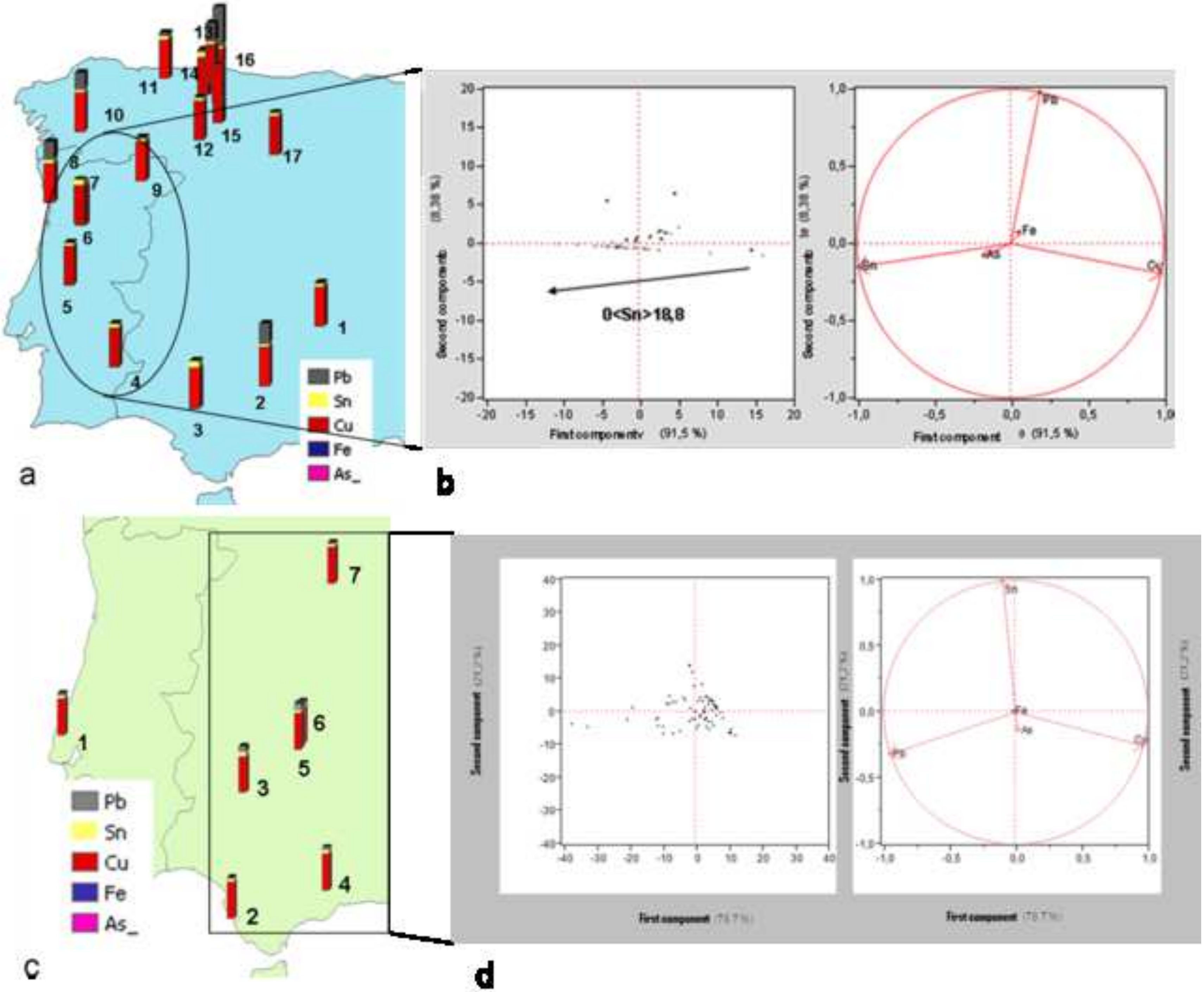

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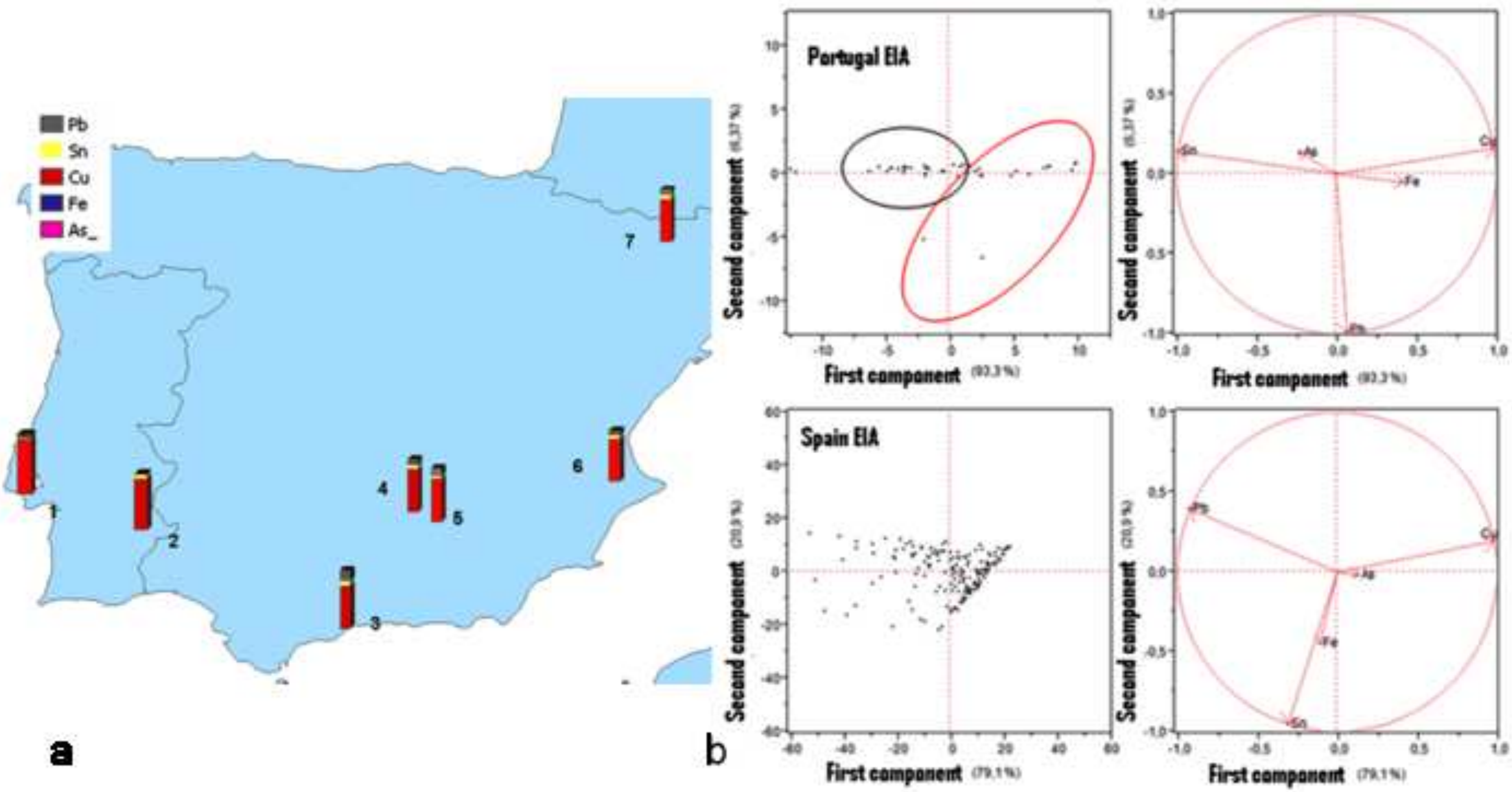

Figure 10

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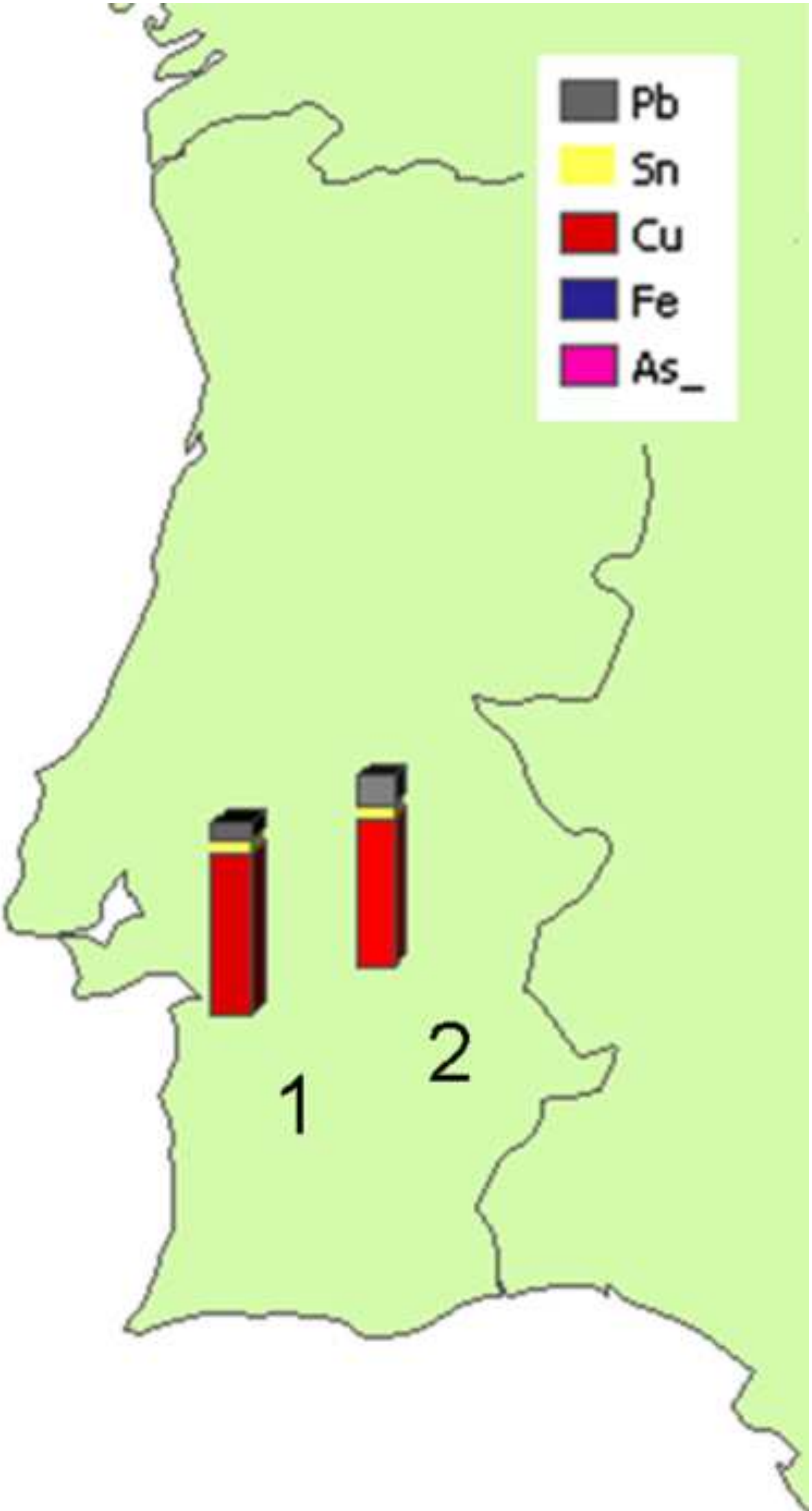

Figure 1
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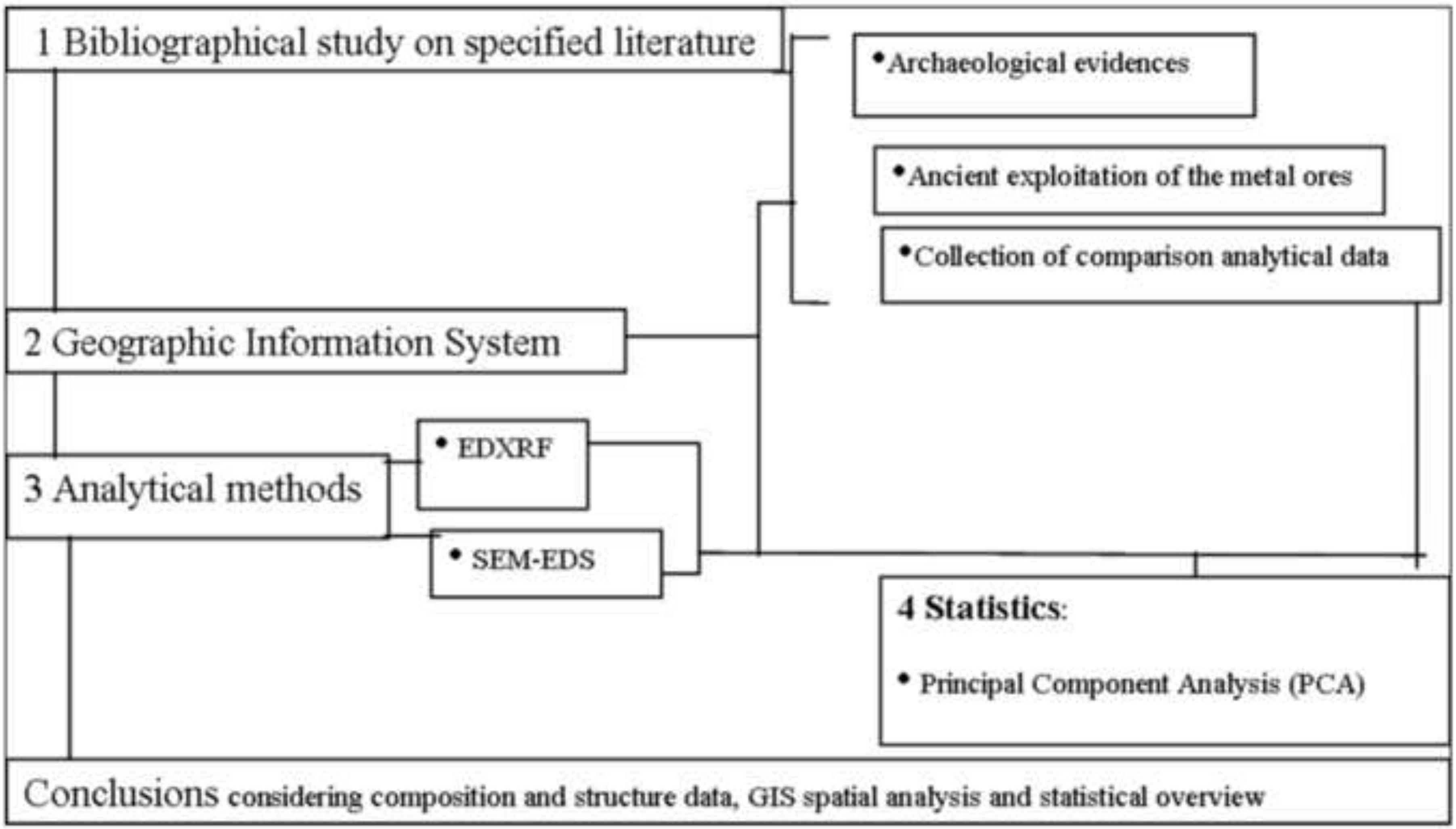

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Combining chemical data with GIS and PCA to investigate Phoenician-Punic Cu-metallurgy

Keywords: GIS, SEM-EDS, EDXRF, PCA, bronze artifacts, Phoenician-Punic

1. Introduction

It is well-known that Phoenicians and later Punics established several colonies in the Mediterranean Basin and in many Atlantic areas [1] acting as innovators and dispensers of technical skills and know-how during the contacts with local cultures, resulting in the production of a wealth of artifacts made up of different archaeomaterials [1]. The production of Cu-based alloy artifacts in particular flourished between the Bronze Age and the Iron Age, reaching a climax during Early - Middle Iron Age (MIA-EIA). From an archaeological point of view, distinguishing between objects of different origins, i.e. Phoenician, locally produced and/or resulting from the mixing up of both elements, is of particular relevance and cannot be achieved using solely “archaeological” tools. The term “orientalising” [2] is here used to identify the mixing production showing local and allochthonous features. In this meaning the term then includes also artifacts belonging to a wider chronological period respect to the usual definition. An integrated approach combining archaeological evidence, analytical data acquisition, statistical processing and spatial analysis tools is therefore needed to achieve more objective conclusions.

As part of a larger, multidisciplinary project on Phoenician and Punic influence on the Copper-archaeometallurgy of the Mediterranean Basin and surrounding regions, this paper focuses on the development and application of an innovative protocol based on PCA (Principal Component Analysis) involving numeric and non numeric variables and GIS (Geographical Information System) as a simple aid to the interpretation of chemical-analytical data of Cu-based alloy artifacts. The protocol is a useful tool for highlighting existing connections between specific alloy chemical compositions, the provenance settlements of the artefact, has been produced and the proximity to mining resources, waterways and allochtonous presence such as the Phoenician/Punic influence in the Iberian bronze technology.

Either the chronological range, Late Bronze Age (LBA) and Iron Age, and a narrower period Early - Middle Iron Age (EIA-MIA), have been selected because of the large amount of “orientalising” artifacts produced during this historical timespan.

The application of GIS [3] has already proved to be a very useful tool especially by considering it as an adjunct to PCA statistical method. The procedure has been applied for the treatment of already published data, available in the literature on bronze archaeo-metallurgy in the Iberian Peninsula [4-28], combined with additional data obtained by EDXRF and SEM+EDS on Phoenician bronze artifacts representing anthropomorphic and zoomorphic figures from Museum collections: Evora (unknown origin), Alcacer do Sal (archaeological provenance). Evora artifacts have been dated to the Iron Age (V century BC), but their precise chronological collocation, geographical provenance, and typology remained uncertain. A previous study [29] put forward the hypothesis of a common origin from the Phoenician site of Alcacer do Sal [30], for all statuettes. Alcacer do Sal (VII century BC) is considered Phoenician from two points of view: the settlement features and the evidence of religious cults connected to Baal and Astarte. Dr. Esmeralda Gomes (personal communication, master thesis) and Prof. Ana Margarida Arruda, were the first to note this similarity and the proximity of the two sites and to assume a common provenance. One aim of the present study is to apply the new multidisciplinary protocol to confirm the hypothesis, by comparing the analytical data from Portuguese items with a chronologically and geographically restrained extended dataset encompassing Phoenician sites from the Iberian Peninsula.

The Portuguese case studies and the Iberian published database points have been selected as a function of: a) the relevance of their strategic geographical positions; b) the presence of important copper, tin and lead mining resources in the surrounding region.

The basic steps of the protocol can be summarized as follows:

1) chemical and physical analysis on local and “orientalising” artifacts and comparison data collection from literature [4-28];

2) creation of databases including archaeological sites and location of ancient mines and ores [31-33];

3) development of GIS thematic maps [3] on the following parameters: a) location of Phoenician/Punic settlements; b) location of ancient mines exploited during the selected timespan; c) location of bronze findings; d) compositional data visualised in connection with geographical positioning;

4) PCA statistical treatment [34], to obtain a representation of Phoenician-Punic bronze production in the Mediterranean basin and neighbouring areas.

2. Methods

The analytical protocol required a flexible formulation with the adoption of a multidisciplinary approach combining archaeology (i.e. the geographical collocation of archaeological settlements), archaeometry (interpretation of bronze chemical compositions), geology (localization and characterization of ancient mines).

The detailed protocol flow-diagram, shown in fig.1 can be described in a sequence of steps.

2.1 Bibliographical study for the elaboration of different databases

Following a thorough bibliographical study, three databases have been compiled as Excel worksheets based on the following data: a) location of archaeological Phoenician settlements; b) chemical data from ancient mines focusing on bronze major elements: Cu, Sn, Pb; c) literature chemical data on bronze archaeological specimens from the same historical period.

Each database was fed with the geographical coordinates of the considered sites for the acquisition in GIS environment.

2.2 Geographic Information System (GIS)

The three different databases have been geo-referenced through the use of GIS to obtain, after conversion in geodatabases, thematic maps of archaeological settlements, mines, waterways and evidence of bronze artifacts. The integration of the databases with GIS let to perform an extensive analysis of the spatial phenomenon under study, by using the various searching options the system offers such as: visual information on the distribution, frequencies and relation between different features under study, database searching tool to create a subset of selected features, using the statistic and editing tools to display on the maps also compositional information or to digitalize geographic features like rivers or paths), georeferencing raster cartography and, finally, creating thematic maps.

2.3 Chemical analysis

The chemical database from published data is based on analyses carried out using a variety of techniques (EDXRF, µ-XRF, AAS). The alloys composition of selected bronze artifacts from Evora and Alcacer do Sal objects in Southern Portugal have been characterized by using a combination of portable and fixed Energy Dispersive X-ray Fluorescence (EDXRF) and Scanning Electron Microscopy coupled with Energy Dispersive Spectroscopy (SEM-EDS). The combined use of the two techniques have already provided useful information for the characterization of a wide variety of historical materials of complex, heterogeneous composition such as mortars, glass, bricks and stone [35-44]. The experimental procedure was selected according to the following requirements:

- both EDXRF and SEM-EDS don’t require sampling of the specimen thus being micro- or non-destructive techniques, an essential requirement when dealing with archaeological objects.

- EDXRF and SEM-EDS are particularly appropriate as complementary techniques to investigate archaeo-metallurgic materials [29].
-The possibility to use EDXRF in portable mode let to perform non destructive analyses directly in museum locations.

The detailed EDXRF and SEM+EDS analytical methodology and operating conditions including quantification methods are described in previous papers [29] and [45-56].

2.4 Statistical analysis (PCA)

Due to the large number of compositional data and the complexity of archaeological information available regarding the artifacts from the Iberian dataset, the application of statistics is useful to obtain a description of their distribution and a macro-grouping according to the distribution of selected variables. PCA statistical analysis has been here used to find connections within multicomponent system, this last described by alloys compositional data (Cu, Sn, Pb, As, Fe) and other non numeric categorical and ordinal features like: typology of object, archaeological context, chronology and geographical areas, with the aim to highlight their similarities and differences [4-28]. In the data treatment was taken into account that the compositional values were obtained by different techniques by evaluating the acquisition conditions and associated error. In any case the data dispersion is wide over the respective confidence interval allowing to work with large groups. The mathematical procedure uses a linear transformation to convert a set of observations of possibly correlated variables, into a set of values of orthogonal (uncorrelated) variables called “principal components”. In this way is possible to have a simplified, linear and two-dimensional grouping of a multivariate system [34].

3. Results and discussion

As mentioned before, the combined archaeological/archaeometrical protocol described above has been tested on a combined dataset, collectively gathered from newly acquired chemical data: votive statuettes of unknown origin from the Evora and Alcacer Museum. The artifacts consisting of four anthropomorphic figures (three male and one female), three zoomorphic representations (three goats) and one anthropomorphic tripod [29, 57-59], while the considered published data are related to several sites on the Iberian Peninsula bronze production of the selected Late Bronze and Iron Age periods.

Archaeological, geographical and chemical data from 101 archaeological sites [1, 30, 7, 11-13, 19, 25-28], 40 mineral ores and more that 500 chemical analyses by EDXRF, μ-EDXRF, atomic absorption spectroscopy from literature [4, 5, 7, 9-15, 18-20, 22, 25-28] were considered and inserted in three separate databases yielding to the creation of comprehensive GIS thematic maps. The chemical data from the EDXRF and SEM+EDS analyses of Evora and Alcacer do Sal statuettes were added into the chemical values database.

As a first approach, the protocol produced a thematic map by using the archaeological databases on the Phoenician-Punic sites in the Iberian Peninsula with the data related to the presence of Cu, Pb, Sn ore deposits (Fig. 2) and the presence of waterways that may have favoured the access from the coast to the hinterland (Fig. 3). This map confirmed that the Phoenician presence in the Iberian Peninsula was intimately connected to mining activities and waterways these last useful for the access from the coastal regions.

As an example of the integration and potentiality between the archaeological and chemical analytical dataset, the GIS was also used to confirm the site Alcacer do Sal as the place where bronze statuettes of similar tipology to those ones in the Evora Museum have been found (fig. 4). The analyses EDXRF were then planned and carried out on eight votive statuettes from Evora Museum and six statuettes from the Alcacer do Sal site to verify the reliability of the two datasets. About all bronze statuettes show a ternary composition with Sn values ranging from 2.2 and 12.3 wt % [29]. In the following step, these compositions have been compared with the database values on bronze artifacts in the Iberian Peninsula as a whole [4,5,7, 9-14, 18-20, 22, 25-28], to identify the possible chronological range in which Evora/Alcacer do Sal are included.

Subsequently, PCA analyses have been carried out on artifacts compositional database to establish trends in the alloys employed in this period (figs 5-9). The compositional data have been selected through PCA as a function of: 1) chronology, 2) type of object, 3) geographical area, in order to understand the evolution of bronze objects production in relation with these three features. The plot in Fig. 5 shows the alloys distribution according with the chronology, while the inserted representation shows the loading plot of the selected variables (Cu, Pb, Fe, Sn, As). In details, the green dots and triangles represent LBA and EIA bronze artifacts reveal a technological evolution from binary bronze (Cu-Sn) to ternary bronze (Cu-Sn-Pb) with iron content respect to Early and Middle Bronze Age alloys characterized by arsenic as trace element. The presence of iron in detectable amount can be ascribed to reducing condition in the smelting furnace [27], a technological improvement imported probably in the Iberian peninsula from Phoenicians skilled metalworkers. The significant amount of lead in the alloy can be seen also as a technological change in the traditional Iberian local metallurgical practice: in fact, around the beginning of the Iron Age in the LBA for some Spanish sites, especially the ones located in Tartessian area, a ternary alloy started to be employed. Comparing compositional data of the Evora statuettes with the compositional trends in the plots of fig. 5, it is possible to suggest a dating between LBA and EIA for the Evora collection and consequently to confirm Alcacer do Sal as the most likely production site. Another PCA plot (fig.6) have been constructed by using non numeric features as variables such as tools or decorative objects. In fact, another important technological evolution was the introduction of the use of specific alloys for the production of distinct classes of objects. Tools (knives, swords, rasps, axes, etc ...), which were expected to have good mechanical properties, were produced mainly with binary alloy, as the presence of lead in the alloy, totally immiscible in the solid phase Cu-Sn, creates structural discontinuities causing possible fracture points in the metal. On the other hand, decorative objects, which did not require strong mechanical performance, but required a serial production with easy procedure involving low temperatures. This could be the reason of the presence of an increase of Pb in the votive statuettes. This is confirmed in fig. 6, where votive axes and the objects of common use (with no practical use) contain lead [15,18].

This confirms the evolution trend in metallurgy causing differentiation in relation to the object category. Since Evora/Alcacer do Sal artifacts are all decorative and votive objects, the ternary bronze composition is in accordance with their production procedure.

Another set of PCA diagrams has been produced checking the trend of the compositional values in relation to the type of the object: local (Iberian) or “orientalising” (fig. 7a,b). While the first category shows a binary (Cu-Sn) bronze composition, the “orientalising” artifacts clearly show a trend towards ternary bronze alloy compositions rich in Pb. Unfortunately is difficult to state with certainty if this change was due to external (Phoenician/Punic) influence or if it could be considered as a locally produced, local technological evolution. From this second plot is however possible to further suggest that the statuettes in the Évora Museum collection originated from a site where the Phoenician presence was quite well established, such as the Alcàcer do Sal one [11, 25-27].

To confirm more firmly the Alcacer do Sal hypothesis can be useful to narrow down the chronological and compositional range. In this way the PCA analysis coupled with the GIS spatial study proceeded focusing, temporally (on shorter timespans) and, spatially, (distinguishing between Spanish and Portuguese regions).

The GIS thematic map (fig. 8a) shows artifacts from Portugal and Spain belonging to Late Bronze Age (LBA). In southern-central Portugal and in Central and Southern Spain, a Cu-Sn binary alloy was commonly used. In Northern Iberia, in the so called “Atlantic area” [26], a ternary bronze was already used [9-13, 25-27]. In fig. 8a it is possible to evidence the exception of artifacts constituted by the ternary bronzes from the Andalusian region of Southern Spain (site 2 in fig. 8a). A critical analysis of the specific literature demonstrates that the artifacts originated from the Atlantic regions of Northern Iberia [15]. As concerns the tin (fig.8b) it is possible to relieve that the

Portuguese artifacts (LBA) present Sn values ranging from "ND" to a maximum of 18,8wt%. Most of artifacts presents Sn values ranging about 5 and 15 wt%, by indicating a specific phase in the Cu-Sn diagram. [10].

For the LBA-EIA transitional period, (fig.8 c,d), the GIS and PCA protocol confirms a trend towards a ternary alloy composition in Spain, while in Portugal (despite one orientalising object showing a small amount of lead - 3.52% - site 1 in fig. 8c [4]) metal artifacts show a binary bronze composition (Maximum Pb: 0.97 % and Sn values between 7.8 and 14.3% [25,26]). Therefore GIS and PCA combined protocol enabled us to better show how compositional changes are more notable in Spain during EIA (fig.8c,d), while in Portugal, the evolution is slower and only during Iron Age a difference of values can be better appreciated.

The GIS map coupled with the PCA plots relating to the full Early Iron Age period (fig.9) shows an increase in lead content and an increase of variability of tin values for the bronze artifacts from Mediterranean Spain. In Portugal, two Iron Age sites have been investigated: Castro dos Ratinhos and Quinta do Almaraz ("orientalising"). While in Castro dos Ratinhos, [25,26], located in the hinterland, the compositions show quite the same values for Sn and Pb as compared to LBA bronzes (Sn content ranging from 6.2wt% to 15.91wt%), in Quinta do Almaraz[27], positioned nearby the sea, Sn decreases to 8.1% and leaded bronze artifacts are present (Pb 4.60 and 5.9wt %). These compositional differences could be explained by considering how a location closer to the sea favoured the technological transfer brought by Phoenician traders [27]).

Coming back to the original "archaeological " problem regarding the provenance and dating of the bronze statuettes from the Evora Museum, the combined GIS and PCA analyses discussed above led us to to assume that, being the Evora figurines composed by a binary bronze with high lead content (reaching up to 26.1wt% [29]), their chronological range should be assigned to an advanced phase of the Iron Age. Querying the GIS system about the presence of similar artifacts from a EIA/MIA phoenician settlements, the system select the Alcacer do Sal site, where analogous artifacts belonging to V century BC have been found. The analysis on six artifacts from Alcacer do Sal [29] (Pb max 24.5wt% and tin between 1.15 and 14.7wt%) show that Alcacer do Sal and Evora compositional data belong to the same type of alloy (fig.10), leading to hypothesize a common origin from Alcacer do Sal as suggested by E. Gomes. The analyses SEM+EDS on both the set of artifacts show the presence of large lead globules [29], suggesting a voluntary addition of this element to fluidify the alloy with a consequent a slow cooling phase. The interpretation on Evora collection is therefore coherent with the affirmation that considerable variation in the Sn content and the presence of Pb in quantity higher than 2%, become common features at the beginning of the Iron Age, a period that coincides with the founding of the first Phoenician settlements on Portugal coasts, during the X and IX centuries BC [12,27,28]. The reason of this unusual high Pb content detected in these artifacts could be manifold: archaeological evidence assigns the Alcacer do Sal statuettes to the V century BC, therefore placing them two to four centuries after those of other settlements in the map (IX -VII century BC). It may therefore be assumed that the propensity by the local material culture to adopt the "new" imported leaded bronze technique, somewhat low at the start [27], increased with time with the result that, by the beginning of Early Iron Age not only an increasing number of artifacts were being produced with leaded bronze, but also that the mastery of the technique allowed the use of higher amounts of lead during bronze production discriminating among different artifacts classes.

In summary, the combined GIS + PCA protocol adopted in this study represents an useful tool to integrate archaeological and archaeometric multivariate databases containing both non numeric and numeric variables. In the case study examined here, it has been successfully used to corroborate the dating (V century BC) and provenance (Alcacer do Sal) of the Evora Museum bronze statuettes, by demonstrating that the same procedure can be applied widely to solve provenance and dating issues emerging in the modern archaeological research.

5. Conclusions

The protocol adopted in this study, combining data from chemical analytical techniques with spatial analysis by GIS system and statistical analysis by PCA, permits the handling, interpretation and integration of a large amount of archaeological/archaeometric analytical variables both numeric (chemical composition) and non numeric (chronological, geographical, typological). The finalresult is the production of thematic GIS maps and PCA plots which may be used as a easy to read, fast tool for comprehensive comparison studies across time periods and geographical locations so essential in archaeological research. The protocol was tested successfully in the case study concerning the precise dating and provenance of the bronze statuettes from Evora, where it confirmed and further refined earlier hypotheses based separately on archaeological or chemical studies i.e. that Phoenician/Punic presence as an allochthonous component in a foreign area is bearer of technological improvements and an activator of more pronounced exploitation of natural resources. Using a process of analysis/comparison of analytical data, it was possible to support hypothesis and suggestions about provenance and chronology for the not contextualized collection in the Museum of Evora. The provenance has been allocated to Alcacer do Sal Phoenician settlement, near Rio Sado. From a more general point of view, instead, it was possible to confirm the use of ternary alloys for the production of these objects, which typology are considered as influenced by Phoenicians. Furthermore it can also be stated that the level of metallurgical skills was already distinguished in relation to artifact classes. The site of Alcacer do Sal, near the coast and near the mouth of the Rio Sado, seems to have been a very much frequented site, because of the technological skills acquired by the local artisans, probably influenced by Phoenicians metalworkers. Further study are intended to extend the validation of the present protocol to other sites and other archeo-materials influenced by Phoenician culture across the Mediterranean Basin and beyond.

The obtained results in the archaeological field demonstrated the scientific procedure adopted in the present work can be considered suitable for the treatment of data derived from different discipline but all concerning an unique historical aspect. The protocol based on available software programs and physico-chemical determinations with traditional methods, can easily exported by reaching one of the essential aim of archaeometry: to create new tools to help solve archaeological dilemmas.

Acknowledgement

This work has been supported by the precious help of Dr. Antonio Alegria (Evora Museum), Dr. Esmeralda Gomes (Alentejo Directorate General for Culture) and Prof. Ana Margarida Arruda (University of Lisbon) for authorizing the use of the artifacts from Evora's collection and Alcacer do Sal and for useful discussions. Thanks are also due to Dr. Federico Marini, Sapienza Università di Roma, for help with PCA. NS and the Hercules Laboratory of the University of Evora would like to thank the EEA and the FCT for financial support.

Figure captions

Fig. 1 protocol flow-diagram
Fig. 2 GIS thematic map on the ancient mines (dots and red numbers) and archaeological settlements (blue triangles and numbers)
Mines [31-33]:
1) Alcoutim **Cu**, 2) Neves Corvo **Cu**, 3) Aljustrel **Cu**, 4) Sao Domingos **Cu**, 5) Lousal **Cu**, 6) Lagoa **Cu**, 7) Salgada **Cu**, 8) Santa Eulalia **Sn**, 9) Argemela **Sn**, 10) Caeira **Cu**, 11) Linares de Cartagena **Cu**, 12) Cerro, Muriano **Cu**, 13) Llamo **Cu**, 14) Siviglia **Cu**,15) Aznalcollar **Cu**, 16) Los Millares **Cu**, 17) Guadalquivir **Sn**, 18) Murcia **Sn**, 19) Ria De Huelva **Sn**, 20) Caceres **Sn**, 21) Aljustrel **Cu**, 22) Canal Caveira **Pb**, 23) Vila Nova de Milfontes **Pb**, 24) Zamora **Sn**, 25) Folgadouro **Sn**, 26) Belmonte **Sn**, 27) Vela **Sn**, 28) Aveiro **Pb**, 29) Zambujal **Fe**, 30) Mertola **Pb**, 31) Moura **Cu**, 32) Vinhais **Sn**, 33) Alandroal **Cu**, 34) Alcoutim **Cu**, 35) Loulè **Cu**, 36) Silves **Cu**, 37) Sabugal **Cu**, 38) Arronches **Cu**, 39) Setubal **Cu**, 40) Viana do Castelo **Cu**
Archaeological settlemets [1,7,11-13,19, 25-28, 30]:
1) Madeira, 2) Castro dos Ratinhos, 3) Cancho Roano, 4) Fraga dos Corvos, 5) Baioes, 6) Figuereido das Donas, 7) Castro de Leceia, 8) Sao Bento, 9) Castro de Sao Romao, 10) Evora (reference point), 11)Ourique, 12) Almogrebe,13) Rocha Branca, 14) Safara, 15) Sines, 16) Castro de Praganca (Estremadura-Cadaval), 17) Castro Marim, 18) Cerro de Rocha Branca, 19) Fonte Velha de Bensafrim (Lagos), 20)Comoros da Portela (San Bartolomeu de Messines -Silves), 21) Pere Jaques (Aljezur), 22) Alagoas (Salir-Loule), 23) Santa Olaia, 24) Castro de Tavarede, 25) Choes, 26)Pardinheiros, 27) Conimbriga, 28) Fonte de Cabanas, 29) Alcacer do Sal, 30) El Castillo (Alcacer do Sal), 31) La Necropoli di Senhor dos Martires, 32) Abul, 33) Setubal, 34) Moinhos da Atalaia, 35) Cerradinha (Lagoa de Santo Andrè, Santiago do Cacem, 36))Quinta do Almaraz, 37) Lisboa (Olisipo), 38) Outorela, 39) Santa Eufemia, 40) Freiria, 41) La Alcacova de Santarem, 42) Torres Vedras, 43) Faiao (Sintra), 44) Verdolay, 45) Aldovesta, 46) Vinarregel, 47) La Fonteta, 48) Cerro de la Mora, 49) Mesa de Fornes, 50) Chorreras, 51) Coll del Moro, 52) Toscanos, 53) Cerro del Prado, 54) Aratispi, 55) Villanueva de la Vera, 56) La Aliseda, 57) Siruela, 58) Medellin, 59) Zarza de Alanje, 60) Aljucen, 61) Morro de Mezquitilla (Algarrobo-Malaga), 62) Huelva (La Joya, San pedro, Esperanza), 63) Niebla 64) Riotinto, 65) Tejada la Vieja, 66) Aznalcollar, 67) Gadir (Cadice), 68) Castillo de Dona Blanca, 69) Mesas de Asta, 70) Lebrija, 71)El Carambolo, 72) Sevilla (Cerro Macareno Valencina), 73) Setefilla (Lora de Rio), 74) Carmona, 75) Osuna, 76) Ecija, 77) Quemados (Cordoba), 78) Castulo, 79) Montilla, 80) Acinipo, 81) Malaka, 82) Frigiliana, 84) Galera, 85) Almunecar (Sexi Laurita, Puente de Noi), 86) Salobrena, 87) Villaricos, 88)Gavilanes, 89) Valdegamas, 90) Tartesso (uncertain location), 91) Carteia, 92) Abdera (Cerro de Montecristo-Adra-Almeria), 93) Alicante, 94) Murcia, 95) Matachel, 96) Lujan, 97) Formentera, 98) Sa Caleta, 99) Ebusus, 100) Puig Des Molins, 101)Mallorca [1,10,

Fig. 3 spatial query for the selection of settlements nearby the rivers (red stars)
Fig. 4 localizations and statuettes of Evora and Alcacer do Sal
Fig. 5 PCA diagrams showing distribution of artifacts in relation with chronological periods **([4,5,7,9-15,15,18,19,22,25-27]**
Fig. 6 PCA plot PCA plot for the tools and non-decorative objects[([4,5,7,9-15,15,18,19,22,25-27]
Fig. 7 PCA plot for local and "orientalising" objects ([4,5,7,9-15,15,18,19,22,25-27]
Fig. 8: a) GIS thematic map with compositional histograms of bronze objects (LBA) in: 1) Almagro; 2) Andalucia; 3) El Carambolo, 4) Castro dos Ratinhos, 5) Central Portugal, 6) Baioes, 7) Figueredo das Donas, 8) Porto, 9) Fraga dos Corvos, 10) Carballino, 11) Navelgas, 12) La Banela, 13) Langreo, 14) Lena, 15) Carmenes, 16) Pola de Laviana, 17) Palencia. **b)** PCA plot for Portuguese artifacts belonging to LBA period **[7,9-13, 15,18,25**; **c)** thematic map on bronze artifacts from LBA-EIA settlements: 1) Torres Vedras, 2) Cadiz,3) Villagarcia de la Torre, 4) la Joya, 5) El Risco, 6) Siruela, 7) Coca; **d)** PCA for LBA-EIA artifacts in Spain **[4,15,25].**
Fig. 9 a) GIS thematic map for EIA bronze artifacts: 1) Quinta do Almaraz, 2) Castro dos Ratinhos, 3) La Luz, 4) Collado de los Jardines, 5) Castellar de Santisteban, 6) El Palomar,7) Lujan. **b)** PCA plots for EIA Portuguese and Spanish artifacts: the points surrounded by red circle belong to Quinta do Almaraz while the black oval contains Castro dos Ratinhos artifacts **[5,20,22,26,27]**
Fig. 10 GIS thematic map from Iron Age artifacts from 1) Alcacer do Sal, 2) Evora [29]

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Combining chemical data with GIS and PCA to investigate Phoenician-Punic Cu-metallurgy

Keywords: GIS, SEM-EDS, EDXRF, PCA, bronze artifacts, Phoenician-Punic

1. Introduction

It is well-known that Phoenicians and later Punics established several colonies in the Mediterranean Basin and in many Atlantic areas [1] acting as innovators and dispensers of technical skills and know-how during the contacts with local cultures, resulting in the production of a wealth of artifacts made up of different archaeomaterials [1]. The production of Cu-based alloy artifacts in particular flourished between the Bronze Age and the Iron Age, reaching a climax during Early - Middle Iron Age (MIA-EIA). From an archaeological point of view, distinguishing between objects of different origins, i.e. Phoenician, locally produced and/or resulting from the mixing up of both elements, is of particular relevance and cannot be achieved using solely "archaeological" tools. The term "orientalising" [2] is here used to identify the mixing production showing local and allochthonous features. In this meaning the term then includes also artifacts belonging to a wider chronological period respect to the usual definition. An integrated approach combining archaeological evidence, analytical data acquisition, statistical processing and spatial analysis tools is therefore needed to achieve more objective conclusions.

As part of a larger, multidisciplinary project on Phoenician and Punic influence on the Copper-archaeometallurgy of the Mediterranean Basin and surrounding regions, this paper focuses on the development and application of an innovative protocol based on PCA (Principal Component Analysis) involving numeric and non numeric variables and GIS (Geographical Information System) as a simple aid to the interpretation of chemical-analytical data of Cu-based alloy artifacts. The protocol is a useful tool for highlighting existing connections between specific alloy chemical compositions, the provenance settlements of the artefact, has been produced and the proximity to mining resources, waterways and allochtonous presence such as the Phoenician/Punic influence in the Iberian bronze technology.

Either the chronological range, Late Bronze Age (LBA) and Iron Age, and a narrower period Early - Middle Iron Age (EIA-MIA), have been selected because of the large amount of "orientalising" artifacts produced during this historical timespan.

The application of GIS [3] has already proved to be a very useful tool especially by considering it as an adjunct to PCA statistical method. The procedure has been applied for the treatment of already published data, available in the literature on bronze archaeo-metallurgy in the Iberian Peninsula [4-28], combined with additional data obtained by EDXRF and SEM+EDS on Phoenician bronze artifacts representing anthropomorphic and zoomorphic figures from Museum collections: Evora (unknown origin), Alcacer do Sal (archaeological provenance). Evora artifacts have been dated to the Iron Age (V century BC), but their precise chronological collocation, geographical provenance, and typology remained uncertain. A previous study [29] put forward the hypothesis of a common origin from the Phoenician site of Alcacer do Sal [30], for all statuettes. Alcacer do Sal (VII century BC) is considered Phoenician from two points of view: the settlement features and the evidence of religious cults connected to Baal and Astarte. Dr. Esmeralda Gomes (personal communication, master thesis) and Prof. Ana Margarida Arruda, were the first to note this similarity and the proximity of the two sites and to assume a common provenance. One aim of the present study is to apply the new multidisciplinary protocol to confirm the hypothesis, by comparing the analytical data from Portuguese items with a chronologically and geographically restrained extended dataset encompassing Phoenician sites from the Iberian Peninsula.

The Portuguese case studies and the Iberian published database points have been selected as a function of: a) the relevance of their strategic geographical positions; b) the presence of important copper, tin and lead mining resources in the surrounding region.

The basic steps of the protocol can be summarized as follows:

1) chemical and physical analysis on local and “orientalising” artifacts and comparison data collection from literature [4-28];

2) creation of databases including archaeological sites and location of ancient mines and ores [31-33];

3) development of GIS thematic maps [3] on the following parameters: a) location of Phoenician/Punic settlements; b) location of ancient mines exploited during the selected timespan; c) location of bronze findings; d) compositional data visualised in connection with geographical positioning;

4) PCA statistical treatment [34], to obtain a representation of Phoenician-Punic bronze production in the Mediterranean basin and neighbouring areas.

2. Methods

The analytical protocol required a flexible formulation with the adoption of a multidisciplinary approach combining archaeology (i.e. the geographical collocation of archaeological settlements), archaeometry (interpretation of bronze chemical compositions), geology (localization and characterization of ancient mines).

The detailed protocol flow-diagram, shown in fig.1 can be described in a sequence of steps.

2.1 Bibliographical study for the elaboration of different databases

Following a thorough bibliographical study, three databases have been compiled as Excel worksheets based on the following data: a) location of archaeological Phoenician settlements; b) chemical data from ancient mines focusing on bronze major elements: Cu, Sn, Pb; c) literature chemical data on bronze archaeological specimens from the same historical period.

Each database was fed with the geographical coordinates of the considered sites for the acquisition in GIS environment.

2.2 Geographic Information System (GIS)

The three different databases have been geo-referenced through the use of GIS to obtain, after conversion in geodatabases, thematic maps of archaeological settlements, mines, waterways and evidence of bronze artifacts. The integration of the databases with GIS let to perform an extensive analysis of the spatial phenomenon under study, by using the various searching options the system offers such as: visual information on the distribution, frequencies and relation between different features under study, database searching tool to create a subset of selected features, using the statistic and editing tools to display on the maps also compositional information or to digitalize geographic features like rivers or paths), georeferencing raster cartography and, finally, creating thematic maps.

2.3 Chemical analysis

The chemical database from published data is based on analyses carried out using a variety of techniques (EDXRF, µ-XRF, AAS). The alloys composition of selected bronze artifacts from Evora and Alcacer do Sal objects in Southern Portugal have been characterized by using a combination of portable and fixed Energy Dispersive X-ray Fluorescence (EDXRF) and Scanning Electron Microscopy coupled with Energy Dispersive Spectroscopy (SEM-EDS). The combined use of the two techniques have already provided useful information for the characterization of a wide variety of historical materials of complex, heterogeneous composition such as mortars, glass, bricks and stone [35-44]. The experimental procedure was selected according to the following requirements:

- both EDXRF and SEM-EDS don’t require sampling of the specimen thus being micro- or non-destructive techniques, an essential requirement when dealing with archaeological objects.

- EDXRF and SEM-EDS are particularly appropriate as complementary techniques to investigate archaeo-metallurgic materials [29].

-The possibility to use EDXRF in portable mode let to perform non destructive analyses directly in museum locations.

The detailed EDXRF and SEM+EDS analytical methodology and operating conditions including quantification methods are described in previous papers [29] and [45-56].

2.4 Statistical analysis (PCA)

Due to the large number of compositional data and the complexity of archaeological information available regarding the artifacts from the Iberian dataset, the application of statistics is useful to obtain a description of their distribution and a macro-grouping according to the distribution of selected variables. PCA statistical analysis has been here used to find connections within multicomponent system, this last described by alloys compositional data (Cu, Sn, Pb, As, Fe) and other non numeric categorical and ordinal features like: typology of object, archaeological context, chronology and geographical areas, with the aim to highlight their similarities and differences [4-28]. In the data treatment was taken into account that the compositional values were obtained by different techniques by evaluating the acquisition conditions and associated error. In any case the data dispersion is wide over the respective confidence interval allowing to work with large groups. The mathematical procedure uses a linear transformation to convert a set of observations of possibly correlated variables, into a set of values of orthogonal (uncorrelated) variables called “principal components”. In this way is possible to have a simplified, linear and two-dimensional grouping of a multivariate system [34].

3. Results and discussion

As mentioned before, the combined archaeological/archaeometrical protocol described above has been tested on a combined dataset, collectively gathered from newly acquired chemical data: votive statuettes of unknown origin from the Evora and Alcacer Museum. The artifacts consisting of four anthropomorphic figures (three male and one female), three zoomorphic representations (three goats) and one anthropomorphic tripod [29, 57-59], while the considered published data are related to several sites on the Iberian Peninsula bronze production of the selected Late Bronze and Iron Age periods.

Archaeological, geographical and chemical data from 101 archaeological sites [1, 30, 7, 11-13, 19, 25-28], 40 mineral ores and more that 500 chemical analyses by EDXRF, μ-EDXRF, atomic absorption spectroscopy from literature [4, 5, 7, 9-15, 18-20, 22, 25-28] were considered and inserted in three separate databases yielding to the creation of comprehensive GIS thematic maps. The chemical data from the EDXRF and SEM+EDS analyses of Evora and Alcacer do Sal statuettes were added into the chemical values database.

As a first approach, the protocol produced a thematic map by using the archaeological databases on the Phoenician-Punic sites in the Iberian Peninsula with the data related to the presence of Cu, Pb, Sn ore deposits (Fig. 2) and the presence of waterways that may have favoured the access from the coast to the hinterland (Fig. 3). This map confirmed that the Phoenician presence in the Iberian Peninsula was intimately connected to mining activities and waterways these last useful for the access from the coastal regions.

As an example of the integration and potentiality between the archaeological and chemical analytical dataset, the GIS was also used to confirm the site Alcacer do Sal as the place where bronze statuettes of similar tipology to those ones in the Evora Museum have been found (fig. 4). The analyses EDXRF were then planned and carried out on eight votive statuettes from Evora Museum and six statuettes from the Alcacer do Sal site to verify the reliability of the two datasets. About all bronze statuettes show a ternary composition with Sn values ranging from 2.2 and 12.3 wt % [29]. In the following step, these compositions have been compared with the database values on bronze artifacts in the Iberian Peninsula as a whole [4,5,7, 9-14, 18-20, 22, 25-28], to identify the possible chronological range in which Evora/Alcacer do Sal are included.

Subsequently, PCA analyses have been carried out on artifacts compositional database to establish trends in the alloys employed in this period (figs 5-9). The compositional data have been selected through PCA as a function of: 1) chronology, 2) type of object, 3) geographical area, in order to understand the evolution of bronze objects production in relation with these three features. The plot in Fig. 5 shows the alloys distribution according with the chronology, while the inserted representation shows the loading plot of the selected variables (Cu, Pb, Fe, Sn, As). In details, the green dots and triangles represent LBA and EIA bronze artifacts reveal a technological evolution from binary bronze (Cu-Sn) to ternary bronze (Cu-Sn-Pb) with iron content respect to Early and Middle Bronze Age alloys characterized by arsenic as trace element. The presence of iron in detectable amount can be ascribed to reducing condition in the smelting furnace [27], a technological improvement imported probably in the Iberian peninsula from Phoenicians skilled metalworkers. The significant amount of lead in the alloy can be seen also as a technological change in the traditional Iberian local metallurgical practice: in fact, around the beginning of the Iron Age in the LBA for some Spanish sites, especially the ones located in Tartessian area, a ternary alloy started to be employed. Comparing compositional data of the Evora statuettes with the compositional trends in the plots of fig. 5, it is possible to suggest a dating between LBA and EIA for the Evora collection and consequently to confirm Alcacer do Sal as the most likely production site. Another PCA plot (fig.6) have been constructed by using non numeric features as variables such as tools or decorative objects. In fact, another important technological evolution was the introduction of the use of specific alloys for the production of distinct classes of objects. Tools (knives, swords, rasps, axes, etc ...), which were expected to have good mechanical properties, were produced mainly with binary alloy, as the presence of lead in the alloy, totally immiscible in the solid phase Cu-Sn, creates structural discontinuities causing possible fracture points in the metal. On the other hand, decorative objects, which did not require strong mechanical performance, but required a serial production with easy procedure involving low temperatures. This could be the reason of the presence of an increase of Pb in the votive statuettes. This is confirmed in fig. 6, where votive axes and the objects of common use (with no practical use) contain lead [15,18].

This confirms the evolution trend in metallurgy causing differentiation in relation to the object category. Since Evora/Alcacer do Sal artifacts are all decorative and votive objects, the ternary bronze composition is in accordance with their production procedure.

Another set of PCA diagrams has been produced checking the trend of the compositional values in relation to the type of the object: local (Iberian) or “orientalising” (fig. 7a,b). While the first category shows a binary (Cu-Sn) bronze composition, the “orientalising” artifacts clearly show a trend towards ternary bronze alloy compositions rich in Pb. Unfortunately is difficult to state with certainty if this change was due to external (Phoenician/Punic) influence or if it could be considered as a locally produced, local technological evolution. From this second plot is however possible to further suggest that the statuettes in the Évora Museum collection originated from a site where the Phoenician presence was quite well established, such as the Alcàcer do Sal one [11, 25-27].

To confirm more firmly the Alcacer do Sal hypothesis can be useful to narrow down the chronological and compositional range. In this way the PCA analysis coupled with the GIS spatial study proceeded focusing, temporally (on shorter timespans) and, spatially, (distinguishing between Spanish and Portuguese regions).

The GIS thematic map (fig. 8a) shows artifacts from Portugal and Spain belonging to Late Bronze Age (LBA). In southern-central Portugal and in Central and Southern Spain, a Cu-Sn binary alloy was commonly used. In Northern Iberia, in the so called “Atlantic area” [26], a ternary bronze was already used [9-13, 25-27]. In fig. 8a it is possible to evidence the exception of artifacts constituted by the ternary bronzes from the Andalusian region of Southern Spain (site 2 in fig. 8a). A critical analysis of the specific literature demonstrates that the artifacts originated from the Atlantic regions of Northern Iberia [15]. As concerns the tin (fig.8b) it is possible to relieve that the

Portuguese artifacts (LBA) present Sn values ranging from "ND" to a maximum of 18,8wt%. Most of artifacts presents Sn values ranging about 5 and 15 wt%, by indicating a specific phase in the Cu-Sn diagram. [10].

For the LBA-EIA transitional period, (fig.8 c,d), the GIS and PCA protocol confirms a trend towards a ternary alloy composition in Spain, while in Portugal (despite one orientalising object showing a small amount of lead - 3.52% - site 1 in fig. 8c [4]) metal artifacts show a binary bronze composition (Maximum Pb: 0.97 % and Sn values between 7.8 and 14.3% [25,26]). Therefore GIS and PCA combined protocol enabled us to better show how compositional changes are more notable in Spain during EIA (fig.8c,d), while in Portugal, the evolution is slower and only during Iron Age a difference of values can be better appreciated.

The GIS map coupled with the PCA plots relating to the full Early Iron Age period (fig.9) shows an increase in lead content and an increase of variability of tin values for the bronze artifacts from Mediterranean Spain. In Portugal, two Iron Age sites have been investigated: Castro dos Ratinhos and Quinta do Almaraz ("orientalising"). While in Castro dos Ratinhos, [25,26], located in the hinterland, the compositions show quite the same values for Sn and Pb as compared to LBA bronzes (Sn content ranging from 6.2wt% to 15.91wt%), in Quinta do Almaraz[27], positioned nearby the sea, Sn decreases to 8.1% and leaded bronze artifacts are present (Pb 4.60 and 5.9wt %). These compositional differences could be explained by considering how a location closer to the sea favoured the technological transfer brought by Phoenician traders [27]).

Coming back to the original "archaeological " problem regarding the provenance and dating of the bronze statuettes from the Evora Museum, the combined GIS and PCA analyses discussed above led us to to assume that, being the Evora figurines composed by a binary bronze with high lead content (reaching up to 26.1wt% [29]), their chronological range should be assigned to an advanced phase of the Iron Age. Querying the GIS system about the presence of similar artifacts from a EIA/MIA phoenician settlements, the system select the Alcacer do Sal site, where analogous artifacts belonging to V century BC have been found. The analysis on six artifacts from Alcacer do Sal [29] (Pb max 24.5wt% and tin between 1.15 and 14.7wt%) show that Alcacer do Sal and Evora compositional data belong to the same type of alloy (fig.10), leading to hypothesize a common origin from Alcacer do Sal as suggested by E. Gomes. The analyses SEM+EDS on both the set of artifacts show the presence of large lead globules [29], suggesting a voluntary addition of this element to fluidify the alloy with a consequent a slow cooling phase. The interpretation on Evora collection is therefore coherent with the affirmation that considerable variation in the Sn content and the presence of Pb in quantity higher than 2%, become common features at the beginning of the Iron Age, a period that coincides with the founding of the first Phoenician settlements on Portugal coasts, during the X and IX centuries BC [12,27,28]. The reason of this unusual high Pb content detected in these artifacts could be manifold: archaeological evidence assigns the Alcacer do Sal statuettes to the V century BC, therefore placing them two to four centuries after those of other settlements in the map (IX -VII century BC). It may therefore be assumed that the propensity by the local material culture to adopt the "new" imported leaded bronze technique, somewhat low at the start [27], increased with time with the result that, by the beginning of Early Iron Age not only an increasing number of artifacts were being produced with leaded bronze, but also that the mastery of the technique allowed the use of higher amounts of lead during bronze production discriminating among different artifacts classes.

In summary, the combined GIS + PCA protocol adopted in this study represents an useful tool to integrate archaeological and archaeometric multivariate databases containing both non numeric and numeric variables. In the case study examined here, it has been successfully used to corroborate the dating (V century BC) and provenance (Alcacer do Sal) of the Evora Museum bronze statuettes, by demonstrating that the same procedure can be applied widely to solve provenance and dating issues emerging in the modern archaeological research.

5. Conclusions

The protocol adopted in this study, combining data from chemical analytical techniques with spatial analysis by GIS system and statistical analysis by PCA, permits the handling, interpretation and integration of a large amount of archaeological/archaeometric analytical variables both numeric (chemical composition) and non numeric (chronological, geographical, typological). The finalresult is the production of thematic GIS maps and PCA plots which may be used as a easy to read, fast tool for comprehensive comparison studies across time periods and geographical locations so essential in archaeological research. The protocol was tested successfully in the case study concerning the precise dating and provenance of the bronze statuettes from Evora, where it confirmed and further refined earlier hypotheses based separately on archaeological or chemical studies i.e. that Phoenician/Punic presence as an allochthonous component in a foreign area is bearer of technological improvements and an activator of more pronounced exploitation of natural resources. Using a process of analysis/comparison of analytical data, it was possible to support hypothesis and suggestions about provenance and chronology for the not contextualized collection in the Museum of Evora. The provenance has been allocated to Alcacer do Sal Phoenician settlement, near Rio Sado. From a more general point of view, instead, it was possible to confirm the use of ternary alloys for the production of these objects, which typology are considered as influenced by Phoenicians. Furthermore it can also be stated that the level of metallurgical skills was already distinguished in relation to artifact classes. The site of Alcacer do Sal, near the coast and near the mouth of the Rio Sado, seems to have been a very much frequented site, because of the technological skills acquired by the local artisans, probably influenced by Phoenicians metalworkers. Further study are intended to extend the validation of the present protocol to other sites and other archeo-materials influenced by Phoenician culture across the Mediterranean Basin and beyond.

The obtained results in the archaeological field demonstrated the scientific procedure adopted in the present work can be considered suitable for the treatment of data derived from different discipline but all concerning an unique historical aspect. The protocol based on available software programs and physico-chemical determinations with traditional methods, can easily exported by reaching one of the essential aim of archaeometry: to create new tools to help solve archaeological dilemmas.

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Figure captions

Fig. 1 protocol flow-diagram

Fig. 2 GIS thematic map on the ancient mines (dots and red numbers) and archaeological settlements (blue triangles and numbers)

Mines [31-33]:

1) Alcoutim **Cu**, 2) Neves Corvo **Cu**, 3) Aljustrel **Cu**, 4) Sao Domingos **Cu**, 5) Lousal **Cu**, 6) Lagoa **Cu**, 7) Salgada **Cu**, 8) Santa Eulalia **Sn**, 9) Argemela **Sn**, 10) Caeira **Cu**, 11) Linares de Cartagena **Cu**, 12) Cerro, Muriano **Cu**, 13) Llamo **Cu**, 14) Siviglia **Cu**,15) Aznalcollar **Cu**, 16) Los Millares **Cu**, 17) Guadalquivir **Sn**, 18) Murcia **Sn**, 19) Ria De Huelva **Sn**, 20) Caceres **Sn**, 21) Aljustrel **Cu**, 22) Canal Caveira **Pb**, 23) Vila Nova de Milfontes **Pb**, 24) Zamora **Sn**, 25) Folgadouro **Sn**, 26) Belmonte **Sn**, 27) Vela **Sn**, 28) Aveiro **Pb**, 29) Zambujal **Fe**, 30) Mertola **Pb**, 31) Moura **Cu**, 32) Vinhais **Sn**, 33) Alandroal **Cu**, 34) Alcoutim **Cu**, 35) Loulè **Cu**, 36) Silves **Cu**, 37) Sabugal **Cu**, 38) Arronches **Cu**, 39) Setubal **Cu**, 40) Viana do Castelo **Cu**

Archaeological settlemets [1,7,11-13,19, 25-28, 30]:

1) Madeira, 2) Castro dos Ratinhos, 3) Cancho Roano, 4) Fraga dos Corvos, 5) Baioes, 6) Figuereido das Donas, 7) Castro de Leceia, 8) Sao Bento, 9) Castro de Sao Romao, 10) Evora (reference point), 11)Ourique, 12) Almogrebe,13) Rocha Branca, 14) Safara, 15) Sines, 16) Castro de Praganca (Estremadura-Cadaval), 17) Castro Marim, 18) Cerro de Rocha Branca, 19) Fonte Velha de Bensafrim (Lagos), 20)Comoros da Portela (San Bartolomeu de Messines -Silves), 21) Pere Jaques (Aljezur), 22) Alagoas (Salir-Loule), 23) Santa Olaia, 24) Castro de Tavarede, 25) Choes, 26)Pardinheiros, 27) Conimbriga, 28) Fonte de Cabanas, 29) Alcacer do Sal, 30) El Castillo (Alcacer do Sal), 31) La Necropoli di Senhor dos Martires, 32) Abul, 33) Setubal, 34) Moinhos da Atalaia, 35) Cerradinha (Lagoa de Santo Andrè, Santiago do Cacem, 36))Quinta do Almaraz, 37) Lisboa (Olisipo), 38) Outorela, 39) Santa Eufemia, 40) Freiria, 41) La Alcacova de Santarem, 42) Torres Vedras, 43) Faiao (Sintra), 44) Verdolay, 45) Aldovesta, 46) Vinarregel, 47) La Fonteta, 48) Cerro de la Mora, 49) Mesa de Fornes, 50) Chorreras, 51) Coll del Moro, 52) Toscanos, 53) Cerro del Prado, 54) Aratispi, 55) Villanueva de la Vera, 56) La Aliseda, 57) Siruela, 58) Medellin, 59) Zarza de Alanje, 60) Aljucen, 61) Morro de Mezquitilla (Algarrobo-Malaga), 62) Huelva (La Joya, San pedro, Esperanza), 63) Niebla 64) Riotinto, 65) Tejada la Vieja, 66) Aznalcollar, 67) Gadir (Cadice), 68) Castillo de Dona Blanca, 69) Mesas de Asta, 70) Lebrija, 71)El Carambolo, 72) Sevilla (Cerro Macareno Valencina), 73) Setefilla (Lora de Rio), 74) Carmona, 75) Osuna, 76) Ecija, 77) Quemados (Cordoba), 78) Castulo, 79) Montilla, 80) Acinipo, 81) Malaka, 82) Frigiliana, 84) Galera, 85) Almunecar (Sexi Laurita, Puente de Noi), 86) Salobrena, 87) Villaricos, 88)Gavilanes, 89) Valdegamas, 90) Tartesso (uncertain location), 91) Carteia, 92) Abdera (Cerro de Montecristo-Adra-Almeria), 93) Alicante, 94) Murcia, 95) Matachel, 96) Lujan, 97) Formentera, 98) Sa Caleta, 99) Ebusus, 100) Puig Des Molins, 101)Mallorca [1,10,

Fig. 3 spatial query for the selection of settlements nearby the rivers (red stars)
Fig. 4 localizations and statuettes of Evora and Alcacer do Sal
Fig. 5 PCA diagrams showing distribution of artifacts in relation with chronological periods **([4,5,7,9-15,15,18,19,22,25-27]**
Fig. 6 PCA plot PCA plot for the tools and non-decorative objects[([4,5,7,9-15,15,18,19,22,25-27]
Fig. 7 PCA plot for local and "orientalising" objects ([4,5,7,9-15,15,18,19,22,25-27]
Fig. 8: a) GIS thematic map with compositional histograms of bronze objects (LBA) in: 1) Almagro; 2) Andalucia; 3) El Carambolo, 4) Castro dos Ratinhos, 5) Central Portugal, 6) Baioes, 7) Figueredo das Donas, 8) Porto, 9) Fraga dos Corvos, 10) Carballino, 11) Navelgas, 12) La Banela, 13) Langreo, 14) Lena, 15) Carmenes, 16) Pola de Laviana, 17) Palencia. **b)** PCA plot for Portuguese artifacts belonging to LBA period **[7,9-13, 15,18,25**; **c)** thematic map on bronze artifacts from LBA-EIA settlements: 1) Torres Vedras, 2) Cadiz,3) Villagarcia de la Torre, 4) la Joya, 5) El Risco, 6) Siruela, 7) Coca; **d)** PCA for LBA-EIA artifacts in Spain **[4,15,25].**
Fig. 9 a) GIS thematic map for EIA bronze artifacts: 1) Quinta do Almaraz, 2) Castro dos Ratinhos, 3) La Luz, 4) Collado de los Jardines, 5) Castellar de Santisteban, 6) El Palomar,7) Lujan. **b)** PCA plots for EIA Portuguese and Spanish artifacts: the points surrounded by red circle belong to Quinta do Almaraz while the black oval contains Castro dos Ratinhos artifacts **[5,20,22,26,27]**
Fig. 10 GIS thematic map from Iron Age artifacts from 1) Alcacer do Sal, 2) Evora [29]

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