



Dottorato di ricerca: Scienze Applicate per la Protezione dell'Ambiente e dei Beni Culturali Dipartimento di Scienze della Terra Coordinatore: Prof.ssa Adriana MARAS

Curso de Doutoramento: Ciências da Terra e do Espaço (Centro de Geofisica) Universidade de Evora Coordinatore: Prof. Mourad BEZZEGHOUD

Tutori: Prof. Luigi CAMPANELLA Dr.ssa Daniela FERRO Dr. Nicola SCHIAVON

Revisori: Prof.ssa Luciana DRAGO Dr. Giuseppe GUIDA

Docenti Esaminatori: Prof. Luigi DEI Prof. Antonio CANDEIAS Prof. José Antonio MIRÃO Prof. Fabrizio PASSARINI

Archaeometric Investigation for Provenance Studies about Copper Metallurgy in the Phoenician and Punic Cultures

Indagine archeometrica per studi di provenienza della metallurgia del rame nelle culture fenicia e punica

Angela CELAURO

XXV Ciclo

Riassunto

Lo sviluppo della ricerca scientifica indirizzata a coadiuvare con elementi diagnostici studi umanistici, ha oggi come funzione prioritaria quella di fornire nuovi strumenti per l'interpretazione, collocazione, contestualizzazione di problematiche storiche. In quest'ottica il presente lavoro è volto alla preparazione e collaudo di una procedura basata sull'acquisizione di dati di composizione di reperti di leghe a base-rame integrata con programmi informatici, con il fine di fornire elementi innovativi per la localizzazione ed interpretazione della presenza e influenza dei Fenici e Punici in aree del Bacino del Mediterraneo, dell'Atlantico e aree continentali. Il percorso progettuale è stato applicato ad un periodo cronologico di interesse (fine dell'Età del Bronzo fino all'Età del Ferro) e considerando in particolare le seguenti aree geografiche: Portogallo, Sardegna, Marocco, Francia e Belgio. Dati di confronto sono stati invece valutati per un periodo cronologico e una area geografica più ampi. La procedura combina all'analisi spaziale delle relazioni tra entità prettamente di natura archeologica, quali la localizzazione di siti, miniere, rotte terrestri e marittime, e presenza di manufatti metallici (eseguita con il GIS), lo studio di dati composizionali ottenuti dalle analisi di manufatti la cui peculiarità di produzione è attribuibile ad un' influenza fenicia. Oltre ad un trattamento con il sistema di georeferenziazione, i dati analitici sono elaborati con metodi statistici (Principal Component Analysis). Tale trattamento statistico, costituito in parte anche dallo studio delle distribuzioni di frequenze per alcuni set di dati, ha permesso lo studio di confronti tra produzioni metallurgiche di tipologia e periodo differente, migliorando la conoscenza storica riguardo la comprensione dei trend evolutivi. Il protocollo analitico è stato applicato a vari insiemi di oggetti bronzei scelti per diversa tipologia, per diverse leghe a base-rame (soprattutto bronzo binario, ternario ma anche manufatti in rame) ed allo studio di prodotti intermedi collegabili al ciclo piro-metallurgico quali scorie e semilavorati. I risultati ottenuti in questa ricerca sono da considerare come un contributo archeometrico agli studi archeologici e storici miranti alla definizione della presenza e influenza della cultura fenicio-punica nel bacino del Mediterraneo, aree Atlantiche e aree continentali con l'obiettivo di procedere all'applicazione del protocollo ad altre specificità che gli archeologi sapranno individuare.

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Official presentations of the research for the Ph.D. thesis and activated collaborations

(Conferences, Articles and Projects)

Articles reporting parts of the research:

- Title: Early Iron Age bronze statuettes in Southern Portugal: Combining archaeological data with EDXRF and BSEM+EDS to assess provenance and production technology. N.Schiavon, A.Celauro, M.Manso, A. Brunetti, F.Susanna Journal: Applied Physics Part A , DOI: 10.1007/s00339-013-7747-7

- Title: A roman bronze statuette with gilded silver mask from Sardinia: an EDXRF study. R. Cesareo, A. Brunetti, R. D'Oriano, A. Canu, G.M. Demontis, A. Celauro Journal: Applied Physics Part A , DOI: 10.1007/s00339-013-7721-4

- Title: Archaeometric investigation for provenance studies about copper metallurgy in the Phoenician Punic Culture.

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Conferences (oral contributions and posters) in which parts of the research have been presented:

- Poster accepted for the conference: E-MRS Spring Conference, Strasbourg, may 2013. Title: Archaeometric Investigation using EDXRF, SEM-EDS and GIS System for Provenance Studies about Copper Metallurgy in the Phoenician-Punic Cultures D. Ferro, A. Celauro, N. Schiavon, L. Manfredi, F. Susanna, A. Dekayir, A. Brunetti, D. Pargny

- Presentation of the research work for the Ph.D. thesis during the conference: "Workshop em Ciencias da Terras e do Espaco", Evora, Portugal, 14.02.2013.

Role: presenter of the work and chairwoman of one of the sessions.

Title of the presentation: Archaeometric investigation for provenance studies about copper metallurgy in the Phoenician-Punic culture

A. Celauro, N.Schiavon, D. Ferro, L. Campanella.

- Presentation during the conference: "Numismatica Punica: un'identità ritrovata", Cagliari, 13-15 december, Italy.

Title of the presentation: Analisi spaziale delle monete puniche rinvenute nell'Europa centrale con il sistema d'informazione territoriale GIS

L.Manfredi (presented by l.Manfredi)

- Presentation during the conference: "Numismatica Punica: un'identità ritrovata ", Cagliari, 13-15 december, Italy.

Title of presentation: Dall'analisi chimico-fisica un contributo allo studio di monete puniche.

D.Ferro, A.Celauro (presented by D. Ferro)

- Official presentation of the Biennal Bilateral Agreement CNR/CNRST (Morocco) on ancient mines exploitations, 5 september 2012, CNR Montelibretti. L.Manfredi, D.Ferro, A.Celauro, F.Susanna

- Presentation during conference: Physical and Chemical Analytical Techniques in Cultural Heritage, Lisbon, 4-5 June 2012.

Title of the presentations: Early Iron Age Phoenician Bronze ex votos from Southern Iberia. N.Schiavon, A.Celauro (presented by N. Schiavon)

- Presentation of a Poster during the conference: "IV International Meeting on Experimental Archaeology: metal as a measure of wealth. Experimentation with ancient minting technologies", 12-15 april 2012, Blera.

Title of the poster: Georeferencing and Imaging Techniques for the archaeometric study of Punic Coins"

A. Celauro, L. Manfredi, F. Susanna

Official projects and collaborations activated to accomplish the Doctoral Thesis and for future developments

- Bilateral Scientific Cooperation with ISMA and IIA -CNR and FCT (Fundação para a Ciência e a Tecnologia)- Ministerio da Educação e da Ciencia.

Name of the project: Archaeological, minero-petrographic and geophysical prospection in the Alentejo Area and in the lower district of Rio Guadiana for the recognition of ancient penetration pathways towards the mining basin of Almada de Ouro.

Formally activated around April 2013

Bilateral agreement for the individuation of ancient ore basins exploited by the Phoenicians in the Portuguese and Spanish territories following the Rio Guadiana and with a special attention to Aljustrel basin (famous for the finding of the Vipasca's Tables) and the Almada de Ouro area. The project is carried out using GIS system for the production of thematic maps on ancient mines, SEM-EDS and XRF for the analytical approach and Mass Spectroscopy for the identification of isotopic fingerprint of the ores and for the study of artefacts provenance. Geological and archaeological prospection will be done for the collection of mineral and slag samples. This project is strictly connected with the Moroccan Bilateral Agreement, explained below.

-Sapienza Università di Roma and Universidade de Evora and Hercules Laboratory: agreement for joint Ph.D.

Collaboration for the execution of common research projects for the Ph.D. thesis

-ISCIMA (Institute for the Study of the Italic and Mediterranean Civilization)-CNR (National Research Council), Montelibretti (Italy).

Collaboration of the archaeometric studies of archaeological materials (SEM-EDS, XRF. XRD and other techniques), preparation of thematic maps via Geographic Information System, statistical elaboration of data with PCA and parametric and non parametric tests

- Bilateral scientific cooperation with CNR (Institute ISCIMA and IIA) / CNRST Morocco (Université Moulay Ismail-Meknes).

Name of the Project: "Ancient Mines of Morocco: Archaeological and Archaeometric study from the minerals to the artefacts"

Formally activated: March 2012

Study of ancient mines exploited by Punics through geophysical and archaeological surveys, remote sensing and interpretation of traces of ancient land exploitation, organization of a geodatabase and use of GIS system for the production of thematic maps on old mining areas

- Bilateral agreement with ISCIMA-CNR, GEGENA2 (Groupe d'Etude sur les Géomatériaux et environnements Naturels, Anthropiques et archéologiques) and CRESTIC (Centre de Recherche en Science et Technologie de l'Information et de la Communication) -Université de Reims- Champagne Ardenne

Development of geodatabase on the numismatic Punic and neo-Punic findings discovered in France and Belgium.

Permission to perform non-destructive or micro-destructive analysis given by :

- National Archaeological Museum of Cagliari (Italy);

- Archaeological Museum A.Sanna Sassari (Italy);
- Evora Museum (Portugal);
- Alcacer do Sal Museum (Portugal) and Dr. Esmeralda Gomes;

- Prof Ana Margarida Arruda, Auxiliar researcher for Uniarq – Centro de Arqueologia, Facultade de Letras, Universidade de Lisboa;

- Museo del Vicino Oriente, Sapienza Università di Roma (Italy);
- Sig. Mauro Viola, private collector of numismatics.

Abstract

This doctoral thesis regards an innovative method based on an elaboration of chemical-physical data in terms of statistics and georeferencing of copper-based alloys artefacts in the Mediterranean and Atlantic regions which underwent the influence of Phoenician-Punic cultures.

Fundamental aim of the research is the localization of areas where this influence is evidenced by the presence of artefacts which characteristics can be attributable to Phoenician and Punic presence. As the argument is fairly large, both geographically and chronologically speaking, focalized areas and types of archaeological finds have been selected, in order to have an as wide as possible view of the overall theme. The chronological range considered is mostly between the Late Bronze Age (LBA) and the Early and Middle Iron Age (EIA- MIA), even if more ancient and more recent periods have been considered for coparison. The study of Phoenician-Punic archaeological sites, trade routes and the type of bronze processing have been considered broadly throughout the Mediterranean Basin and surrounding areas (but also atlantic and continental areas), leading to the production of databases containing information on archaeological settlements, mines and ore basins, and compositional data coming from literature or obtained from the analyses of bronze artefacts (SEM-EDS and EDXRF analyses), managed through an unprecedented application of Geographic Information System (GIS tool). So the GIS application includes data coming from archaeometric analytical sessions and from analogous researches published on specialised literature, carried out on samples of different typologies. The collected databases were elaborated on the basis of statistics-mathematic methods, in particular Principal Component Analysis (PCA), while frequency distributions and Box-Whisker diagrams have been used for the study of smaller datasets. The production of manufactures (small size metal statuary), connected to religious worship, were addressed in Alentejo, Portugal (Evora and Alcacer do Sal) and in Sardinia, Italy (Cagliari and Sassari). This part of the thesis has been also further developed thanks to the activation of the master thesis of Valerio Graziani (Sapienza University of Rome, Master Degree in Science Applied to Cultural Heritage), under my tutoring and the supervision of Dr. Daniela Ferro (ISMN-CNR) and Dr. Federico Marini (Sapienza University of Rome), who tested additional applications of GIS and PCA on a more extensive database of analytical data coming from the sites of greatest interest in the Mediterranean. The mining and the production of slags and semi-finished products have been examined for Moroccan area (especially in Meknes Region) while metal artefacts of Roman period from Volubilis archaeological settlement have been analysed in the light of a probable technological conservatism between Punics and Romans. The production of bronze Punic coins was deepened for a private collection (Collection M. Viola) and through the study of Punic and neo-Punic finds in France, Belgium and in lesser extent in Northern continental Europe areas. The results have been interpreted with a unique perspective, so as to allow a new vision on the Phoenician presence an bronze production in the areas where they settled. Further result is the elaboration of a useful tool for the archaeological research that is reproducible in several fields, even different from the ancient metallurgy studies.

Topics

Localization of Phoenician and Punic presences in the Mediterranean and some Atlantic areas; Archaeometallurgy of copper-based alloys; Technological production of Phoenician-Punic manufactory in the Mediterranean Basin, Atlantic and continental areas compared to local productions.

Keywords

Phoenician - Punic metallurgy, EDXRF, SEM-EDS, GIS, XRD, statistic elaborations, bronze artefacts, coins, slags, ingots

Preface

The research carried out is part of a large multidisciplinary study, aimed at the investigation of cultural centres of the Mediterranean regions affected by the influence and presence of the Phoenicians and Punics, by focusing on the localizations of copper-based artefacts, found out of the oriental "motherland", whose production were influenced or ascribable to these cultures, and by the identification of common elements in metallurgical production (some of the results are already published in A.Celauro *et al.*, 2013; N. Schiavon *et al.* 2013, R.Cesareo *et al.* 2013).

One of the aims was the creation of an analytical protocol, suitable for the identification of a connection between copper-based artefacts and a Phoenician, and later Punic presence (viewed in the light of the exploitation of mines, activation of commercial exchange and the foundation of settlements), derived from the results of geographical, analytical and statistical investigations performed on objects from the Late Bronze Age onwards. This research is thus focused on those features that allow the distinction of areas affected by the Phoenician and Punic presence as an allocthonous element. Moreover, as in the most frequented areas the two distinct types of metal production, the local and Phoenician one, merged together producing an intermediate production, which in this thesis will be often called "orientalizing¹", the approach has been addressed in an attempt to isolate elements that can distinguish this third type of production. In fact the Phoenicians and later the Punics, as is well known, established colonies in the coastal areas of the Mediterranean Basin and in many oceanic areas, acting as innovators and dispensers of skills and experience in the artisanal production, during the various contacts with other cultures. The distinction of these types of products is very difficult to perform only from the archaeological point of view. That's why one of the largest efforts has been done in the application of classical methods of archaeometry, statistics and a geographical analysis to achieve more meaningful results, especially in the attempt to distinguish these different productions, even if very often the technological evolution and the mixing of different cultures produces peculiar results which are difficult to standardize. Those results are interpreted in the light of the localization of archaeological settlements, mining areas and diffusion paths that is suggested to have been the purpose and the way of the Phoenician-Punic spreads.

All the artefacts that are part of the whole production "chain", as slags, ingots, semifinished products and metal objects or anything else that is related to the metallurgical process, were therefore selected as objects of the study.

Part of the scientific work planned for the Ph.D. thesis was carried out during these three years in agreement with archaeometric research centres and museums in Italy and also outside the national area. In fact, since each area shows local peculiarity in the relationship established with the allochthonous presence, it was essential to obtain data from the widest possible range of geographical locations.

¹ In this thesis the term orientalizing is sometimes used to identify a typology of objects (solely for aesthetic and plastic aspects), characterized by a non-native, oriental, influence (to be more precise in this thesis the meaning of the term is connected to Phoenician influence); so this definition is, in some cases, disidentified to the chronological sense used in archaeological studies, that's why this word is reported in some paragraph of the text under quotation marks. In some case the definition "Phoenician inspired" artefacts is used to identify artefacts , belonging to a chronological span more recent then the proper Orientalising period

The collaborations activated during the research period are listed below and the connected areas are shown in the picture (fig.1):

- Portugal: the University of Evora, Laboratory HERCULES-Herança Cultural, Estudos and Salvaguarda (joint supervision of doctoral theses), Museu de Evora, Museu de Alcacer do Sal;

Morocco: bilateral agreement with CNR and CNRST, in detail with the Université Moulay Ismail, Meknes, Equipe de Geoxploration & Géotechniques, Faculté des Sciences;
Sardinia: National Archaeological Museum of Cagliari and Museum A. Sanna of Sassari and Sassari University;

France: Université de Reims Champagne-Ardenne, institutes CRESTIC and GEGENA²
ISMA: Istituto Studi Mediterraneo Antico – CNR, Montelibretti, Italy.

In the Museums of Evora and Alcacer do Sal, collections of anthropomorphic and



zoomorphic small bronze statuary were studied while, in the Museums of Cagliari and Sassari, nuragic bronze statuettes, one thymiaterion and also tools, such as axes and razors, and half-finished objects as plano-convex and oxhide ingots, have been selected and studied. In Rome it was possible to analyse one small object from the

collection of the Near East Museum, Sapienza University of Rome. With regard to Punic production, it was chosen to perform archaeometric and spatial investigations of coins collections of different types: private collection in Rome (M. Viola collection); Punic and neo-Punic coins found in France and Belgium and northern areas (work conducted in collaboration with the Université de Reims, in the Champagne-Ardenne region). In Morocco a field research has been planned for the localization of mineral basins of ancient exploitation.

The research required activity of a highly itinerant aspect, for this reason portable equipment such as XRF instrumentation have been used, with Monte Carlo simulation and

method of quantification and WinAxil X-Ray analysis software, while non-portable classic instrumentations such as the SEM-EDS or XRD were used only where these equipment were available or when it was possible to move samples. The application of mass spectrometry has been limited to analysis of a single sample (a fragment of lead leaf from Volubilis). Furthermore, because of the primary requirement, in some of the cases under study, to not minimally damage the objects conserved in museums, it was necessary to take into account also the total non-destructiveness or the minimal invasiveness of the analytical method applied. The main results of the sessions, all the steps of the analytical study and the conclusions that have been reached are exposed in the following sections. First of all the historical and geographical background will be reported, in the introduction, in order to set a generic platform that allows to understand the choices made in the selection of research areas and classes of artefacts. Right after the introduction, the research procedures and guidelines will be briefly explained. Whereupon each chapter will have a short but focused introduction to archaeological sites and objects under study. The sequence of chapters and treatments of the data is organized in chronological order. First of all the thesis reports the results obtained for small statuary in Portugal and Sardinia. After the exposition of these topics the discussion will face the research on ancient mines in Morocco and the study of artefacts from Volubilis archaeological settlement. In conclusion will be presented the study on massive presence of Punic and neo-Punic coins in France and Belgium and lastly the archaeometric study on Punic coins of private collection. Following this order, the research covers a chronological period ranging from the Late Bronze Age to the Roman Empire, in other word to the almost total disappearance of Punic presence from the Mediterranean.

Insights on the thesis title and further exploration of the thesis topics

Given the multitude of these purposes, the choice of a title that explains all aspects of the research was an issue that has been widely discussed. We decided to define these different themes under the term "*provenance studies*", even if the term "provenance" has a different meaning in each chapter. Explaining these aspects, a deepen glance on this research is also proposed.

First of all, the analyses on Evora votive statuary collection, whose origin was lost since it was a private collection in the XVIII century, have been carried out with the precise intent to reconnect those artefacts to an archaeological settlement of provenance. In order to

succeed in this intent it has been necessary to enlarge the studied topic to the bronze metallurgy in the Iberian Peninsula, allowing to reach more general conclusions. Another aspect, which can be referred to the theme of provenance, was the study of a cultural attribution of these artefacts, since they show features that are not typically local (allochtonous) but which can be ascribed partly to Phoenician bronze production. Another "provenance" aspect was the localization of ore basins that probably were the aim of Phoenician and Punic voyages of exploration in Portugal.

The cultural provenance is the element that guided also the selection of nuragic bronze statuettes in Sardinia. In fact some objects have been chosen from the local types while the major part manifests features in common with Phoenician votive artefacts. Also in this case the compositional data were interpreted in the light of Phoenician presence, archaeological settlements, mines and harbours location in the island.

A study of provenance of the raw materials from the ores has been executed for the Moroccan area, since the research aims is the localization of the ancient mines exploited by Punics.

At last the research on Punic coins has been faced with a double purpose: first of all their localization in foreigner areas (especially France, Belgium, but also Estonia, Ireland, UK, Poland, Germany to list some examples), far from the minting sites, has been used to advance hypothesis of Punic frequentation. The presence in those areas is not widely contemplated by the classical approaches, and here is connected again with the Punic commercial purpose (for minerals but also other precious goods) that pushed them in very far away areas. Secondly an archaeometric approach has been applied for the study of a widespread type of Punic coinage, for which the classical numismatic interpretation propose Carthage as emitting authority, with the aim of discovering the existence of subspecies, emitted in secondary mints and under the control of Carthage. So, in the first case, the provenance is intended as the study of the reasons why an high amount of foreigner coins can be found in far-off areas (again in connection with route and paths used to penetrate in foreign areas, and with the localization of ore basins which are often the reason of the Phoenician-Punic spread), while in the second case-study, the discovery of coinage subspecies can suggest the provenance from different emitting areas.

A last clarification is made to explain the used term: copper metallurgy. Since this research involves different typologies of artefacts, coming from different geographical areas and chronological period, the study regards a wide compositional range, from copper artefacts

to the production of ternary bronze. In this way a generic wording is used allowing to involve also very different kind of productions. Since this research is to be meant as a preliminary approach to a wider project that we hope will be long lasting, especially considering the vastness of the subject, this thesis proved to be a good testing ground for the proposed research protocol.

Bibliography of this chapter

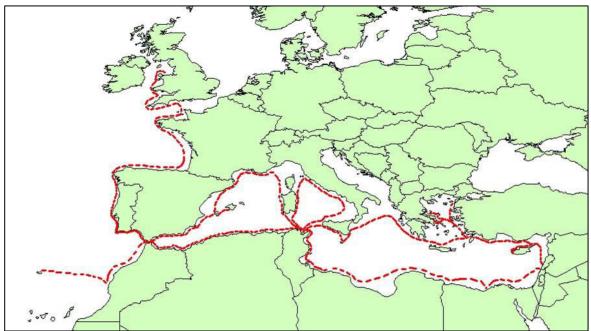
A. Celauro, N. Schiavon, L. Campanella, D. Ferro, *Archaeometric investigation for provenance studies about copper metallurgy in the Phoenician Punic Culture*, in Conference Acta: Workshop em Ciencias da Terras e do Espaco, Livro de Acta 2013, programa Doutoral en Ciencias da Terra e do espaco, Universidade de Evora, Coordenandor: Mourad Bezzeghoud, fevreiro 2013, © Universidade de Evora, ISBN: 978-989-98196-2-7

N.Schiavon, A.Celauro, M.Manso, A. Brunetti, F.Susanna, *Early Iron Age bronze statuettes in Southern Portugal: Combining archaeological data with EDXRF and BSEM+EDS to assess provenance and production technology*, in Applied Physics Part A , DOI: 10.1007/s00339-013-7747-7

R. Cesareo, A. Brunetti, R. D'Oriano, A. Canu, G.M. Demontis, A. Celauro, *A roman bronze statuette with gilded silver mask from Sardinia: an EDXRF study*, in Applied Physics Part A, DOI: 10.1007/s00339-013-7721-4

CHAPTER I

HISTORICAL AND GEOGRAPHICAL OUTLINES PHOENICIAN AND PUNIC CULTURES FROM THE MOTHERLAND TO THE WESTERN AREAS



Phoenician and Punic naval routes in the Mediterranean basin and in Atlantic areas.

Thematic map created with GIS system for the Ph.D. thesis

To pursue the aim of the thesis is necessary to identify the places where the Phoenicians and Punics came into contact with the local populations, in the chronological period identified as the greatest expansion of the Phoenicians-Punics in the Mediterranean basin (IX-III BC). The term Phoenician refers to the population which, coming from the coast of Lebanon, expanded commercial trade to the Mediterranean, without ever establishing a true colonizing power, but rather a constellation of *emporia*. Phoenicians of Tyre settled in Tunisia founding Carthage which became one of the most important powers in the Mediterranean, expanding their influence to all Northern Africa. This huge establishment is called Punic Culture, which had properly colonizing characteristics. This short aside, just to state a common cultural origin of these two separates identities, will be explained in a more detailed outlook in the following sections,

Many problems arise from the interpretation and definition of the identity of the Phoenician population, who are just well known as merchants of the seas; much more difficult is to establish if they had a country of their own and if they had any geographical boundaries. According to Herodotus (VIII, 89), the Phoenicians came from the Persian Gulf, belonging to ethnic group of Semites (Gras *et al.*, 2000). The discovery of Phoenician inscriptions provides a clearer idea of the situation: terms like "Phoenician" or "Phoenicia" never occur, but other terms linking to narrower identities are mentioned, like Sidonian, Tyrians, Ghiblytes etc... The Phoenicians never refer to themselves as Phoenicians. This population did not have a self-referential term. Another used term, Canaanites, has a geographically expanded meaning and appears in the texts of the II millennium BC and in the Old Testament (Gras *et al.*, 2000).

The greek translation of the term Canaanites means Phoinix and Canaan: these two words mean both the red color, indicating their most important product of trade, the murex, and the purple made from its powdering, remarking their commercial attitude. But this wasn't the only product of their trades, they were also famous for the precious cedar wood and other luxury goods, which they exchanged with many products they were lacking, such as ores. It is well known that the Phoenicians considered themselves a maritime confederation and shared a common cultural heritage, which could have appeared as fairly homogeneous to a foreigner observer (Gras et al., 2000). In conclusion, for the people of foreign countries the word "Phoenicians" had functional meanings rather than geographic and ethnical ones.

They were considered essentially the non-Greeks, the Orientals, since the East was truly distant to the West, not only geographically speaking (Bonnet, 2005).

1.1 Chronology

With regard to the chronology, several evidences indicate an occupation of the Phoenician coast since prehistoric times. Already in the III millennium BC there was a very rich kingdom in Byblos, the center of a commercial network with Egypt. Historians are not sure if at that time it is already possible to speak of the Phoenicians in *strictu sensu*. Several periods in the history of this civilization can be distinguished:

the first from 1200 BC to the IX century BC, which was the period of political autonomy,
the second, from the IX century to 612 BC, in which the Phoenicia was part of the Assyrian orbit,

- a third period, up to 539 BC, of Babylonian domination

- fourth and last period, until 330 BC, of Persian domination.

Since about 1200 BC the Phoenicians expanded in the Mediterranean in search of new mining sources and new markets: throughout the whole chronological span they were protagonists of the maritime commercial trade (Bonnet, 2005; Moscati, 1992).



Tightened in a small area between the mountains of Lebanon and the Mediterranean, the Phoenicians very early projected themselves towards the sea, seen as an extension of motherland, and this is the reason why they pushed themselves more and more into the West.

The Phoenician country was composed by a series of city-state, located on the coastal areas, with one or more commercial and military ports, and a less extended inland area: their "nation" approximately corresponded to the Lebanon and Anti-Lebanon mountains, separated by the Beqaa valley (Gras *et al.*, 2005). In this land there were vast forests of cedar wood, a precious commodity. The horizontal circulation in the West-East direction, towards Syria and Mesopotamia, was difficult, despite there were several passages such as Homs, in the northern part of the kingdom, near Arwad. So Phoenicia was in a way

protected, but also isolated from the big eastern centers of power, a situation that forced them to go westward. The North-South vertical communications were facilitated seaborne, this is why the contacts with Egypt were constant. The southern route then continued in the direction of Cyrenaica and North Africa by sea; this path also involved the southern islands of the Aegean, in particular Crete. Towards the North, the Phoenicians reached Syria, Cyprus, Minor Asia, and from there they reached Greece. The presence of names such as Phoinix, Phoinike or Phoinicus along these routes in Lycia, Cilicia, Caria, Rhodes, Crete, indicates that relationships were intense. The Phoenician territory is at the crossroads of two major trade routes linking Egypt to Anatolia NS and Mesopotamia/Iran and the Mediterranean EW. This was a strategic position of the Syrian-Palestinian interface since the Bronze Age. During the Iron Age only four major areas were reported, to which the others referred: Arwad, Byblos, Sidon and Tyre, whilst, during the Late Bronze Age, around twenty locations can be enucleated. In the northern Phoenicia, Arwad (today Ruad in Syria) was an important city located on an island like Tyre. Unfortunately the archaeological traces are almost non-existent, since the island has never been subjected to systematic excavation. Tell Kazel in Syria, today known as Sumura, was frequented till the Middle Bronze Age; in the territory of this city were also included the sites of Tell Arqa and Tripoli, both in Lebanon. Byblos, today Gebeil, had intense commercial relations with Egypt for the selling of cedar, juniper and pine, materials absent in Egypt, while Beirut acquired from the Egyptians the precious papyrus. Biruta, currently Beirut, hosted port activities. In southern Phoenicia, Sidon and Tyre were the main cities, but also Sarepta was an important economic center, where is archaeologically documented the economic activity of the purchase of purple, textile, metal, ceramics and jewelry. During the Late Period, some merchants of Sarepta went so far as to reach the Tyrrhenian Sea, as documented by the idol of Sarepta mentioned in an inscription in Pozzuoli. The Phoenician presence in Akhziv and Akko in Israel shows that it is impossible to draw a line between Phoenicia and Israel/Palestine (Aubet, 2007; Barreca et al., 1971).

All the listed areas are the maritime capitals whose survival was ensured by a network of secondary settlements with an agricultural vocation. The Phoenician economy was then made of commercial relations with the Mediterranean but also with Syria, Anatolia, Palestine and Mesopotamia. Metallurgical production was remarkable although Phoenicia was almost devoid of metal sources, and this is the fundamental clue for this thesis. The Phoenicians were known as specialists in metallurgy and their expansion was mainly

related to the research of metals: Cyprus, Anatolia, in Sardinia in the Iberian peninsula and later, with the Punic also in North Africa (Barreca *et al.* 1971; Ruiz Mata 2000). The Phoenicians left the extraction of minerals to local people, while reserved for themselves the processing and export of metals.

The Phoenicians taught the techniques of metalworking in many places (Crete and Euboea Pitecussai), to the Greeks and the other local populations, in a continuous technological transfer, thus starting the so-called Orientalizing Art, characteristic of the VIII and VI centuries BC in the Mediterranean basin. In fact, these objects of great value were in the earliest periods donated to princes of the local aristocracy and elites, in exchange for access to mineral resources (iron, lead, silver, copper, tin, gold), and on the other hand, these objects constituted the final product of the same raw materials processing, closing in this way the circle of commercial circuits in which the Phoenicians offered technologies (marine, metal, jewelery) and finished products (Giuntoli & Pedrazzi, 2006). The maritime expansion could take two different forms: on one side an expansion of trade and on the other the creation of real colonies. In these settlements the Phoenicians lived in close contact with local populations, introducing their religious uses and the technical expertise that was gradually assimilated by the local population (eg. granulation technique in jewelry). The Phoenician applied a fairly homogeneous settlement pattern throughout the Mediterranean, Egypt, Libya, Anatolia, Aegean, Sardinia, Tyrrhenian and Sicily, North Africa and Iberian Peninsula. Sometimes the expansion became a proper colonization, whereas the structures went permanent as the exchanges became perennials: these are Cadiz, Lixus and Carthage in Tunisia. These are strategic locations that close access to Sicily and Gibraltar (Barreca et al., 1971).

1.3 The trade routes in the Mediterranean

Since the Phoenician expansion was carried through coastal shipping, the landing points were set near headlands and islands, lagoons or estuaries where it was easier to land and find shelter. At first these were nothing but thinly populated commercial ports, in which many events took place: water and food supply, exchange of manufactured objects and goods of various kinds with local products. Through these routes, the Phoenicians gave rise to a great civilization, contributing to the creation of a Mediterranean cultural koine ("commonality") (Aubet, 2007).

Syria, Palestine and Mesopotamia



These are the earliest affected areas because these were the closest ones, and served as a granary for cities with limited hinterland. The Phoenician footprint in Israel is very strong in the building techniques, the alphabet, cults and ceramic processing. In Syria, the Phoenician presence is certifiable at least from the Bronze Final. The excavations at Ras el Bassin and Al Mina detect the presence of numerous Phoenician and Cypriot objects mixed with Greek materials. With regard to Mesopotamia, the Assyrians were so much interested in the Phoenician harbors that these territories were then subjected to a heavy tribute but also a strong commercial incentive. In fact, the Assyrians opened the market to the Phoenicians and encouraged the exploration of metal in Anatolia and the Mediterranean. The exceptional skill of the Phoenician craftsmen in the processing of metal, wood and ivory ensured their presence in the Assyrians kingdom. The two most important colonial cities in this area were Ruad and Ugarit (Bonnet, 2005; Grass, 2000).

Anatolia



The Phoenician presence in Anatolia is documented by a series of Phoenician or Phoenician-Luvites inscriptions. From the harbors of northern Syria, Phoenician sailors reached the Cilician coast easily. There was also a land route to reach the luvites realms or Aramaic Anatolia, and finally arrive at the vast mineral resources of Amano and Taurus. There was a cultural Koinè between Phoenicia, Syria and southern Anatolia. It is noted that in fact existed a well-documented commonality of various cults in Anatolia such as Malqart and Baal (Bonnet, 2005).



Egypt

Since the time of the Second dynasty of Egypt, at the beginning of the third millennium BC the relations between Egypt and the centers of the Phoenicia flourished. One of the mainly

contacted sites was Byblos, which provided wood and resin, essential for mummification, metals and precious goods. Numerous objects found in Egypt bear inscriptions of the Phoenician members of foreign communities in Memphis, from the Delta and the oasis near the east side of the Nile. The Phoenician district of Memphis has, for example, a sanctuary dedicated to the Phoenician goddess Astarte with a characteristic style of the hegemony of the Libyan Pharaohs XXII, XXIII and XXIV dynasty that allows to assume the presence of Phoenician artists at service of the Pharaohs. Between 664 and 525 BC the presence of Phoenician sailors and mercenaries was attested in Egypt: according to tradition, the first ones, upon request of the Pharaoh Nação, undertook the exploration that lead to the circumnavigation of Africa, while the latter accompanied the Pharaohs in Nubia. The social revolution that shook Egypt around the late III millennium BC, interrupted for more than a century the relations with Phoenicia. Between the XX and XVIII centuries BC, the Egyptian rebirth makes again possible relations between Egypt and the Phoenician centers. But in the XVIII century BC, the advent to the throne of the Pharaohs Hyksos, resulting in a further period of slackening in the relationship between the Phoenician area and the Egyptian empire (Aubet, 2007; Bonnet, 2005).

Cyprus and Aegean



The strategic location of Cyprus in the heart of the Gulf of Alexandretta, combined with its wealth of copper in the Mount Troodo, explain the great interest of the Phoenicians in this area. The excavations of Enkomi, Paphos, Kition revealed a composite civilization. Kition is the location of a real Phoenician-Cypriot kingdom which can be called as the "cypriot Carthage". Other Phoenician kingdoms appear in literary sources: Paphos, Amathus or Lapithos. The Aegean is therefore an area where the Phoenicians moved with ease. Their presence in Rhodes, Samos, the Dodecanese was, without doubt, gave to Phoenicians the way to reach Attica and Euboea. The findings of Lefkandi, Eretriao and of the Athenians necropolis demonstrate the presence of very stable trade. The island of Crete has a remarkable role in the trade between the Mycenaean, Phoenician, Syria and with Egypt. In the necropolis of Tekke (Knossos) there is a Phoenician inscription dating back to 900 BC probably attributable to a Phoenician goldsmith settled there (Barreca, 1971).

Carthage and North Africa

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The Phoenician presence in North Africa in a short time reached rapidly Tunisia. Phoenician sites have been found in large numbers both on the coast and inland. It is possible, therefore, to speak, in the case of Tunisia, of a total penetration. Among the main settlements can be cited Utica, Hadrumetum, Kerkouane in the region of Cap Bon, Thapsus, Djerba, Maktar, Dougga and Sicca, but despite this massive presence in Tunisia, Carthage (which means "new city"), in near today's Tunis, however, remained the undisputed capital of a brilliant civilization. The findings from the latest excavations date the foundation of the city in 814 BC, confirming the date provided by the classical sources that attribute the foundation to the princess of Tyre, Elissa. At the origin of Carthage edification, in that land there was mainly the Assyrian and Greek authority, which created a constant menace to the Phoenician colonies along the Mediterranean coast, but Carthage always maintained friendly relations with its neighbors and quickly assumed a position of hegemony, and later became an economic and commercial empire, creator in its own turn of new colonies (these new colonies are Punic, meaning with this term a Carthaginian belonging). In 264 BC Carthage clashed with the Roman power: this is how the Punic Wars started, which lasted for about a century and will be completed in 146 BC, marking the end of Carthaginian power by Scipio Aemilianus. Both the port and the city was burnt completely. In fact, during recent excavations, archaeologists have found remains in damaged condition under a thick layer of ash. Whether were serious destruction on major Tunisian cities, Kerkouane, dating to the IV century BC, however, is the best preserved city in the Punic world. The oldest evidence brought to light show the existence of an urban plan, with houses richly decorated and equipped with every comfort and a large sanctuary and necropolis of Areg El-Ghazouani have been excavated, in addition. This city is now

Libya



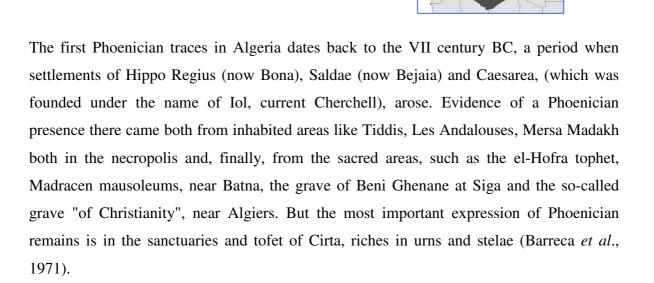
The Libyan coast, since the II

millennium BC, played an important role in the development of Mediterranean civilization,

included among the World Heritage Sites of UNESCO (Gras, 2000).

in fact the traders, with their insecure boats and in need of frequent stops, kept close to the coast line, creating several landing points. Around 1000 BC, the Phoenicians landed on the coast of Libya, founding several ports including Leptis, the Oea (now Tripoli) and Sabratha, which experienced the greatest period of splendor. These three sites soon turned into commercial ports (emporia) and, finally, in real cities. From these three centers hence the name Tripoli, the 'three cities', which together with the Cyrenaica, experienced higher growth than other Libyan areas. Little is left of defensive walls and the urban structure of emporia founded by the Phoenicians and Carthaginians, as these were later transformed into the well known Roman towns. The most important finds that testify the Phoenician presence in Libya, are those related to the cult of the dead: funerary stelae, pools the offerings to the dead, sarcophagi, engraved stones and bas-reliefs representing at times the icon of Tanit, the goddess of fertility (Barreca *et al.*, 1971; Gras, 2000).

Algeria



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Morocco



The Phoenician presence in Morocco is evidenced by the classical authors, telling of the foundation of numerous sites. According to Strabo, the Phoenicians founded 300 colonies along the Atlantic coast of Morocco including Rusaddir (now Melilla), Emsa, Tamuda, Tangier, Lixus and Mogador (now Larache). Excavations have brought to light numerous

necropolises identified and excavated in Tangier, Lixus, Melilla and Tit in which were found pottery, jewelry and amulets. In the other sites the Phoenician presence is shown by other artefacts such as pottery, tools, jewelry, amulets and inscriptions. In 1966 was published a collection of Punic and neo-Punic inscriptions discovered in Morocco: from this work was deduced that the language of the Phoenicians and the Carthaginians was very prevalent in this country. Furthermore the Phoenicians influenced all fields of the Moroccan daily life: language, religion and even the world of the dead and the afterlife (Barreca *et al.*, 1971).

Italian Peninsula and islands

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The Phoenician interest in the Tyrrhenian area, in Etruria, the coast of Lazio, Campania and Calabria and Sardinia, is derived from the presence of significant mineral resources which attracted many other populations. From the VIII century. BC the Phoenicians in Sardinia create many settlements as Nora, Tharros, Sulcis, Monte Sirai, Olbia etc ... In Sicily, the local communities had many contacts with the Greek populations. There are many testimonies of conflict in the VI and V century, which saw the Greeks opposed to Carthaginians for the control of the island. In fact, Sicily, placed in the middle of the Mediterranean, has a strategic position that arouses the desire of both entities. Overall, however, prevailed the Phoenicians and their presence in Sicily is concentrated in the west of the island: Mozia, Palermo, Solunto, Erice, Trapani, Marsala. The Phoenicians are not only present on the territory of Sicily and Sardinia, but also in other areas of the peninsula. Is enough to remember the discovery of three gold *laminae* in Pyrgi (now Santa Severa in the municipality of Santa Marinella, on the Lazio coast north of Rome) written in Phoenician and Etruscan, constituting a previously unimaginable evidence of a Phoenician influence on the Etruscans. Also the findings at Ischia and Liguria which indicate a very active trade within the Tyrrhenian Sea (Barreca et al., 1971; Bonnet, 2005).

Malta



The geographical position of the Maltese archipelago, its ports and its cultural history

enable us to understand why the Phoenician occupied these places. Malta was a strategic point for trade, hence its name, originally Malet, which means "shelter". The country had a projected outward economy, by the way of sea, in the form of barter trade and piracy, due to lack of mineral resources and the limited land available for agricultural use. The multiplicity of external relations may be encountered in the presence, at the site of Bahirija, of foreign settlers, groups of Maltese reaching the eastern Sicily and the finding of Mycenaean pottery. In addition, the Phoenician presence in Malta is confirmed by the discovery of pottery with red slip of the VIII century BC in indigenous settlements, proof of cohabitation between the two groups. The two main cities the geographer Ptolemy mentioned for Malta were: Marsaxlokk, Rabat-Mdina and for the island of Gozo, the site of Victoria. Among the most famous places of worship there is the great suburban sanctuary of Tas-Silg, dedicated to the Phoenician goddess Astarte, dating to the Eneolithic period where the structures and systems continues to be used for all the Phoenician and Punic centuries, at least until the I century BC (Barreca *et al.*, 1971; Bonnet, 2005).

Spain

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The Phoenician presence is concentrated on the coasts of Andalusia, close to the Tartessos areas, rich in mines. The Phoenician colonization in Spain responds to a fundamental aim: to gain control of the sources of the metal trade. One of the main centers in Spain is the ancient Phoenician Gadir, which overlaps the modern city of Cadiz, which was a colony of great importance, as evidenced by its sphere of economic and cultural influence that extended into the Atlantic Morocco (Lixus and Mogador) to Rochgoun and Ibiza. Its foundation was responding to a purely economic need for gain of silver ore in a period when there was a strong demand in the East by the great powers of the inner area, and in particular Assyria. The ancient Gadir occupied a small archipelago located in the center of a bay, overlooking the estuary of the river Guadalete: the smallest islands housed the Phoenician village, while the greater were the Punic necropolis and the temple of Melqart. Along the eastern coast of Andalusia there was an enormous concentration of Phoenician settlements, in the area of Tartessos (this area is well explained in Chapter III). Colonies, dated between 750 and 550 BC, have in common the location on a low coastal promontory, at the mouth of a river (Toscanos, Morro de Mezquitilla, Charreras), on a peninsula

overlooking a floodplain (Almuñécar), or on an island in the middle of an estuary as Cerro del Villar del Guadalhorce. This position gave the *emporia* a double strategic advantage: ensured landing condition, as ships could be moored in shelters and protected from winds and currents, and to guarantee settlers the easy exploitation of alluvial soils close to the river, which even today are very fertile soil. The necropolises stood a short distance from the town. Towards the beginning of the VI century BC is noted the emergence of a crisis that culminated around 550 BC with the general abandonment of the area probably due to a reorganization of the settlements in the eastern Andalusia. The causes of the crisis are to be found in the fall of Tyre in the hands of Nebuchadnezzar (586-573 BC), or should be connected to the fall of Tartessos in Eastern Andalusia. After the crisis Spain returns to be occupied in the towns of eastern Andalusia, but the culture is, in that period, properly Punic. Great strategic importance had the island of Ibiza, an obligatory stop in the shipping routes through the western Mediterranean. In the second half of the VII century BC Phoenician population groups, from Gadir, settled in different parts of the southern coast of the island. The colonization coincides with the moment of greatest economic prosperity of the Phoenician colonies of Andalusia, around 630 BC, attempted to expand their sphere of business, going to the Gulf of Lyon, in search of tin and other raw materials. Towards the middle of the sixth century BC this circuit is interrupted by the commercial crisis of the Phoenician colonies in Spain and, shortly after, the island enters the orbit of political influence of Carthage. The most famous evidence of Punic presence in Ibiza are the vast necropolis of Puig d 'es Molins, and the two major sanctuaries of Isla Plana and the Cave Es Cuyram. Later, under the influence of Carthage, other cities in Spain pointed out: as Cartagena and Sagunto. Finally is to be mentioned Malaka, today Malaga, a commercial center of great importance (Barreca et al., 2002; Bonnet, 2005; Ruiz Mata, 2000; Schubart et al., 2002).

Portugal



Through an enviable knowledge of the navigation art, the Phoenicians made journeys of exploration for commercial purposes, in search of precious metals reaching also Portugal, which is on the extreme West side of their usual paths. In an attempt to open a new trade

routes for these ores as an alternative to continental path through France and Marseille, the Phoenicians undertook other trips to distant lands; some ancient authors speak of possible routes to the Azores isles (at a considerable distance off the Portuguese coast) and also Madera archipelago (off the Moroccan coast). The most recent archaeological investigations testify, moreover, the presence of Phoenician settlements along the coast of Portugal for reasons of commercial expansion (Abul, Sines, Alcacer do Sal etc...). After the Phoenicians also the Punic vessels started to frequent this area. Surely the Carthaginians in 500 BC had become the undisputed masters of the Mediterranean, replacing the Greeks, following the destruction of the city of Tartessos, which was the key commercial center on the Spanish coast beyond the Straits of Gibraltar. Masters of the only way of connecting with the Atlantic, the Carthaginians took possession of the northern route that led to Cornwall, the land of the tin. So during the succession of the centuries the influence of Phoenicians and Punics has always characterized these territories (Arruda, 2000; Bonnet, 2005).

France and Great Britain

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Must not be forget that the Phoenicians were also present in France and Britain. The southern part of France and the area around the present city of Marseilles, were affected about the VIII-VII century BC from voyages of exploration for commercial purposes made by the Phoenicians in search of precious metals, tin and other raw materials. The Phoenicians, in fact, after the founding of Carthage in 814 BC, attempted to expand their sphere of business, going to the west coast of Sardinia, in the North, the Balearic Islands and the Gulf of Lyon. Certainly they frequented the coasts of southern Provence since the mythical city called Heraclea is famous, identified in the Espeyran, between the lagoons of the Camargue (Saint-Gilles-du-Gard). As far as the presence in Britain is not so clear, there are lots of traces of Phoenician-Punic settlements in this area. It can be, however, mentioned in this regard the journey made during the V century BC by Carthaginian Imilcone that, pointing to the north and along the ocean coast of France, reached Britain, and perhaps the cold Cassiterides islands (Isles of Scilly, Cornwall and Ireland) for the highly coveted deposits of tin (Aubet, 2007).

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It is said that the Phoenicians have visited also the American continent. The alleged most sensational discovery was announced in 1874, when the "at that time director" of the National Museum of Rio de Janeiro, Professor Ladislao Netto discovered an incision with Phoenician characters discovered at the currently named Joao Pessoa, Brazil, at the northern Atlantic coast, in which was reported the following story:

"We are children of the Land of Canaan from Sidon, the city of the king. The commerce has thrown us on this distant shore, a land of mountains. We sacrificed a young man to the Very Highs and now in the 19th year of Hiram, our mighty king. We embarked from Ezion Geber, in the Red Sea and traveled with 10 ships. We were into the sea together for two years around the land of Ham (Africa), but we were separated from the hand of Baal"

The inscription was denounced as a fake by the major scholars of the Semitic language of the time and than it was forgotten. In 1968 the inscription was re-examined by a Semitist, Gordon, who declared the authenticity. It remains in doubt the veracity of this precious finding, which moreover has never been possible to recover the original. On a purely theoretical point of view the arrival of a traditional boat from the Atlantic coast of Africa to the north of Brazil is not excluded by geographers (Moscati,1992).

1.4 The Phoenician and Punic artistic productions

To fully understand the purpose of the thesis, in this chapter are also briefly discussed the classes of metal artefacts that were commonly produced by Phoenician and Punic artisans.

The main activity with which the Phoenicians appear in the Mediterranean world is the metallurgical handicrafts, mostly composed of small objects made of the finest materials that were traded or sold along the Mediterranean routes. The question arises whether it should be consider art or not, particularly given the anonymity of the authors that contribute to give a collective and serial feature to the production. The problem is not only related to the formal aspect but also to the repetition, through time and space, of same typologies and iconography that does not change, for which is difficult to understand where an object is created and when it was produced. So the repetition of subjects obstructs the recognition of individual artistic personalities but makes it easy an assignment of a finding to the

Phoenician archeology. In addition, the lack of study of the Phoenician production has hindered the reading of these processes in the light of authors autonomy, that a careful study of the object may indicate. The Phoenicians products have the characteristic to be articulate for genres and categories: *stelae*, terracotta figurines, proteomes and masks, jewelry and statuettes (Moscati, 1992). From ancient literary sources it appears that the Phoenicians were excellent artists of metal working and that their ateliers produced a refined quality. From a Ugaritic text (KTU 1.4 1 23) are listed many beautiful objects produced for the deity Athirat from the divine Kothar, including a finely chiseled cup. From the Homeric poems (II. 23.741, Od. 4, 615, XV 115) are famous vessels of damascene silver and Sidonic workmanship. Neo-Assyrian texts tell about different objects in gold, silver and bronze appear as paid tribute to the rulers of Assour from the cities of the Amurru coast (Moscati, 1992).

From the Old Testament (1R.7, 13-14; 2Chr.2, 13) are described admirable cast works made by a old Tyrian artisan for the Temple of Solomon in Jerusalem. Finally, Ez.27 .12-22 appears to show the importance that covered the metals in trade of Tyre with distant countries for the supply of mineral resources. The search of metals is one of the main reasons of the Phoenician expansion in the Mediterranean, and it is this reason that leads the Phoenicians in Etruria, in Sardinia and the Iberian Peninsula and North Africa, and this is always the goal that characterizes their maritime activities and their business strategies. This has encouraged the development of a thriving metallurgical industry in the homeland during the first half of the I millennium BC, industry which finds its foundations in the important know-how that they took from the Canaanite civilization of the II millennium. Apart from precious metals like gold and silver, the Phoenician metallurgists continued to use mainly bronze and seldom lead. Iron, introduced in the region after 1200 BC with the arrival of the Sea Peoples, was very useful for forging weapons and other tools but were unusable in the decorative arts. This explains the flourishing of an industry of which remains archaeological records confirming the specialist literary information (in general Tylecote, 1976; Muhly, Maddin & Karageorghis 1982; Rothenberg & Blanco Freijeiro, 1982; Curtis, 1988; Mitchell, 1988; Lipinsky, 1992). The first discoveries of metal products manufacturing date from the XIX century BC, in the East and in the Mediterranean. In 1849, A.H. Layard, discovered at Nimroud, in the North East Palace of Assournassirpal II a rich deposit of bronzes including more than a hundred cantilevered cups attributed to Phoenician art. Some findings were discovered in Etruria in the graves of Caere and Preneste and also in Cyprus, Crete and Greece during the excavations of the grout of Ida and in the cells of Olympia. All these discoveries caught the attention of the Hellenists and etruscologists who pointed out the key role played by Phoenician and Oriental metalworking in the spread of the Orientalizing phenomenon. The discoveries continued until the present day and prove that these products were objects of a great diffusion and were not just luxury goods for the Phoenicians themselves, but were also massively exported. These weren't only the objects of trading but were also used as gifts or tributes paid to kings, war booty, votive offerings in the great sanctuaries and funerary offerings for deceased members of high rank.

The arts of metals in Phoenician world can be divided into three categories: toreutics, the small size statuary, and tools and objects of different type. It should be mentioned that the examination of these categories is not easy, either because the archaeological evidence is insufficient in the Phoenician Motherland and also because many discoveries came from illicit excavations and the information from the context is completely lost. As a result is not possible to have no more than the typological and stylistic grouping to try a cultural attribution of the metalworking(Giuntoli & Pedrazzi, 2006).

Jewelry



Jewelry in gold, silver, bronze, glass and precious stones represent a significant sector of Phoenician art (Gras, 2000). The development of this kind of artefacts, thanks to the possibilities offered by the trading of metals along the routes of the Mediterranean, is linked to a rich artistic tradition that precedes the Phoenicians in the Syrian-Palestinian area (Moscati, 1992). The conservatism, that manifests itself in the types and technologies applied in the decorative repertoire, makes it difficult to distinguish between the production of both East and West, where for example Tharros appears to be a particularly active center of production (Gras, 2000). Regarding the materials, the jewelry are mostly made of gold and silver as well as bronze, precious stones and colored glass especially for bracelets and necklaces. The prominent techniques is the embossing, granulation and filigree that generally characterize the oldest objects (Giuntoli & Pedrazzi, 2006).

Cups



A production of refined elegance divided into small numbers of high value objects, is that of the cups of gold, silver and bronze, embossed with complex ornamental figures. These engraved Phoenician cups show a high technical capacity. No evidence is currently found in the Phoenician motherland and period. These were found, however, in the immediate precedents time, at Ugarit site and also from Assyria, in Cyprus, Greece and Italy, in parallel to the diffusion Phoenician craft. It is interesting to follow the irradiation of this typology in the Aegean: in Olympia, Athens, Sparta, Delphi, and especially the island of Crete. The cups discovered in Assyria at Nimrud have common characteristics: the presence of a central rosette The cups discovered in Assyria at Nimrud have common characteristics: the presence of a rosette in the center, decoration in concentric bands, animals, humans, but also geometric patterns, the latter with a not only decorative function but also a dividing mean. In the simpler cups prevail ornaments with no the development of narrative figuration. A very rich documentation comes from Italy: six cups comes from Preneste, four from Cerveteri, one from Pontecagnano (Salerno), one from Macchiabate at Sybaris and a fragmentary specimen from Vetulonia. The cups of Vetulonia, Cerveteri and Preneste have something in common, while two examples from Pontecagnano and Macchiabate are autonomous from the production of Cerveteri and resulted by the businesses from the way of Magna Grecia. Over all this western production has weighed some reserves on their Phoenician origin, but rather an orientalizing typology, or in other words, imitation production made by local people. Today, the common opinion is returned to the Phoenician workmanship. Another type of Phoenician embossed metalwork are the skyphos. In Italy this type is represented by those in bronze from the Barberini tomb of Preneste (Moscati, 1992).

Coins



Coins in gold, silver, copper, bronze, lead and the much rarer alloys like potin and bullion are another type of metal finding, very useful for the archaeological and archaeometric studies and interpretations. In the mid-fifth century BC the coinage was adopted in synchrony in Phoenicia and Carthage, with more than a century of delay than the micro-Asiatic and Greek experiences. The coinage of the Phoenician cities cut its role from the Persian field, as the coinage of Carthage placed immediately as an alternative to the Greek coinage system of Sicily and had such a political charge to stand out in comparison with the Roman coinage. The obverse and reverse of the Phoenician and Punic coins had often religious, warlike or maritime themes. For example, on the obverse of the earliest coinage of Sidon in silver, wrought on Phoenician foot, dating back to 450-435 BC, usually was reproduced the type of warship with triangular sail, on the reverse alternatively the Persian king who tends the bow and the wagon with royal personage. To this latter figure was joined in 395-375 BC a third character who is following the wagon on foot representing, according to some source, the king of Sidon himself. The turreted walls, the two lions and the firsts Phoenician abbreviations are characteristic of Phoenician coinage. With the reign of Abdashtart, 375-362 BC, the bronze numeral stands alongside the silver coinage. The coinage of Tyre, that also starts in autonomy with silver numeral, uses it for its obverse and reverse a general marine environment. But in Tyrian mints are used many more themes of Egyptian tradition, indicating the strong connection with this area: an example is the obverse with the owl with flagellum and scepter. Between 435-400 BC the first coinage has the dolphin and the murex followed by the adoption of the type of the divinity on the sea horse. Between 377 and 357 BC is introduced the first coinage in bronze, that reflect the types of numerals of silver, with on the obverse the head of a satyr, the ram, the dolphin, the murex, the cedar tree (the last two themes indicate commercial products, emphasizing again the great commercial vocation of this population).

From the monetary point of view the activity of the mint of Carthage is fundamental because it covers a very wide temporal range beginning with regularity in the IV century BC, and persisting until the fall of Carthage, even if the cultural influence continues throughout in the Roman period. The foot adopted was the Phoenician and the series were minted in silver, bronze and electro. The types assumed both for the gold and for bronze were: the head of the Core, galloping horse, the standing horse, the horse behind the palm tree, the palm tree. These types will remain the same until the fall of 146 BC, and will be the most used currency for commercial transactions, which is why it is possible to find these coinage all around the Mediterranean and in many areas of the inland.

The situation for the Punic coinage in Sicily became complex for the continued interconnection with the Greek system: beside the coinage of Phoenician settlements as

Mozia and Panormo, starting around 480 BC, other emission are located in Sicily, but under the direct control of Carthage. For example Mozia and Panormo align the ponderal system to the Euboean-Attic one and the emissions in silver of Syracuse, Agrigento, Selinunte and Gela. The common types are: head of Gorgon, palm, dog and female head and dog tearing deer etc... The series of gold and bronze with on the obverse the head of Core and reverse the standing bull are typical of coinage in Sardinia, but also the virile head and the ear of wheat are Punic elements in Sardinia. The few known Maltese numerals show the persistence of Punic themes also in the subsequent coinage in Latin legend. In Iberia the high efficiency of mining is immediately reflected in the fine silver emissions that arise generally between 238 and 206 BC. There are two coin series, the first produced in fixed mints as Cadiz, Ibiza and Sexi, the second referring to occasional mints, as Ampurias and Sagunto. The most common iconography is the obverse the head of Core and the head of Heracles-Melgart with lion's skin or laureate, to the reverse the horse with or without palm and the elephant. The coin types of Cadiz and Ibiza reflect greater autonomy, accepting the obverse and the reverse iconography that enhance economic and religious themes such as tuna and the God Bes (Moscati, 1992). In conclusion, as already mentioned for other types of objects produced by the Phoenicians and Punics, the study of coins allows to understand several aspects of the connection with other population, but also allows to perceive how much the cultural influence remains even after their end.

Small size statuary



The bronze metallurgy in the Phoenician Punic world is composed of many categories but two of these emerge significantly among the other and placed themselves at the beginning and at the late age of its Western culture: the small size statuary and the votive razors. While the small plastic, with its antecedents in Syria and Palestine from the late III millennium comes to the early Iron Age (XII XI century BC) and is a valuable clue to the oldest Phoenician frequency in the West, the razors bind tightly to Carthage and are the prestigious vehicle of culture between the VII and II centuries BC. Limited are the basic types of small size plastic: the fighting deity (smiting god), the god in throne with an Egyptian crown, the striding and blessing character, the woman with heavy decorative ring. Important is the role of Cyprus in the diffusion, in particular of the types of fighting gods and the striding king. Both schemes are derived from Egyptian prototypes: the first one was much more mediated by Syro-Palestinian tradition, tougher in the second the Egyptian mark. The divine figure has comparisons with similar bronze statuettes of Syrian production discovered in Byblos (XIII-XIV century BC). This artefact was probably brought to the West by the Phoenicians. The documentation of this type of statuary in Sicily is limited to the findings in 1955, in the waters of Selinunte, of a bronze statuette, 36 cm tall reproducing the kind of fighting divinity Reshef while in Sardinia some typology of bronzetti nuragici shows connection with these oriental figurative scheme. For Sardinia, the discovery of "Phoenician like" statuary falls within a little more recent horizon then the bronze of Selinunte; these votive statuettes, which show the typical nuragic scheme mixed with Phoenician features, come from many archaeological sites such as nuraghe Flumenelongu of Alghero, from Olmedo and the sacred well of Santa Cristina Paulilatino, just to mention some of the most famous. These figurines have therefore features in common with the typical Sardinian Bronzetti Nuragici, but show some discordant elements. Nevertheless these productions are anyhow identified by the local term nuragic. Even for Sardinia, Cyprus has played the role of the decisive intermediary is testified by the finding of the bronze tripod at Santadi, local product on Cypriot models of XII-XI century BC.

Also in Spain, the Syrian and Phoenician bronzework influence the art of Tartessos. Of particular significance is the recent discovery in Cadiz of two statuettes of fighting deities type. The two statuettes are completely in line with Syrian production of XIII-XIV century BC recovered from Cyprus in the XII and XI centuries BC. Another statue from Cadiz, preserved in the Museum of Madrid, is a standing character, probably a God, with gold mask on his face, long tunic and bare feet. The idea of the golden mask and a taste for the multi-material can be found in the production of statuettes of Ugarit in the second half of the II millennium (Giuntoli & Pedrazzi, 2006). In Portugal, in the numerous archaeological sites of Phoenician cutoff, is possible to find abundant evidence of this contact by the observation of votive statuettes. Generically speaking in Spain and Portugal there are lots of examples of the smiting gods, blessing divinity, the praying figure. These can be both imported phoenician artefacts or a local production in imitation of these new oriental decorative style (Avila, 2002).

Weapons, tools and equestrian vestments



Another interesting category of metalwork consist of weapons, and equestrian ornaments and tools.

A unique example is provided by the well-known Amatunte embossed shield (British Museum) which shows, around the central ombo, a frieze of lions attacking a bull with palms figures in the gaps, and the edge decorated with a pattern of trees. The shield has been attributed to a Phoenician laboratory of the second half of the VIII century BC. Another remarkable piece is the bronze axe conserved in the National Museum of Beirut. The horn-shaped blade presents on the two faces an identical finely engraved scene: a warrior goddess probably Astarte or Anat, flanked by two genuflecting worshiper. Also to be mentioned are the tools produced by bronze embossed plates including the frontal and blinkers that reminds, for their shape and style, similar objects in ivory found in large quantities in Nimrud. The ornaments of horses, found in Cyprus and the Aegean style could be of North-Syrian or Phoenician style. To the latter current is attributed without doubt a group of blinkers found in the tombs of Salamis, which bring the usual vegetable and zoomorphic motifs (Giuntoli & Pedrazzi, 2006).

Besides these famous examples, there are a huge number of tools and weapons in the museums of all around the Oriental areas and the Meditarranean Basin. The study of this kind of object is interesting because, since both the weapons and the tools should guarantee a mechanical and sometimes thermal resistance, they mirrored the real technological skill of a culture.

Razors

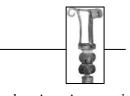


Few products of the Punic production, like the so called bronze razors, can highlight the autonomy of artisanal choices the world of the West made on the regards of the oriental motherland.

Originality of typological solutions, study and functionality of the visual space, used to emphasize the eschatological purpose, are signs of independence from the Aegean and Egyptian models. Found in the main funerary contexts from Iberia to North Africa and Sardinia, razors from the VII to II century BC are the elements that depict the religious piety; are symbols of the purifying depilation of the corpse and perhaps also of all the people who came in contact with it. Placed next to the deceased, razors had the function to collecting the impurity ripped by the deceased and by the rituals manipulators. The razor also had the function to simplify the transition to the afterlife. Hence the appearance on both sides with engravings (Moscati, 1992).

Indeed, the objects made of a metal blade (weapons, etc.) did not allow the embossing work but could be decorated by engraving. An already mentioned example is the bronze axe of Beirut; around this eastern prototype forms an important category of bronzes attested in that period only in the Punic world, namely the razors (C. Picard, 1966; Acquaro, 1971; Cecchini, 1992). Some of the oldest specimens are devoid of decoration, but from the V century razors have figurative scenes finely engraved on both sides. Alongside the distinctly Greek style, iconographic themes of oriental and Egyptian inspiration are of great interest: the birth of Orus, the goddess nursing the baby, the falcon resting on a lotus flower and bearded character with a high tiara and an ax identified as Melqart. About 200 specimens have been found in the Punic tombs of Carthage, in Spain and Sardinia. (Giuntoli & Pedrazzi, 2006). They are symbols of a rather narrow burial ritual, in fact there are a small number of findings, so the razors had a symbolic function only for a small slice of the Punic society. The examination of razors presence for geographic areas identified as the main production center Carthage: Sardinia and Spain are rarely able to set up their own language typology. Commonly the razors are long objects of rectangular shape with the half-moon blade on the short side and the swan's head handle, with a suspension ring on the opposite side.

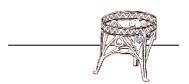
Incense burners



The bronze censers also called candlesticks, perfume burner or *thymiateria* constitutes a particular class of metal objects. These objects were evidently the typical religious tools of the Phoenician Punic world. These consist of a high column-shaped stelae, often not preserved, adorned with a double or triple set of corollas with pending floral petals to which overlap three tall and curved elements ending in volutes. These had to support one or two cups containing the incense. Another type has, instead of the floral capital, an Egyptian type in papyrus-shape. These censers were often found in tombs and some sanctuaries in contexts dating before the end of the VIII and V centuries BC. Around 90 pieces are

attested as these knew a large spread in the Mediterranean and in addition, in some areas of Sidon. These incense burner were certificates in large quantities in Cyprus and Samos and in the West especially in Malta, Sardinia and Spain. In addition to metal prototypes are known also rare copies in ivory (those of Salamis) and terracotta. Given the large number of discoveries it has been argued that Cyprus had been more than Carthage a major center of production (Avila, 2002).

Tripods



Other furniture items are the Phoenician metal tripods, generally associated with pots and bowls. Some specimens of Phoenician production probably comes from Cyprus as well as the tripods of wrought iron decorated with a series of lilies discovered in the royal necropolis of Salamis. From this necropolis comes a large amount of oriental metal tools of different types and origins of which some of Phoenician type (Karageorghis, 1969). To these is possible to add some bronze decoration of wagon, like the sphinx-head type of Egyptian style (Avila, 2002).

1.5 Orientalizing Art

There are various ways local and alien cultures can meet: the Phoenician production may, for instance, be present in many geographic areas because it came in the form of commercial items of exchange. It is also established that the Phoenicians used to implant manufacturing workshops in the most important colonies where contacts were more frequent. This is the reason why in the excavations, in the mentioned geographical areas where the phoenician influence were stronger, is likely to find objects not belonging to the same culture. The native production of a given place and the allochthonous one, in this case the Phoenician, can merge and give origin to a new production, as the so-called Orientalizing art (Avila, 2002). So the distinction between Phoenician production, the native production of a given area and the orientalizing production is quite difficult to perform only from the typological study of the objects. The difficulties in the interpretation manifest themselves if there is the necessity to figure out when an object is truly Phoenician or if it was produced in workshop in which the Phoenician component is one of several possible, and was not received in a direct way. The objects for which this difficulty in the interpretation is higher are, above all with regard to metals, the jewelry, the Iberian and Sardinian bronzes, but also the cups found in Italy in non-Phoenician context: this doubt therefore raises the question on two alternatives: 1) if the artefact was truly Phoenician and was imported, 2) if the production is local but under an oriental influence, which is the element that makes it difficult to interpret the origin of the metalwork. Initially the archaeological main interpretation was to attribute all production cited above to the Phoenician metalworking. The change in style was interpreted as an evolution of characters in the inside of Phoenician crafts, as a result of long-range movements of artisans. Then the common opinion was reversed: the recognition of foreign intervention contribution coupled with the fact the environment was Phoenician, led to the creation of this new attribution: the orientalizing art, a merging production which joined local and allochthonous (Oriental) elements. This classification was then enlarged to all the material in question. Now prevails a third phase of judgment, to be referred to the study of Maria Eugenia Aubet, more complicated but more convincing, valid especially for Iberian Peninsula. On one side there is the wide penetration of Phoenician on the southern coast of the Iberian Peninsula and the subsequent graft of Phoenician workshops in these area: there is no doubt that part of the production produced by these workshops were totally Phoenician. On the other side there is to be considered the spread of Phoenician objects in the trade routes that reached much more inside the occupied zone and the subsequent undeniability of reception of the products by a local craftsman who assimilates, reworks them, and therefore gives rise to the Orientalizing art phenomenon. But perhaps the case is still different: Phoenician craftsmen may have settled in the interior part of Iberia, and have worked here: in this case, the processing should be attributed to themselves, in whole or in part. With some confidence can be said that firstly the Phoenician objects reach the receiving areas (due to the ethnic penetration and because of the trade and commerce) and then the local developments were determined. Once set the issue, the Orientalizing question invests many craft categories some of which as been already mentioned: jewelry, ivory objects, figurines, cups, biconical jugs just to enumerate some of the most important categories (Moscati, 1992). Since the orientalizing factor is configured as an element of interest on the margins of the Phoenician production, with which is compared as a development and a interference (Moscati, 1992), is very interesting to understand what kind of interface is generated between the local populations and the "settlers" if they can be defined like this, and what kind of technological transfer has been activated or, alternatively, if the exchange interested only the figurative point of view and no production techniques has been exchanged.

Short specification on Orientalizing term in the thesis: the term, originally conceived by F. Poulsen, designated an artistic trend, found in Greek objects, of Near – Eastern forms and models imitation. A later use of the term has been generalized to all the classes of objects which show imitations or influences from Near East iconography. The use of the term " orientalizing craftsmanship " is related to Phoenician art and crafts since traders of Levantine cities were the main vectors responsible for the spread of the near-east influence across the Mediterranean. For Avila (2002) the meaning of the term can be approached for three variables that determine its use under different archaeological aspects: artistic-formal, chronological and cultural-historical factors. In this paragraph and often in the whole thesis the term will concern especially the first and third aspects. In fact the term is used here to denote not typically local objects, whose external characteristics demonstrate a connection to Phoenician and later Punic cultures, indicating also change in local cultural assetts. Here instead is not always respected the chronological characteristic of the term that refers to objects produced during the Orientalizing period (VIII-VI century BC), that's why the term is sometimes used below quotation marks, to refer to a meaning that is used in this thesis, or replaced by the definition "of Phoenician inspiration" manufacturing, indicating a phoenician-punic influence, or in other words a production in imitation of the Phoenician-Punic one. For Sardinian production another definition is used, "Oriental" or Phoenician", following Bernardini & Botto (2010) who adoperated these terms to indicate similar classes of votive objects influenced by oriental productions.

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Chapter II

RESEARCH PROTOCOL



Classes of objects under investigation

2.1 The procedure

The finality of this research is the setting out of a suitable procedure applicable to archaeometric studies where are required characteristics of itinerancy, non-destructiveness of the methods and necessities to make quick comparisons between cultural realities, very far among themselves. In parallel, as the chronological, geographical and material range is quite broad, it is necessary to provide the protocol of procedures that allow the treatment of analytical results in a flexible way.

Being all the artefacts part of the whole production "chain" of metal objects, but also halfworked materials such as the ingots or half-worked materials linked to the smelting, slags or anything else that is related to the metallurgical process, were selected as object of the study and the protocol changes slightly in relation of what kind of material is under study (a more detailed explanation of the metallurgical cycle can be found in Appendix A). The procedure, explained below, has produced good results of reliability in different fields of metallurgical research on the Bronze Age - Iron Age Mediterranean and Atlantic Areas, whereby it is proposed as a good investigative procedure, that is suggested to be applied in this type of archaeometric study.

The block diagram below exemplifies the fundamental phases through which this procedure is carried out (fig.1):

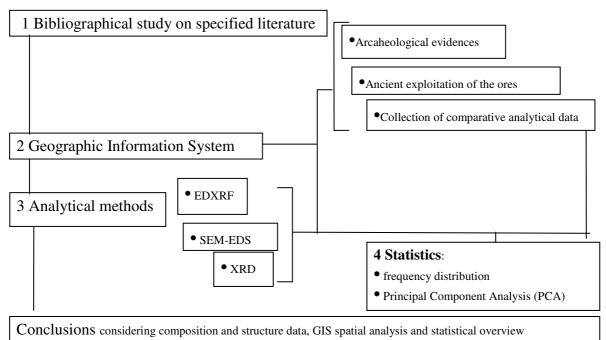


Fig. 1: block diagram to summarize the steps of the procedure

Each numbered phase will be explained in detail in the following paragraphs, following their numerical order.

Phase 1) Bibliographical study for the elaboration of different databases

Following a throughout bibliographical study, three types of database have been collected in simple excel sheets on:

- archaeological settlements;
- ancient mines for the extraction of metal ores, especially primary elements for bronze alloying (Cu, Sn, Pb);
- collection of analytical data on copper and bronze archaeological objects, coming from specialised archaeometric literature.

These databases have been collected separately for every different geographical area under study such as Morocco, Portugal, Sardinia, while for Punic coins from France and Belgium another particular database has been created and will be explained separately, in the dedicated chapter (Cap.VI). The third database on compositional data have been collected only for spanish and portuguese bronze production.

Each database has two columns for the geographic coordinates, longitude and latitude, for the acquisition in a GIS environment which permits to transfer all the information from the excel sheet to the geodatabase, using a Geographic Information System. These two fundamental data (longitude and latitude have to be expressed in decimal degrees) are found searching each place in the database using a web mapping service application such as Google Maps or Google Earth. Beside these two fundamental column, the other features are shown below. Another column, which is always specified in all the three types of tables, is the indication of the geographical area, namely the grouping of sites in broad clusters of neighbouring locations.

Archaeological settlements:

<u>Name</u>: current name of the settlement or the ancient name (very useful because in many case it is a toponym);

Longitude and latitude: the geographic coordinates in decimal degrees;

Chronology: Period of settlements foundation and ancient frequentation;

Area: Division in sectors (North, South, East, West of the concerned area);

Type of settlement and archaeological context : Local or Phoenician settlement;

Presence of Bronze artefacts: indication of bronze artefacts or slags in the stratigraphy.

Ancient Mines:

<u>Name</u>: current name of the mine, of the basin or the ancient name ; <u>Longitude and latitude</u>: the geographic coordinates in decimal degrees; <u>Area</u>: Division in geographical sectors (North, South, East, West of the concerned area); <u>Ore</u>: characterization of extracted mineral (if this information is available); <u>Elements</u>: Metal element extracted after mechanical and pyromentallurgical treatments;

Archaeological bronze artefact:

<u>Name or number of inventory</u>: Description of the object or the number of museums inventory;

Longitude and latitude: the geographic coordinates in decimal degrees;

Chronology: Period of production;

Area: Division in sectors (North, South, East West of an area);

<u>Function of the object</u>: Type of artefact (subdivision in votive, decorative objects or tools which should provide mechanical strength);

Type: Local, Phoenician or "orientalizing" production;

Composition: Composition normalized for Fe, Cu, Sn, As, Pb elements;

Using these tables is easier to simplify the large number of information coming from extensive bibliography available on the arguments.

Phase 2) Geographic Information System (GIS):

GIS system allows the enrichment of mapping with all the suggestions resulting from analysis of different components which compose a given space, thus becoming a fundamental mean to evaluate, analyse and represent any kind of spatial phenomenon. The geographic data have specific characteristics compared to other data types because their information are characterized by a specific position in the space and also in time. The geographic datum is composed by the coordinates, their values give the position of a site in the explicate time and from the other attributes and columns of the database is possible to have all the other information. By associating to that simple pair of geographic data become information that, represented on the map by means of the technologies offered by the GIS, appears to have a very high potential of use. The three different databases are georeferenced through the use of GIS system to obtain, after a conversion in geodatabases, thematic maps of archaeological settlements, mines and bronze artefacts (Migani & Salerno, 2008). In the geodatabase are included all analytical results on bronze artefacts from archaeological sites, extrapolated from archaeometric literature. All of these information are, in these way, attached to the geographic coordinates of the settlement where the samples have been found.

Using ArcMap, an ArcGIS Desktop application dedicated to the development of cartographies, analysis and editing of geographical data, is possible to use toolbars and commands to interact with the data. Each database and cartographical image consists of separate layer that can be overlaid to provide a layering of information.

To georeference information or a point is necessary to express the spatial position of the item through a sequence of coordinates that relates to a reference system. A coordinate reference system (CRS) is a coordinate-based local, regional or global system used to locate geographical entities. A spatial reference system defines a specific map projection, as well as transformations between different spatial reference systems. Is necessary to start from the definition of the ellipsoid of rotation that is a bi-axial ellipsoid. One of the main reference ellipsoids is the World Geodetic System (WGS), a standard for use in cartography and geodesy. The WGS constitutes a modern mathematical model of the earth characterized by having the origin coinciding with the centre of earth mass. The latest revision is WGS 84 (dating from 1984 and last revised in 2004), which was valid up to about 2010. The WGS 84 system is a conventional terrestrial system (CTS) a definition which means that the geodetic system is in solidarity with the earth and refers to elements (such as Greenwich etc...) fixed with the coordinates origin and coincident with the Earth mass centre. So the first step is to set the GIS system with the correct geographic reference system to allow proper georeferencing of maps and the correct positioning of the points.

Once the database is entered into the GIS, and a cartography has been georeferenced, is then possible to perform a real analysis of the spatial phenomenon under study, using the various exploratory possibilities the system offers. Some of the most useful tools are explained as follows.

1) Organization and enrichment of territorial information: having assigned to each item a geographical coordinate, it is possible to visualize its correct position and the relation with similar findings. GIS system allows the analysis of data of different nature. The presence of different information like geographic data and alphanumeric data, makes it necessary to use, inside a GIS system, models of structured data in different formats. Layers can be raster images, cartographies, digital data, texts, tables and GPS (Global Positioning System). Thanks to the union of these data it is possible to prepare a geographic database browsable through queries for the production of maps and reports. Vector data are used for the description of the geographic components of spatial information. The geometric characteristics of a given object are recorded as a sequence of coordinates (x, y)of the points that form the vector. Typical vector data are those that come from manual digitization of maps, from topographic surveys, from CAD and GPS. In the vector data models a set of primitives is used to represent objects: point, line and area. As in the case of vector data, raster data are as well used for the description of the geographic component of spatial information: the geometric characteristics of an object are represented as a grid (grid format) or as a set of pixels (picture formats). Typical raster data are those generated by scanners and programs for the interpretation of the images. The various types of data correspond to different formats in GIS environment, as well of composite type. The shapefile format is organized on the basis of multiple files that have the same name but with different extension. A shapefile consists of at least three different files: one for the attribute management (.dbf), a file containing geometric information with .shp extension and a file containing the metadata extension (.shx).

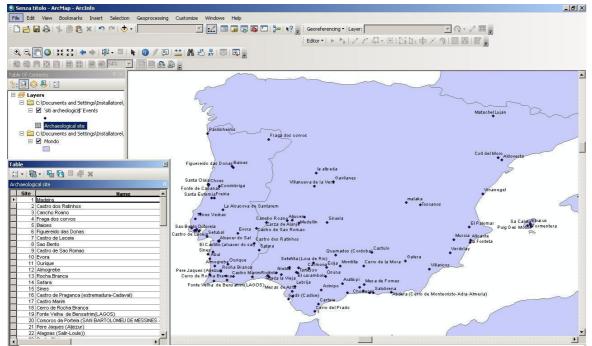


Fig. 2: the figure shows a screenshot of the GIS system where the layers, the attribute table and the layout view window can be seen

The geodatabase (DBMS) is another important data format which is the standard ArcGIS application logic to access and work with all files of geographic data. It is not a simple data format as shapefile, but a data model. The difference lies in the fact that a geodatabase, besides the data, can also store rules and relationships (on the features or attributes). The geodatabase architecture is organized in tables that contain rows, and columns with the same domain (an integer, decimal, character, date, etc ...). The rows of a table can be related with rows of another if the two tables have columns in common (called primary key in the principal table and external key in the related table). The attributes organized in the table, can still be accessed and consulted by opening the attribute table of which the layer is constituted (fig.2).

2) Communicating the information: The catalogue has been georeferenced to provide visual information on the distribution of different features under study. Giving each macrogroup a different symbology it is possible to monitor how these are positioned, what is the frequency of occurrence and the relationship between different entities (fig.3).



Fig. 3. example of thematic map with its legend

3) Query the database: the management of information in a DBMS is carried out through a special language, the SQL (Structured Query Language), developed in the late 70s by IBM and has become the reference language for relational databases. It is based upon relational algebra and Tuple relational calculus, SQL consists of a data definition language and a data manipulation language. The SQL language is not only a query language, it presents in an integrated form the functionality of both classes of languages used in databases: the DDL (Data Definition Language) and DML (Data Manipulation Language). The SQL operators are: =, >, <, >=, BETWEEN, LIKE, IN, OR, AND, NOT, IS. By using this simple set of operators following the arithmetic logic it is possible to make any kind of selection. The layer containing the information can be queried by selecting objects that have particular characteristics: is possible then to create a subset. On the map will be highlighted the areas where the selected feature has been found, and the attribute table displays information related only to the identified features (fig.4).

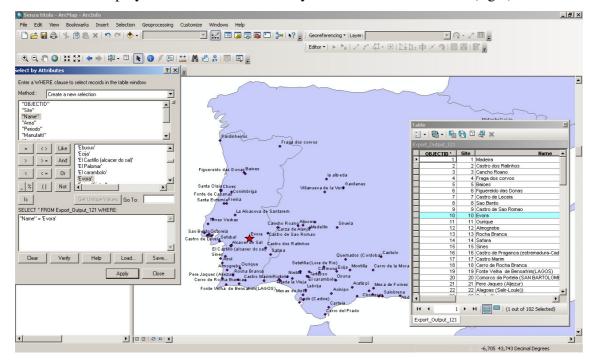


Fig. 4: screenshot of GIS SQL "selection by attributes"

4) Using GIS drawing and statistic instruments: GIS system is also equipped with drawing instruments with which is possible to create new shapefiles (point, line or area used for digitalization of rivers or routes) that allow to highlight important features. The statistic means allow to display on the maps also compositional information, for example, on artefacts compositions, like histograms.

5) Georeferencing raster cartography: using georeferencing instruments it is also possible to put cartographies such as ancient maps or satellite images in the GIS system, assigning geographic coordinates to the raster data and overlap geodatabase as is shown in the picture (fig.5).



Fig. 5: a XVII century Iberian Peninsula maps, georeferenced with GIS system to allow the localization of Phoenician-Punic archaeological sites found with bibliographical research during the period of doctoral research

6) Creation of thematic maps: all these operations that have been described in the previous sections give as results the thematic maps, which thematism is understandable trough the use of legend and symbology.

All thematic maps that appear in this chapter and throughout the thesis have been created *ad hoc* by the use of GIS for the research purposes and archaeometric scopes of the Ph.D. research.

Phase 3) Analytical method

A simple, fast and non destructive analytical methodology, combining Energy Dispersive X-ray Fluorescence (EDXRF) and Scanning Electron Microscopy coupled with Energy Dispersive Spectroscopy (SEM+EDS), has been applied to characterize the alloys composition of the bronze artefacts under study. On the slags from Morocco also XRD analysis has been applied to provide information of their compositional nature. The choice of analytical techniques responded to certain characteristics which are listed below:

- the easy availability in different research institutes that have hosted the work;

- both EDXRF and SEM-EDS don't require a sampling of the specimen thus being as much as possible non-destructive methods (it was necessary to take samples of slags only to subdue them to XRD analysis);

- instrumentation choices are appropriate to the study of metals and slag metallurgy since the EDXRF is able to reveal the chemical elements having atomic weight greater than or equal to that of sodium (conducting the analysis under room conditions the visible elements often ae the ones from Si or P onward, also considering the attenuation effect of berillium window) and using a beam with a voltage of 35 KV is possible to collect the Sn K β line. SEM-EDS, due to the conductivity of the samples, is the most suitable technique to obtain magnified images with both the chemical and morphological contrast, choosing between the use of backscattered electron detector or the Everhart-Thornley secondary electron detector. The EDS (Energy dispersive X-Ray spectroscopy) allows to detect the characteristic X-rays formed by the electronic probe (of elements with an atomic number from carbon forward).

- the use of portable EDXRF has allowed us to perform many analyses directly in the rooms of museums, allowing to study collections that could not be moved.

- XRD and TIMS analytical methods complete the protocol, being fundamental for the study of slags composition (XRD) and to attempt a provenance study.

The combined use of these techniques has proved useful in unravelling technological processes and help to shed light on the characterization of the artefacts.

Preparation of the specimens: the artefacts have been analyzed either on polished and on unclean areas. This difference in treatment could have caused some anomalies in the EDXRF data but, since all the artefacts have been submitted to mechanical cleaning during restoration in the pre-musealization phase, the contribution of the patina should be minimized. In addition to this observation, it should be considered that the aim was to find compositional macro-groups of artefacts, starting from qualitative compositional data, which permit to state easily what kind of alloy characterizes the artefacts (arsenical copper, binary bronze, ternary bronze etc..).

When it was possible, small areas were cleaned and the procedure used was: grinding with an emery and abrasive paper of different grit sizes (up to 1500) to obtain a cleaned area of about around 25 mm^2 .

Since the protocol is proposed as a research tool to be used also by non experts in chemistry/physics, it is necessary a brief, indicative excursus on the characteristics and functioning of the applied methods. The presentation of the analytical methos, proposed in list form, furnishes key information useful for other professional figures who work in archaeometric research, and allow to apply the protocol in relation to the kind of samples and desired results.

EDXRF portable instrument

X-Ray fluorescence is a widely used method for elemental analysis, based on characteristic emission of an atom under the incidence of intense electromagnetic radiation. More in detail, when any material is exposed to short-wavelength X-rays or gamma rays, ionization of their component atoms may take place. The electron removal, in this way, makes the atomic electronic structure unstable and during the subsequent process of electrons readjustment, from the upper orbits to the vacant ones, the secondary and characteristic X-Rays occurs. The main transitions have specific classification as follow: the transition from L to K shells is called K_{α} , the M to K transition is called K_{β} , while an M to L transition is called L_{α} , just to enumerate the most important ones. Each of these transitions yields a fluorescent photon with a characteristic energy equal to the difference in energy of the initial and final orbital. The wavelength of the secondary rays are related to the atomic number of the element concerned and shall allow in this way the identification of compositions. The X-Rays generator is mainly a vacuum tube composed of a cathode which produces electrons for thermoelectric effect, and an anode (or anticathode) on which the electrons are accelerated, because of a potential difference. When the electrons collide the target, the primary X-Ray beam is produced. The range of analyzable elements depends in part on the values of the potential difference at which the device can operate. X-ray generators in the range 35-40 kV are often used in the cultural heritage analysis, because those values allow excitation of a broad range of atoms. Berillium window is located at the exit of the tube and entrance of detector (as it has low density and atomic mass, Be is relatively transparent to X-rays and other forms of ionizing radiation. In new instrumentations berillium windows can be replaced by plastic windows reducing

furthermore the attenuation effect) and a collimation system is used to focalize the beam on the specimen. Also the detector has a collimation system at the entrance, between the sample holder and the analyzer crystal, in order to reduce the divergence of the outgoing rays (Polesello & Guenzi, 2006). The analyzing crystal is a monochromator consisting of diffraction grating that separates the outgoing beam from the sample in the contributions relating to the individual wavelengths. The outgoing beam, once divided into its individual constituents, is revealed by the detector (Matteini & Moles, 2003). In energy dispersive analysis, dispersion and detection are a single operation, commonly performed by Silicon Drift Detectors (SDD). The X-ray photon activates a large number of SDD detector atoms, with the amount of charge produced being proportional to the energy of the incoming photon. The charge is then collected and the process repeats itself for the next photon. This simple apparatus consists of a few elementd of riduced dimensions, which must of course be mounted respecting the geometry, allowing the execution of non-destructive testing. Obviously the use of foucalized x-ray beam required particular safety conditions since the highly penetrating X-Ray can be harmful for the operator, since with this portable instrumentation the analysis is conducted in open air. This technique proved to be extremely useful, for a type of research that is highly itinerant like this and also because many museums cannot allow a moving of the artefacts out of their rooms. The easy handling of the instrumentations have to be associated with an accurate choice of the investigated area, requiring, if is possible, opportune artefact cleaning procedure.

The analytical EDXRF sessions were interspersed always by the analysis of bronze standards of composition as similar as possible to the object under study, to achieve correct quantification of the spectra. All measurements were carried out at room condition performing about three measurements per each statuette.

Data have been processed and quantified by using the WinAxil 4.5.2 and PyMCA softwares, comprising fundamental parameters and experimental calibration factors, but also an innovative reverse Monte Carlo simulation quantification approach has been used. This method is based on the reproduction of a simulated spectra, feeding the system with all the information on instrumental characteristics and apparatus geometry. This phase is foundamental for a correct calculation of the simulated spectra. Overlapping the experimental spectra with the simulated ones is possible to achieve a good quantification of EDXRF outputs. Therefore it is necessary to proceed by "trial and error" simulations, assuming compositions that can describe well the experimental spectrum, as long as the

overlap is approximately the best. This approach allows to significantly improve the sensitivity of the spectrometers, taking in account also the non planar nature of the surface. Other Monte Carlo codes are capable to cope with rough surfaces by using regular structures such as hemispheres or stripes. In our case, the rough surface could be modeled using any irregular shape. Monte Carlo code used in the thesis makes use of a constantly updated X-ray library for any X-Ray data (X-raylib). The Monte Carlo code has been tested on a variety of cultural heritage items. The theoretical LOD of the system is about 10 ppm (Bottigli *et al.*, 2004) while the real one will be higher considering the peculiar conditions of measurements.

SEM-EDS analysis

Scanning Electron Microscopy, coupled with X-Rays microanalysis, is one of the most useful technique used in cultural heritage research because it allows the collection of high magnification images of the surface of samples with compositional information as well. Its operation is rather complex, but it can be broadly described, for non-specialist users, in order to show how much it is a versatile apparatus in this type of research. So this section provide a brief overview of SEM-EDS describing the function of its various subsystems. As is common knowledge the resolving power of an optical microscope is more dependent on the wavelength of the light used for observation (the resolution increases with decreasing wavelength). Using electrons instead of light is possible to get, therefore, a higher resolving power and a magnification value much higher than that of an optical microscope. Being negatively charged, electrons can be manipulated using magnetic fields, also called magnetic lenses and an electromagnetic optic (Matteini & Moles, 2003). The two major components of the SEM is the electron column and the detectors. The electron gun generates electrons and accelerates them to energy in the range of 0.1-30 KeV. The spotsize from a tungsten hairpin is demagnified and focused by electron lenses. The beam emerges from the final lens into the specimen chamber, where it interacts with the sample to a depht of approximatelly 1 micron and generates the signal used to form the images and the EDS results. The scanned images is formed point by point. The deflection system causes the beam to move along lines, until a rectangular raster is generated. Two pairs of electromagnetic deflection coils are used to move the beam across the sample. The magnification is the ratio of the raster length in the screen and the real one on the specimen. So the microscope column is mainly made of the electron gun, electron lenses and scan

coils (Goldstein et al. 2003). The sample chamber can be setted on high vacuum mode or variable pressure (depending on the characteristic of the sample and its conductive behaviour) while the column is always kept at high vacuum values. When the electrons strike the sample, a variety of signals are generated, and it is the detection of specific signals which produces an image or a sample elemental composition. The three signals which provide the greatest amount of information in SEM are the secondary electrons, backscattered electrons, and X-rays. Secondary electrons are emitted from the atoms occupying the top surface and produce a readily interpretable image of the surface. The contrast in the image is determined by the sample morphology. A high resolution image can be obtained because of the small diameter of the primary electron beam. Backscattered electrons are primary beam electrons which are "reflected" from atoms in the solid. The contrast in the image produced is determined by the atomic number of the elements in the sample. The composition of the sample can be derive from the backscatter phenomenon because the likelihood of the electron elastic scattering is directly proportional to the atomic number of the impressed atom. The backscattering coefficient thus increases with the atomic number. The image will therefore show the distribution of different chemical phases in the sample. To get the best from the contribution of backscattered electrons it is useful to have as flat as possible sample surface to eliminate the topography produced by rough surfaces. Because these electrons are emitted from a depth in the sample, the resolution in the image is not as good as for secondary electron. The third signal is produced by the interaction of the primary beam with atoms in the sample, resulting in shell transitions and the emission of an X-ray. EDS can provide rapid qualitative, or with adequate standards, quantitative analysis of elemental composition with a sampling depth of 1-2 microns. Xrays may also be used to form maps or line profiles, showing the elemental distribution in a sample surface. For each of these signals (secondary electrons, backscattered electron and X-Rays), SEM-EDS instruments have a dedicated detector. Everhart-Thornley Detector is a secondary electrons detector consisting of a scintillator in the Faraday cage, positioned in the SEM chamber, with a low positive voltage applied to attract the relatively low energy (less than 50 eVs) secondary electrons. So this detector allows to collect the weak signal of the secondary electrons and to separate it from the BSE signal. BSE detectors are usually either of scintillator or of semiconductor types while the EDS detector, mounted in the sample chamber of the instrument at the end of a long arm, which is itself cooled by liquid nitrogen, is commonly made of Si(Li) crystals that operate at low voltages to improve

sensitivity or the previously described "silicon drift detectors" that operate at higher count rates without liquid nitrogen cooling. This brief description introduce some specific information on samples analyses.

Cleaned and unclean areas were analyzed by SEM-EDS performing at least three measurements per each statuette. The spatial resolution and phase contrast of SEM Backscattered electron imaging proved to be useful in the investigation of the micro-texture and distinction of phases of the bronzes, while the EDS system provided 2D major elements maps highlighting compositional heterogeneities.

SEM-EDS analysis after etching with ferric chlorides solution: for the study of the bronze jar from Alcacer do Sal (Portugal) a soft etching has been performed on a polished small area of the object. The recipe (Scott, 2000) is particularly used for copper and copper-based alloys and the compounds portions are as follows:

Alcoholic Ferric Chloride:

- 120 ml of ethanol C_2H_5OH
- 30 ml of hydrochloric acid HCl
- 10 g ferric chloride FeCl₃

A drop of the solution has been poured on the cleaned area and left for few seconds. Immediately after, the action of the reagent is blocked by rinsing with distilled water and dabbing with a sheet of absorbing paper the surrounding area (not directly on the sample to prevent scratches) and spraying thoroughly with compressed air.

XRD analysis

This instrument has been used for the analyses conducted on slags from Morocco, to define the chemical composition of this complicate class of findings. The X rays diffractometer used is a Miniflex Rigaku on powdered samples. Also for this kind of analytical apparatus a short introduction is proposed below. X-Ray diffraction method is basedt on the diffraction of X-ray beam produced through a X-Ray tube (as seen previously for XRF). In the range of 0.2 to 2.5 Angstrom, wavelengths are approximately of the same order of magnitude as the interatomic distances. So the crystal lattice of the solid substances is capable of causing diffraction phenomena against a beam of appropriate X-rays that invests it, according to a precise angle. The diffraction of X-rays, coming from the various crystal planes, form a series of reflections which constitute a characteristic profile of the analysed crystal. This phenomenon is related to Bragg equation:

$n\lambda = 2d\sin(\theta)$

where the wavelength is the value belonging to incident X-Ray beam, *d* is the distances between the cristal lattice planes and *teta* is half the angle between the direction of incidence and refraction of the beam. As the interplanar distance is characteristic of a given lattice, it is possible to use this technique for the study of crystalline substances, while it is not fruitfully applicable to amorphous solids. Even if the foundaments of the technique is unique, the instrumentations today available are various, in some equipments the sample has to be finely grinded, therefore the technique can be defined destructive or microdestructive, given the small quantity of sample require. Otherwise some new instrumentation allow to analize surfaces without any sampling. Besides the X-rays generator, the XRD apparatus is made up of collimation system to obtain parallel X-rays, a chamber for the samples and a detector for the diffracted rays which is generally a goniometer Geiger counter (Matteini & Moles, 2003).

The results of this kind of analysis is fundamental for the understanding of slags samples, because the use of the sole SEM-EDS cannot give unique information on the crystalline nature of the samples. The combined use of SEM-EDS and XRD instead allows an easier interpretation of complicated pyrometallurgical samples.

Thermal Ionization Mass Spectrometry:

Thermal Ionization Mass Spectrometry (TIMS) analysis has been performed only for one lead fragment from Volubilis. Also for this last kind of analytical apparatus a brief explanation is given, dedicating it to a non-specialist audience, to explain briefly the reasons that have led to its use for the study of a lead artefact from Volubilis. This method has been applied for the detection of isotopic ratios used for provenance studies.

Mass spectrometry, generically, is based on a process of fragmentation of the molecules of a substance and the subsequent separation of the charged particles on the basis of mass / charge ratio. The charged particles we are dealing with are isotopic ions, consisting of ionized atoms, but other types of ions can be studied with this method (Matteini & Moles, 2003). In TIMS instruments, the sample is heated to stimulate the ionization of the atoms. Then the ions are accelerated into a beam by a electromagnete and separated into individual beams, based on the mass/charge ratio of the ions. This mass-resolved beam is then directed into collectors where the ion beam is converted into voltage. Comparison of voltages corresponding to individual ion beam yields to measure the intensity of the signal of each

mass bringing to the values of isotope ratios. In this thesis the lead isotopic ratios of a Volubilis sample has been obtained with a provenance purpose because the relative proportions of the individual isotopic species is bound to specific petrogenetic phenomenon. To obtain reliable results the sample must be chemically purified and then is destroyed once submitted to analysis. This method is micro-distructive because a small sampling of micrograms of sample (equivalent to the tip of a pin) is required for the analysis. In any case, this is the fundamental technique for provenance studies although there are some limitations, especially if the obtained results are not interpreted in the light of other analytical methods. In fact, since the petrogenesis of distant geographical areas may be the same, and since every mining area has an isotopic variability which causes the overlap of the isotopic fields belonging to mineral deposits also very far from each other, it is necessary to carry out also trace elements analyses like PIXE (for example) to have a clearer and more reliable provenance interpretation of a sample results.

Binary and Ternary Diagrams: compositional analytical results were then displayed for easy reading on ternary diagrams drawn with the free program Triplot for the primary alloying elements (Cu, Sn, Pb).

Phase 4) Statistics

Due to the large number of both analytical data and measurements done on the artefacts, the application of statistics is useful to obtain a description of the distribution and a macro-grouping of the artefacts. When the system is characterized by more than three components, like in the case of Iberian bronze artefacts (where each artefacts is specified for its content in Cu, Sn, Pb, As, Fe elements, but also for their typology and chronology aspects) the multivariate Principal Component Analysis (PCA) has been applied while, when it was necessary to describe the shape of measurements distribution and the grade of the fluctuation around a mean value, the distribution of frequencies with histograms has been conducted, in such a way to understand if the distribution is Gauss-shaped or bimodal. The use of statistical tools for the study of copper-based alloys must always take into account the heterogeneity of the alloy itself, so sometimes the data are not fully representative of a given object. In any case, since PCA is used to obtain a qualitative separation into large compositional groups, this observation can be regarded as relatively unimportant.

A short description of the selected methods is as follows, but for a detailed explanation of PCA method is reported in Appendix B.

Principal Component Analysis: is a useful statistical technique that has found application in cultural heritage fields and is a common technique for finding patterns in high dimension data. PCA statistical analysis has been used to find connections between this multicomponent system, connecting information like composition, expressed for five elements (Cu, Sn, Pb, Fe, As) and for other nominal features like the classes of objects, the archaeological context, the chronology and the geographical areas, in such a way as to highlight their similarities and differences. This is a mathematical procedure that uses an orthogonal transformation to convert a set of observations of possibly correlated variables into a set of values of linearly uncorrelated variables called principal components. In this way a simplified, linear and two-dimensional grouping of a multivariate system can be obtained (Sadocchi, 1993). A much more detailed revue of this method is reported in Appendix B.

Frequency distributions and Box-Whisker diagrams: used for the archaeometric study of the Punic coins in chapter VI. The frequency distribution permit to describe a set of measurements and to identify the various parameters that describe the distribution as: the central value, the symmetry, the mean, median, mode, the range of variation etc. ..

Box-Whisker diagrams serve to highlight the structure of a univariate distribution. Depending on the software used to generate them, these diagrams are presented in many different way. The key points are: a middle line represents the median, the box (box) that extends from the tenth to ninetieth percentile, whiskers (whisker) which goes to the sides of the box connecting the fifth and ninetieth percentile of the box. The dots beyond the whiskers are the most faraway data from the mean, the minimum and maximum outliers in some cases or like in our case, artefacts belonging to different sub-species.

This general protocol has been slightly changed in relation on the peculiarity of each geographical area and class of objects under study. In fact it was not possible to use the same instrumentation in every location, due to the availability of the assets but also taking into account the museums provisions or the possibility of moving the archaeological objects. The availability of data in the literature has instead determined the number of databases produced for every section. The following part will summarize the steps performed during each session:

Votive statuary in Portugal: Collection of the three databases, GIS, SEM-EDS, EDXRF and statistics.

Bronzetti Nuragici in Sardinia: Collection of two databases (archaeological settlements and ancient mines; the third database was not collected because of the lack of a large number of comparison data), GIS, EDXRF.

Slags and metal leafs in Morocco: collection of two database (archaeological settlements and ancient mines; the third database was not built because too few information has been found from archaeometric studies), GIS, SEM-EDS, (it was not possible to try PCA elaboration because of the absence of comparison data from literature) XRD and TIMS.

Mauro Viola's Punic Coins Collection: SEM-EDS and statistics.

Punic Coins in France and Belgium: collection of a different kind of geodatabase in GIS environment that will be freely available in the internet at the webpage of GEGENA2 departement of Universitè de Reims.

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CHAPTER III

ARCHAEOMETRIC STUDY OF PORTUGUESE BRONZE STATUETTES

Class of object: Bronze votive, small-size statuary

Area: Portugal

Artefacts: Museu de Evora and Museu de Alcacer do Sal collections



Localization of Evora and Alcacer do Sal

Highlights

Eight bronze statuettes (ex-votos) of unknown provenience and dating, belonging to the Evora Museum, has been studied to determine the original archaeological sites. The goal of the research is, in this specific case, to suggest a dating and provenance for the collection and, in a more general point of view, to attempt an identification of elements that may help to distinguish between the autochthonous artefacts and the production affected by the presence of "colonizing" Phoenicians. This type of manufacturing was selected because of their potential link with the local cult of the goddess Adaegina in the Iberian Peninsula, and because of the presence of some features belonging to the Phoenician cult of Baal and Astarte. The research started from the study of metallurgical resources of the Iberian Peninsula and the location of archaeological sites in which evidences of the Phoenician presence are attested. In parallel a geodatabase was constructed, containing both the archaeological sites dating from the Late Bronze Age and Iron Age, and the presence of metal objects and, if present, compositional data found in literature. Statistical analysis has been conducted on these collection of data, in order to have a general understanding of bronze production in Portugal. The analytical study was performed with XRF and SEM-EDS analyses on cleaned areas of the statuettes. By comparing the compositions of the statuettes (an alloy of copper, tin and lead) with the analytical data from the geodatabase, it was possible to suggest a dating of the statuettes to the Iron Age. The presence of iron in percentage higher than 0,05% can suggest many hypothesis, that range from the smelting of ferrous minerals to the use of fluxing agent and thermal operation in reducing environment attainable in high technology furnaces. Querying the geodatabase on the presence of metallic statuettes in Phoenician settlements it was possible to identify the area of Alcacer do Sal (already suggested by archaeologists), where more than 50 very similar votive offerings were found, as the original archeological site. These Iron-Age artefacts of similar typology were recovered from this well known Phoenician settlement (V century BC) located about 50 km W to Évora. The concession for the analysis of 6 artefacts from this archaeological site was therefore asked, in order to compare the two set of XRF and SEM-EDS data. The alloy compositional data is consistent with a provenance of the Évora statuettes from the known Phoenician settlement of Alcàcer do Sal in Southern Portugal.

3.1 Historical and Archaeological Introduction of Portugal and Spain during the *"Phoenician and Punic period"* (LBA/EIA)

Archaeologically speaking Portugal is a discontinuous area: this statement can be made especially considering the relations with the Eastern world. The valleys of Mondego, Tagus and Sado constituted important areas relating to trade and therefore connected with Phoenician commercial network (Arruda 2000). The Phoenician presence in Portugal may be defined between the VIII and VI centuries BC, although this does not mean that there are cases in which this range broadens (Shubart et al., 2002). Thus the Phoenicians presence has mostly a shoreline character, bounded on some specific areas of the coast, specifically near the estuary of the three major rivers (previously mentioned Sado, Mondego and Tagus) located on the Algarve and on the western coasts. The presence of the Phoenicians in Portugal seems to be thus reduced, indicating that the whole territory remained mostly alien to their commerce and did not participate directly, forming a periphery which kept a number of different relationships with the seaside settlements. It should be pointed that the Phoenician presence in the littoral areas implied, especially in the VII century BC, the permanent establishment of populations, first of all near the Strait of Gibraltar. The available data indicate Santa Olalia and Abul as fundamental colonies, founded in order to structure and organize commercial activities. On the other hand the large amount of orientalizing pottery in Alcacer do Sal, Almaraz, Lisbon and Santarem and certain building techniques may indicate that in these settlements a segment of population was Phoenician. In Santarem there are certain technologies that can hardly be acquired without direct knowledge. The metallurgy of silver, the glass paste, the application of engobes, are hardly teachable without a direct contact and through direct practice that presupposes the presence of artisans who were particularly skilled in these production. The arrival of the Phoenicians causes in Portuguese territory, in terms of social, economic and technological development, the formation of deep regional differences. The south of Portugal did not correspond to a unity, even though is evident a common Mediterranean presence. This diversification was the result of the presence, in this very large geographical area, of a constellation of human groups organized into distinct social establishments. Conimbriga, Santarem, Almaraz, Alcacer do Sal, Setubal, Castro Marim, are settlements whose occupation was continuous and uninterrupted throughout the I millennium BC. This continuity is not visible only in the type of settlement, in the size of the occupied area, but also from the point of view of the cultural matrix of recognized materials. Deep relationships existed between these Portuguese areas with the Mediterranean, a connection that was constant throughout the Iron Age. These orientalizing material productions were maintained for the entire Iron Age, till the Roman arrival: this is why this phenomenon was called "orientalizing" conservatism". In Santarem, Setubal, Alcacer do Sal, Conimbriga and Almaraz the gray ceramic, pithoi, the painted bands decorations, are identical throughout the Iron Age. It is not fully known how the indigenous society was organized at the time of Phoenicians' arrival in the western coast, since the archaeological data are scarce or completely nonexistent. About the social and economic context it is not possible to say exactly what kind of structure there was in the Late Bronze Age: the stratification of Portuguese local culture and Near East ones were not comparable. The Phoenicians had a highly hierarchical structure that has no equals in the entire Iberian Peninsula. The relationships established between the Phoenician traders and the natives who lived in the south of Portugal were asymmetric, as it was the trade itself. The most important object of commerce were the metal ores, exchanged with foodstuffs and manufactured items. The Phoenicians used to contract their business with the local elites that started to be created in the Late Bronze Age. The purchased goods then become prestige objects found in cemeteries as burial objects. Conimbriga, Almaraz, Santarem, Lisbon, Setubal, Alcacer do Sal, and Castro Marim played a vital role in the colonial process and in the only known necropolis, the one of Senhor dos Martires at Alcacer do Sal, where many oriental objects integrated the afterlife scenarios (Arruda, 2000).

The geographic location of these villages allowed direct access to a hinterland rich in metal ores and a control of the trajectory of these materials extracted in the inland. It is in these settlements that all the artefacts and foods, carried from the Phoenician ships, were distributed to the nearby hinterland *via* rivers and inland paths, and from which they received the raw materials (tin from Beiras and silver and copper from Alentejo) that were sold to the Phoenicians. These major settlements are the responsible for the creation of other villages under their direct political control. This is the case of Setubal, near the estuary of Sado river and of Crasto in the Mondego valley, which foundation could depend respectively to Alcacer do Sal and Conimbriga. The archaeological data suggest that Santa Olaia and Abul were certainly preceded by previous commercial contacts, so it can be thought that in these area there were some resources that particularly interested the Phoenicians, in other words, the possibility to exploit mineral ores here embedded stimulated their presence.

Same procedures and reasons brought Phoenician colonies in Spain and since the division of the Iberian Peninsula in the two sectors, Portuguese and Spanish, are only fictitious from a archaeological perspective, a brief tour of the Spanish Phoenician sites is useful for the full understanding of the phenomenon. Also in this case the Phoenician colonization in Spain responds to a fundamental aim: to gain control of the metal trade sources. Ancient historians such as Strabo referred to the riches of a far away land called Tartessos (Huelva) and from the 1960 the archaeologists began to carry an information explosion on this ancient settlement and the Phoenician colonies that appears here from the VIII century BC. Huelva, just to show an example, was probably located downstream from the Riotinto mining region, rich in metal ores. Other sites in Spain of major interest, following the coastline from west to east were: Gadir, Castillo de Dona Blanca, Cerro del Prado, Montilla, Cerro del Villar, Malaka, Morro de Mezquitilla, Almunecar (Sexi Laurita), Abdera, Villaricos and La Fonteta just to enumerate some of the most important. From these first colonies, more than 50 other sites have been developed, both on the coast and inland, especially near the mouths of mines (Schubart, 2002). Given the large number of archaeological sites and the fundamental geographic location of these, it was decided to establish a database of all sites, in order to produce thematic maps with GIS (fig.1, collected sites come mainly from Schubart *et al.* 2002; Arruda, 2000).

As explained previously in Chapter II, the collection of the databases and all the GIS elaborations have been personally executed during the research period. In the thematic map in fig.1 all the settlements are indicated with a black dot, while Evora and Alcacer do Sal, areas of study for this chapter, are highlighted with a red dot. In fig.2 a hydrographic shapefile, containing the Portuguese major rivers, has been used as a background of the archaeological settlements thematic map. In this way it was possible to recognize the close connection between the localization of the sites and the distance from the coastline and the course of navigable rivers, one of the preferential paths for the penetration in the hinterland. In fact sailing upriver (upstream) was a well known navigation skill in ancient time (when the wind was behind the sail, the river can be navigated easily upstream but with the wind against it was possible to use some modality like: tacking from side to side; hauling, kedging and also towing with horses or oxen from the banks). Digitization of the Rio Sado was performed separately and is shown in the image using a dark blue color, as it is located in the vicinity of the archaeological site of Alcacer do Sal, indicated with a larger red dot. In fig. 3 the result of a selection by location (spatial query) is shown, and it has been executed asking to GIS system how many settlements were close to rivers. The system answers pointing out with a red star all the sites in proximity to rivers. As can be seen in fig. 3 almost many dots are now marked with a star, except for sites founded on coast. The result of this spatial query is to indicate the shipping paths as available and conceivable input lines used for both the Phoenicians and Punics later, to settle in this territory. Also, the transport of the traded products was performed precisely through the vessels. In this way the traders could arrive in the vicinity of mining areas and transship ingots or halfworked materials sailing following the river current. Given the close connection between the mineral resources of the Iberian Peninsula and the strategic location of these sites, the discussion proceeds with the geological description of this area and the georeferencing of the both old mines and ore basins (for which an ancient exploitation is unsure but not excludable).



Fig. 1: phoenician main archaeological settlements in Portugal and Spain:

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

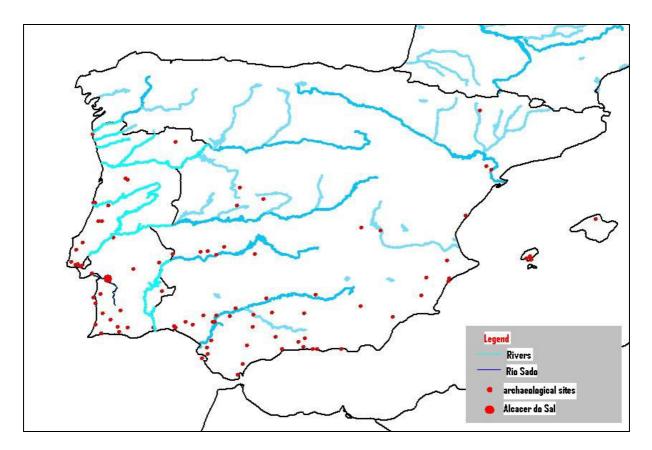


Fig. 2: Rivers, Rio Sado (in dark blue) and Alcacer do Sal site

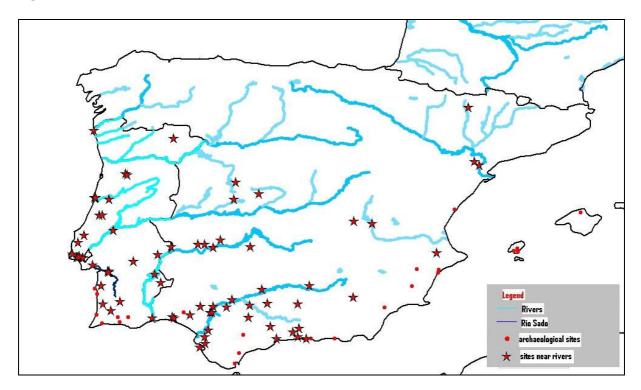


Fig. 3: spatial query for the selection of settlements nearby the rivers

3.1.1 Some more specification on Spain and especially on Tartessos

A small digression is dedicated to the Spanish area on the border with Portugal, focalizing on ancient Tartessos and the Phoenician colonies which settled here affecting a large section of the South-West Spain and the neighboring South of Portugal. Tartesso was probably placed in a area where a cultural facies was present during the II millennium BC, extending to southern Portugal and Extremadura. Signs of this material culture appear between the Late Bronze Age and the Iron Age. Now is well known that an indigenous cultural horizon rooted in the Bronze Age came into contact with Phoenician during VIII-VI century BC. By that time Phoenicians played an important role in the evolution of that whole area (involving also Southern Portugal). Traditionally the Phoenician presence in that area is associated with the exploitation and trade of important metals, especially silver. For example the mining of silver in Rio Tinto region reached a climax during the VIII-VII century BC and in this huge exploitation justify for example the foundation for example of Gadir settlement, the principal harbour for metal trade obtained from Tartessian area. Many settlements of that area where in close contact to mines, while several others had a function of landing points and emporia. Phoenician colonies on the shores of Granada, Malaga and Cadiz had already been founded in VIII century BC, and early activity is also found in Toscanos, Chorreras and Mezquitilla. That allochthonous influence and the development and flourishing of a complex culture here is also attested by the appearance of tartessian gold, silver and bronze treasures like the ones of Aliseda, Carambolo, Ebora and Sines and ivories from Carmona (M.E. Aubet in Schubart et al., 2002). Indigenous settlements were located in strategic areas that dominated the communication routes near Guadalquivir river (Carmona, Carambolo, Lebrija, Montemolin, Ecija) and many local settlements were also found at point of access to silver and copper mines (Cabezo de San Pedro and San Bartolomè de Almonte in Huelva). The zone of Huelva is a mining area which contains a complex ore formed by conglomeration of minerals. Pyrite predominates but there are also large quantities of copper, lead, zinc, gold and silver (J. Fernandez Jurado in Schubart et al. 2002). In Rio Tinto the silver extraction took place since the 2nd millennium and the technique of cupellation was already known (M.E. Aubet in Schubart et al., 2002). The Phoenician presence probably exert a triggering effect in the exploitation of mines in that area. The shoreline that goes between Abdera (Adra in Almeria) and the ancient Malaka (Malaga) contains for that reason a considerable density of Phoenician settlementstat were

established by the VIII century BC. Malaga, Granada and Almeria are located in areas rich in metals. In Baria ancient silver mining activity are attested (M.E. Aubet in Schubart et al., 2002). The archaeological findings in the mining center of Tejada la Vieja, San Bartolomè de Almonte and Penalosa reveal the existence of extraordinary activities in the mining and processing of silver (M.E. Aubet in Schubart et al., 2002). The excavation carried on here have provided information on the mineral ores, which was principally gossan while the metal was silver. It mustn't be forgotten that copper, silver and gold was found also in Sierra Morena and Northwestern area. The introduction by the Phoenician of technological innovations into indigenous metallurgy would have increased the yield of local mining and metal working. The VII century constituted the boom of Phoenician trade in the west. In this period the spread of Phoenician goods reached the entire Guadalquivir and Guadalhorce valleys and also territories in the inland, and the cultural exchange that took place with that oriental presence can be easily seen for example in the grave goods and planning of necropolis like La Joja and Setefilla. Between 1100-700 BC the coastal territories of central southern Portugal were inhabited by indigenous communities who dominates river traffic. These settlements channelled copper from Alentejo, tin from Beira Alta and gold from Portuguese Extremadura toward the coast, as well as Phoenician product to the interior (toward Medellin, Aliseda and Merida). Tejo provided a natural route for accessing tin and copper in the interior. In the VII century BC Abul had the function of commercial settlement. This network was also controlled from the indigenous centers of Alcacer do Sal and Setubal. In eastern Spain Vinarregel, Pena Negra and Saladares, but also near Segura river, indigenous centers controlled the Phoenician commerce. The boom of the Phoenician trade culminated in the middle of VII century BC. with the founding of small trading colonies in Sa Caleta (Ibiza). During the V century the Tartessian crisis started, due probably to the fall of Tyre or the exhaustion of the mines of Huelva or the native hostility but none of these reasons are currently accredited. This period is characterized by the abandonment of many Phoenician settlements on the shore such as Cerro del Villar and Toscanos and the movement of the activities towards the inland (M.E. Aubet in Schubart et al., 2002). This paragraph is intended to be a further focalization on the connection between Phoenician presence, mining activities and local population. The thesis go on taking into account a lager number of mines and the study of compositional data on metal artefacts coming from Portugal and Spain and collected from from literature.

3.2 Mineralogical characterization of Iberian Peninsula

The geological characteristics of Portugal follow the general features of the Iberian Peninsula: the prevailing rocks are the granitic and metamorphic strands of Precambrian and Paleozoic age, which form the base of the Meseta. Relatively recent volcanic formations are present between Sintra and Lisbon and the Algarve. Exclusively sedimentary formations of the marine environment, mainly limestone outcrops, characterizes Estremadura and a smaller part of Algarve. Also sedimentary rocks of continental or sea origin belong Neogene Era, with alluvial deposits, which are present in the valleys of the Tagus and Sado, emerge in a narrow band along the coast north of Capo Carvoeiro. In order to explain briefly the localization of anciently exploited mining areas in Portugal, it is easier to divide Portugal in four different mineralogical zones: South Portuguese plate, Ossa Morena Zone, Central Iberia zone and the Iberian Pyrite Belt. In the north of Iberian Peninsula another important area is the Iberian Tin Belt. Each of these areas was rich in metallic ores usable also with the ancient metallurgical techniques, especially in the Baixo Alentejo, Estremadura, Beira Alta and Algarve.

The Iberian Pyrite Belt extends from Portugal to Spain in southwest Iberia and constitutes one of the world largest reservoirs of massive multi-metal sulphide deposits (Fe, Cu, Zn and Pb) (Barriga *et al.*,1990, Saez *et al.*, 1990). This area was already exploited in ancient times for the extraction of lead and copper sulphides, easily smelted for the production of copper and lead trough a roasting phase (to convert the sulphide in oxide generally: metal sulphides + molecular oxigen => metal oxides + sulfur dioxide), and then the reducing of the oxide to metal. Archaeological evidences enlightened the presence of Canal Caveira ancient mine for the smelting of lead sulphide, where a slags assemblage has been found. Always in the Alentejo there is Aljustrel, well known area for the discovery of two metallic tables (Vipasca Tables) of Adrian period, where a ancient epigraphy explains the techniques of mining and the related fiscal charges, but also for the discovery, between the archaeological settlement and the Guadiana river, of a large amount of slags. Aljustrel mineral deposit was exploited from Eneolithic Age (II millenium BC) and is composed of copper oxide, carbonate and primary and secondary copper sulphate (calcopyrite and calcosite) (Domergue, 1987).

From the Sul Portuguese must be enumerated Sines, Castro Marim and Vila Nova de Milfontes Phoenician archaeological sites, were metallurgical operation were executed.

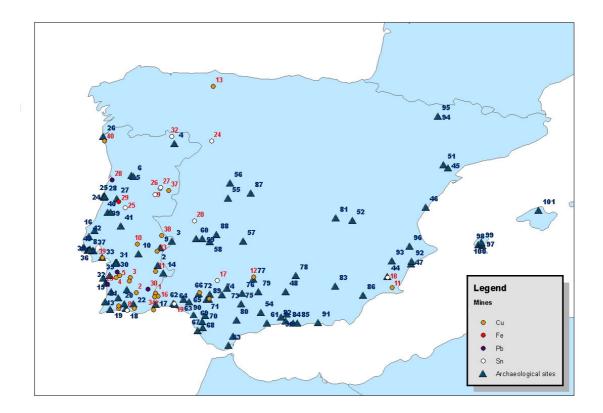


Fig. 4: GIS thematic map on mines (dots and red numbers) and archaeological settlements (blue triangles and numbers)

Mines: 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) 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

In Ossa Morena, veins of lead and copper were widely exploited in ancient times and near Santa Olaia there was an exploitable mine of tin ore (cassiterite), which was one of the most sought ore because of the highly difficulty in its extraction (the mineral was either too finely dispersed in the embedding rock and the richest veins localized at unreachable depths) and the scarcity of mineral deposits accessible with the ships. As it was said in the previous paragraph, Santa Olaia was one of the most controlled Phoenician sites in the Portuguese panorama.

In the Central Iberia zone veins of copper, tin and lead ores were exploited between the Bronze Age and Iron Age.

These are some of the major mines in Portugal. In this case it was decided to produce a database of mines (fig.4 in the previous page) exploited in antiquity (and the unsure reservoirs) in the Iberian Peninsula (40 mines collected from literature, mainly in Portugal), which are specified for the three important elements for the bronze alloying (Cu, Sn, Pb). From the observation of the map which contains both the archaeological sites and mines exploited, is possible to note the close connection between mines and sites.

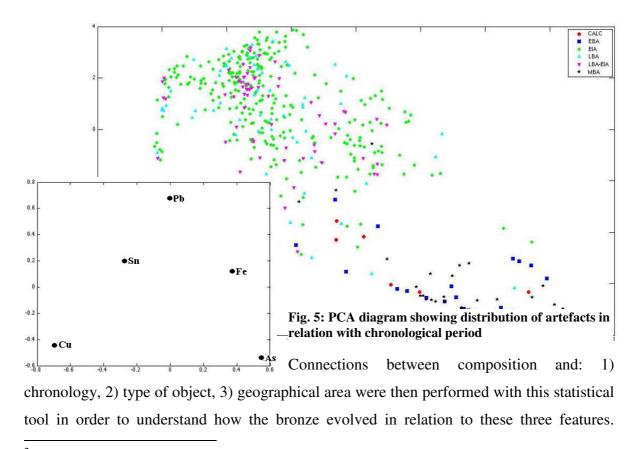
3.3 The archaeometallurgy in Portugal between LBA-IA

The ancient metallurgy in the Iberian Peninsula is described by numerous archaeological finds that dot this area in a very broad chronological range. Before starting the specific discussion of the bronze artefacts between the LBA-IA particularly in Portugal a small digression is made again for Tartessos area. The localization of the main harbor city is unsure and broadly discussed. The discussion on metallurgy starts with Tartessos because, as said previously, this was a main important center for metal commerce (especially the trade of tin, silver and copper). That's the reason why this area became an important landing point for Phoenician ships. The presence of huge amount of metal artefacts in Huelva, El Carambolo, Medellin, La Joya, let interpretations propend for a localization of this cultural area in Western Andalusia, Estremadura and Southern Portugal. Phoenician metalworkers produced a huge quantity of pyrometallurgical residues in the Rio Tinto. Mining and smelting in this area was furthermore stimulated by Phoenician commercial interest, and whose presence let an orientalizing phase to start.

Bronze artefacts of local, Phoenician and mixed origin, from the Mediterranean Basin sites as a whole, and from the Iberian Peninsula in particular, have been the subject of several archaeological/archaeometric studies combining typological characterization with a variety of advanced chemical and physical analytical techniques (Avila, 2002, 2004, 2005; Harrison & Craddock, 1981; Braganca, 1979; De Caro et al., 2002; Delibes de Castro et al. 2001, Figueredo et al. 2007-2011; Gomez Ramos et al., 1997; Melo et al., 2007; Monge Soares 1976, Montero Ruiz, 2003 a,b; Perez & Iglesia, 2008; Rovira, 1985, Rovira et al., 2005; Romero & Albeda, 2010; Tavares da Silva ,1978; Valerio et al., 2010-2012). Some of these studies suggest that, during Late Bronze Age, Iberian metallurgy (between the end of the II millennium to the early I millenium BC) was traditionally characterized by the use of binary Cu/Sn alloys with Pb < 2%, and with Sn content between 8-14%. On the counterpart bronze artefacts from Northern area such as British Isles, Western France and North Western Iberia (the so called Atlantic area), already in LBA present ternary bronze compositions. Higher variability in Sn and higher Pb contents (together with Fe content > 0.05%) became common at the beginning of the Iron Age (about 800-400 BC), concurrently with the establishment of the first Phoenician settlement in the South Western coastal regions of the Peninsula. It is well known that the Phoenicians used to introduce new technological know-how in their most important colonies. In the metallurgical field, P. Valerio et al. (Valerio et al. 2010-2012) and E. Figueredo et al. (2009) indicate that the evolution toward the use of ternary alloy (Cu-Sn-Pb) started in the south central areas of Iberian peninsula during orientalising period. Lead enhances the fluidity of the molten bronze alloy and increases the temperature solidification range, thus facilitating the casting of decorative objects (Figueredo, 2007-2011; Valerio, 2010-2012). The presence of iron can give a lot of information, from the smelting of ferrous minerals to the use of high technology in the pyrometallurgic process (De Caro et al., 2002). From this point of view, the presence of iron should suggest that the furnace used for the ore smelting operated in a reducing environment, so the presence of this element can be found in the metallic bath (Valerio 2012). This kind of furnace, as the use of fluxing agent, were probable results of technological transfer from allochthonous influences, and in other words could also be introduced by the Phoenician metalworkers during their prolonged contact with local cultures (Valerio 2010-2012; Figueredo, 2007-2011). It is however important to note that iron presence could suggest the nature of smelted ores (i.e. chalcopyrite), so is important to understand where iron is found in the object. Autochthonous Iberian productions and imported Phoenician technology and rendering therefore frequently merged and gave rise to a new, "hybrid" typology of production, especially during orientalising period, that could

be characterized archaeometrically by distinct alloys compositions and archaeologically by a distinct tipology (Avila, 2000; Valerio, 2010-2012). In the case of the southwesternmost regions of the Iberian Peninsula, including what is now Central-Southern Portugal, archaeometallurgical studies on Late Bronze Age-Early Iron Age artefacts are increasing in recent years (Figueredo *et al.* 2007; Valerio *et al.*, 2010-2012;); in these regions and in general in the Iberian Peninsula there are important examples that will be used herein to make consideration on productions (local or allochthonous).

At this step of the research a database containing analytical data coming from similar studies on bronze artefacts from the Chalcolithic era to the EIA (Early Iron Age) has been collected from literature (specified for the location of archaeological settlement, type of settlement (Phoenician, "orientalizing" or local), chronology, classes of object (tools or decorative object) and composition (Cu, Sn, Pb, As, Fe). More than 500 compositional data² has been collected and statistically analyzed by the mean of PCA (see appendix B).



² The core database has been collected from literature (Avila, 2002, 2004, 2005; Harrison & Craddock, 1981; Braganca, 1979; De Caro *et al.*, 2002; Delibes de Castro et al. 2001, Figueredo *et al.* 2007-2011; Gomez Ramos *et al.*, 1997; Melo *et al.*, 2007; Monge Soares 1976, Montero Ruiz, 2003 a,b; Perez & Iglesia, 2008; Rovira, 1985, Rovira *et al.*, 2005; Romero & Albeda, 2010; Tavares da Silva ,1978; Valerio *et al.*, 2010-2012) during my Ph.D. A Master Degree thesis on Conservation of Culturale Heritage has been activated (student name: Valerio Graziani in cooperation with my PhD thesis) on this topic, and the database currently reaches more than 2000 compositional data. The implementation has been carried on by V. Graziani and a PCA elaboration is now under processing.

Geometrically the PCA aims to presenting the data in a reference system that better illustrates their structure (reference exchange). Axes will then be translated to describe the dataset, according to the direction of biggest explained variance (see appendix B for a detailed description of PCA). The data in the first plot show the distribution of samples (score plot, where the values of the scores are the coordinates of the objects for each main component) in relation with the chronology (fig.5) and the second image is the loading plot that shows the allocation of the variables components (Cu, Pb, Fe, Sn, As), the direction of their growth and the importance (the weight, the number associated to each element considered in absolute value) that each numeric variable has on the determination of the two principal components. The plot in fig. 5 shows the evolution trend from Chalcolitic (main use of arsenical copper) to the adoption of bronze alloy during advanced phases of BA and IA (leaded bronze especially found in EIA). The green dots and triangles used to represent LBA and EIA bronze artefacts show this technological evolution from binary bronze to ternary bronze with higher level of iron. Since the data are fluctuating into a wide range of composition, other PCA plots have been elaborated to distinguish between tools and decorative objects. In fact another important technological evolution was the use of different alloys for the production of different classes of objects.

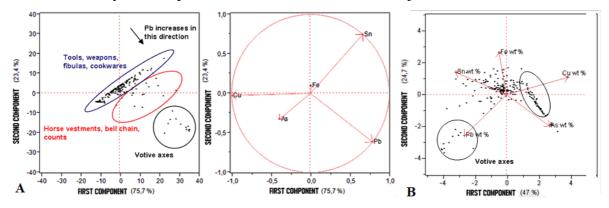


Fig. 6: A) PCA plot on covariance for the tools and non-decorative objects. The second picture shows the loading plot of the variables. B) PCA biplot on correlation

Tools (knives, swords, rasps, axes, etc ...), which were expected to have a better mechanical performance, were produced mainly with binary alloy, since the presence of lead in the alloy, totally immiscible in the solid phase Cu-Sn, created structural discontinuities along the edges of the globules of lead, causing weaknesses in the object (and fractures). Instead decorative objects, whose required mechanical performance was minimal, were often produced with the available and less expensive metal, which is the reason why an increase of the lead in the alloy and an increase in the levels of arsenic is expected, generated by the

reuse of metal through recasting of old objects. From the PCA graphics (fig.6), several information have been obtained. The plot A have been elaborated on covariance to better show the variability for the main elements in the alloy (Cu, Sn, Pb) while B biplot (produced by an overlapping of scores and loadings plots) is calculated on correlation, better showing the contribution of minor elements (Fe, As). Is important to bear in mind that PCA plots on covariance and correlations must be read separately, using their own loading plot (for the PCA on covariance it was created a separate loading plot, located to the right of the score plot, while for the correlation has been decided to use a biplot that overlaps the loadings and the scores values). From the plot is possible to notice the unexpected presence of some tools which contain lead, although the majority are made of binary bronze. The leaded tools (surrounded by a oval in the two diagrams, toward the direction of lead), from a deeper study of the plots, seem to be almost votive axes and objects of common use (not "tools" in the strict sense of the word, in the red oval in plot A). This could be explained deducing that metallurgy was differentiated in relation to the type of object. Another set of PCA diagrams has been produced checking the trend of the compositional values in relation to the type of the object: local, "orientalizing" or "of Phoenician inspiration". In the first diagram the local artefacts show binary bronze composition (fig.7). In fact the Pb variable does not show high loading values on the PC1, both considering the PCA plot calculated on covariance (A) and correlation (B). This demonstrate that this element is not very decisive in explaining the dataset variability for local artefacts. Furthermore the point cloud is positioned preferentially toward the direction of copper and tin and not toward lead. In the plot, surrounded by the black oval, are located the artifacts made of copper and the others are characterized by binary bronze with values

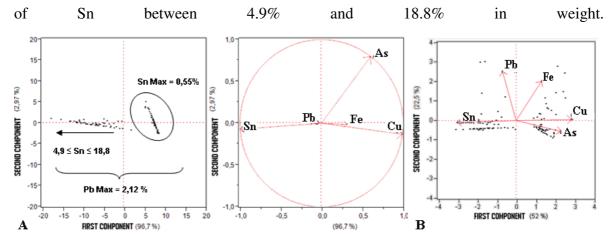
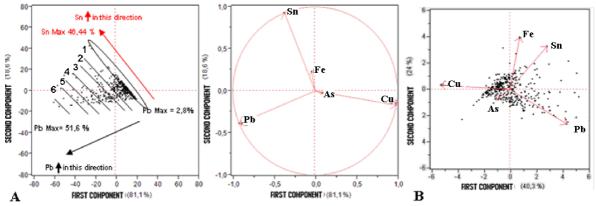


Fig. 7: PCA plot for local objects, A) PCA on covariance; B) PCA biplot on correlation

In conclusion by plots in fig.7 is possible to see that local Iberian production was traditionally connected with copper and binary bronze production (as reported by the literature on this argument).

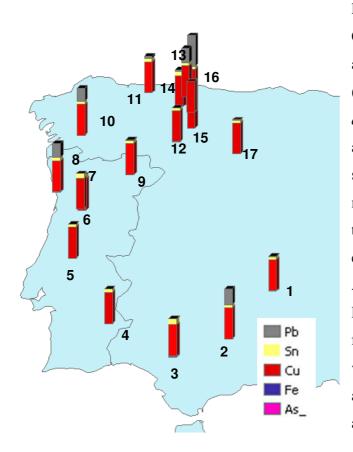
Fig. 8: PCA plot for "orientalising/ Phoenician-inspired"objects, A) PCA on covariance; B) PCA biplot on correlation. In the score plot A the oval collects all the Pb values which are up to a maximum of 2, 8/ wt%. The lines and the adjacent numbers indicate classes of artefacts with comparable Pb values. Pb grows till a maximum of 51,6 wt %. The numbered classes are explained in the text.



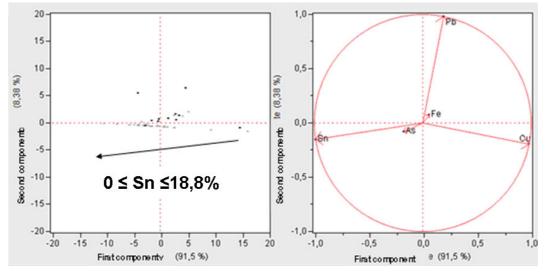
The diagram above shows the compositional trend of the "orientalizing" and "Phoenicianinspired" artefacts going towards ternary bronze alloy (fig.8). In fig. 8 A (PCA on convariance) and B (PCA on correlation) some features have been enlightened and using the loading plots (on the right for A plot and inside the biplot B) it is easy to read the PCA results. Also in this case distinction between this two calculating methods of PCA have been executed to show how much major and minor elements in the alloys describe the explained variatiation of the dataset. These artefacts are composed of ternary bronze alloy for the part of the cloud of points that grows following a route that goes from the centre to the left corner at the bottom in plot A (with increasing values of Pb for increasingly negative values of first and second principal components). In biplot B the ternary alloys are located mainly in the right-bottom of the plot. Using both the methods is possible to notice higher loading values (considering absolute volues) for Pb variable, showing that for "orientalising" or "of Phoenician inspiration" artefacts this element is importante in the determination of explained variance of the dataset. Arsenic for example is not important for the description of compositional values as it is described by low values on both PC1 and PC2 also considering correlation matrix. All the points surrounded by the oval (in fig. 8A) are the scores for of artefacts that do not contain Pb in the alloy. From the oval toward the bottom left angle (fig. 8A), lead values increase till the maximum (51.6 wt %). Equidistant lines in fig. 8 A have been drawn to separate roughly and to distinguish classes of artefacts

in which the values of lead are similar. The classes have been numbered to identify unambiguously these different Pb values: area n.1) Pb content approximately between 3-5 wt %; area 2) Pb content approximately between 6-9 wt %; area 3) Pb content approximately between 10-15 wt %; area 4) Pb content approximately between 16-29%; area 5) Pb content approximately between 30-45%. So, within two adjacent equidistant lines, Pb can be considered fairly constant. Increasing values of tin follow the tin arrow in the loading plot on the left. From the statistical study of these data, it is possible to note a prevalent use of the ternary alloy in objects of "orientalizing/Phoenician-inspired" type. Unfortunately is difficult to state with certainty if this change was due to external triggering or if it was a self - produced technological evolution. Further study can be done through additional PCA analysis for distinct chronology and through a spatial study with GIS system. The distinction between Spain and Portugal, fictitious if considered ancient times, it is actually useful to distinguish approximately coastal areas on both sides of the Strait of Gibraltar and considering Tartessos.

Fig. 9: GIS thematic map for compositions of bronze objects (LBA) in: 1 Amagro; 2Andalucia; 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



The GIS thematic map (fig. 9) shows artefacts from Portugal and Spain. During Late Bronze Age in Portugal the mainly used alloy was Cu-Sn. In Northern Spain and Porto, already evidenced by authors as (Figueredo et al.2007,2011; Valerio et al. 2010-2012) ternary bronze was already used, while in Central and south of Spain, as in Portugal, the main used bronze alloy was Cu-Sn. In the site n.2 the high amount of lead is explained since the artefacts are Atlantic Axes. In fig. 10 PCA plot for Portuguese LBA artefacts shows a first slight change of compositional values with few artefacts with a small amount of lead: three "orientalizing" artefacts from Fraga dos Corvos have



amount of lead of respectively 2.01, 4.8 and 6.14 % in weight (Figueredo et al. 2007).



The tin values (fig.10) from the compositional data collected from literature for Portuguese artefacts (LBA) are between the "non-detection of the element" to a maximum of 18.8%. Besides these extremes, more common values are ranging between about 5 and 15 percent, typical of a "good bronze" with high mechanical properties(Figueredo *et al.* 2009). Going across the span of time between LBA and IA a pause on the hinge period of passage from LBA and EIA can be useful (fig.11,12).

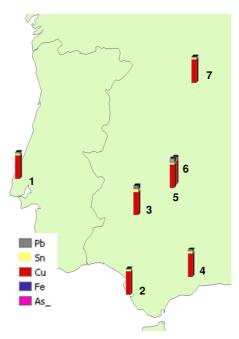


Fig. 11: GIS thematic map on bronze artefacts from LBA-EIA settlments: 1 Torres Vedras, 2 Cadiz,3 Villagarcia de la Torre, la Joya, 5 El Risco, 6 Siruela, 7 Coca

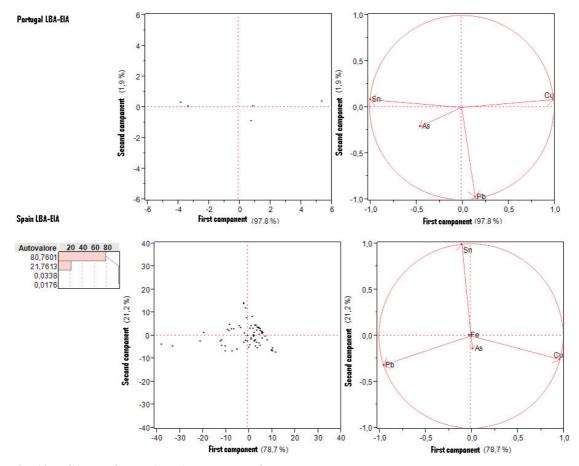


Fig. 12: PCA plot for LBA-EIA bronze artefacts From GIS map and PCA plots is possible to observe that in Spain is noticeable an evolution through ternary alloy, while in Portugal (Torres Vedras) one orientalising object shows a

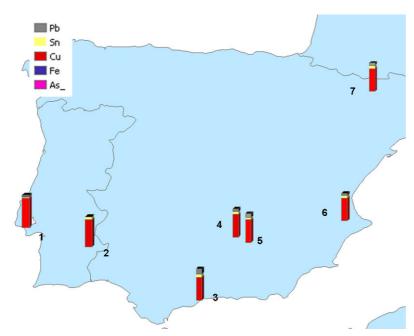


Fig. 13: GIS thematic map for EIA bronze artefacts: 1 Quinta do Almaraz, 2 Castro dos Ratinhos, 3 La Luz, 4 Collado de los Jardines, 5 Castellar de Santisteban, 6 El Palomar,7 Lujan

small amount of lead (3.52%) while in objects from Castro dos Ratinhos, datable to period, same the composition is a binary bronze (maximum Pb: 0.97 % and Sn values between 7.8 and 14.3%). During EIA (fig.13, 14) stronger compositional changes are notable in Spain, while in Portugal, as we will see later, the evolution is slower and only around the Middle Iron Age vastly different values can be appreciated. Is important to remember here that stronger commercial contacts were established between Phoenicans and Tartessians, that could suggest a faster assimilation of thechological transfer.

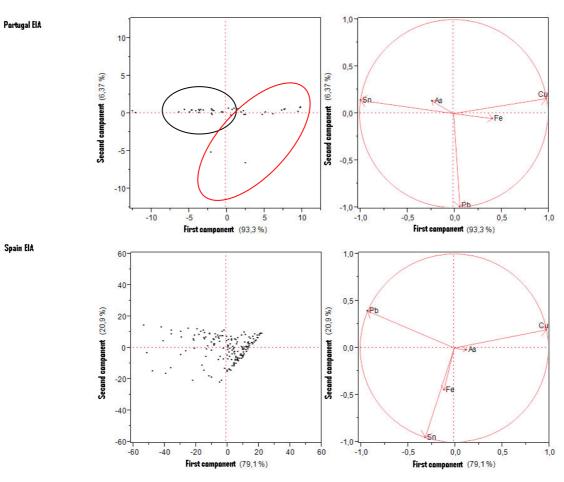


Fig. 14: PCA Plots for EIA Portuguese and Spanish artefacts: the points surrounded by red circle belong to Quinta do Almaraz while the black oval contains Castro dos Ratinhos artefacts From the PCA plots for Early Iron Age in fact is possible to note that a high increase in the values of the lead and an increase in the variation of those of the tin for the bronze artefacts from Spain. In Portugal two Iron Age sites have been inspected, Castro dos Ratinhos and Quinta do Almaraz ("orientalising"), evidencing different information. While in Castro dos Ratinhos, located in the inland, the compositions show quite the same values for Sn and Pb to the LBA (maximum tin content around 15.9 % and a minimum of 6.2%) in Quinta do Almaraz, positioned nearby the sea, the bronze composition fairly change. Tin decreased to very lower levels (maximum around 8.1%) and two leaded bronze artefacts appear (Pb 4.6 and 5.9 %). An observation in this regard was made by Valerio et al. (2012), who suggested that, generally, technological transfer from allochthonous presence could have been

received faster from all the sites in close contact with this culture (in other words the sites near the harbour and landing points). The inland sites received that stimuli with a delay of several decades.

All of this preliminary study was used in the following paragraphs for the study of a museum collection in the Museum of Evora. Is important to note conclusively that in Biblos the ternary alloy was already used from Middle Bronze Age (El Morr & Pernot, 2011), favouring the hypothesis of the technological transfer from the Phoenicians.

3.4 Introduction to the case study: votive bronze statuettes from Evora Museum collection

The introductory paragraphs explained which were the archaeological, mineralogical and metallurgical peculiarities of Portugal before and during the period of Phoenician and Punic presence. Now is possible to better understand which was the thematic under study in this chapter.

Eight bronze statuettes (ex-voto) of unknown provenience and dating, belonging to the Evora Museum (fig.15), have been studied to determine the original archaeological sites (this part of the research is published in N.Schiavon *et al.* 2013). The problems related to the collection are the total absence of information on the archaeological provenance of the 8 pieces: the only reliable information is that they had belonged to the private collection of Arcebispo Manuel Do Cenaculo (XVIII century).



Fig. 15: Evora collection of votive statuettes

In the collection there are 4 anthropomorphic figures, three males and one female, that show disproportionate bodies: the trunks are too long compared to the legs and the head is diamond-shaped. One of the male figures has a hand near his mouth posed in the open position, as if the little man was throwing a warning. All the figurines are considered by authors as produced with the lost wax technique and cannot stand without a support. This is why is possible to hypothesize that they were decorative elements of votive lances, swords or banners. The three animal figures are goats, also produced with the lost wax techniques, show a geometric design on the mantle and a relatively more naturalistic bodies. The last artefacts is an anthropomorphic part of a tripod. These statuettes where studied as a comparison to Alcacer do Sal similar artefacts by the archaeologist Esmeralda Gomes in her master thesis, proposing a probable common provenance of the two collections.

This type of ex-votos is present throughout the Portuguese Estremadura and the Alentejo, but also in Spain. These zoomorphic statuettes were defined in 1895 by J. Leite de Vasconcellos (Leite de Vasconcellos, 1895) as votive objects consecrated to Adaegina, who had her cult in Lusitania and Spanish Extremadura. In fact some of these goat statuettes, found in Caceres, show an inscription on the base, consecrated to the Lusitanian deity Adaegina, synonym for Proserpine. The archaeologist Augustus Filippe Simoes (1878) played the 4 human figures as idols belonging to religions from East (Phoenicians and Egyptians) who had cults linked to masculine and feminine elements, symbolized by Baal and Astarte. The goats, according to Simoes, were objects of worship in Egypt. The human figures are then interpreted as smiting Gods in 1992 by L. Berrocal-Rangel (Berrocal, 1992), which binds well to the cult of the Phoenician god Baal and Astarte. Another thing to keep in mind is that the type is fully equal to many zoomorphic figurines found in Luristan. The collection was chosen for the study because it presented exactly the archaeometric problem under study: the need to distinguish between Phoenician, local or "orientalizing/of Phoenician inspiration" production. Aim of the study was therefore twofolded: a) provide for the first time analytical data on the Evora statuettes in order to obtain their provenance; b) assess whether the combined, non destructive and relatively simple methodological approach adopted in this study could shed light on the interaction between local (Iberian Peninsula) and allochthonous ("orientalizing"-Phoenician) bronze making technology and, in turn, be useful to understand if/how the "orientalizing" influence had been locally modulated.

3.5 Materials and Methods

Preparation of the samples

To avoid contamination from superficial corrosion patinas, the surface area to be analysed of all the artefacts under study were cleaned by grinding with abrasive paper of different grit sizes and diamond paste to obtain a cleaned area of about 30 mm². The cleaned areas were then analyzed by EDXRF performing at least three measurements per each statuette; care was applied to analyse only the middle/top areas of each statuette in order to obtain meaningful elemental alloy compositional data least affected by compositional heterogeneity, caused for instance by the migration of Pb in peripheral areas. As it has been the case in the study of other archaeomaterials from Southwestern Iberia, the spatial resolution and phase contrast of SEM Backscattered electron imaging proved to be useful in the investigation of the micro-texture of the bronzes, while the EDS system provided 2D major elements maps highlighting compositional heterogeneities.

EDXRF

The spectrometer was an Eclipse IV Oxford Instruments X-Ray tube (45 kV, 50 μ A, 2.25 W) with a Rh anode. The out-coming radiation was collimated by a tantalum collimator resulting in a 5 mm diameter beam at the sample surface, placed at 55 mm distance from the tube and 10 mm from the detector. The Super silicon drift detector (SDD) was an Amptek XR-100SDD with a 25 mm² detection area (collimated on 17,2 mm²) and 500 μ m thickness, and with a 12.5 μ m Be window. Energy resolution was 140 eV at MnKa FWHM 5.9 keV and the acquisition system was an Amptek DppPMCA. The angle between the incident and emitted beam was 90°. This geometry allows for a high background reduction due to Compton scattering. The sample was positioned at the focal point of two laser beams. The X-ray generator was operated at 40 kV and 20 μ A, with an acquisition time of 150 s. All measurements undertaken were carried out in ambient air. Data were processed and quantified using the WinAxil 4.5.2 software comprising fundamental parameters and experimental calibration factors.

SEM+EDS

A HITACHI S3700N Variable Pressure-SEM, interfaced with a Quantax EDS microanalysis system has been used. The Quantax system was equipped with a Bruker AXS X-Flash® Silicon Drift Detector (129 eV Spectral Resolution at FWHM/Mn Kα). Standardless PB/ZAF quantitative elemental analysis was performed using the Bruker

ESPRIT software. The operating conditions for EDS analysis were as follows: backscattered electron mode, 20 kV accelerating voltage, 10 mm working distance, 120 mA emission current. The detection limits with this configuration for major elements (> Na) were in the order of 0.1 wt%. The large chamber of the HITACHI S3700N allowed the direct analysis of the small statuettes without the need of sub-sampling therefore permitting to perform a totally Non Destructive (ND) type of investigation.

3.6 Analytical results

Tab. 1: XRF quantitative data

By the XRF (tab 1) qualitative analysis is possible to establish that the 8 statuettes are made of the same kind of alloy, a leaded bronze. The overlapped spectra of the prayers n.3296 and 3300, show that their trend is similar; the same has been observed between 3298 and 3299, in this case the leaded bronze has less tin. All the spectra obtained from the goats statuettes and from the tripod element are also similar. The computation of the quantitative results (tab. 1) with Win Axil software shows that the eight statuettes are made of a ternary alloy consisting of copper, tin and lead. The presence of iron is higher than the XRF instrumentation limit of detection.

Results from EDXRF analyses of artefacts from Evora (Axil X-Ray analysis software). Values in wt.%.								
Artefact (n.inv)	Description	Fe	Cu	Sn	Pb			
Evora 3296	prayer (male)	1.50 ± 0.02	66.60 ± 0.10	5.80 ± 0.20	26.10 ± 0.20			
Evora 3297	Goat	0.09 ± 0.01	74.43 ± 0.10	6.99 ± 0.20	18.48± 0.10			
Evora 3299	prayer (male)	0.08 ± 0.01	86.23 ±0,10	2.20 ±0,08	11.49 ± 0.10			
Evora 5062	Tripod	0.30 ± 0.07	86.20 ± 0.10	9.20 ± 0.20	4.30 ± 0.10			
Evora 3300	prayer (female)	0,46 ± 0.01	63,63 ± 0,10	$12,30 \pm 0,30$	23,61 ± 0,20			
Evora 3295	Goat	0.04 ± 0.01	80.27 ± 0,10	11.00 ± 0,20	8.70 ± 0.10			
Evora 3294	Goat	0.04 ± 0.01	84.36 ± 0.10	6.80 ± 0.20	8.80 ± 0.10			
Evora 3298	prayer (male)	0.21 ± 0.01	73.20 ± 0.10	2.29 ± 0.10	24.30 ± 0.20			

By the XRF qualitative analysis we can establish that the 8 statuettes are made of the same kind of alloy, a leaded bronze. In figure 16 is shown the overlapped spectra of the prayers n.3297 and 3300, to show that the trend is similar for them; for the same reason in figure 17 the spectrum 3298 has been over imposed on the spectrum n.3299, in this case the leaded bronze has less tin. In figure 18 the anthropomorphic and zoomorphic artefacts are compared overlapping the prayer 3296 and the goat 3297 while in figure 19 the tripod 5062 is compared with a goat. At last in fig.20 the spectra obtained from the goats statuettes 3294 and 3295 has been displayed all together with the tripod element spectrum because the composition is in a median position between the two groups.

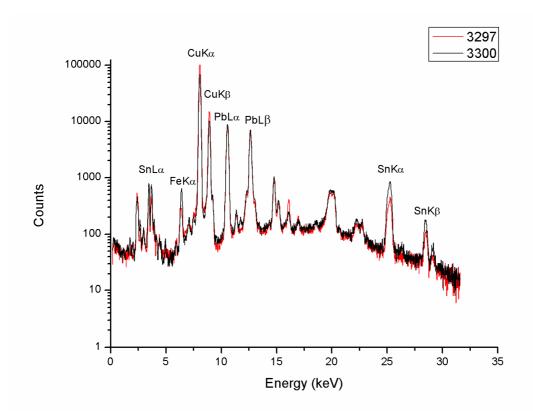
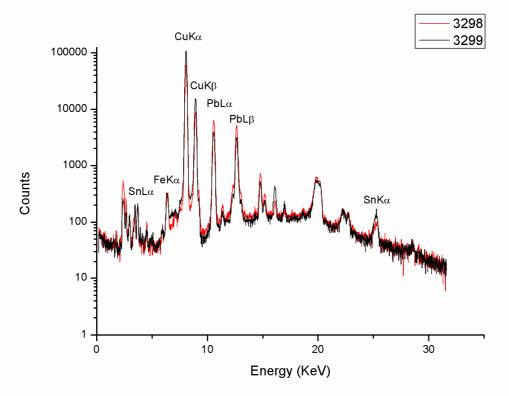


Fig. 16: EDXRF overlapped spectra of artefacts n. 3297-3300 Fig. 17: EDXRF overlapped spectra of artefacts n. 3298 and 3299



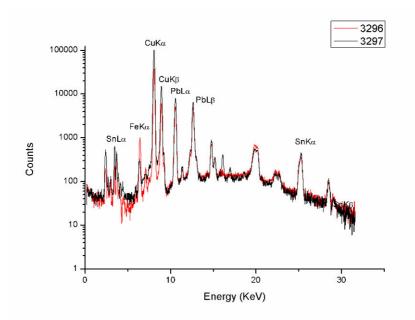


Fig. 18: EDXRF overlapped spectra of artefacts n. 3296 and 3297

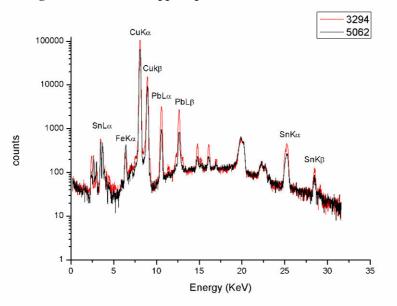


Fig. 19: EDXRF overlapped spectra of artefacts n. 3294 and 5062

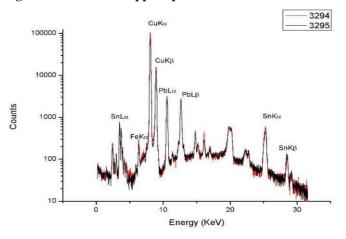
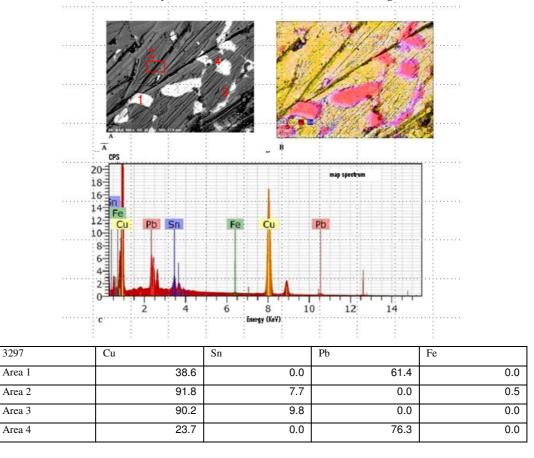
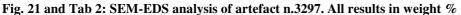


Fig. 20: EDXRF overlapped spectra of artefacts n. 3294 and 3295

The eight bronze artefacts have a similar composition, characterized as ternary alloy copper-tin-lead, in which fluctuations of the presence of lead, which is separated from the matrix, and the heterogeneity of the bronze itself, justify the presence of slightly dissimilar compositional data. It must be taken into account the common practice of reusing old metal for the melting of new objects and the non-standardization of the alloy when the object to be produced was decorative and non-functional such as a tool or a weapon that had to maintain certain technical qualities.

By observation of the image obtained with the scanning electron microscope it is possible to note how much the presence of lead characterize the alloy to the extent that it is not accidental but voluntary added. The quite large size of the lead globules (fig.21 and tab.2) of the sample 3297 could demonstrate that the cooling has been slow, probably using warmed or well insulated moulds, because the lead, insoluble inside the bronze in the solid phase, has had much time to separate from the matrix of bronze and congregate in large globules (for a general interpretation on grain size of ancient metals see Scott, 1992; this observation is here indicated as a hypothesis since that information in Scott is not given specifically for lead globules).





Similar lead globules has been noticed in the other artefacts SEM images and in the EDS compositional table a similar trend is shown, where the tin is mostly within the 8% in weight and is never higher that the 13% in weight.

Assigning a color to each element present it was possible to understand how the elements were distributed in the object, as the false color image carried over for the 3298 sample (fig.21 on the right). In a bronze matrix composed by copper and tin there are lead globules of big dimension showing a deliberate addition of lead in the alloy.

3.7 Using GIS system to find the provenance of Evora statuettes

The compositions of the eight statuettes are the starting evidence for the attribution of the collection to an archaeological site. Referring and consulting the two databases exposed previously, the ternary alloy seems to reach Portugal from the LBA/EIA, previously in the coastal areas. Asking the GIS system, in order to find matches in the nearby areas, about the presence of an LBA/EIA settlement where similar artefacts were discovered, the system answered founding Alcacer do Sal Phoenician settlement, where more than 50 votive statuettes, fully similar to the Evora's collection, have been found (fig.22).



Fig. 22: localization of Evora and Alcacer do Sal and pictures of two statuettes per each area

In order to validate the hypothesis of the origin of the Arcebispo Cenaculo's collection at this site, six statuettes (fig.23) preserved at the Museo de Alcacer do Sal were analyzed with EDXRF and SEM-EDS. Doc. Esmeralda Gomes (Gomes, Mestrado em Arqueologia) and Prof. Ana Margarida Arruda 2000). Portuguese (Arruda archaeologists who worked in this settlement, had for first noticed this similarity and the proximity of the two sites (Evora and Alcacer do Sal). Dr.ssa Gomes has also given

permission to conduct the analysis of six of the statuettes coming from the site, for SEM-EDS and EDXRF analytical sessions.

3.8 Alcacer do Sal artefacts

Alcácer do Sal is a settlement where the IPPAR (Portuguese governmental institution for the architectonic patrimony) undertook excavations since the year 1993 until 1997 (Gomes, Mestrado em Arqueologia, unpublished). Alcacer do Sal was in a strategic position because the Rio Sado was seen as a way of penetration for the traders vessels in these areas. The trade of products derived from mining of iron and copper suggests that here can be conjectured also the metal processing. In Alcacer do Sal area, the archaeological level attributable to the Iron Age II contains sacred areas because of the presence of an important assemblage of bronze statuettes, human and animal figures dated to the V century BC. The human figures represent warriors (with the shield and probably a dagger in the hands) and worshipers (personification of the prayers with the arms raised, most of them are naked and with very evident genitals). The animal representations include horse (or donkey), caw, ox (or bull) and a single representation of dog (fig.23).

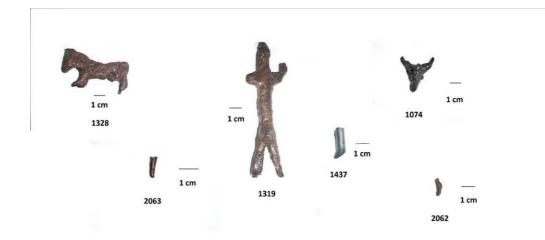


Fig. 23: Alcacer do Sal artefacts

According to their characteristics, they are ascribed to the Iron Age (V/IV centuries BC). Nicolini (1968) studied all the kind of votive offering in the Iberian Peninsula, and lists the positions of the offerings in relation to the position of the arms and legs. The statuettes of Evora's number 3298, 3300 and 3296 belong to the same type of prayers, the 3A (arms bent with hands at head level). The prayer of Alcacer (no. inventory 1319), also belongs to this category. Evora's statuette no. 3299, similar to the statue of Alcacer do Sal n.1 ,belongs to the position 15 D (left arm on his chest and right arm raised)(Gomes, Mestrado em Arqueologia). Six artefacts were selected from this archaeological site for XRF and SEM-EDS analysis. With the same analytical apparatus the six artefacts from Alcacer do Sal have

been investigated. The composition of these statuette can be ascribed as ternary alloy Cu-Sn-Pb, as for the Museu de Evora's collection, except for the artefact n. 1319, where the Pb content is 0.17: this low amount is confirmed by SEM analysis as is said in the next section, can be explained in the light of the common practice in the ancient time of recycling the metals. By the way, that spectrum is associated to the 1328 one, as they present a similar trend (fig.24); the same overlapping is made for 1047 and 1437 spectra (fig.25), and for the 2062 and 2063 ones (fig.26).

The type of the two productions is already very similar to a superficial study of the figurines. In the next paragraph the results of the XRF and SEM-EDS analysis will be shown and the SEM EDS false color images obtained on cleaned areas of the artefacts.

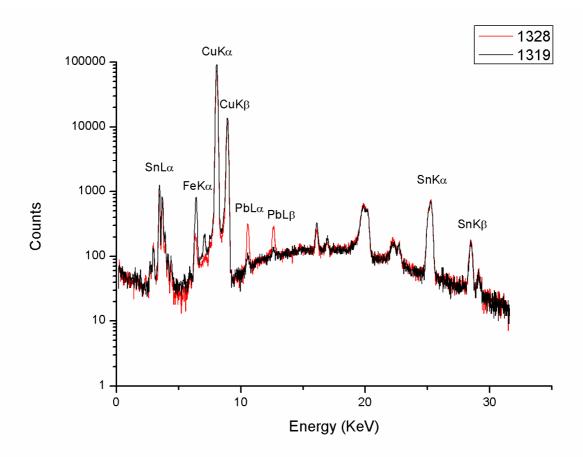


Fig. 24: EDXRF overlapped spectra of artefacts n. 1328 and 1319

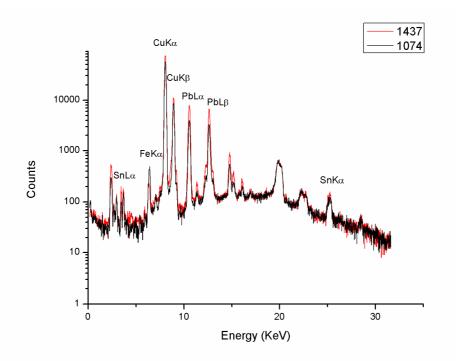


Fig. 25: EDXRF overlapped spectra of artefacts n. 1437 and 1074

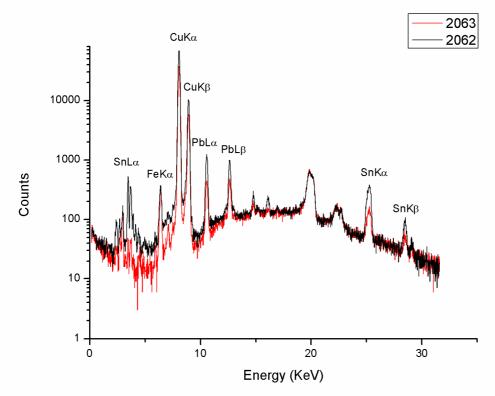


Fig. 26: EDXRF overlapped spectra of artefacts n. 2062 and 2063

3.9 Analytical results of Alcacer do Sal bronze statuettes in comparison with Evora's collection data

Textural analysis by SEM +EDS analysis of cleaned areas of the bronze samples (fig.27 and 28) shows that the Pb, due to its highly low miscibility with Cu in the melt and the immiscibility in the solid state, occurs as segregated, scattered inclusions with globular and inter-dendrite morphology: the large size and the very sharp and distinguishable edges of the lead globules observed could demonstrate that the bronze production process can be hypothesized considering a slow cooling phase also for Alcacer do Sal artefacts (for a general interpretation on grain size of ancient metals see Scott, 1992).

On the other hand, SEM + EDS analysis (backscattered electron imaging) of the two samples from Alcacer do Sal (1319-1328) with low Pb content confirms the absence of Pb Fig. 27: A) SEM Backscattered electron image and B) EDS spectrum of an Alcacer do Sal statuette

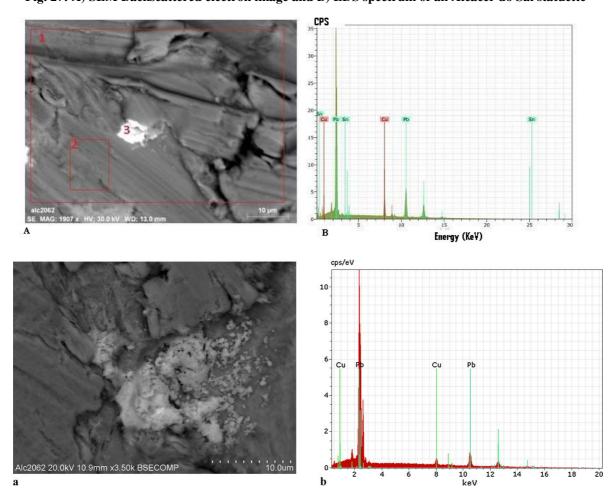


Fig. 28: A) SEM Backscattered electron image and B) EDS spectrum of an Alcacer do Sal statuette (2062)

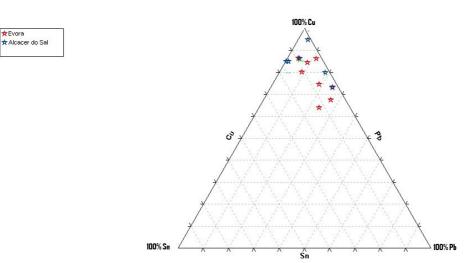
enrichment areas in the alloy.

In the table below the EDXRF quantitative data (tab.3) are given both for Alcacer do Sal and Evora's collections and a ternary diagram has been produced to compare easily XRF data (fig. 29). Qualitative EDXRF data interpretation performed by comparing superimposed spectra from the two sites confirms that the 8 statuettes belonging to the Evora museum's collection and the 6 selected ones from Alcacer do Sal are mostly made of the same type of alloy, i.e. a leaded bronze (Cu-Sn-Pb) alloy with variable Sn content.

Results from EDXRF analyses of artefacts from Evora, Alcacer do Sal (Axil X-Ray analysis software) Values in wt.%.							
Artefact (n.inv)	Description	Fe Cu		Sn	Pb		
Evora 3296	prayer (male)	1.50 ± 0.02	66.60 ± 0.10	5.80 ± 0.20	26.10 ± 0.20		
Evora 3297	Goat	0.09 ± 0.01	74.43 ± 0.10	6.99 ± 0.20	18.48± 0.10		
Evora 3299	prayer (male)	0.08 ± 0.01	86.23 ±0,10	2.20 ±0,08	11.49 ± 0.10		
Evora 5062	tripod	0.30 ± 0.07	86.20 ± 0.10	9.20 ± 0.20	4.30 ± 0.10		
Evora 3300	prayer (female)	0,46 ± 0.01	63,63 ± 0,10	12,30 ± 0,30	23,61 ± 0,20		
Evora 3295	goat	0.04 ± 0.01	80.27 ± 0,10	11.00 ± 0,20	8.70 ± 0.10		
Evora 3294	goat	0.04 ± 0.01	84.36 ± 0.10	6.80 ± 0.20	8.80 ± 0.10		
Evora 3298	prayer (male)	0.21 ± 0.01	73.20 ± 0.10	2.29 ± 0.10	24.30 ± 0.20		
Alcacer do Sal 1074	bull's head	0.42 ± 0.01	79.50 ± 0.10	1.88 ± 0.10	18.20 ± 0.1		
Alcacer do Sal 1437	human leg	0.06 ± 0.01	72.95 ± 0.1	2.50 ± 0.10	24.48 ± 0.20		
Alcacer do Sal 2063	bovine horn	0.40 ± 0.01	94.45 ± 0.20	1.15 ± 0.10	4.00 ± 0.07		
Alcacer do Sal 2062	human foot	0.24 ± 0.01	85.97 ± 0.10	8.80 ± 0.20	5.00 ± 0.05		
Alcacer do Sal 1328	bull (entire figure)	0.46 ± 0.01	84.70 ± 0.10	14.70 ± 0.30	0.14 ± 0.01		
Alcacer do Sal 1319	prayer (male)	0.07 ± 0.01	85.04 ± 0.10	13.79 ± 0.30	1.10 ± 0.02		

Tab.3: EDXRF quantitative data (Axill and Monte Carlo method) from the entire set of artefacts in the chapter (Evora, Alcacer do Sal)

Quantitative data (tab. 3) confirms the compositional similarities between the two sites. In particular, notwithstanding the fairly high variability in the range of Pb contents observed across the dataset (tab. 3), all bronzes from Evora and four out of six artefacts from Alcacer



collections show significantly high Pb contents with an average Pb value of 15,7 wt % in Evora and of 8.8 wt% in Alcàcer do Sal. Normalizing the quantitative XRF data for the Cu, Sn

Fig. 29: triplot of Evora's and Alcacer do Sal's XRF data normalized for Cu-Sn-Pb

and Pb elements has been possible to study both the collections using a ternary diagram (fig.29) which has allowed to identify groups of similar alloy. In detail is important to underline that these groups are composed from artefacts coming both from Evora and Alcacer do Sal, that show a very similar composition. The groups of artefacts are as follow and are related with the spectra in brackets: prayer 3298 3A, leg 1437 (fig.30); horn 2062, goat 3294, goat 3295 (fig.31); foot 2063 and tripod element 5062 (fig.32). This grouping is not connected on the typology or shape of the artefacts; this heterogeneity could be probably caused by the availability of raw materials or scrap metals use.

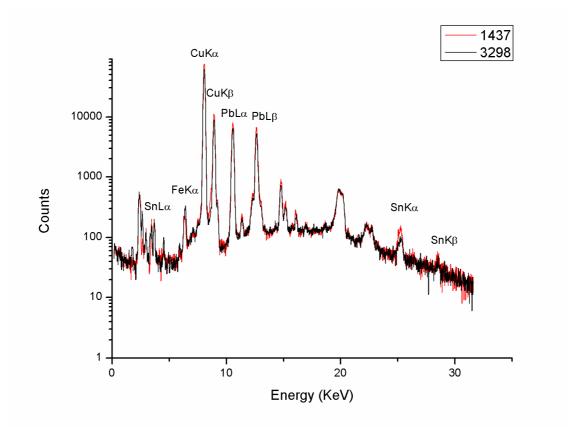


Fig. 30: overlapped EDXRF spectra of artefacts n. 1437 and 3298

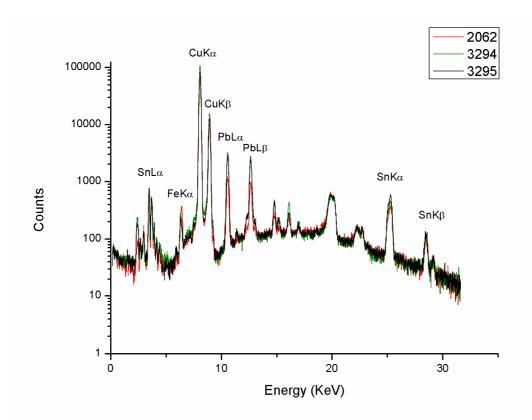


Fig. 31: overlapped EDXRF spectra of artefacts n. 2062, 3294 and 3295

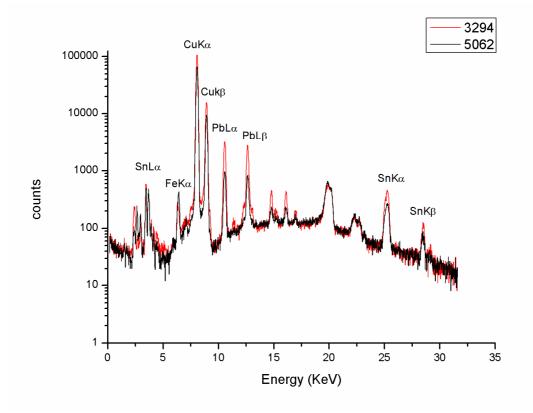


Fig. 32: overlapped EDXRF spectra of artefacts n. 5062 and 3294

This non standardized composition confirms that the craftsmen knew that this kind of decorative objects didn't need a particular mechanical performance or other particular technological features, also considering their small dimensions; on the other hand the biggest effort in a smithery was dedicated to the production of tools and weapons which was mechanically, thermally produced and whose composition was strictly controlled.

The significative presence of leaded bronze (i.e. Pb > 2wt%) in the objects under study is not unexpected as is noted on other bronze artefacts from Iron Age. Phoenician settlements in the Southwestern regions of the Iberian Peninsula revealed a significant usage of leaded bronzes, and indeed an increased use of leaded bronze was identified (and linked to a "orientalizing" production) also in the Southeast regions of the Peninsula (Perez & Iglesia, 2008; Rovira *et al.*, 2005; Prado Torreira, 1982). Nevertheless, the lead values in the Evora and Alcacer bronzes appear much higher than the ones reported for leaded bronze objects from other Phoenician settlements in the South of Portugal (Valerio, 2012). Pb contents in small bronze artefacts recovered from the Early Iron Age Phoenician/"orientalizing" Quinta do Almaraz settlement (9th–7th centuries BC), for instance, show values ranging between 4.6 and 5.9 wt% (Valerio, 2012). The reason behind the unusually high Pb content detected in this study could be manifold:

a) archaeological evidence assigns the Alcacer do Sal statuettes to the 5th century BC, therefore placing them two to four centuries after those of the Quinta da Almaraz settlement (9th -7th century BC). It may therefore be assumed that the propensity by the local material culture to adopt the "new" leaded bronze technique, somewhat low at the start (Valerio 2012), increased with the result that, by the beginning of Early Iron Age not only an increasing number of artefacts 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. In fact whereas only a few bronzes from the Quinta da Almaraz site contained Pb (the rest being binary Cu-Sn alloys), in the cases here investigated all (but two) samples were made of a Cu/Sn/Pb ternary alloys.

b) another factor that could explain the unusually high Pb content of the bronzes hereby investigated may be related to the post-Phoenician increasing rate of extraction of Pb from the abundant galena-bearing deposits present within the local Iberian Pyrite Belt ore (Barrica *et al.*, 1990; Saez *et al.*, 1999);

c) a third explanation may be more related to the specific functionality of the objects: it is well known that abundant low melting point Pb inclusions in bronzes, such as the ones encountered in this study, are responsible not only for a higher degree of castability during the bronze making process but also a flow hardness final metal product. The intentional addition of Pb, therefore, is often associated with the need to produce objects with complex decorative features, i.e. not requiring high strength, and a somewhat standardized alloy composition such as it is the case in the production of object destined to be used as weapons such as daggers, spears etc., (Valerio, 2012). This is the case of the statuettes examined in this study which, although in a somehow simplified way, do show attention to figurative details; moreover, their archaeological interpretation as ex-votos (dedicated either to local or to Phoenician "gods" (Simoes, 1878; Vasconcello, 1895), suggests that these were objects of "decorative or worshipping" production, again not requiring high resistance to mechanical stress or material strength: in this respect, the addition of lead could have facilitated the production process allowing faster (i.e. low temperature) casting which is typical of "as cast" technology in small statuary bronze production in the Iberian Peninsula (Monteiro Ruiz *et al.*, 2003).

In conclusion these high amount of lead for the production of decorative artefacts could be explained clearly with the development of a more skilled diversification and higher technological level attained for bronzemaking in relation to the kind and functionality of the objects themselves (Valerio, 2012). Except in one case (ref. 3295, goat in Evora), significant impurities of Fe are present in the bronzes from both sites investigated: content ranges from 430 ppm up to 1.5 wt % averaging in Evora 0.34 wt% and in Alcacer 0.37 wt%. These values are higher when compared with those of pre-Phoenician Late Bronze age artefacts in the Iberian Peninsula, which have been found not to exceed 0.05 wt %(Valerio, 2012), whereas appear fully consistent with increased Fe contents as reported in Early Iron-Age bronzes across Portugal (Valerio, 2012), Spain (Perez & Iglesia, 2008; Rovira et al., 2005; Craddock, 1987), and indeed the whole of the Mediterranean (Ingo et al., 2006). The widespread Fe increase from LBA to EIA has been explained by some authors with the introduction by the Phoenicians of a more efficient copper smelting technology, associated with higher reducing conditions attainable only in high technology furnaces (Ingo, 2002 a-b, 2006, Figueredo, 2010, Craddock, 1987). Additionally, it has to be noted that the bronze makers could have extracted copper from local mineral deposits containing high impurities of Fe, such it is the case with the sulphide deposits in the IPB where the mineral chalcopyrite is known to be present in significant amounts (Barriga, 1990).

The Sn content of the bronze statuettes investigated shows in both sites a high degree of variability ranging from 2,20 wt % to 12,3 wt % in Évora and from 1,15 wt % to 14,7 wt % in Alcàcer do Sal. Two compositionally groups may be identified: a low Sn one (Evora 3298, 3299 and Alcacer 1074, 1437 and 2063) and a high Sn one (Evora 3294, 3295, 3296, 3297, 3300, 5062; Alcacer 1319, 1328 and 2062). The low tin bronzes group is consistent with reported alloy compositional data from Late Bronze Age and early Iron Age bronzes in the southwestern end of the Iberian Peninsula (Valerio, 2012; Rovira, 1995). The low Sn content in these artefacts has been interpreted as due to an increasing usage of recycled bronze scrap by the ancient metal makers: in this case, the low tin content can be explained by preferential oxidation of this element during repeated melting cycles, a process which has been reproduced experimentally in melting tests in the laboratory where significant Sn depletion in the final alloy product were found (Sarabia, 1992). The practice of using scrap bronze was indeed common in Late Bronze-Early Iron Age metallurgy workshops throughout sites located in the western end of the Iberian Peninsula either in Portugal (Castro dos Ratinhos, Quinta da Almaraz) or Spain (Cancho Roano, Talavera La Vieia) and has been commonly associated with "orientalizing" production technologies (Valerio 2012). It has to be said that low tin in bronzes could also be associated with historical periods of shortages in tin supply, as it occurred in Northern Italy (De Castro, 2001) and in central Iberian Peninsula during the LBA (De Castro, 2001). It is, however, unlikely that such a shortage occurred in the examples examined here as abundant Sn-rich mineral resources (from cassiterite-bearing granitic rocks) were readily available from nearby regions in the North of Portugal (Barriga, 1990; Figueredo, 2007; Pennhallurick, 1986).

The observed high variability in Sn content in the studied bronze artefacts with the identification of low-Sn bronze artefacts associated with high-Sn ones have been related with the introduction during the Iberian Early Iron Age of a different mode of bronze production involving the direct co-smelting of copper and tin ores (Figueredo, 2010). In fact, in the Iberian Peninsula as a whole, mixed Cu-Sn ores are not as scarce as initially accepted (Hunt Ortiz, 2003) having been identified in several mining areas exploited since ancient times (e.g. Toledo, Murcia and Sierra Morena). In the portuguese territory, though, while Cu mining resources were mainly concentrated in the South, being associated with the IPB ore deposits, tin (extracted mainly from cassiterite, a Cu-Sn mineral) were mainly located in the northwestern regions of the Iberian Peninsula as already mentioned (Pennhallurick, 1986). It is therefore unlikely that Cu-Sn co-smelting played a major role in

the high Sn variability observed in this study whereas a diversified network of local raw materials supply centers from the North and from the South of Portugal coupled with the varying rate of use of scrap bronze may provide a better way to explain the analytical data. In fact, the increasing use of local raw materials by the indigenous southwestern Iberian populations for the production of everyday ornaments such as the "ex-votos" bronze figurines in this study has been reported since LBA (Valerio, 2012).

3.10 Results discussion

The EDXRF analytical results suggest that the bronze artefacts from Evora and Alcacer not only share common typological and functional features but also a common alloys composition, i.e. they are made of a ternary leaded bronze alloy with variable Pb and Sn contents. The provenance of the Evora bronze statuettes collection from the Phoenician Alcacer do Sal settlement and their dating (V sec. BC), already hypothesized on the basis of archaeological evidence alone, can then be furtherly proposed³. The micromorphology analysis by SEM-EDS Backscattered electron images of the Portuguese samples shows that the Pb was added deliberately during the bronze making production to lower the melting temperature and to increase the fluidity of the melt. The bronze production technology was characterized by a slow cooling phase allowing the segregation of lead into well defined globular and inter-granular areas. The relatively high iron content of the artefacts founds correspondence on EIA bronzes in the Phoenician-influenced Mediterranean Basin and could be interpreted, besides the use of iron bearing minerals, as a result from an initial smelting and processing of the raw materials under a strong reductive atmosphere. To support this thesis with some confidence, it should be necessary to take samples of statuettes for metallographic analysis, which is currently and correctly avoided by museums curators this interpretative hypothesis on iron presence is only reported as a point of view of some authors. The high variability both in Sn and in Pb contents reflects in most cases the combined influence of local availability of geological raw materials for metal production and of indigenous versus Phoenician-Punic technology of production. In fact, results do not indicate a unique mode of production but rather the use of mixed

 $^{^{3}}$ To confirm this result, a future (highly probable) developing of this research will be the investigation of the statuettes with PIXE instrument, to obtain results on minor and trace elements. Also the analysis on other similar votive production in the Iberian peninsula, if it will be possible to continue in the study, could be considered in the future.

technologies which can be considered the hallmark of this kind of "mixed" bronze manufacturing. Notwithstanding the need for expanding the dataset (currently the thesis of V. Graziani has been activated with this purpose, see footnote on page 58 and introductory paragraph), both in terms of number and geographical distribution of samples to get a wider perspective of bronze metallurgy during Phoenician presence in the Mediterranean, the simple, fast and non destructive analytical approach used in this study proved useful in providing new analytical data to complement archaeological evidence. It has to be stressed that a ND methodological approach such as the one adopted in this investigation become an essential prerequisites of archaeometric research, especially when dealing with important archaeological objects stored in Museum collections around the world.

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CHAPTER III

Part B

The pear-shaped Bronze Jar from Alcacer do Sal

Area: Alcacer do Sal



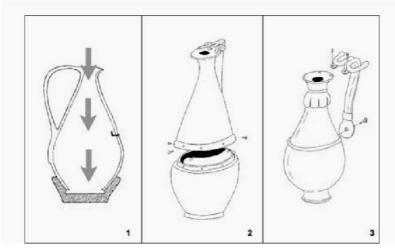
3b. 1 On the "orientalising" bronze pear-shaped (piriform) jars

A huge number of bronze jars, produced between the end of VIII and VI century BC, have been traditionally mentioned under the generic group of Tartessian Bronze. This name accepts, more or less, a native production for this orientalising finds. A global approach allows to order different craftsmanship traditions which display a cultural and chronological distribution. The most important group are the western Phoenician bronzes where it must be included the typical "orientalising" bronze jugs. This colonial group seems to disappear at the end of VII century BC (Avila, 2002).

Many interpretative problems arise from the attempt to ascribe this production to a particular population in fact, as explained previously, the orientalising handicraft turns around the three classical grouping: importation from Phoenician traders, local colonial production and the indigenous imitation, with connections with social and cultural derivations.

This is why an analytical study of the bronze pear-shaped jars (fig.1) can shed light on the production characteristics. The pear-shaped jars are related, for their shape and decorations with Phoenician traditions, since this is a type of artefacts that is commonly found in Palestine, but also in Cyprus, central Italy and, of course, Iberian Peninsula, in orientalising areas. This particular shape is shared also with the ceramic jars but, if the potteries are found in the coastal areas, the metallic jars are much more common in the inland, probably produced by local populations under the strong influence of Phoenician presence, as a part of funeral equipment.

J.J. Avila (2002, 2004, 2005), one of the most expert author on the Iberian Peninsula



"orientalising" metal object, summarized the production of the jars as a rupture with the local tradition, first of all by the use of the more and more frequent ternary alloy. For Avila also other changes are observable, such as the exclusive use of

Fig. 1: pear shaped jar

lost wax technique, which was almost unused in the previous time. Since the pear-shaped jars consisted of an upper and lower part, joined together, Avila proposed for this object the use of pins for a mechanical union followed by a casting-on (*sobrefundido*) method.

The author suggests for this kind of jars the following conclusions:

- a disruption with the artisanal local traditions of the LBA;

- homogeneity of the technical procedures;

- uncommon use of a real welding and use of mechanical junction and casting-on technique;

- formal relationship with the Phoenician objects;

- shared behaviour with other Mediterranean similar objects;

- local peculiarities.

So the conclusion suggested is that the pear-shaped objects were a colonial productions.

Avila also explains in his works other two classes of bronze jars: the Egyptianising and the globular typology, the first is connected with Phoenician tradition as well, while the second group is much more connected with Italics and Greeks traditions.

For the globular jars, bound to greek-Etruscan metallurgy of the first half of the VI century BC, there exist a separate technological panorama, characterised by the presence of real welding and hammered parts of the vase.

3b.2 Materials and Methods



A bronze pear-shaped jar (fig.2) from Alcacer do Sal (Arruda, 2000) Phoenician settlement, dated to the VII century BC has been analysed by the EDXRF and SEM-EDS

instrumentations used for the analysis on the Alcacer do Sal statuettes on cleaned microareas of the artefact.



Fig. 2: Alcacer do sal pear-shaped jar and a detail of the inner pin

The jar is composed of two parts delimited by a rim (easily visible) and a pin in the internal part is evident.

EDXRF analysis as been performed on the pin and on cleaned areas of the jar. The SEM-EDS analysis has been conducted on a cleaned area near the rim of the jar, on the upper part. The cleaning has been conducted in as much as possible mimetic way, so it is possible that the compositional results are slightly affected by the presence, if even the slightest, of the patina. It should be necessary to proceed with the study using a metallographic microscope for a sure identification of metallic phases in the jar, but the absence of this tool and the inability to move the jar too did favour the exclusive use of EDXRF and SEM-EDS.

3b.3 Results

The first set of measurement has been conducted with a DELTA Alloys and Metals Handheld XRF Analyzer, a portable XRF instrument with a recent invented configuration called "gun like" that, thanks to the reduced size of the measuring area and its maneuverability, allows the analysis of the small nail in the inner concave surface of the vase. The analysis performed with this type of XRF is more superficial due to further lower sensitivity to low atomic numbers; since it has been used for analysis of the bronzes this weakness appears to be irrelevant in the study of the composition of the vase. With this portable instrument other two areas of the jar has been analysed and the quantitative data are showed in tab.1 (in weight %).

Area	Cu	Pb	Sn	Fe	Si	Al	Р
Pin	26.82	24.53	17.38	13.42	1.85	3.21	2.24
Central upper area	29.15	36.63	20.67	5.75	4.85	nd	2.90
Lateral lower area	44.82	27.22	14.17	3.35	7.48	2.04	0.57

Tab. 1 XRF data (weight %) of Alcacer do Sal Jar

Is important to report (for the final interpretation) that the central upper area is a cleaned spot located in the close vicinity of the rim.

From the EDXRF quantitative data can be inferred that the pin is made of quaternary alloy, composed of Cu-Pb-Sn-Fe.

The upper and lower sectors of the jar is made of different ternary alloys (Cu-Sn-Pb), confirming the separation of the object into distinct areas, produced in two different steps of the production chain. The alloy can be described as a ternary alloy with a strong addition of lead in both side of the jar and a high presence of iron.

Another set of measurement has been executed with the same XRF instrument used for the other Portuguese artefacts, the Eclipse IV Oxford Instruments X-Ray tube (45 kV, 50 μ A, 2.25 W) with a Rh anode. With this instruments have been executed analysis on cleaned lower and upper areas of the jar, on the rim and on the decorative palm that adorns the handle attachment. The spectra have been elaborated, in order to obtain quantitative compositional data (tab.2), with the software PyMCA, using bronze standards as a reference of the correct quantification of each element. Instead of the central upper part, the upper- side cleaned area is quantified (thus avoiding that the quantitative result is influenced by the presence of the rim).

Tab. 2: EDXRF results in weight %.

	Fe	Cu	Sn	Pb
Lower Part	0.77	72.75	6.65	19.83
Upper-side				
Part	0.78	69.51	10.47	19.24
Rim	3.0	38.94	20.78	36.78
Palm	3.75	26.14	21.66	48.44

Four combination of overlapping spectra have been set out in order to show the differences and similarities in this four different areas of the jar (fig.3, 4,5, 6)

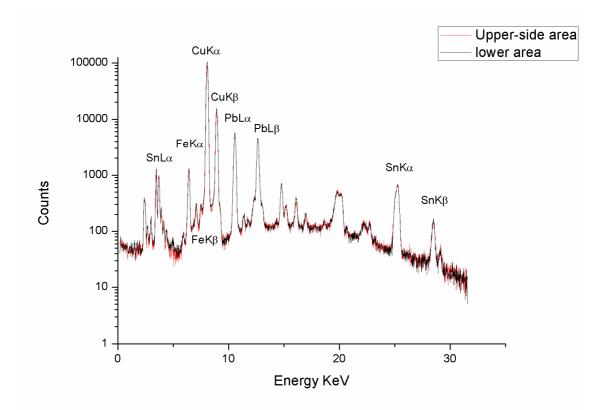


Fig. 3: EDXRF spectra of upper-side and lower areas overlapped

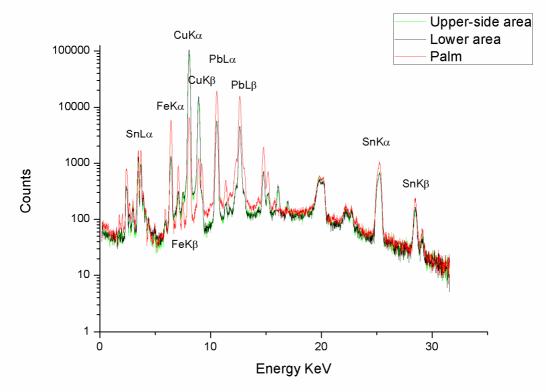


Fig. 4: EDXRF spectra of upper and lower areas overlapped on the palm spectrum

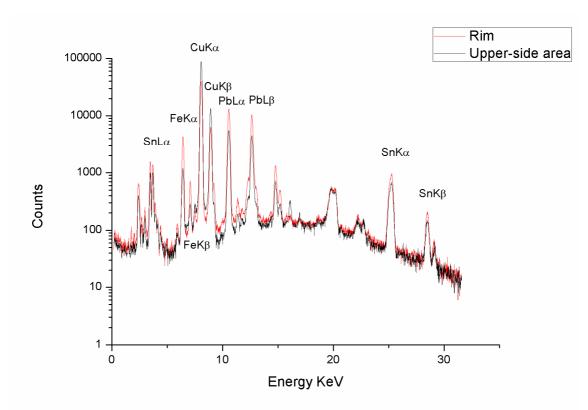


Fig. 5 EDXRF spectra of upper area overlapped on the rim spectrum

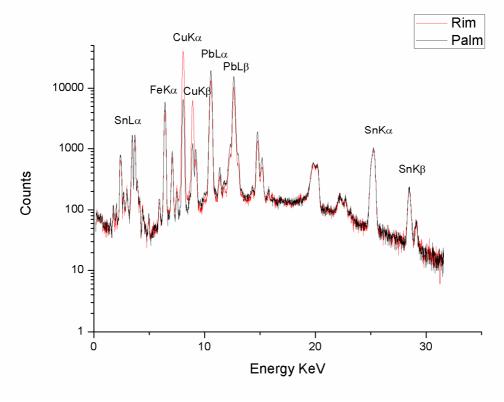


Fig. 6: EDXRF spectra of palm and rim areas overlapped

Upper and lower areas composition of the jar are similar. The palm (not cleaned to avoid any damage on the decorative area) and the rim have a similar composition between them (palm compositional data obtained on the uncleaned area could have to much higher values of tin and lead), whilst are not completely comparable with the above and below areas (tab.2). This evidence may suggest the assemblage of composite vase and the use of a soldering in the junction point of the upper and lower part. These observations are clarified below, thanks to the execution of SEM-EDS analysis on cleaned and etched areas.

From SEM-EDS analysis (fig.7 tab. 3) on cleaned upper area some information on production technology used for the object can be obtained. This area is near adjacent to the rim.

	weight 9	70					
		Cl	Fe	Cu 	As	Sn 	Pb
and the second second	1	16.3		4.1		-	79.4
	2	-	_	93.9	-	6.0	_
· · · · · · · · · · · · · · · · · · ·	3	-	2.0	18.5	3.8	6.6	69.0
N. VA	4	-	0.4	90.2	_	6.96	2.3
SE (μ15 (140 X, W/) 20.0 kV, WD: 13.1 mm							

Fig. 7and tab.3 : SEM-EDS analysis on cleaned upper area

This analysis confirms the alloy as a ternary leaded bronze with large lead globules

separated by liquation. The second picture has been taken going towards the rim.

ALL THE ALL	weight 9	76				
		Ρ	Fe	Cu	Sn	Pb
	-					
	1	-	-	100.0	-	-
	2	-	-	7.1	-	92.8
BACK TALL	3	-	0.5	74.5	17.0	7.9
	4	-	1.1	40.9	21.5	36.4
SE MAG: 2300 x HV: 20.0 kV WD: 13.1 mm	5	-	-	55.3	-	44.6
	6	2.3	4.2	24.8	42.5	26.0
	7	-	1.6	51.0	26.4	20.9

Fig. 82 and tab 4: SEM EDS analysis of cleaned upper part

From this SEM backscattered electron image it can be hypothesized that the jar was soldered by a tin rich alloy with tin content higher than 17%, probably quickly cooled.

This type of jars was commonly produced, following Avila (2002,2004,2005) by lost wax procedure and a casting-on method was used to produce the upper part of the jar and to join the two parts. From the following SEM-EDS analysis, made on a cleaned and etched part towards the rim is possible to suggest again the use of a welding for the case of Alcacer do Sal vase.

The bronze cleaned area has been etched with ferric chloride in hydrochloric acid, ethylene and distilled water solution (Scott, 1992), to show better the phase structure (fig. 9-10 and tab. 5-6).

	weight 9	%						
		С	P	Fe	Cu	As	Sn	Pb
	1	_	-	_	1.8	_	0.5	97.6
	2	7.	1-	-	89.	2-	-	3.6
	3	-	3.5	3.2	6.0	-	43.5	43.6
	4	-	-	4.5	8.4	-	44.6	42.4
ANALIZIA	5	-	1.3	5.1	5.8	1.4	54.4	31.8
CENTRAC, 2000, NO. 200 AV NO. TRAININ	6	-	_	4.0	5.6	-	49.5	40.7
	7	_	_	4.8	5.4	-	52.5	37.1
	8	_	-	5.0	6.6	-	47.9	40.2

Fig. 9 and tab.5: etching of cleaned upper part towards the rim

weight	%						
Mg	P	Fe	Cu 	As	Ag	Sn	Pb
			- 11.2 8.9			54.8 54.3	
			0.9 12.1			48.2	
4 –	-	-	96.2	_	_	1.0	2.7
5 -	-	-	1.5	_	-	1.0	97.4
6 –	-	-	90.9	-	-	6.2	2.8
7 -	-	-	69.0	-	16.	8 7.9	6.2

Fig. 10 and tab. 6: etching of cleaned upper part towards the rim

The etched area suggests that a welding composed by a brazing alloy Sn-Pb were used to join the upper and lower parts of the jar, beside the use of quaternary alloy pin. The presence of elongated structure also suggest that the cooling has been achieved quickly.

3b. 4 Discussion of the results

In conclusion, from the EDXRF and SEM-EDS set of analysis can be hypothesized that the bronze was made out of a ternary alloy, one of the alloys dedicated to the production of these "orientalizing" objects, but while an as-cast and lost wax procedure with the use of casting-on is commonly found in the piriform jars found in Iberian peninsula, the analyses seems to tend more to the use of a brazing alloy to solder upper and lower part.

In this light this jar could be a particular case, showing both Phoenician features (typical of pear shaped jar as the presence of rim and pin) but also Greek and italic features (such as the welding). In this way it is possible to conclude that in Alcacer do Sal, a cultural melting pot produced mixed-type of bronze metalworking, that can be defined particular in its kind.

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CHAPTER IV

SARDINIAN BRONZE PRODUCTION FROM THE LATE BRONZE AGE

"BRONZETTI NURAGICI" AND THE ORIENTAL SUBGROUP

Area: Sardinia

Classes of Objects: votive, small-size statuary



Gis thematic map of the archaeological provenance settlements of the artefacts under study

Highlights

At this point it was decided to go on studying Sardinian bronze production. In fact, also Sardinia, because of its great mineral wealth, was a landing point and a centre of commercial exchange for the Phoenician ships. In Sardinia there was, since the Late Bronze Age, a flourishing production of statuettes which are known as "bronzetti nuragici". These production shows very typical and local characteristics. Among these huge production it is possible to distinguish other sub-species of statuettes whose characteristics can be defined as imitations of Phoenician production.

Aim of the study was to assess whether the combined archaeometric approach could shed light on the interaction between local (Sardinian) and allochtonous (Phoenician) technological know-how and on how the Phoenician influence had been modulated locally in the making of metal artefacts. In this respect, for comparative purposes, selected bronze statuettes, displaying Phoenician features, from the Nuragic collection of the National Archaeological Museum in Cagliari and the G.A. Sanna Museum of Sassari, Sardinia were analysed by EDXRF.

The quantitative processing of the spectra has been conducted with Monte Carlo Simulation Method, while the connection between archaeological Phoenician settlements and the localization of ancient exploited mines for Cu, Sn, Pb metals has been studied with GIS system. The results has been interpreted in the light of localization of mines, archaeological settlements and landing points.

4.1 Archaeological Introduction

The history of Phoenician and Punic Sardinia deals with two different historical periods ranging from the IX century BC and the III century BC, concerning the peaceful arrival on the island of the first Phoenician traders and their integration into the nuragic civilization, bringing new knowledge and technologies, and the subsequent Carthaginian presence aimed at the exploitation of mineral resources and the control of the fertile Sardinian lands (Barreca *et al.* 1971).

The advent of the Phoenicians in Sardinia is inserted in the trail of commercial frequentations by Cypriots and Oriental merchants; this is the reason why indigenous peoples had come into contact with the culture of the Near East, as early as the Late Bronze Age, receiving stimuli and suggestions that, from the Iron Age, lead to complex processes of sociological, ideological but also technological change. The first real proof of the

Phoenician presence in Sardinia goes back to the IX century BC with the stele of Nora and the allocation of the commercial area of Sant' Imbenia in the Gulf of Alghero, but in this period their settlements had to be still provisional, up to the point that is difficult to find consistent traces. The research of raw materials, in particular metals, is one of the main reasons for the Phoenician expansion in the Mediterranean and it is this reason that brings them up in Etruria, in the Iberian Peninsula and North Africa, and that characterizes their maritime activities and their business strategies. Only after the mid-VIII century BC their presence is more stable and the confirmation is the creation of real settlements and outreaches in the Gulf of Palmas, in the Gulf of Oristano, along the coast of Olbia and in the Gulf of Cagliari. Into the first half of VIII century BC, the coastal regions of North, Central and South Sardinia are marked by the Phoenician structure settlements, from the Gulf of Palmas (Sulky, Monte Sirai, San Giorgio in Portoscuso, San Vittorio of Carloforte) to the Gulf of Oristano (Othoca, Tharros, Neapolis) from the landing place of Olbia to the Gulf of Cagliari (Barreca et al., 1971). The most popular sites of this period are those of Tharros, Sulky and Monte Sirai, set on a well-defined urban contest. After the VIII century BC new centers established themselves on the coasts and inland. So in the nuragic Phase IV, between the XII and the IV century BC (900-500 BC), there are various accounts of eastern acquaintances in Sardinia, recorded in the material culture of various areas of archaeological interest. The evidence can be found in the ceramics and jewelry, but a strong indication is provided by the study of metal objects, particularly in small bronze statuettes, which shows some features connectable with Phoenician manufacturing (Flumenelongu, Olmedo, Oschiri S. Cristina in Paulilatinu etc. ..). In this category we can also add the bronze supports for lamps (typically Phoenician-Cypriot productions), the votive razors, the ox-hide ingots but also the bronze swords with antennas, numerous types of buckles and the bowls, just to list a few examples of metal artefacts that change in this period, introducing oriental characteristic in Sardinia.

Between IX and VII century BC the Sardo-Phoenician communities were born: that articulated society will be broken in the VI century BC by the Carthaginian expansion in the Mediterranean. The Battle of Mare Sardo marked the change caused by the growth of Carthage, which in the first half of the century, organized a big structured state in the Northern Africa. Between 540 and 510 BC Carthage controls Sardinia; in the ancient Phoenician settlements the change of the funerary costumes (from cremation to inhumation), the new funerary structure (chamber tombs), the new production in

craftsmanship (pottery, metallurgy etc...) evidence the importance of that change. Carthage looks for strategic targets in Sardinia: the direct control of fertile agricultural lands and metallurgical districts through a massive penetration and presence (Bernadini & Botto, 2010).

In both periods the oriental presence in Sardinia is testified by the changes in the material cultures of the local populations.

Thematic maps on main archaeological settlements of these two separate periods have been prepared from two different databases, and thematic maps on Phoenician and Punic presence in Sardinia have been arranged (Fig.1).



Fig. 1: GIS thematic maps on Phoenician (on the left) and Punic (on the right) presence in Sardinia

Since the main triggering reason for their presence in Sardinia for a so long span of time was to be found in the ore richness of the region, a brief paragraph on the mineral characteristic of Sardinia is due, for a better understanding of the connection between the archaeological settlements and mines.

4.2 Ancient mines in Sardinia and metallurgy

A geological outline of Sardinia shows a wide choice of metalliferous georesources, which were certainly well known to the inhabitants in ancient times. The lead sulfides, so widespread in the crystalline basement of the island, is the best example. The appearance of the first metal objects (Cu, Ag) are attributed to the Ozieri culture (second half of the IV millenium BC); Ozieri culture is assumed to be the starting point of the first exploitation of Sardinian Pb and Cu ores, the presence of silver was limited by the mastery of the cupellation technology (Lo Schiavo et al., 2005). Actually, argentiferous galena was the only available silver ore, since silver minerals were not accessible in ancient times. Tin is present on the island but is difficult to demonstrate the presence of exploitable basins in ancient times. The possibility of the exploitation of easily accessible tin basin that could be completely exhausted in ancient times, leaving any evidence of their previous presence, must be remembered. Tin was considered a precious metal and its value depended largely on the distance between known deposits and ancient users. The closest tin districts were the area of Monte Valerio (Tuscany), the region of Castilla and Leon in Spain and Santa Olaia in Portugal. The Nuragic culture flourished in the Bronze Age with an intense development of mining and metallurgy activities. Between Late Bronze Age and Iron Age the extraction activities were also increased and triggered by Phoenician merchants, and by Punic colonizers later (Bartoloni).

Neglecting the indication of modern mineral basins, a list of the mineralization which could have attracted and stimulated the interest of ancient metalworkers can be done from the study of ancient evidences of ancient mines (Baldracco, 1985; Lo Schiavo, 2005; Manconi, 1986). In this view the gossan formed by the weathering products, characterizing the basemetal sulfide mineralization from the outcrops to some meters in depth, is important, being a possible source of useful ores, like primary sulfide remains, regenerated sulfides, native metals, oxides, carbonates etc.. In copper deposits malachite can be particularly abundant and easily recognizable because of its vivid color, but also chalcopyrite was exploited in ancient times. Some of the most important ancient mines were Calabona, Funtana Raminosa, Baccu Locci, Rosas just to enumerate some examples. Lead and silver was present and exploited in the Metalliferous Ring (Iglesiente), but also in Montevecchio, Argentiera, the Sarrabus Silver Lode (Campidano), Baccu Locci, Fluminimaggiore, Silius, Sos Enattos, Monte Arbu, Gennargentu, Corr'e Boi, Gonnosfanadiga, Villacidro, Monte Zippiri and others of less importance. Tin was available in Sa Bumbarda, Matzanni (Genna Cantoni), Perdu Cara (Monte Linas), Canale Serci(Villacidro), Su Suergio (Martalai), Corti Rosas (Ballao), even if their exploitations were unsure in ancient times. Since Abini (Teti) is also known for the discovery of metallic tin scraps and tin minerals, and since this rare

finding is attested also in other three settlement: in the hoard of Forraxi Nioi (Nuragus), in the foundry of La Maddalena (Silanus/Lei) and S'Arcu' e is Forrus (Villagrande Strisaili), it would seem that the exploitation of mineral nearby resources is believable (Lo Schiavo *et al* 2005). Collecting the localization in a database for the ancient mines in Sardinia, another thematic maps has been elaborated with GIS system (fig.2).

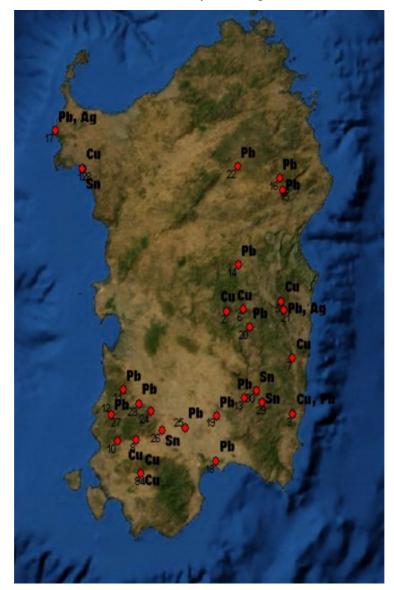


Fig. 2: GIS thematic map on ancient mines (Cu, Sn, Pb): 1) Calabona,2) Funtana Raminosa, 3) Baccu Locci,4) Rosas, 5) Correboi, 6) Monte Nieddu, 7) Talentino, 8) Sa Duchessa, 9) Barisonis, 10) Iglesias, 11) Montevecchio, 12) S'Oreri, 13) Genna Tres Montis, 14) Correboi, 15) Sos Enattos, 16) Guzzurra, 17) Argentiera, 18) Sarrabus Silver Lode, 19) S'Ortu Becciu, 20) Monte Arbu, 21) Gennargentu, 22) Corr'e Boi, 23) Gonnosfanadiga, 24) Villacidro, 25) Monte Zippiri, 26) Matzanni, 27) Perdu Cara, 28) Sa Bumbarda, 29) Su Suergiu, 30) Corti Rosas Digitizing the major rivers in Sardinia it was possible to obtain a richer map in order to correlate the presence of these sites with the ability to penetrate in the hinterland through navigation (fig.3).

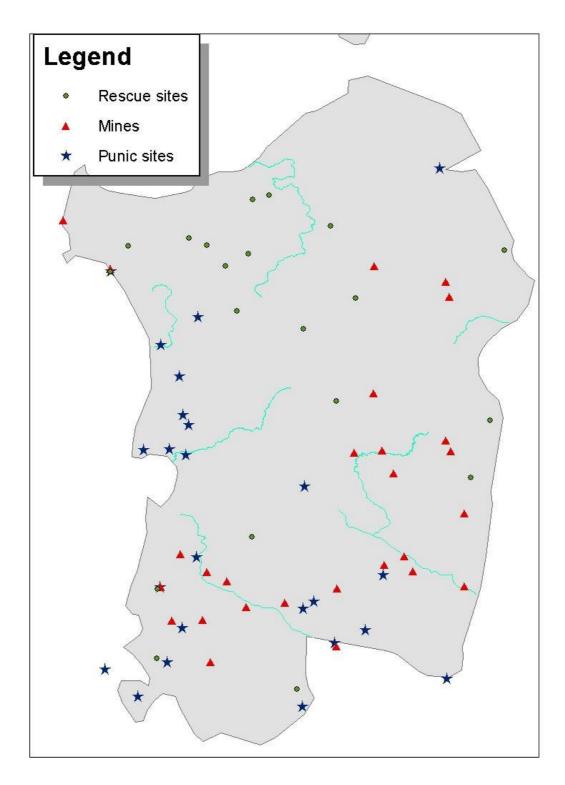


Fig. 3: thematic map with main digitalized rivers, settlements and mines

4.3 Nuragic Bronzes and the "Phoenician" subgroup

Nuraghic bronze statues are typical sardinian bronze sculpture probably produced during the final phase of the Bronze Age and the Iron Age. Archaeologists have not yet succeeded to accurately date the figures and a lot of chronological interpretations exist (Lilliu, 1966) indicating more ancient or more recent datings. They were allegedly made by the IX century BC the VI century BC, but the recent findings from Orroli of bronze fragments dated to the XIII century BC have questioned their actual dating.

The small size statuary, with its antecedents in Syria and Palestine from the late III millennium, coming to the Early Iron Age, is a valuable clue to the recognition of the oldest Phoenician frequency in the West. The Phoenician and Syrian bronze statuary, with Cyprus as intermediary which is increasingly recognized, was one of the first cultural evidence of transmissions that the Phoenician culture operates in the West. The stimulations arising there to the indigenous world of Sardinia are of particular interest for the interpretation of Nuragic bronze statuettes (Moscati, 1997). If the Syrian and Phoenician bronze production come to influence the Iberian production, in Sardinia the arrival of Eastern models, perhaps through Cyprus, plays an important role for the evolution of Nuragic art. For Sardinia, the findings coming from the vicinity of Nuraghe Flumenelongu of Alghero, from Olmedo and the sacred area of Santa Cristina Paulilatino, are some of the most egregious examples: these bronzes are a clear testimony of the resumption of relations between the nuragic environment and the Near East, after the fall of the Mycenaean Emperor. Even for this aspect Cyprus has a decisive role, and the bronze tripod found at Santadi, local product on Cypriot models of XII-XI century BC, is a perfect example. Equally obvious is the contribution given to the Phoenician iconographic production of Nuragic bronze statuettes.

The circulation of ceremonial bronze statuettes, of Eastern production and tradition, in the Italian Peninsula and the presence of the ceremonial and anthropomorphic bronze in the Isle of Sardinia, which accompany the extraordinary flowering of local bronze is therefore subject of discussion. In order to search for Phoenician specificities in the eastern centre of production and in the expansion in the Mediterranean, the study of the production of bronze artefacts is a preferential approach, as in the techniques of production lie the material culture of a unique craftsmanship. There is a unanimous recognition of the interweaving of craft workers and cultural influences that converge towards the Sardinia between the IX and VII century BC and the active role of the Sardinian centers, who welcomed the new

inspirations processing them in a particular way (Bernardini & Botto, 2010). It is possible to identify a coherent set of Phoenician bronze figures who is located in correspondence with the recognized attendance of mercantile and colonial settlements in the Mediterranean and Atlantic coasts, affected by the arrival of this allochthonous presence.

Paolo Bernardini and Massimo Botto (2010) explained profusely this issue, clarifying many points of discussion on this argument. They suggest for that production the denomination "Phoenician bronzes" or "Oriental bronzes", underlining a common Phoenician influence but meaning with those terms a genesis in areas of miscegenation (presence of local and oriental elements merging togheter). They suggest for this particular subgroups of statuettes a general dating between VIII and VII century BC, while in their interpretation Sardinian Nuragic bronzes belong to a wider span of time between IX and VI century BC.

The main features that allow to separate them from local production are the hats and clothing, with a marked Egyptian imprint, as well as in the overall scheme of the figure, essentially summed up in the hieratic or fighting position (Avila 2002). In detail there are three basic types of headgear (white crown of Upper Egypt, Atef crown and miter) with four positions of the figure (arms close to the body, arms bent in the position of god fighter, the arm bent at chest in hieratic position, bent arm with the palm open in the gesture of praying or blessing). Even the domed hair is to be considered as a variant for hats. (Bernardini & Botto, 2010). Is currently being discussed whether the production is local or in imitation of the oriental production. Also Avila (2002) suggest to insert two Sardinian statuettes form Flumenlongu and Galtellì between a Phoenician-Mediterranean group, composed of Iberian artefacts. For Bernardini and Botto (2010) there is an important difference between Iberian and Sardinian "oriental" production: while the Iberian statuettes are found in areas of direct Phoenician settlement, Sardinian figurines are also found in indigenous locations, especially near shrines. These observations lead the authors to indicate a local production, made in culturally composite work shops, with the presence and activities of craftsmen of Phoenician tradition. The presence of Eastern culture craftsmen in symbiosis with the indigenous workshops explains the circulation of aliens models, postures and furnitures.

A selection of local and "oriental" statuettes in the National Archaeological Museum of Cagliari was made to perform EDXRF analysis in order to investigate a possible difference even in the alloy, between this statuettes and those of certain local production. Other classes of artefacts has been chosen, such as a *thymiaterion*, to get a glimpse of the local metallurgy and of the presence of oriental (Phoenician or Phoenician-Cypriot) elements, but also some tools, to obtain information on the level of productive specialization, and some ingots, for the research on half-worked materials.

4.4 Materials and Methods

Bronze statuettes from the Nuragic collection of National Archaeological Museum of Cagliari and A. Sanna Archaeological Museum of Sassari, were analysed by EDXRF. The Sardinian bronze artefacts come from a wide selection of Nuragic settlements with attested Oriental (Phoenician and Punic) influence in the island, such as Bonorva, Flumenelongu, Anthas, Sardara and Laerru. Some typically local nuragic bronzes has been analyzed as a meter of comparison, in order to understand if the metal alloy is different between the two groups. As a result of the selection on *bronzetti nuragici*, between the available ones, 29 figurines (fig.4) coming from settlements equally distributed in the island territory (fig.5), of both anthropomorphic and zoomorphic type, were picked out for EDXRF analysis (Lilliu, 1966; D'Oriano & Sanciu, 2000). Also other categories have been selected to broaden the variety of statuettes under study. Due to the tight rules of the museums, it was not possible to clean even small surfaces of the artefacts, and since it was not possible to move the statuettes from the rooms of the museums, it was not possible to perform SEM-EDS analysis. The best compromise was to use a portable EDXRF to analyze the statuettes, choosing the more homogeneous and cleaned areas. It is, however, important to note that all the bronzetti have been restored in pre-musealization phase, then the figurines did not show severe incrustations or residues of burial earth. Since it is assumed that an overestimation of Sn and Pb content, due to the presence of oxidized compounds, could occur as demonstrated by Figueredo et al. (2007), only values of Pb higher than 10 -15 wt %, have been considered as a probable voluntary addition in the alloy. Currently we consider these data as a qualitative description of the statuettes and one of the future development of this part of the research will be aimed to the possibility to reconstruct the layered structures of the objects and to determine the thickness of the patina. It this way, using the internal ratios of X-ray lines, and considering the attenuation of the outgoing Xray intensity due to the patina, the attempt will be to obtain information about internal layers composition, in such a way to attain the composition of the metal, without the necessity to clean it.

The permission to remove mechanically the oxidized layers on small areas of the object has been obtained only for the statuette with the silver mask from Florinas (G.A. Sanna Museum), whose composition of the mask and the welding has been analysed directly on cleaned metal (Cesareo *et al.*, 2013). Another artefact analysed on cleaned areas of the metal is a dog shaped circular harness, belonging to the Museum of the Near East in the University of Rome Sapienza, whose provenance is Sardinia as well. This object will be exposed separately since we carried out SEM-EDS analysis on it.

The spectrometer used is an Amptek Mini X-ray tube equipped with Ag anode (50 kV, 80 μ A). The X-ray detector was a X-123SDD by Amptek with again a resolution of 140 eV at MnK α FWHM 5.9 keV. All measurements were carried out under ambient air. An innovative reverse Monte Carlo simulation (Brunetti *et al.*, 2004; Bottigli *et al.*, 2004) quantification approach has been used. This approach allows to significantly improve the sensitivity of the spectrometers, taking into account also the non planar nature of the surface. Other Monte Carlo codes are capable to cope with rough surfaces by using regular structures such as hemispheres or stripes. In our case, the rough surface could be modeled using any irregular shape. Our Monte Carlo code makes use of a constantly updated X-ray library for any X-Ray data (X-raylib) (Brunetti *et al.*, 2004; Schoonjans *et al.*, 2011 a-b). The Monte Carlo code has been tested on a variety of cultural heritage items. The LOD of the system is about 10 ppm.

Catalogue of the artefacts (Tab.1). When it is available, the inventory number proposed by Lilliu (1966) is reported. "Oriental" term is used to indicate a Phoenician influence (as proposed by Bernardini & Botto, 2010)

Tribal leader (5)Teti, AbiniLocalCagliari1Archer with directional plume (16)Teti, AbiniLocalCagliari2Demon (four eyes/four harms) (104)Teti, AbiniLocalCagliari3OffererTeti, AbiniOrientalCagliari4Figure during libationsMonte SiraiOrientalCagliari5Lyre playerMonte SiraiOrientalCagliari6Truncated figure (166)BonorvaOrientalCagliari8Offerer (61)VillacidroOrientalCagliari10Figure with a daggerAntasOrientalCagliari11Figure with shield (137)BauneiOrientalCagliari12Archer (25)SardaraOrientalCagliari13ArkOschiriOrientalCagliari14CentaurNuleOrientalCagliari15ThymiaterionUnknownCyprioteCagliari16WarriorNuraghe PitzinnuLocalSassari17Bull (194)PerfugasOrientalSassari20Bull (202)IlloraiOrientalSassari21Offerer (175)OlmedoOrientalSassari22Offerer (important figure) (50)AntasLocalSassari23Small boat with bovine proteome (280)ArdaraOrientalSassari24Offerer (important figure) (50)AntasLocalSassari24Of	Description and n. inv. (Lilliu, 1966)	Provenance	Tipology	Museum	Fig.
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Archer (25)SardaraOrientalCagliari13ArkOschiriOrientalCagliari14CentaurNuleOrientalCagliari15ThymiaterionUnknownCyprioteCagliari16WarriorOssi sa MandraLocalSassari17WarriorNuraghe PitzinnuLocalSassari18Warrior with rapier and shield (88)Ossi sa MandraLocalSassari19Bull (194)PerfugasOrientalSassari20Bull (202)IlloraiOrientalSassari21Offerer (175)OlmedoOrientalSassari23KourosOlmedoGreekSassari24Offerer (important figure) (50)AntasLocalSassari25Small boat with bovine proteome (280)ArdaraOrientalSassari27Mufflon (221)LaerruOrientalSassari27	Figure with shield (137)	Baunei	Oriental	Cagliari	11
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Small boat with bovine proteome (280)ArdaraOrientalSassari26Deer head proteomeunknownOrientalSassari27Mufflon (221)LaerruOrientalSassari28	Kouros	Olmedo	Greek	Sassari	24
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Mufflon (221)LaerruOrientalSassari28	Small boat with bovine proteome (280)	Ardara	Oriental	Sassari	26
	Deer head proteome	unknown	Oriental	Sassari	27
Statuette with silver maskGiorrè Florinasno local feedbackSassari29	Mufflon (221)	Laerru	Oriental	Sassari	28
	Statuette with silver mask	Giorrè Florinas	no local feedback	Sassari	29

Illustrations of the artefacts (fig.4)













































Localization of the archaeological settlements of provenance (fig.5)

4.5 Brief descriptions of *Bronzetti Nuragici* under study and of the archaeological context the findings

Since the choice of the figurines was performed on the basis of particular features which characterize the type of statuette as local or "Oriental" production (using the term of Bernardini & Botto, 2010), a short description of the artefacts and of the archaeological settlements (localization in fig.5) is due to permit a better understanding of the aim of this part of the research.

Local Artefacts:

1) Teti (Abini)

Abini is a sanctuary of Nuragic Period on the territory of Teti. The word Abini has surely Oriental origin, *Abi*, *Ab* (father or lord) and, *Ini* (Response, prayer), suggesting that this was the place where fathers and ancestors prayed or answered to requests. The first discovery was made during excavations made by some local shepherds-farmers around 1865, which brought to light a lytic cist full of votive objects of bronze, which gave the clue to the interpretation of the center of Abini as an extended settlement dating back the VIII and VII centuries BC. The sanctuary chronology is made complex by the presence of the early Bronze Age findings amongst the material; however it can be assumed that the chronological span is between the Recent and the Final Bronze Age, not excluding the presence of a more ancient settlement and later occupations (Lo Schiavo *et al.*, 2005). Some of the nuragic bronzes from this area are considered a Sardinian local production because they present an iconography that is typically local.

Chieftain: this is one of the very popular iconographic series in the local figurative bronze production, the tribal chief. It is the n $^{\circ}$ 5 in the Lilliu catalogue (1966), called Praying Chieftain, 20 cm high. Caught in the position of greeting, the character has a peaked cap in front of which there is a thin band decorated with median streaks.

Archer with directional pen: The statue No. 16 of the of Lilliu catalogue (1966) is classified as "archer with directional pen rod". It is 23.5 cm high, and the right wrist has the brassard. The character wear a hat and tunic. On the chest is present the rectangular plate from which the dagger gets out. On the back there are the case for the conical arrowheads, a sword and scabbard with a long rod that ends with three rings that fasten it to a triangular striated pen, considered as an identification sign. The headgear cap has long broken horns.

Demon soldier: the statue No. 104 in the Lilliu catalogue (1966), called by the author "demon soldier ", is 15 cm high. The helmet consists of a hemispherical dome with median

ridge and horizontal horns. The body is covered by a heavy jacket with short sleeves and hands held tight two shields each provided with three short daggers in the inner part. The figure shows a dagger near the chest with a rope hanging over its shoulder. Two long braids descend to his chest and the general massive bronze form gives the appearance of a warrior.

2) Ossi (nuragic complex of Sa Mandra e' Sa Giua)

The nuragic complex is the only in the territory to have been the object of systematic excavations. In the village melting activities are also attested, as evidenced by a fusion fund with copper ingots (which analyses results are reported below in the text), pins and axes incorporated into the metallic mass .

Warrior with jeweled hilt dagger and the Warrior with rapier and shield : this type belongs to the category "warriors", a type of Sardinian local production.

3) Nuraghe Pizzinnu

In the nuragic site of Pizzinnu, Posada, were found also bracelets, daggers and pins along with the statuettes of a warrior and a bull.

Warrior: the characteristics of the statuette are typically local.

Bronzetti Nuragici with oriental features:

1) Teti Abini

Offerer: the statuette has his right arm with the palm open, in the typical oriental blessing gesture.

2) Monte Sirai

The oldest settlement on Monte Sirai consists of a series of *nuraghi* (VIII-VII centuries. BC.) scattered on the hillside and at the top of the hill. The small town center underwent substantial renovations that gave the still visible shape also after excavation. The site was abandoned in the I century AD. The settlement has been partially excavated; the city walls have been identified, where a door opens towards the plateau, flanked by square towers. Behind the door there is a large space occupied partially by a small Punic temple dedicated to Astarte. In this section there are two small cells: in one of these was found a statuette of worship with the offerings still in place (I sec. BC) and in the other were found objects of worship like a clay male proteome of Carthaginian type (V sec. BC.), a female proteome of Greek type (V or IV century BC.), and two little bronze statuettes of "oriental" style. The foundation of the Punic settlement is classically positioned to the VI century BC to the IV-III century BC. Currently a more ancient dating, placed around the end of the VIII century

BC, has been attributed to the shrine and sacred area, and the same chronology is shared by the statuettes.

Figure during libation and Lyre player: The two statuettes reproduce two sitting characters: one is the figure of a man pouring wine in a *patera* from a "*askos* like" jug, and the other is a lyre player. The two statuettes have similar characteristics: not well defined bodies, the heads incongruent to the body, dorsal flatness and undifferentiated legs, apart from the presence of a slight groove These two little characters, in which remains the mounting nails, had to belong to the same artefact, perhaps a ritual bowl that was part of the liturgical furnishings of the temple. These statuettes have been defined a colonial production, due to the presence of the "*askos* like" jug, of near-eastern and Greek type, but also the figure of the lyre player, that falls under the category of musical entertainer, is not local. The fact that these figures are part of a liturgical object in a sacred local space, highlights the interrelationship between Eastern artisans with local customers (Bernardini & Botto, 2010).

3) Bonorva

The necropolis of Domus de Janas-Sant'Anderea Priu is related to the Ozieri culture (3500 BC). This area has been frequented continuously, by all subsequent cultures, till the end of the Middle Ages: from nuragic populations first, then Romans, and in the final stage by early Christians who transformed it into a church.

Truncated figure: From a cave in the village of Riu Mulinu of Bonorva comes this statuette, truncated at the skirt: it represent a character who bends his right arm with the palm open in the typical oriental blessing gesture.

4) Flumenelongu

The nuraghe Flumenlongu is a tower with at least two superimposed layers which bind, as well as an open space, a secondary tower and some ancillary rooms. In its margins lies a village of round huts, and from one of them comes an important storage room that returned many objects in bronze, ingots, axes, a hatchet and few bracelets.

Truncated figure: The statuette was the result of illegal excavations made in this area, this is why there are no other data on the context. The statuette is of the type of blessing characters, a Phoenician variant of the Syrian god – fighter and represents the face and chest of a man who wears a lengthened hat, interpreted as an approximate representation of the white crown of Upper Egypt. The bust is slightly twisted and his arm bents at an almost straight angle and ends with the fist. The right arm is raised forward with an open hand.

The general hypothesis is that the statue was originally a full-length figure while the twisting of the body suggests the movement of the legs, with one leg overtaken. These characteristics suggest a dependence to the Eastern iconography distributed between the middle of the second and the beginning of the I millennium. The statue seems to be a combination of local and oriental features. The presence in the settlement of Sant' Imbenia of Phoenician merchants and craftsmen, that between the end of the IX and the beginning of VIII centruy BC, developed a strong interrelation with the indigenous community of Nurra, seems to be the explanation of this pastiche (Bernardini & Botto, 2010).

5) Antas

The **Temple of Antas** is an ancient Carthaginian-Roman temple located in an area colonised by the Carthaginians and then by the Romans, attracted by its iron and lead deposits. The punic temple (around 500 BC) was dedicated to *Sid Addir*, an interpretation of the local *Sardus Pater Babai*. The individual graves pit covered with stones in Antas contained bronzes of clear Oriental suggestions, adapted to local production to indicate the status of the deceased, seen as a god-hero. In the necropolis of Antas was also recovered a pin on which some Phoenician letters appear, further confirmation of the Phoenician presence in this area.

Figure with a dagger: The elements that build and identify the figure are a dome that covers the head, the gesture of blessing, the nudity with explicit representation of sex, the overall pattern of smiting god with the weapon in hand, but also an element from Crete, the flexed legs. This combination of features is plausible in the repertoire of Phoenician bronze circulating in the Mediterranean in the early centuries of the Iron Age (Bernardini & Botto, 2010).

6) Villacidro, archaeological area of Matzanni

Nuragic shrine composed by three well temples and the remains of twelve huts dating back to the XII and IX century BC. The statuette has been found in the main well. 200 meters of distance is localized a Punic shrine.

Offerer: it is an offerer with in the right hand an hemispherical bowl with a form of cheese on the bottom. In the left hand the figure has a plate with five pieces of dried fruit. A wide band placed on the head leaves uncovered the hair at the top and the locks also fall on the forehead and neck. The facial expression is different from the common bronzes. Chin and pointed nose, eyes and eyebrows globular arc are unique among those known in Sardinia. Recognitive element is the sharp shape goatee, connecting that artefact to oriental influence.

7) Baunei

In the Baunei territory there are many nuragic findings, documented by the remains of 19 *nuraghi*, both the tholos type and the corridor type. Around the *nuraghi* are conserved the remains of villages and the "giants tombs" (S'Olloli, Perdu Saccu, Olovette Cannas, on Scusorgiu, Fonnacesus, Nuraxi Albu, Orgoduri, Sat Tiria, St. Pietro, Golgo' and Mesu, Latalai, Co 'and Serra) designed to contain the deceased, especially of small communities that came to settle in the valleys. From this area come some evidences, such as the elegant ship in bronze with bovine proteome (or an antelope), from Golgo, and another little ship with a figurine of monkey and bovine proteome handle, which suggest that in Baunei territory was practiced melting activity, probably favored by the presence of small mine of Genna Arràmene. The representation of the antelope, monkey and also an African man representation suggest that the production was done by artisans who visited Africa.

Figure with a shield: bronze figurine of a soldier (represents a person with typical facial features of an African man) with shield and sword on his back.

8) Lanusei

The archaeological park of Seleni is located in the region of Lanusei and is consisting of the nuraghe "Gennacili", a village and two graves of giants. The *nuraghe*, built on a rocky outcrop, is not in excellent condition. Its cylindrical structure is surrounded by a cluster of two hundred huts and there are remains of defensive walls. The first is dated from the XV century BC, while the second tomb is dated back to the XII-XIV century BC. Some bronzes from Seleni park are exposed in the National Archaeological Museum of Cagliari: lacking of an exact location of the discovery, however, they seem to be related to religious contexts. The three bronzes of Nuragic production seem to be a warrior with bow, a praying woman and a leader / priest dressed in scarf and hat. These three *ex-votos* were left in the vicinity of religious places (sources, sacred wells) or embedded with lead in the stones that formed the enclosure of temples. These statuettes have been found together with punic coins.

Figure with scarf: this statuette represent a priest dressed with scarf

The next three statuettes have been found in association with individual graves, a clear sign of the articulation, in aristocratic sense, of nuragic society during the early centuries of the Iron Age, more and more influenced by the Phoenician presence on the island (Bernardini & Botto, 2010).

9) Sardara

Among the most important monuments of Sardara must be counted the sacred temple of S. Anastasia, one of the oldest and best preserved of the island, that seems to have been built around the IX and VIII centuries BC. Around it is well visible the Nuragic village and, at short distance from the archaeological site of Santa Anastasia, in "Sa Costa", were found the famous "archers of bronze" of exceptional workmanship. In the village Sa Costa was found a pit tomb built of large slabs of stone. The deceased, buried, was located on a large sheet of bronze and next to the body the two famous figurines of armored archers were found. This type of burial is very different from the typical Sardinian tombs of megalithic tradition. The eastern origin therefore seems obvious considering the spread of this type of burial in the attested Phoenicians tradition in Sardinia between the VIII and VI centuries BC.

Archers: The two archers wear a armored garment that finds comparison with representations of armed men in the Assyrian bas-reliefs (Bernardini & Botto, 2010)

10) Oschiri

This artefact was found in the only pit grave found in the site of Nuraghe Lughenia, in the territory of Oschiri.

The ark: It is a casket of bronze dated to the Nuragic Age, found in half of the '800 by Alberto La Marmora and is particular interesting because is part of a funeral equipment. In detail it is a miniature representation of an ark on wheels, which refers to the tradition of Homeric Keimelia into which converge oriental objects (Bernardini & Botto 2010). This examples of memorabilia, preserved near the deceased, to enlight the rank and social prestige, is interesting because it reproduces, on a small scale, the ark of traditional wood. From this bronze it can be assume that the prototype of the '*cascias*' dates back to the nuragic age. Cascias is openable from the top and the four faces of the box are divided by horizontal ridges, reminiscence of the wooden structure of the case. Reliable sources say that the culture of the ark had begun in the center of Sardinia, in the heart of Barbagia ("*cascias barbaricina*") because this area, very rich in chestnut woods, could produce a large amount of raw material

11) Nule

The site consists of a large quadrilobated *nuraghe* with a triangle convex plant, formed by a central keep and three towers joined from a bastion, very similar in type to Nuraghe Santu of Torralba.

Centaur: In this territory of Nule, in a area of difficult interpretation, which was read as a burial, was found a bronze figure representing a centaur. This model comes indisputable from Aegean and the near-east area of the Mediterranean Sea and qualify and emphasize the changes of customers taste, in that period open to new representative models, no longer exclusively native (Bernardini & Botto 2010). Or maybe this statuette could represent the *Boe Muliache*, a man who become bull, mythical character of Sardinian folklore

12) Unknown

Candlesticks or *Thymiaterion*

The analyzed *thymiaterion*, of unknown provenience, lack of context information. The bronze censers, also called candlesticks or *thymiateria*, constitutes a particular class of bronze artefacts. These objects were evidently typical tools of worship from the Phoenician -Punic world. They consist of a high stele-shaped column, adorned with a double or triple set of floral corollas with petals pending, to which overlap three long and curved elements, ending in volutes. These columns were meant to support one or two cups containing the incense. Another type, of Egyptian style, has a *capitello*, shaped in a papyrus leaves- shape. The censers were often found in tombs and some sanctuary, in some contexts dating before the end of the VIII to the V centuries BC. Ninety copies are known in the Mediterranean area, as this kind of artefacts had a great spread. Besides some areas of Sidon, the thymiateria were certificates in large quantities in Cyprus and Samos, while for the West Mediterranean were found especially in Malta, Sardinia and Spain. Given the large number of discoveries it has been argued that Cyprus had been, more than Carthage, a major center of production (Avila 2002).

13 and 14) Perfugas and Illorai

Perfugas is a nuragic village with a big sized temple dedicated to the worship of water.

The archaeological site of Illorai shows some allocthnous features represented by the presence of basalt picks for the excavation of the graves (inhumation was not local) and the plaster used on the interior walls and the red paint are elements that were very rarely found in these cemeteries.

Bull: this kind of small size statuary, very well attested also in the Iberian Peninsula, has a clear oriental matrix.

15) Siligo

Important nuragic settlement of big dimension consists of several structures distributed around a well temple.

Offerer: the statuette has his right arm with the palm open in the typical oriental blessing gesture

16) Olmedo

Well nuragic temple.

Offerer: the statuette is one of the most discussed artefacts about the item: is it a local or "oriental" production? In fact it present the typical structure of the oriental offerer, with the palm open right harm in the typical oriental blessing gesture, while the left arm is bent at 90° and ends with the closed fist. The domed wig and the pointed beard are other oriental elements as the position of the legs (Bernardini & Botto 2010).

17) Ardara

Scala de Boes Nuraghe, the artefact belongs to VIII-VII century BC. The Punic period of this site is suggested by the discovery of punic coins.

Little boat: La Marmora in 1840, classified them as "votive objects of oriental origin". La Marmora suggests that the ships were dedicated to Astarte, who represented the characters of Isis and Artemis.

18) Laerru

This artefact has been found in a shrine inside an hoard of bronze objects of nuragic and roman typology. The mufflon is interpreted here as an oriental production.

Mufflon: this statuette shows analogous shape to the goat of Alcacer do Sal and also quite similar features to many other statuettes from Nuristan (oriental origin). Also in this case this bronze product was the terminal element of a banner, as in the case of Alcacer do Sal. This nuragic statuette further emphasizes the connection between Sardinia and the Iberian Peninsula, through the recognition of a common matrix, the Phoenician presence.

19) Giorrè

This statuette was recovered from a nuragic sanctuary close to Florinas: this nuragic settlement is made up of several rooms surrounded by a wall, connectable with the cult of water.

Statuette with the silver mask: this is a unique artefacts in Sardinian which, both for the position of legs and harms and for the presence of a silver mask on the face. The facial

portion of the statuette is covered by a silver mask, partially gilded and welded to the bronze body. Unfortunately the context data are scarce since it was found in a area subjected to illegal excavation together with fragments of ceramic dated to II century BC and with some Punic coins of local and Siceliot brand dating to the IV and III century BC. This statuette is particular because it shows connection with the punic bronze production (some parallel use of silver or gold leafs on bronze objects is to be found in North Africa and Spain) but also to the Roman pantheon and decorative style. Currently this statuettes has been dated to the II century (Cesareo *et al.*, 2013 D'Oriano, 1977).

After this detailed catalogue of statuettes, the entire set of EDXRF quantitative data are displayed in the following tab.2. The statuette from Giorrè will be reported separately since on the layered structured has been carried on it.

Results from EDXRF analyses of artefacts from archaeological settlements in Sardinia (Monte Carlo Simulation Method). Values in wt. $^{ m M}$	of artefacts from	archaeological	settlements in S	ardinia (Monte	Carlo Simulat	ion Method). V	'alues in wt.%
Artefact	Provenance	Fe	Cu	As	Sn	Ag	Pb
Tribal leader	Teti, Abini	0.29 ± 0.01	82.94 ± 0.1	0.29 ± 0.01	6.03 ± 0.1	10.29 ± 0.1	0.15±0.01
Archer with directional plume	Teti, Abini	1.97 ± 0.01	79.16±0.1	2.75 ± 0.01	3.93 ± 0.1	pu	12.19±0.1
Demon (four eyes/four arms)	Teti, Abini	0.97 ± 0.01	81.55 ± 0.1	0.65 ± 0.01	6.47 ± 0.1	pu	10.36±0.1
Offerer	Teti, Abini	1.05 ± 0.01	72.89 ± 0.1	0.60 ± 0.01	12.05±0.1	pu	13.40±0.1
Figure during libations	Monte Sirai	2.60 ± 0.01	62.42 ± 0.1	1.36 ± 0.01	11.12±0.1	pu	22.50 ± 0.1
Lyre player	Monte Sirai	0.92 ± 0.01	77.12±0.1	1.33 ± 0.01	7.15±0.1	pu	13.48 ± 0.1
Truncated figure	Bonorva	2.00 ± 0.01	79.00 ± 0.1	pu	16.00±0.1	pu	3.00±0.1
Truncated figure	Flumenelongu	0.40±0.01	78.10±0.1	pu	4.00±0.1	pu	17.50 ± 0.1
Offerer	Villacidro	0.84 ± 0.01	97.70 ± 0.1	pu	0.04 ± 0.01	pu	1.40±0.1
Figure with a dagger	Antas	0.73 ± 0.01	62.70 ± 0.1	1.51 ± 0.01	16.98±0.1	pu	15.84 ± 0.1
Figure with shield	Baunei	0.49±0.01	74.51 ± 0.1	2.59 ± 0.01	11.08±0.1	pu	11.33 ± 0.1
Figure with scarf	Lanusei	1.26 ± 0.01	72.21 ± 0.1	1.37 ± 0.01	13.40±0.1	pu	11.76±0.1
Archer	Sardara	1.00 ± 0.01	77.50 ± 0.1	pu	8.10±0.01	pu	14.10±0.1
Ark	Oschiri	0.89 ± 0.01	81.85 ± 0.1	1.45 ± 0.01	5.57 ± 0.1	nd	10.24 ± 0.1
Centaur	Nule	0.85 ± 0.01	74.48 ± 0.1	2.55 ± 0.01	10.94 ± 0.1	nd	11.18±0.1
Thymiaterion (1)	Unknown	2.22 ± 0.01	76.26 ± 0.1	1.11 ± 0.01	7.07 ± 0.1	pu	13.33 ± 0.1
Thymiaterion (1)	Unknown	2.45 ± 0.01	72.86 ± 0.1	1.11 ± 0.01	8,90. ± 0.1	pu	14.68 ± 0.1
Thymiaterion (1)	Unknown	2.40 ± 0.01	72.31 ± 0.1	1.14 ± 0.01	10.18±0.1	pu	13.98 ± 0.1
Thymiaterion (4)	Unknown	10.95 ± 0.1	48.21 ± 0.1	2.50 ± 0.01	8.33±0.1	pu	30.00 ± 0.1
Thymiaterion (4)	Unknown	6.03 ± 0.1	48.25±0.1	2.65 ± 0.01	12.06±0.1	pu	31.00±0.1

EDXRF quantitative data processed with Monte Carlo Simulation Method (tab. 2)

Results from EDXRF analyses	s of artefacts from archaeological settlements in Sardinia (Monte Carlo Simulation Method). Values in wt.%	archaeological se	ettlements in Sa	rdinia (Monte C	arlo Simulatior	n Method). Val	lues in wt.%
Artefact	Provenance	Fe	Cu	As	Sn	Ag	Pb
Warrior (breast)	Ossi sa Mandra	2.46 ± 0.01	90.41 ± 0.1	0.62 ± 0.01	0.37 ± 0.01	pu	6.15±0.1
Warrior	Ossi sa Mandra	2.37 ± 0.01	91.02 ± 0.1	0.75±0.01	0.50±0.01	pu	5.36±0.1
Warrior	Nuraghe Pitzinu	0.61 ± 0.01	97.54 ± 0.1	pu	PN	pu	1.84 ± 00.1
Warrior with rapier and shield	Ossi sa Mandra	0.86 ± 0.01	91.24 ± 0.1	1.87 ± 0.01	4.31± 0.1	pu	1.72 ± 0.1
Bull (spine)	Perfugas	2.51 ± 0.01	87.81 ± 0.1	0.72 ± 0.01	0.60 ± 0.01	pu	8.36±0.1
Bull (spine)	Perfugas	1.39 ± 0.01	84.20 ± 0.1	0.12 ± 0.01	6.16±0.1	pu	8.13±0.1
Bull (belly)	Perfugas	1.57 ± 0.01	88.66 ± 0.1	0.60 ± 0.01	0.12±0.01	pu	9.05±0.1
Bull (between the paws)	Illorai	3.62 ± 0.01	78.53 ± 0.1	0.56 ± 0.01	7.12±0.1	pu	10.17 ± 0.1
Bull	Illorai	2.10±0.01	81.70±0.1	0.58 ± 0.01	3.96±0.01	pu	11.66±0.1
Offerer (breast)	Siligo	2.71 ± 0.01	78.32 ± 0.1	0.13±0.01	5.81± 0.01	pu	13.03 ± 0.1
Offerer (mantle)	Siligo	1.44 ± 0.01	79.19±0.1	0.13±0.01	6.15±0.1	pu	13.09 ± 0.1
Offerer	Olmedo	1.77 ± 0.01	80.84 ± 0.1	0.14 ± 0.01	3.67 ± 0.1	pu	13.59 ± 0.1
Kouros	Olmedo	0.28 ± 0.01	71.25 ± 0.1	5.81 ± 0.01	9.49 ± 0.01	pu	13.17 ± 0.1
Kouros	Olmedo	0.58 ± 0.01	78.03 ± 0.1	6.94 ± 0.01	6.94± 0.1	pu	7.51 ± 0.1
Offererer	Unknown	1.04 ± 0.01	87.83 ± 0.1	2.26 ± 0.01	6.09± 0.1	pu	2.78 ± 0.01
Small boat (bottom)	Ardara	0.20±0.01	81.78 ± 0.1	1.61 ± 0.01	4.44 ± 0.01	pu	11.97 ± 0.1
Small boat (side)	Ardara	0.20 ± 0.01	78.98 ± 0.1	1.75 ± 0.01	5.78±0.1	pu	13.30 ± 0.1
Deer head proteome	unknown	0.35±0.01	80.24 ± 0.1	1.52 ± 0.01	0.01 ± 0.01	pu	17.88±0.1
Deer head proteome	unknown	0.41 ± 0.01	78.92 ± 0.1	1.77 ± 0.01	0.03 ± 0.01	pu	18.88 ± 0.1
Mufflon	Laerru	1.51 ± 0.01	83.4 ± 0.1	pu	9.0±0.1	pu	6.0±0.1

EDXRF quantitative data processed with Monte Carlo Simulation Method (tab.2)

4.6 Discussion of the results

Metallurgy related to the Phoenician-Punic presence and mineral richness in Sardinia

The development of metalworking in Sardinia starts from the use of copper, passes through the extensive adoption of bronze alloys, and subsequently goes trough the beginning of iron-working. The earliest metal artefacts include a series of daggers, swords and spearheads whose analyses reported a composition of unalloyed copper. The next stage in the development of metallurgy was the production of arsenical copper (likely to be produced by arsenical copper minerals). The earliest bronze artefacts were all made of tin bronze where the two main impurities were found to be Pb and Fe, probably contained in the copper ores, since the refining process was not particularly efficient to remove them. The first bronze artefact appears in the so called "Bonannaro Culture", (XVIII-XVII century BC, Bronze Age Phase I). The introduction of leaded bronze begun also in Sardinia as a subsequent phase of metalworking, so it is not unusual to find, for the Beginning of Iron Age artefacts, a high lead content accompanied by variable amount of tin. As it has been seen previously stated for the small Iberian statuary, the beginning of the use of leaded bronze can be understood as coinciding with the first Phoenician attestations in Sardinia. In some case a high lead content appears to be accompanied by a small amount of tin. This suggest that, perhaps when tin was not available, it was substituted by lead. But this observation is not always true. The presence of artefacts in which high lead contents are accompanied by high tin contents reveals that in some cases the addition of lead, as an inexpensive substitute of the tin, was sometimes unfounded.

The reason for such a high content of lead in the tin alloys, as already explained in chapter III, is to be found in the cheapness of the alloys thus made and the ease of production (the metal in the molten state is more fluid due to a lowering of the melting point). The lead in the alloy does not have a significant solubility in copper and tin alloy but will form a dispersion of larger or smaller particles, depending on the temperature of cooling. These lead particles, softer than the matrix, impart a structural discontinuity to the artefact, decreasing the hardness and making more difficult any machining operations. This is the reason why such a high deliberate addition of lead is found mostly in ornamental objects. Indeed the addition of lead favors the obtainment of artefacts with articulated surface

details, which were then finished for engraving or torsion (Giardino 1995). The large amount of lead, for example, is not very often found in the ancient Punic coins, who had to survive the minting on their surface (Ingo 1994) even if there are some coins with high lead presence (De Caro *et al.*, 2002; Ingo *et al.*, 1997; Manfredi, 1997).

This wide variation of lead amount in bronze small statuary gives a certain indication of its deliberate addiction but does not allow to say that the added amount was precisely chosen by the artisan.

The arsenic, zinc and antimony presence as secondary elements, comes from the original minerals (copper sulphides) which have not been released during roasting phase. During the process of roasting the mineral sulfides were burned in air at elevated temperatures, so as to convert the sulphides to oxides. This procedure, beyond the production of copper oxide, purified the charge from the impurities of arsenic, zinc and antimony. Sometimes the presence of arsenic may indicate a possible re-use of older metal as the high presence of arsenic was found in many artefacts of the Early Bronze Age (with varying levels between 1-2% and 6-7%). However, the possibility of an intentional addition of arsenic during the melting of the alloy should be taken into account, with the aim of deoxidize and to increase the hardness of the artefacts (Ingo, 1994). The presence of iron can give a lot of information, going from the smelting of ferrous minerals to the use of high technology in the pyrometallurgic process (De Caro *et al.*, 2002). In this case, the presence of iron tells that the furnace used for smelting the ore operated in a reducing environment, and the iron could be present in the metallic bath also because of the use of fluxing agents based on iron oxides, in order to eliminate the residues of siliceous material from the melt.

Some authors suggest that this kind of reducing furnace and the use of fluxing agent have been introduced by the Phoenician metalworkers.

Starting from these introductory information, EDXRF analyses on the statuettes can be now interpreted, attempting to characterize the base alloys.

4.6.1 The small Bronze statuary and the mineral resources

As it is already been said in many occasions, Sardinia in antiquity was known for its vast wealth in minerals (Bartoloni). In the previous paragraph on ancient mines exploitation it was explained that ores of silver, lead and copper were the fundamental goal for commercial traffic in this area. Nowadays, in the light of archaeological discoveries, is interesting to note that ancient centers were located in the immediate vicinity of the most important ore deposits, or to natural harbors. So, on the basis of the presence of mines the reasons that led to the foundation of strategic settlements can be interpreted. The exploitation was organized by the local population while the trading of minerals was mainly carried out by Oriental populations. The main sites of silver extraction in the form of lead deposits and argentiferous galena, often in the inland area, were well connected to the settlements near the sea.

Between the end of the II millennium and the beginning of the I millennium BC, all the Sardinian deposits belonged to different nuragic cantons and the minerals were then purchased with the Phoenician merchants through the practice of exchange of products, donations and purchasing.

In the following part of the paragraph it will be explained the strong relations between EDXRF data of the statuettes, their settlements of provenance and the presence of anciently exploited basins nearby.

The production of silver (from the lead deposits) in **Antas** for example, which exploitation was carried out at least until the IX century BC, is documented by the presence of graves and the village found near the deposits. The interest in the Antas deposits was renewed with the Carthaginian conquest of Sardinia: this is proved by the discovery of an inscription, dating from the V century BC, to Sid Addir Babbay in the sanctuary, a Punic deity associated with a local divine personage, in memory of the presence of Carthaginian and Sardinian in this place. The Carthaginian authority used often to establish ownership of mines with a syncretism of deities. This explains the finding in this area of the statuette, with a noticeable lead content (XRF spectra in fig.6), which was probably coming from the ore deposits in the area of Montevecchio, Villacidro, Iglesias and Monte Zippiri and the area of Fluminimaggiore and Gonnosfanadiga (especially Iglesias is one of the largest deposits of argentiferous galena in the island), while the non-local appearance can be explained by the presence in that settlements of mixed local and oriental features.

From the same mining area also belong to the settlements of **Sardara** and **Monte Sirai**, close to Antas: all the analyzed artefacts coming from this area are characterized by a high lead content (XRF spectra in fig.7). This observation carries to an hypothesis: probably the silver extracted in this area was intended to be traded, whilst the lead, separated by the mean of cupellation, was widely available for the production of manufactured decorative statuary. The lead copper mixtures could be in line with the general technological evolution

that occurred all over Europe in the Early Iron Age and were marked by hoards of artefacts with high lead levels. A low Pb content is conversely found in **Villacidro** offerer, suggesting that this explanation model cannot be widely considered.

Nearby Capo Monte Santo are located the argentiferous mines of Corr'e Boi, of the mining area of Monte Arbu at the Gennargentu. The rich deposit headed to embarkation points near **Baunei.** Even in this case it is an ancient town centered around the presence of a sanctuary dedicated to Astarte, the Phoenician goddess (probably hidden by the presence of a church). The presence of these silver mines explains the presence of this element in the EDXRF analysis of the tribal chief of **Abini**; the nuragic site in fact is located in the vicinity of the Gennargentu Mountain (XRF spectra in fig.8).

These XRF data were compared with the results obtained by the analysis of other twelve nuragic bronzes from Teti, and a similar surprising presence of silver was also found in the worshipper with braids from Teti Abini, where the silver had entirely replaced the tin (Lo Schiavo, 2005; Atzeni *et al.*, 1992). The silver is not commonly added to copper because of the low solubility in the solid form. The curious presence of a large amount of silver can be explained in the antiquity as a lack of expertise in the cupellation method for separating silver from argentiferous lead. Another hypothesis is that this alloy was an experiment carried under foreign suggestion, for the reproduction of *candidum corinthium aes* (Giumlia-Mair, 2000). The results of this lively metallurgy are given by the metal mixtures found all along the history from the Bronze Age to later periods: the addition of silver to the alloy was an attempt to improve the aesthetic characteristic of *corinthium aes*, by the experimental method of subsequent trials (Lo Schiavo *et al.*, 2005).

The nuragic sanctuary of Abini (Teti) is also known for the discovery of metallic tin scraps and tin minerals and these rare findings are attested only in other three settlement in Sardinia: in the hoard of Forraxi Nioi (Nuragus), in the foundry of La Maddalena (Silanus/Lei) and S'Arcu' e is Forrus (Villagrande Strisaili).

At Abini settlement 6 kg tin ore fragments were found, together with bun ingots indicating that this was not a votive area but a metallurgical workshop. The same observation can be assigned to Forraxi Nioi, where many fragments of cassiterite were collected. In the Maddalena, besides tools, ornaments and weapons, a piece of white metal has been found, which underwent analysis and turning out to be pure tin. The last in chronological order was the finding, in 1999, of few fragments of metallic tin from the ground, confirming the belief that a foundry, serving the sanctuary, must have existed also in this site. In these case

tin metal fragments and minerals was found together with copper, lead and iron bars, allowing the supposition that these were foundries, where melting and alloying operation took place. The fact that in these areas no smelting clues were found makes possible that the tin came from outside as a commercial product, here only used for alloying.

In Sardinia there are some tin mineral resources: in Matzanni (Genna Cantoni), Perdu Cara (Monte Linas), Canale Serci (Villacidro), Su Suergio (Martalai), Corti Rosas (Ballao). What is not sure is if the Bronze Age technology were adequate for this exploitation, because in some of these case the cassiterite is scattered in the massive quartz so only flotation process can separate the mineral from its primary mixture. A possibility that need to be much more investigated is if cassiterite placers, with high concentration of ore, easier to be exploited, could be present in these areas (Lo Schiavo, 2005). In fact, if we consider that hypothesis, ancient metallurgists could have exploited all the ore and used it for their alloys, leaving any clue on the placers presence.

Interpreting the mentioned mineral and metallic residues, a local provenience of the source of tin seems not to be possible, based on analysis on the lead isotopes in the tin scraps. Another possibility is the provenance of this tin from Tuscany but the question is still open to debate and the analysis of major number of samples is needed. The possibility of a foreign supply of tin is plausible because the commercial route from Tuscany, Iberia Peninsula, Cassiterides isles and Germany has a long history.

In any case this digression has been made in order to interpret the analytical results found for artefacts from **Baunei**, **Lanusei** (spectra in fig.9), **Oschiri** and **Nule** (fig.10), and partly also for other two statues from **Abini** (offerer and demon) quite rich in tin, as can be seen from the data in tables. Baunei and Lanusei are two very close nuragic sites and the compositions of two artefacts show also similarities in the composition (high level of tin and lead). Considering the proximity of the two sites with both Su Suergio and Corti Rosas cassiterite deposits it is possible to assume in a hypothetical way the exploitation of a local resources. The Baunei site (Lanusei has the same position) is already been identified as a mooring point of merchant ships, so the presence of objects containing high tin levels can be seen both as the results of local resource usage, despite it is also possible that in these two sites the trading of tin ingots from outside took place (e.g. from neighboring Tuscany). Another attestation of tin bronze can be found in Tharros (Ingo *et al.*, 1996) where fragments of melted refractory materials containing high level of tin have been studied. The vitrified zone of the fragments show a noticeable amount of iron and tin, as well as tin and copper metallic dropplet, residues of the pyrometallurgical process for the formation of highly tinned bronze. These results indicate that inside Tharros settlement, a high temperature pyrometallurgical process was carried out to produce bronze by the mixing of cassiterite with metallic copper. Reaching the bronze melting point, the cassiterite is reduced to metallic tin and absorbed by copper through cementation process. This interpretation explains the absence of tin ingots in the Sardinian territories (this absence can be explained alternatively by the scarce durability of tin when exposed to weathering).

In the northern Sardinia are located **Bonorva** and **Flumenelongu** archaeological sites (very close each other) were the two truncated statuettes come from. These two statuettes show very similar iconographic features interpeted, as written previously, as an "oriental" production. Near Flumenelongu are located metalliferous deposits of Nurra, where were located the very famous silver mines (silver-lead). The mining town was so rich to be called Argentaria or Agentiera and was so vast that it covers the entire promontory, jutting out of the sea. The area of Argentiera (Nurra) is divided into different polymetallic areas: the first extends from the mining sites of Argentiera and Canaglia to the promontory of Capo Caccia, and includes the ancient copper mine of Calabona. Mining areas of Argentiera, for the consistency of the ore deposits of lead, zinc, silver, and even iron (Calabona), had been exploited since the pre-Roman time and after, during the colonization. The archaeological sites affected by the presence of the Phoenicians in this area include also the embarkation site of the precious metals, Sant' Imbenia, in which nuragic village were found numerous traces of oriental influence, probably owed by the merchants and correspondents for the transport and trade of minerals and metal half-worked ingots. In a area so much frequented by merchants from the East and so rich in mineral resources, it is easy to interpret the data obtained from the analysis of the two truncated statuettes, one rich in lead and the other in tin, as a product of a mixture of iconographic and technological items, evidence of strict relations between the Oriental and local peoples. It is not clear why the two statues were cut off at the legs and it is impossible to understand the timing of this operation (probably not antique). One hypothesis is that both the statues were originally inserted in holes in the rocks, and soldered with lead. The two statuettes broke on the leg level during the removal from that base.

Olmedo and **Flumenelongu** statuettes, characterized by the same structure and rendering features, which connect strongly to Phoenician production are made of ternary alloy. The two archaeological sites are only 15 km distant, suggesting that this two statuettes were

produce from the same metal workshop, frequented by Phoenician artisans probably connected with the Sant'Imbenia harbor.

The ark and the Centaur, from **Oschiri** and **Nule**, are very interesting because they have similar composition (XRF spectra in fig. 10). The two sites are very close, as the two representations of artefacts are very similar. Both the ark and the centaur are reinterpretations of themes to which the local population was tied (*boe muliache* and *cascias barbaricina*) fused with oriental elements (the centaur and the Homeric *Keimelia*). The two artefacts associated with individual burials of high ranking deceased, give exactly the idea of how the new aristocracy was connected with the allochthonous presence on the island. A similar composition and an analogous merging of local and foreign myths, suggest that the two products have been made from the same metal workshop. Also **Illorai** and **Perfugas** bulls, as the artefacts from **Nule** and **Oschiri** show similar EDXRF data XRF spectra in fig.10 and 11).

A last consideration is made for the statuettes of local typology coming from **Ossi sa Mandra**, **Nuraghe Pizzinnu** and **Teti**. The three warriors from Teti, Nuraghe Pizzinnu and Ossi sa Mandra are made of copper or binary bronze with small amount of lead, in accordance with the general conclusion that especially the "oriental" types were produced with the ternary alloy. On the other hand one warrior from Ossi and the archer and demon from Teti, local artefacts as well, are made of ternary bronze. This discrepancy of data can be explained starting from the type of archaeological settlement. In Teti also an "oriental" type of statuette has been found, the offerer, made of ternary alloy. In the light of this observation Teti was a settlement frequented also by Phoenicians, or in connection with them, so probably the commitance were both local and oriental, as can be said probably for the artisans working in this area, sharing technological skills and decorative features. Ossi sa Mandra on the other hand, is characterized by the presence of a foundry which again were probably influenced by the presence of the much more skilled Phoenician artisans.

The last results are reported for three artefacts coming from nearby areas, the votive ship from Ardara (De Boes area), the offerer from Olmedo and the mantled offerer from Siligo. These three artefacts show similar trend in composition, a ternary alloy with analogous amount of metal content, suggesting, due to the proximity of the three areas, a unique metal foundry or a similar technology of production (XRF spectra in fig.12).

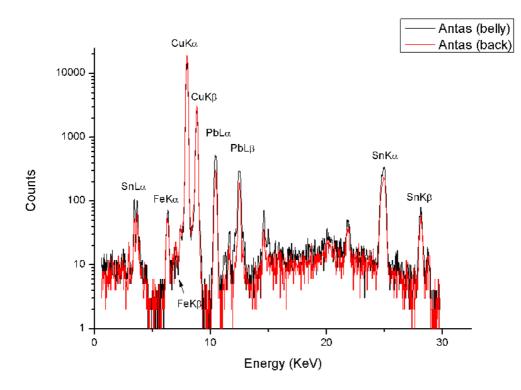
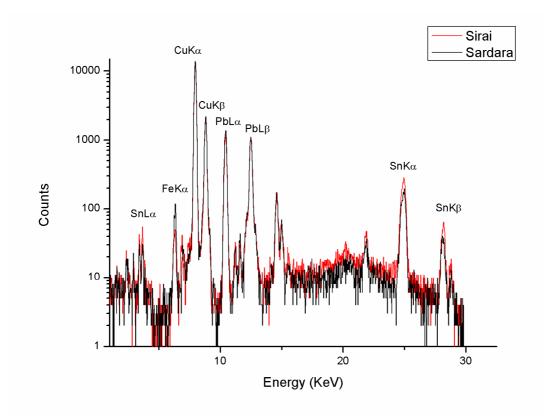


Fig. 6: (above) EDXRF spectra of Antas statuette

Fig. 7: (below) EDXRF overlapped spectra of archer from Sardara and statuette from Monte Sirai



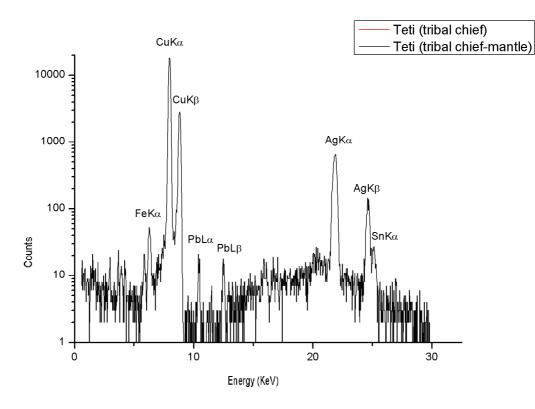
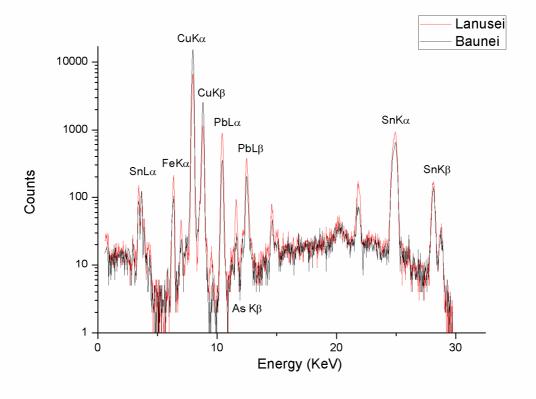


Fig. 8: (above) EDXRF spectra from the tribal chief of Teti

Fig. 9: (above) EDXRF spectra of Lanusei and Baunei artefacts



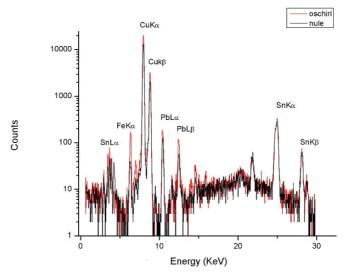


Fig. 10: (below) EDXRF spectra from the centaur of Nule and the ark of Oschiri

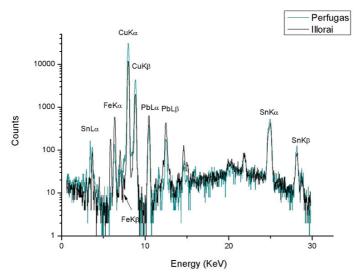


Fig. 11: (above) EDXRF spectra from the bulls of Illorai and Perfugas Fig. 12: (below) EDXRF spectra from the votive ship of Ardara and the offerers of Siligo and Olmedo

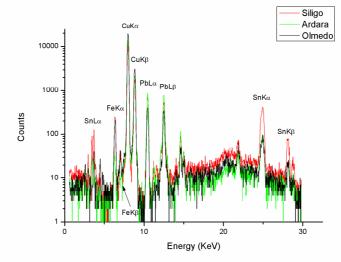




Fig. 13: GIS thematic map with localization of mines and some of the sites mentioned in the paragraph

A GIS thematic map with the main mentioned ore basins of copper lead and tin and some of the most important settlement is proposed in Fig.13. In this way is easier to appreciate the connections exposed previously.

From the table of compositions is possible to realize that a sharp separation of compositions between local and oriental type of artefacts is not highly evident.

Since, as stated by Berbardini & Botto (2010), the interface between Phoenicians and local cultures in Sardinia is different from the Iberian model (the oriental production is more often attested in Phoenician sites in Iberia), and since the oriental subgroup is cronologically collocated in the middle of a longer nuragic period, some new interpretations on these peculiar statuettes can be proposed and in the next paragraph will be exposed. In conclusion of this part, a connection with the exploitaition of mineral resources, especially those directly stimulated by the presence of Phoenicians, can be observed.

4.6. 2. Ternary diagram for Sardinian Artefacts

All the EDXRF produced data has been compared with analytical data from literature (Atzeni, 1995) and a ternary diagram has been produced, to compare a higher number of artefacts. A ternary plot has been produced, containing both analytical data from our sessions in Cagliari and sassari, but also other data (XRF and AAS) from literature (Balmuth 1996; Tylecote *et al.*, 1983; Atzeni, 1995) to have a wider set of data for comparisons. The ternary plot (fig.14) shows a rather sharp separation between local and "orientalizing" *bronzetti* even if some exception cases are found.

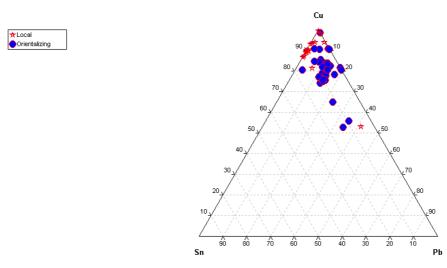


Fig. 14: ternary plot (Cu, Sn, Pb) for nuragic artefacts of local and orientalizing tipology

This could further underline the results obtained during Portuguese analytical session, demonstrating a common trend of evolution in metallurgy even if some differences have been noted. First of all in some case the oriental statuettes in Sardinia were found in local settlements, thus suggesting, as already mentioned by Botto & Bernardini (2010), a fusion of local and allochthonous elements, while in Iberia this kind of artefacts were found in areas of direct Phoenician installation. Furthermore since in Sardinia the Nuragic bronze statuary and in general the metallurgy was already fully developed and the use of lead addition was already tested, is more probable that the Phoenician and Punic influence is absorbed more from stylistic point of view, creating a real merged production.

4.6.3 Comparison between Alcacer do Sal (Evora) and Sardinian statuettes

As already mentioned above, EIA Orientalising bronze objects from Iberian Peninsula Phoenician settlements are quite comparable, both from a typological and from a compositional point of view, to collections from related Mediterranean areas, such as that of Sardinia, but also other areas of Italy and Sicily (La Niece, 2007; Ingo, 2006). Except in one case, the Portuguese data confirm the compositional similarities with the Portuguese samples here investigated in the common use of Cu-Sn-Pb alloy. As it is the case of Evora and Alcacer objects, also in the Sardinian artefacts both Sn and Pb show a high compositional variability ranging respectively from 4.0 to 16 wt% and from 1,4 to 17.5 wt%. Again must be taken into account the heterogeneity of the ternary alloy itself where the lead is immiscible at the solid state. What can be said with some certainty is the presence of similar features in the rendering of Sardinian and Portuguese bronze little statuary, as it is clearly visible from the comparison of the Evora goats and the mufflon from Laerru, although the metallurgy in Sardinia is based on a more populous and detailed nuragic (local) production. Furthermore the compositions of the goat and mufflon are comparable. The introduction of leaded bronze started in Sardinia, as in other Mediterranean area, as a subsequent phase of metalworking, so it is not unusual to find, from Early Iron Age onwards, high lead contents accompanied by variable amounts of tin in bronze making. As seen previously in the case of the Iberian small statuary, the use of leaded bronze is verified especially for decorative or votive objects.



Fig. 15: goat from Evora and and the mufflon from Laerru-Sardinia

So in conclusion, the comparison of bronze statuary between Portuguese and Sardinian production, evident also as a result of a aesthetical rendering for the case of the Portuguese goat and for the Sardinian mufflon (fig.15) (which show perfectly a common cultural matrix: the Oriental presence in both areas), is in any case verified for other statuettes, whose compositions are comparable.

4.6.4 Statuette with gilded mask from Giorrè.

This statuette (n.29 in the catalogue) represents a naked standing figure with a silver mask with traces of gilding on the hair. In this case the analysis were carried on cleaned microareas of the object. We decided to add this artefact to the sample population because, even if it is classically allocated between roman productions (II century BC), it shows connection to the Punic use to decorate the statuettes with silver and gold leafs on the head or other parts of the body (a similar use is also found in North Africa and Spain). In this case the analysis were carried on cleaned micro-areas of the object. Furthermore the presence of Punic and Siceliote coins in the site of provenance could indicate the presence of a mixed Punic and local population whose culture survived to the Roman colonization. The mask was originally a decorative element of another object, as is testified by the presence of the rosette-shaped decorative element on the upper part of the mask, from which it was cut. The mask was then applied on the bronze face of the statuette, which as it is shown by the radiographs performed on the piece (D'Oriano, 1997), had not been outlined by the artisan. Therefore the statuette was made from the start with the intention of applying the mask on the face area. The aim of the whole study of this statuette was to explore the layered structure of the statuette and to determine the homogeneity and thickness of the gold silver and welding sheets. This is possible by using the internal ratios of characteristic X-ray lines Au $(L\alpha/L\beta)$, Ag $(K\alpha/K\beta)$, Au $L\alpha/Ag L\beta$ and Pb $(L\alpha/L\beta)$. The results were compared with those obtained with simulated X-ray spectra obtained both experimentally and both by using Monte Carlo simulation method (this part of the research is fully described and published in Cesareo et al., 2013). From this study has emerged that the bronze contains high level of Pb and Sn and the silver mask, gilded in the forehead, hair and chin, is soldered to the statuette with a lead-tin alloy of approximately 55% Sn and 45% Pb, similar to the brazing alloy found in Alcacer do Sal jar (Chapter III part B).

4.6.5 Discussion of the XRF results for the *Thymiaterion*, ingots and tools

The analytical data of the thymiaterion show a composition of high leaded ternary alloy. The tymiatarion is composed of four pieces showing different composition (fig.16). This composition was compared with the analysis carried out by Jimenez Avila (2004) on Iberian censers. The two sets of measurements are comparable because 60% of the Iberian samples contain an average level of lead of 4.5% with peaks of up to 32.48% (Timiaterio de

la Quejola). Despite the complete lack of context information the artefact is certainly "orientalizing", for its own structure and because of the composition.

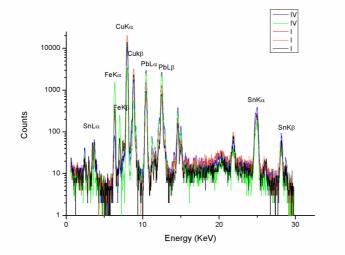
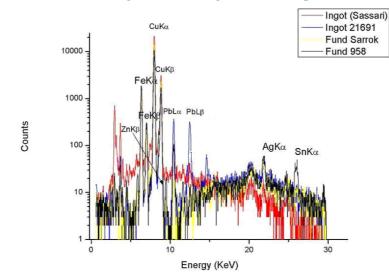


Fig. 16: EDXRF spectra obtained from different dismountable parts of tymiaterion Discussion of the analyses on plano-convex ingots and tools

In conclusion, to remark more the already obtained conclusion on Sardinian bronze production other five examples, analysed during the session in Cagliari and Sassari Museum, are presented: two plano-convex ingots from Nuraghe Sant'Antine and Nuraghe Flumenelongu (table 4 and spectra in fig. 17) two tools from Ossi sa Mandra and Flumenelongu (table 5) and two furnace funds.

Description	Fe	Cu	Zn	Ag	Sn	Pb
Fund 958	14.31	79.82	1.34	4.21	0.28	0.04
fund						
Sarrok	7.72	89.39	0.64	1.99	0.12	0.14
ingot	3.12	94.80	0.79	0.53	0.27	0.49
ingot	12.90	61.45	0.43	2.39	0.39	22.44



 Tab. 3: XRF quantitative data of the ingots;
 Fig. 17: EDXRF spectra of the ingots and funds

Area	Туре	Cu	Zn	Ag	Sn	Pb
Flumenlongu	Axe	96.17	0.38	0.27	1.43	1.75
Ossi sa						
Mandra	Axe	94,48	0.20	0.94	3.23	1.16

Tab.4 : XRF quantitative data of the axes

In the table 3 the analytical results suggest that the ingots can be divided in primary and secondary ingots (as explained in Lo Schiavo *et al.*, 1990). The first three examples are primary ingots in which only one element, copper, is present, beside the impurity. In the secondary ones, written in red, a voluntary addition of lead is recognized, then it can be said that the addition of lead could be premeditated since the production of semi-finished ingots (this is an hypothesis as is difficult to understand the belonging phase of pyro-metallurgical cycle).

In table 4 three axes from Ossi sa Mandra showed standardized compositions which can be considered as binary bronze or better copper artefacts. The absence of lead must be interpreted in the light of the mechanical stresses an axe must resist. The presence of lead separated globule produced a discontinuity that favours the fracture of a metal objects. While submitting a copper object to hard hammering cycles allows to produce a stronger tool. In conclusion in Sardinia a differentiation of alloying and mechanical treatment were already common procedures in the metal working.

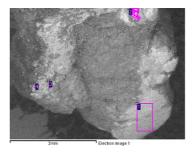
4.6.6 A circular dog-shaped harness from Near East Museum- Sapienza University



Further analysis were carried on on a dog- shaped harness preserved in the Near-East Museum (Sapienza), coming from Sardinia (unknown settlement). These data are reported because they have been obtained on a cleaned surface (mechanical cleaning by the use of scratching utensil and

Fig. 18: dogshaped harness from Sardinia

abrasive paper) of the object to have a reliable compositional percentage of the alloy.



S.	Fe	Cu	Sn	Pb
1	1.8	78.3	16.9	3.0
2	4.1	84.9	7.9	3.1
3	3.4	98.5	4.7	2.7.

Fig. 19 and tab. 5: SEM-EDS analysis on a cleaned area of the artefact

From the interpretation of EDS data the alloy is composed of a binary bronze with a Pb content in the range of 2.7-3.1%. A similar object was also found in Monte Sirai, together with the two statuettes of the Libation and Lyre Player. This object can be interpreted as an horse harness.

4.7 Conclusions

This set of analytical data and archaeological, historical and stylistic considerations, crossed out with the localization of ores and mines in the highlighted areas, lead to the interpretation of a mixed local-oriental production carried out in workshops where, in other words, local artisans were working together with metallurgists of Phoenician tradition and culture, exercising together in areas of indigenous worship and engaged in satisfying oriental and local customers. The presence of Eastern culture artisans collaborating with local artisans explains the strong circulation of patterns and postures not typically local in Sardinia. These Phoenician craftsmen were themselves influenced by the Aegean productions, including Cretan and Levantine, thus carrying with them other foreign traditions. All these influences are absorbed by the Sardinian production with the arrival of the Phoenicians. This explains the difficulty of assigning a specific culture, Phoenician-Punic or local level, owing to the connection between different settlements, exploitation of the mine basins, the external commercial intent, and the cultural melting. It is clear that in Sardinia there was a connection between exploitation of the ore basin, the external commercial intent, and the cultural melting. The metal trade routes were of strategic importance, and were certainly accompanied by an exchange of technical know-how.

Since also the local statuettes from Teti shows a strict connection with the localization and exploitation of ore basins, it can be inferred that in Sardinia the compositional data are more influenced by a territorial characteristic than by the presence of external agents like phoenician presence. In fact, in Sardinia metallurgical industry was highly specialized since the Bronze Age and mineral resources were already known and exploited. The arrival of the Phoenician merchants caused an increase in the exploitation of ores and resulted in a more systematic relationship between settlements, mines and harbors.

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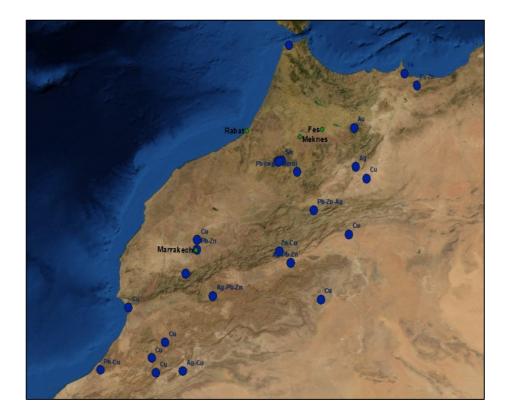
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CHAPTER V

ANCIENT MINES OF MOROCCO AND ARCHAEOLOGICAL EVIDENCES

Class of objects: metallurgical slags and artefacts



GIS thematic map on ancient mines in Morocco

Ancient mines of Morocco : archaeological and archaeometric approaches

Highlights

Since Phoenicians and Punics undertook exploratory trips driven by interest in trading minerals, metal ingots and artefacts, a part of this thesis is devoted to the study of mineral resources in Morocco. In fact this area was certainly affected by their presence and certainly was recognized as one of the richest in mineral deposits, but unfortunately, since currently it is not interested by extensive archaeometric studies, it has been underestimated as area of mining exploitation touched by the Phoenician routes near the Strait of Gibraltar. The depth study of this area is therefore very important for a correct, and relatively unprecedented, interpretation of the Phoenician and Punic metallurgy. This research has involved typical instruments of cartographic research such as the study of satellite images, GIS and interpretation of the territory in the light of bibliographical research, while the archaeometric study of artefacts and slags found during prospection has been carried on with SEM-EDS, XRD and Mass Spectrometry.

5.1 Introduction

From January 2012 a bilateral agreement between the ISCIMA-CNR and the Moroccan CNRST, in detail the team of Geophysics of Universitè Moulay Ismail in Meknes, has been activated for the study of mineral-metallurgical history of Morocco in the Phoenician-Punic period.

The scarcity of studies about the use of mines in Phoenician and Punic times and about processing technologies, let consider of great interest the research on the ancient mines of Morocco. The region is indeed privileged to have the presence of major mining areas, Punic and Phoenician cities and plenty of metal artefacts and coins minted from the autonomous cities. Morocco in fact, due to its geographical location and geological diversity, has interested many civilizations through history (Protohistory, Roman and Islamic period), and this interest was often caused by the great richness in minerals. The different Moroccan regions, and specially the southern part, have delivered many kind of metals (Lead, Zinc, Copper, Silver and Gold). Important, therefore, is the possibility of an integrated study that follows the ore from its mining areas (with the relative changes in the palaeoenvironment and the characterization of anthropogenic frequency), the identification of transport routes, storage of the mineral, the study of the processing areas and of the pyro-metallurgical processes and products obtained. This kind of research leads to the reconstruction of a complete production cycle referred to an historical epoch, the Phoenician-Punic, which represents the connection point between protohistorical metallurgical traditions and the Etruscan-Roman productions.

This research has some innovative features, due to both a wider approach applied to this issue and to the geographical area taken into account, Morocco. In fact, not only studies on mining in antiquity are few for this area, but it is also less regarded the important role that this area has had in the trade circuit nearby of the Strait of Gibraltar, the true focal hub of the Mediterranean basin. In archaeological and archaeometric studies the most known basins, such as the Sardinian, the Cypriot and Iberian ones, are often mentioned, leading, as we shall see later, sometimes to misleading conclusions. The fundamental role of this phase of research is therefore precisely to deepen and assert the importance to the archaeometric studies in North Africa.

This joint research is based on a set of different tasks, that permit to individuate numerous interesting mineralogical areas. These steps are ranging from the identification of the mineralogical basins through literature and through mapping and photointerpretation, GIS and cataloging of data, archaeological prospection and archaeometrical analysis such as SEM-EDS detection of primary and secondary elements and recognition of the structure and morphology of the samples or XRD analysis for the mineralogical composition and the degradation products. Currently these research studies are mainly performed on the region of Meknès, where many interesting site were found: one of this is the well known mine of Aouam and, not very far, the very famous archaeological site of Volubilis. Many samples of slags and metallic artefacts have been taken from these two sites, submitted to SEM-EDS and XRD analysis, the results of which are being presented in this chapter. Also a preliminary analysis with Mass-Specroscopy has been carried out on a metal leaf from Volubilis, to detect the Pb isotopes ratios, in order to assign a basin of origin to this hoard of metal artefacts. This part of the research is currently at a preliminary stage but as the project on the ancient mines of Morocco, approved by CNR and CNRST, is at the beginning phase, and will last in the future, a more thorough search, including all the slag and artefacts from Moroccan ore basins, will be performed, taking into account also the trace elements of the samples (it is expected to perform these analyzes with PIXE-PIGE), and the isotopic abundance of further elements.

4.2 From literature to GIS thematic maps

The ancient mines were located on the basis of data from literature (texts of ancient authors were studied such as El Yaqoubi (IX sec.) El Bekri (XI cent.): Kitab el Istibçar (XII

century.): El Marekeschi (XIII sec.): Leone l'Africano (XVI century).: Ibn Haucal (JA 1842), but also texts on geology of the Moroccan territory) (Gsell, 1928; Julien, 1970; Moret, 1930; Toubal, 1995). Then all this information has been collected in a GIS system and 21 anciently known mining sites, exploited for the Cu, Au, Sn, Fe, Pb, Zn, Ag smelting, were located using this system (fig.2), while fig.1 the digitized rivers have been place in relation with archaeological punic sites.

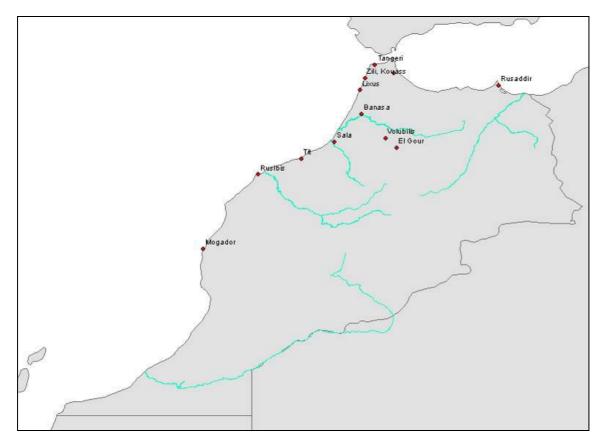


Fig. 1: major Moroccan rivers and Punic sites

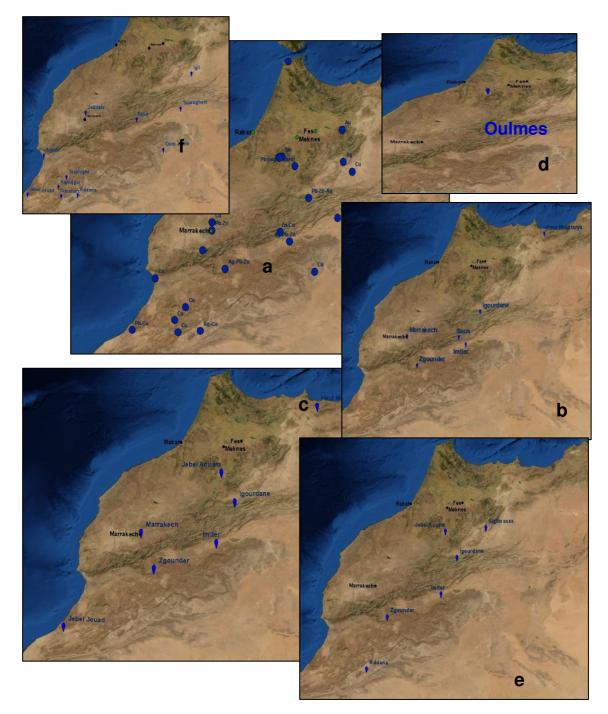


Fig. 2: GIS thematic maps on ancient mines of Morocco. A)Cu, Sn, Pb, Fe Ag; b) Pb; c) Zn; d) Sn, e) Ag, f) Cu

On the base of the thematic maps produced, and with the aid of remote sensing images study (fig.3), in July 2012 (and partly during a second mission in 2013) the mines of



traces of ancient exploitation are easily visible

Fig. 3 remote sensing image for Oulmes mines, where Aoual

Aouam, Oulmes, Zaida, Ait' Ammar and Imiter (fig.4) and archaeological sites of Tayadirt (Lambert, 1968) and El Gour have been prospected superficially. It is interesting to note how much that mines are connected with the presence of rivers, showing a probable pattern of exploitation. From

Aouam site lots of pottery specimens were collected including a fragment of



furnace and tuyere and plenty of slag and mineral fragments (from superficial survey, for these samples dating information is lacking), testifying intense exploitation of the area, probably not only in contemporary times. The samples were analyzed once back SEM-EDS in the

Fig. 4: explored areas and major rivers (in red) laboratory located at Sapienza University of Rome, and the results will be shown in the next paragraphs.

Several findings such as pyro-metallurgical slags and metal artefacts have been studied also from Volubilis archaeological stratigraphy, suggesting that in this area was also dedicated to the processing and smelting of ores to obtain metal. It has been decided to carry out the whole study of these findings with the application of an archaeometric procedure, consisting of the use of SEM-EDS, XRD and mass spectrometry to allow the interpretation of the type and technological level of the forgery herein settled. This study has been undertaken because beside Volubilis Roman stratigraphy in this area also pre-Roman evidences were found, belonging to the Punic occupational period. Even if the artefacts available belong majorly to the Roman Empire period, however it was decided to perform an analytical deepening of these artefacts and slags, because usually the cause of a Roman occupation to an originally Punic site was generally dictated by the intention of conquest and appropriation of exploitable resources. Since Volubilis, as seen in Figure 1, is far from the coastline, it is likely that the interest of the conquerors was to be found in the strategical position nearby a exploited mine. In fact, the Romans conquered the strategically important Punic sites, as can be seen widely in Spain, and since the Punics were excellent knower of their area, their sites were usually allocated in interesting areas from the mineralogical point of view. Also a technological conservatism between the more experienced Punics and Romans let us think that the metallurgical technologies do not extensively change in the Roman period.

In this way, a study of Roman artefacts and slags may give useful clues in the recognition of exploited mines and technologies borrowed from Punics.

Aouam Mine

The mining center of Tighza-Aouam (fig.5,6) is located in the administrative region of Meknes -Tafilalet in the Middle Atlas, 6 km far from the city of Mrirt and geologically is located in the northern



Fig. 6: Aouam mine, Northern vein

part of the Moroccan Meseta and is characterized by a



and is characterized by a multimetallic mineralization Fig. 5: Aouam Mine, Ighrama Ousser

composed by argentiferous galena, mimetite and blend (sphalerite). The metals obtained from the refining of these minerals in contemporary times, actually exploited by CMT (Compagnie Miniere de Tiouissit), are lead, silver but also zinc and bismuth. The gangue that characterize these deposits is composed of barite, calcite,

chalcopyrite, quartz and siderite and the type of rocks are schist and sandstone. In much more details Tighza-Aouam deposits is composed of three successive mineralization events: 1) scheelite, pyrrothite, arsenopyrite and gold in skarns, 2)scheelite, wolframite, arsenopyrite and gold in veins, 3) Ag bearing galena and sphalerite. Lead, zinc and silver are in the form of veins, in the form of argentiferous galena and sphalerite. It is known that this mine was exploited since the IX century (Idrissides Dinasty). A fortified wall (fig.5) dated to the XII and XIII century is still present (Boushaba & Michard, 2011). In fig. 6 an open air exploited vein has been noticed thanks to the presence of numerous tunnels that follow its length. This site, due to the presence of exploitation evidence (fragments of furnace, tuyeres, fragments of ceramics and slags) and due to the vicinity to Volubilis, Tayadirt and El Gour archaeological sites, has been considered of high interest, and the slags coming from superficial prospection (dating information is thus lacking) has been deeply studied to characterize the type of metallurgical operation was carried out in here.

Volubilis

Volubilis, known to be one of the largest roman colonial town in Morocco, is currently a very well preserved archaeological site, positioned in the Middle Atlas and is UNESCO World Heritage Site from 1997. The excavations indicate a foundation in the III century BP by Carthaginian merchants as well attested by the presence of the remains of a temple dedicated to the Punic god Baal, and findings of pottery and stones inscribed in the Phoenician language. After the fall of Carthage the city was a part of the Reign of Mauritania, which became a Roman client state and, from the year 40, a part of the Roman Empire. The Punic influence lasted afterwards, as attested by the great consideration into which the Carthaginians where taken, also after their fall. Volubilis, as the centre of big commercial traffic, became the administrative centre of the Mauritania Tingitana Roman province. Volubilis was than abandoned by the Romans around the year 280 when the Berber tribes start to increase in power in the surrounding areas.

The great history and the important role that had this colony, has focused interest on the study of its trade and the production of local manufactures and "industry". Because of is location nearby the mineral basins in the Middle Atlas, and the fact that it is only about 100 km from the Aouam mine, it was decided to investigate the nature of the metal findings resulted from excavation in this area.

As mentioned earlier, samples of metallurgical slags were taken by both these areas, in order to investigate what kind of processing was performed in the Moroccan area, and which were the traded minerals. This argument is presented below, starting from the general discussion of slags chemistry and then in detail the analytical results obtained with SEM-EDS and XRD on Aouam and Volubilis samples.

5.3 The slags

The slag is one of the most durable and noticeable waste product of the pyrometallurgic process. Furthermore the discovery of slags during a prospecting operation is an important event because the composition is chemically stable and therefore these materials are a snapshot of the metallurgical processes that produced them. In other words the microstructure is closely linked to the pyro-metallurgic operating conditions and, of course, to the type of processed minerals.

Since these wastes can be produced during different step of metallurgical operative chain, there is a great variety of slags types such as the extractive ones (obtained by the smelting of rough minerals) and the refining ones (produced during purification and alloying phase). The processes which produce these slags cannot always be clearly defined, because complicated thermodynamic processes take place in relation to the type of temperature and the type of environment (more or less reducing/oxidizing) of the furnace and crucibles. The discussion will proceed for points, to highlight the most salient elements that allow the understanding of slag (Hauptmann, 2007).

1) Parameters of formation and general composition

Ancient smelting processes are controlled by four parameters: composition of the charge, 2) firing temperature, 3) gas atmosphere during smelting redox conditions and 4) reaction kinetics between the components of the charge and the duration of the reaction. Pyrometallurgical process began with the introduction into the furnace of the charge consisting in the grinded (any large impurities in the ore was crushed and removed, this enrichment was carried out trough grinding and washing) ore and charcoal and, if necessary, the addition of scorifying and fluxing agents. The temperature of fire in a ancient furnace could reach 1200 °C and the air were introduced through bellows and ceramic pipes called tuyeres. The forced addition of air was fundamental to guarantee the controlled combustion

of coal to carbon monoxide, which react with the minerals allowing their transformation into metal, becoming carbon dioxide, according to the general reaction:

MeO + CO => CO2 + Me

Or, in the case of sulfur minerals, a preliminary operation is needed, the roasting of the ore on a ventilated fireplace to convert the sulfur into oxide:

Metal sulphides + molecular oxigen => metal oxides + sulfur dioxide

 $MeS + 2O_2 \Rightarrow MeO_2 + SO_2$

Oxide was then treated as shown in the previous reaction.

During the processing of minerals into metal, one of the more complex step was the removal of those non metallic components contained in the metalliferous rocks, for which separation was not possible previously through the sorting. Generally speaking the separation of gangue produces slags that must have a melting point as low as possible and a good ability of sliding, to obtain the physical separation from the metal. To separate these components it was necessary to optimally balance silicate phases and oxides, in such a way to allow the formation of autonomous low-melting silicate slag. The metallurgist had to arrange a selected addition of materials that favored the formation of fluid slag, which is separated from the metal flowing towards the bottom or the outside of the furnace. Iron oxides combine preferably with the silica and form the slag. If the ore was rich in iron, it was therefore necessary to add mineral silicate or quartz, but if the ore was rich in silica, it was necessary to add iron oxide to produce a smooth and low melting fayalite slag.

Fluxing agent: $Fe_2O_3 + CO = 2FeO+CO_2$

Slugs: $SiO_2 + 2FeO = 2FeO * SiO_2$ fayalite

Fayalite is the major component of the smelting slag in reducing environment, accompanied by other mineral species, in relation to the type of ore and the type of process. The presence of alkali and alkalineearthy metals and phosphorus may decrease the c melting temperature of the slag up to 150 ° C less. This is the reason why very often in the slags bone fragments or ash particles were found.

The identification of the original ore from the study of slag is sometime tricky. For example the main FeOAl2O3-SiO2 (C, f, A, S) (system taken elements in the composition of copper slag belong

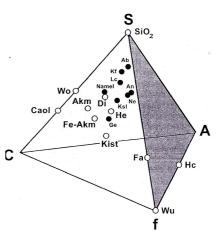


Fig. 7: common phases in iron-rich silicate slags shown in the system CaOfrom Hauptman 2007)

to thernary sistem $FeO-Al_2O_3-SiO_2$ and the iron smelting slags locate in a narrower field in the same ternary diagram, that's why is hardly to distinguish between this two kind of slag (Hauptmann 2007).

Going on with more detail, most of the studied slags, the iron rich silicate slags, mainly from the later periods of the early metallurgy, have compositions in the low melting parts of the quaternary system CaO-SiO₂-FeO-Al₂O₃ (fig.7). As stated previously, in accordance with the type of raw material, a trend towards iron rich silicates component concentration is evident. But not all the slags have a fayalite compositions, there are two different and separate types. In fact this quaternary system for iron rich slags, can be splitted into two ternary diagram: CaO-FeO-SiO₂ (paragenesis fayalite-wuestite-hercynite) and FeO-Al₂O₃- SiO_2 (clinopyroxene-bearing paragenesis) to describe the entire slagging panorama. The major components of ancient slags are spinel (Fe₃O₄ or MgAl₂O₄) and pyroxene kalifeldspar, $(CaFeSi_2O_8),$ feldspars (albite, anorthite), feldspathoids (leucite, kalsilite/kaliophilite, nepheline), melelites, akermanite, clinopyroxenes, calcium silicates (wollastonite, shanonite), olivines (as said fayalite and kirschsteinite), wuestite, hercynite, cristobalite, tridymite. During their formation, iron rich slags react very sensitively to the CO/CO₂ values in the smelting furnace. This leads to the crystallization of different Fe $^{2+/3+}$ ratios. The principle, known as the Bowen-Fenner-Trend in petrology, can be defined by a reaction:

 $3Fe_2SiO_4+O_2 \Leftrightarrow 2FeO^* Fe_2O_3+3SiO_2$

If the pO₂ is sufficiently high, magnetite and silica-rich compound crystallize first, so the

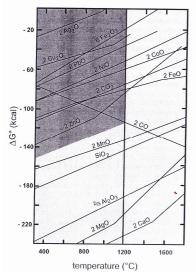


Fig. 8: free entalpy temperature chart for oxide formation, (taken from Hauptman 2007)

main components are Fe oxides. If pO_2 is low, no magnetite will be formed but an iron silicate (fayalite) or even metallic iron precipitates. This conclusion is important in the light of distinguish between different chronology of the smelting process. Plotting the energy of formation ΔG° of archaeometallurgical interesting compounds (fig.8), it is possible to have an idea of which was the reducible one, operating with a furnace with a temperature limit of 1200°C, when using charcoal as reduction agent. Only the oxides with enthalpy of formation above the energy of oxidayion of C to CO can be reduced. All elements that cannot be reduced by the furnace condition remain in the oxide state and form slag constituent: SiO₂, Al₂O₃, MnO, MgO and CaO. Other element are reducible, thus turning into metal and can be separated from the slag (Ag, Cu, As, Sb, Ni, Co, Sn, Zn, Fe, Pb). Additionally, the main, minor and trace elements on the silicate slag matrix can give all information about the origin and nature of the ore. About the liquation of the components it is suggested that slags from ancient periods (IV millennia BC) often contain un-decomposed inclusions which can belong both to the fluxing agent and to the remains of host rock. For example quartz does not take part to the reaction at the liquid state, until a sufficiently high temperature is reached and a long duration is achieved. It is important to note that only a small range of compounds are molten around 1200°C. Other compositions, particularly those with a high SiO₂ content, are not fully molten. In such slags inclusions of undecomposed quartz are likely to be found, cristobalite and trydimite (often observed in ancient slags). Also barite and chromite have a high melting point and are found undecomposed (Hauptman, 2007; Rothemberg, 1990).

2) Smelting and crucibles slags

Another important factor to be explained, important especially for the non ferrous metallurgy, is the distinction between smelting slags and crucible slags (melting/refining and casting). The latter take their name from the small pot where the process were carried out. This kind of slag is often attached to the wall of the crucibles and this is why the composition is commonly alkali-aluminium-silicates, as a results of reactions between the metals and crucibles material, charcoal ash and silica rich fluxes. It is possible to separate these two kind of slags even from a macroscopic study of the external characteristics: crucible slags are heterogeneous, glassy and porous, have small size and weight, while smelting slags (from furnaces) are well crystallized and show only few large pores. There seems to be indications that even in the early stage of metallurgy a highly developed technology was known, for example in Los Millares in southwest Spain or in the chalcolithic site of Shiqmim in Israel, where not only smelting slags but also refining slags were found. The smelting slags are mainly composed of ferrous silicate (fayalite), iron-rich and poor of the extracted metal, while the crucible refining ones are characterized by a higher-valent content of oxides and sometimes residue of wood ash in the matrix.

3) Recognition of smelted ore

The chemical microstructure of slag is, as mentioned earlier, connected to the type of ore used; in other words it is possible to understand, by the presence of certain elements, which

metal was obtained in a peculiar area. With regard to the macroscopic appearance, for instance, a slag with ferrous red-colored external surface, with ocher-brown areas and a black and vitreous fracture surface, could suggest that the slag is the product of a metallurgical process for the transformation of iron oxides into metallic iron without, however, exclude that the slag could be produced during the smelting of lead and copperbased minerals. In the case of iron ore, as for non ferrous metals, if the gangue was rich in silica it was necessary to add iron oxide to produce flowing and low melting slag. To improve separation of slag from solid metallic iron, is used a controlled excess of iron oxides that formed with the silicate contained in the rock a low melting slag. The presence of dendrites phases of iron oxides like wüstite (or more rarely magnetite) and fayalite and a glassy matrix consisting of glassy aluminum-silicate iron of calcium (anorthite), potassium and manganese and the presence of iron microparticles, allow to hypothesize the smelting of iron-based ores. The presence of small metal particles in a minimum quantity indicates a good separation between slag and metal, and then a skilled conduction of the pyrometallurgic procedure (Bultrini, 1995, Ingo et al. 1995). The ancient iron smiths had to use an extraction technique not so different from that used for copper. It is possible that the process was an innovation of the smelting techniques already used to make copper. Iron oxides were used as a flux in the copper smelting process and so with some experimentation the process could have resulted in accidental production of iron, which could have then been developed into purely iron smelting. Iron smelting was originally produced in bloomeries, furnaces where bellows, through the tuyeres, were used to force air through a pile of iron ore and burning charcoal. The carbon monoxide produced by the charcoal reduced the iron oxide from the ore to metallic iron. Since bloomery was selffluxing the addition of fluxing agent was not required. However, the bloomery was not hot enough to melt the iron, so the metal collected in the bottom of the furnace has a spongy mass, or bloom. Is possible that roasting phase in a fire was achieved to remove sulfurs and any moisture in the ore. What was extracted from the oven was not, however in this case, a molten mass but a spongy structure (bloom) consisting of metal mixed with coal and slag, the latter mostly composed of iron oxide and fayalite. The bloom was heated and hammered repeatedly to release the trapped slag: in this way wrought iron was produced. A mass of soft iron was obtained, scattered by micro-slag in the inside, which was then subjected to additional cycles of heating and hammering. To obtain a higher mechanical strength, the iron was subsequently carburated with the enrichment of carbon, through prolonged contact

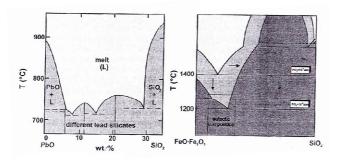
with the coal in the forge. This process is called cementation. This brief excursus has been reported to enlight which were the pyrometallurgical phases that produced slag during iron processing, which could came from the furnace or forging process (Giardino, 2002).

Since the basic composition of the slag coming from the smelting of different types of ore is often similar, it is very helpful to look for the microparticles of metal in the slag, which often remain trapped in the structure of the slag, even if the extraction process is highly efficient.

For example the presence of tin, copper and copper-tin may instead indicates the presence of a slag obtained by a technologically sophisticated process of bronze production, defined co-smelting.

The presence of microparticles of lead or litharge, accompanied by the presence of silver, may indicate respectively the smelting of lead-based ore and the cupellation of galena for the obtaining of silver. About the early lead-silver technology a short digression must be

Fig. 9: systems SiO₂ PbO and SiO₂ Feoxide (taken from Hauptman 2007)



made for the deliberate adding of quartz to the liquid slags. Sometimes this addition is useful to cool down the slag, in order to facilitate the removal of the agglomerate to the molten metals. It is important to understand the reactions that take place between different oxides and with quarts,

because the results change in relation to the metal involved in it. In the case of SiO_2 and PbO, a small amount of lead oxides and alkali oxides is sufficient to dissolve SiO_2 and to lower liquidus/solidus temperature and viscosity (fig.9).

In the furnace this lead to a too early formation of slags, which anticipates the precipitation of metal from ore. In this case the addition of quartz can stop this premature fusion of the charge. In the other case FeO and other divalent oxides (CaO, MgO and MnO) which occur in slag of copper metallurgy, have a lesser ability to form a melt with SiO₂, due to their higher liquidus-solidus temperature. In these conditions a further addition of SiO₂ would lead a rise in liquid/solid temperature, that causes a solidification of the molten materials in the furnace (as the saying goes in the technical jargon: to freeze a furnace). This is why the adding of quartz is understandable in the lead processing and not for example in the copper smelting. In the light of this, in many lead slags, quartz is well distinguishable in the matrix

but it is less easy to understand if it comes from the original charge or have been added afterwards.

These are the basic guidelines to make sensate interpretations in the analysis of this complex material; in the next paragraph the examination of the compositional and structural aspects of our samples.

5.4 Experimental

The study of metallurgical slags from the point of view of the characterization of the composition, but also the interpretation of the process that created them, shows many elements of complexity.

As explained previously, these consist of fine-grained mixture of many different phases. The use of the optical microscope is one of the classical means in this kind of archaeometric research, but sometimes it is difficult to determine mineralogical phases with exception to main components such as olivine, spinel and pyroxene. X-ray diffraction (XRD) is useful to recognize the mineral nature of the slags, except if the solidification happened in a glassy state and a transition from the microcrystalline, *via* the cryptocrystalline up to the hyalocrystalline state take place. It this case XRD cannot detect the phases which constitute the slag, making futile the analysis. Scanning electron microscopy and the microprobe analysis (SEM-EDS) is more suitable for the study of the slags, because it allows both to understand the structure of the sample and provides punctual composition of the phases encountered.

For the investigation of Aouam and Volubilis slags it was decided to use SEM-EDS (LEO 1450 VP) and XRD (Miniflex Rigaku) and cross the data obtained by these two methods, in order to have as much information as possible. The SEM backscattered electron images have proved particularly helpful because allowed us to identify the metallic, glassy and crystalline phases in the samples. In addition to it a future development of this work will be the submission of slags samples to Inductively Coupled Plasma-Mass Spectrometry analysis (ICP-MS) to obtain the isotopic ratio of lead. This data will be compared with the MS analysis of metal artefacts from Volubilis (already done with TIMS instrumentation-Thermal Ionization Mass Spectrometry, for one lead fragment) and the matching or, on the

contrary, the discrepancy between the two results will allow to state if the metals have Moroccan origin or whether they were traded with other country.

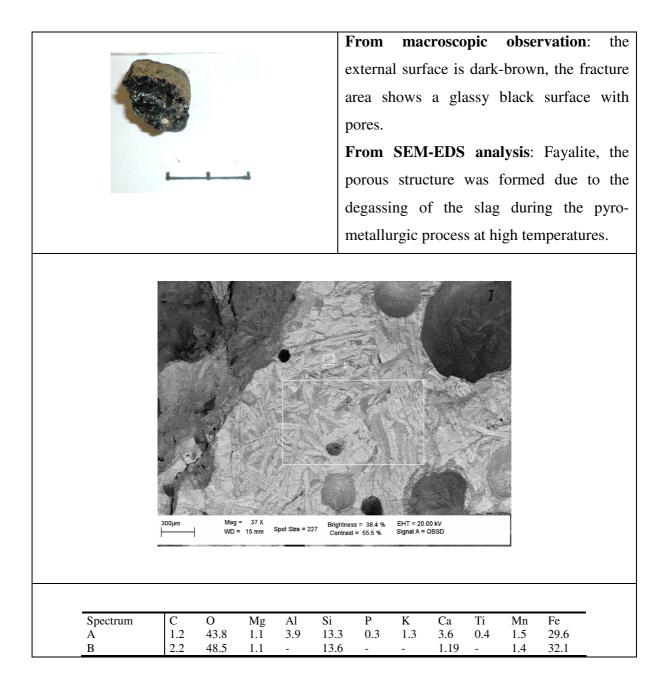
5.5 Results

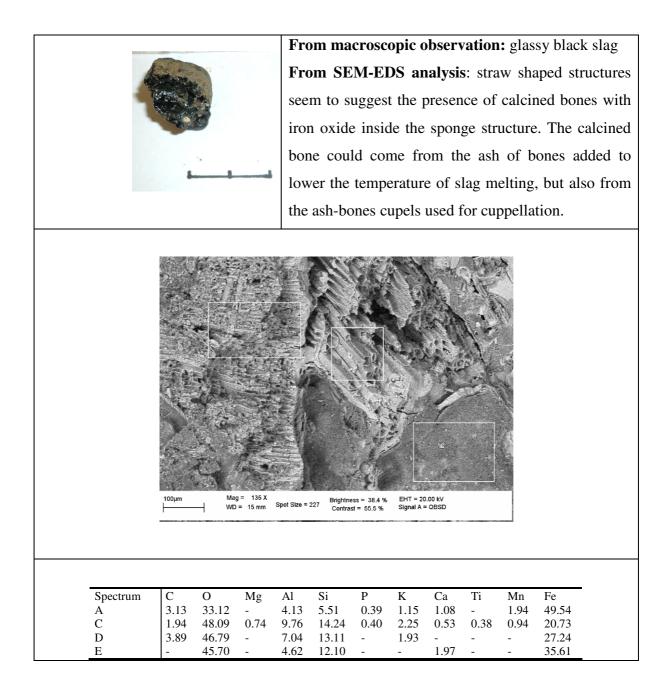
Several fragments of slags were collected from Aouam mine(uncertain dating) and Volubilis site, both small and massive in size. While Aouam slags came for superficial prospection, for Volubilis slags a precise stratigraphic chronology is available (in the table below):

N°	Object	Stratigraphic unit	Localization	Cronology
Vol.03.4198	Slag	46	Ch. 2- Esp.1	II century
Vol.02.4824	Slag	14	Ch. 1-Esp. 2	III century
Vol.03.5606	Slag	00	Ch. 4	150-250

These slags come from a stratigraphic layer belonging to Roman presence in Volubilis. We decided to report the results since is testified that Roman conquerors continued to exploit mineral deposits previously localized by the Punics.

Even macroscopically can be distinguished different types in dimension, textures and colour, and with the aid of SEM-EDS has been performed a classification. The discussion will begin with Aouam samples because their higher number allows to discuss various aspects. Each image of the samples shows the external appearance of the slag and the fracture surface, which allows observation of the slag inside. All EDS results in the tables are intended as weight %.





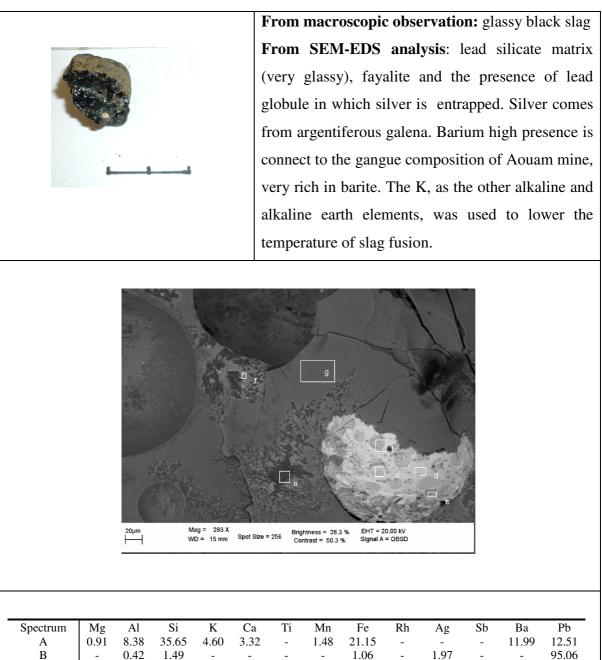
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0.92

1.43

3.71

9.69

13.59

1.64

0.65

0.93

_

0.88

8.28

43.65

1.46

8.34

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-

0.51

2.03

0.97

2.75

1.47

1.72

6.29

14.14

30.36

0.38

0.49

1.47

2.21

5.96

0.80

3/3

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-

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6.86

87.49

96.37

35.88

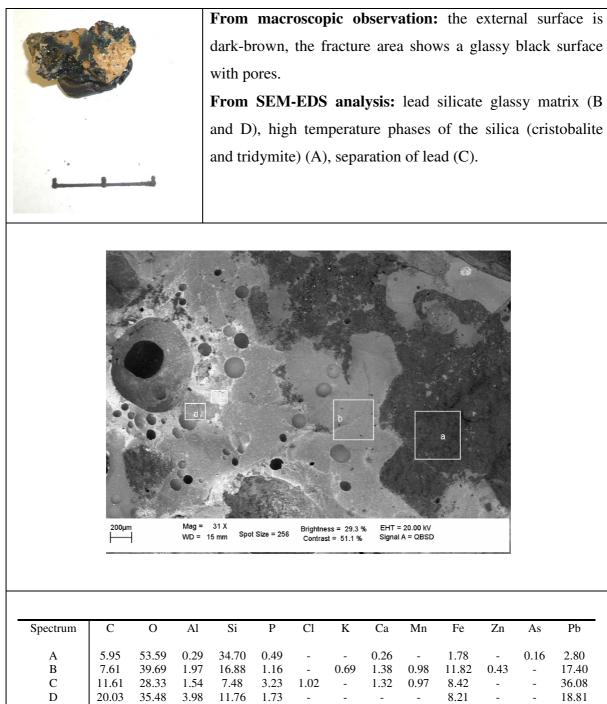
69.91

36.78



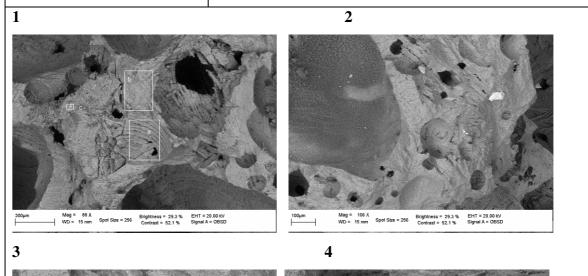
_						SEN	1-ED	S ar	nalys	sis: th	ne m	atrix	is co	ompo	osed	of lead
	Side the					silica	ate v	vith	glass	sy sti	ructu	re. T	he c	lark	com	pounds
							d be	clir	юру	roxen	e (a	ugite	or	hede	rberg	gite) in
		9				whic	ch the	e pre	senc	e of	calci	um p	hosp	hate	is de	etected,
		-						-				-	-			nace or
								-	pre	sence	01 a					
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1									2							
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	1.96 K X 15 rum Spot		htness = 30.8			******	2µm	ł	Mag = 3.34 WD = 15 mm			1ess = 30.8 %, asl = 49.3 %	EHT = 20.00 Signal A = Of		13	
						Si	²µm ⊢ P	K		π Spot Size =					Ba	Pb
1	15 mm Spot	Size = 297 Co	ntrasi = 49.3 %	ຊິ Signal A	= QBSD	Si 14.8	<u>2µт</u> Р 1.0	K 0.8	1/WD = 15 mm	n Spot Size =	297 Contra	asi = 49,3 %	Signal A = OE	BSD	Ba 1.5	Pb 22.7
1 Spectrum A B	C 3.3 4.0	O 40.8 45.7	ntrasi = 49.3 %	Mg 0.4 0.5	Al 3.0 2.1	14.8 9.9	1.0 6.1	0.8 0.6	Ca 2.2 8.9	Ti - 0.4	Mn 0.8 0.6	Fe 8.2 5.3	Zn 0.4	BSD	1.5 -	22.7 15.4
1 Spectrum A	C 3.3	O 40.8	ntrast = 49.3 ° Na -	Mg 0.4	Al 3.0	14.8	1.0	0.8	WD ⁼ 15 nm Ca 2.2	Ti - 0.4 0.2	Mn 0.8	sst = 49.3 % Fe 8.2	Signal A = OF	BSD		22.7
1 Spectrum A B C	C 3.3 4.0 5.2	O 40.8 45.7 39.5	Na - 0.5 -	Mg 0.4 0.5	Al 3.0 2.1 3.1	14.8 9.9 13.9	1.0 6.1 4.4	0.8 0.6 1.6	Ca 2.2 8.9 7.5	Ti - 0.4 0.2	Mn 0.8 0.6 1.2	Fe 8.2 5.3 11.8	Zn 0.4 - 0.4	As - -	1.5 -	22.7 15.4 8.8
1 Spectrum A B C	C 3.3 4.0 5.2	O 40.8 45.7 39.5	Na - 0.5 -	Mg 0.4 0.5	Al 3.0 2.1 3.1 2.5	14.8 9.9 13.9	1.0 6.1 4.4	0.8 0.6 1.6	Ca 2.2 8.9 7.5	Ti - 0.4 0.2	Mn 0.8 0.6 1.2	Fe 8.2 5.3 11.8 6.9	Zn 0.4 - 0.4	As - - 0.5	1.5 - 1.8 -	22.7 15.4 8.8
1 Spectrum A B C D 2 Spectrum dark	C 3.3 4.0 5.2 4.3 C 2.7	O 40.8 45.7 39.5 40.2 O 46.0	Na - 0.5 - 0.3 Na 0.3	Mg 0.4 0.5 - Mg 0.5	Al 3.0 2.1 3.1 2.5 Al 2.5	14.8 9.9 13.9 12.8 Si 10.7	1.0 6.1 4.4 1.4 P 5.4	0.8 0.6 1.6 0.4 K 0.7	Ca 2.2 8.9 7.5 1.8 Ca 8.1	Ti - 0.4 0.2 - Ti 0.2	Mn 0.8 0.6 1.2 0.6 Mn 2 0.6	Fe 8.2 5.3 11.8 6.9 n Fe 5.7	Zn 0.4 - 0.6 Zn 0.2	As - - 0.5 Ba	1.5 - 1.8 - Pb 16.	22.7 15.4 8.8 27.9
1 Spectrum A B C D 2 Spectrum dark Light gray	C 3.3 4.0 5.2 4.3 C 2.7 -	O 40.8 45.7 39.5 40.2 O 46.0 42.6	Na - 0.5 - 0.3 Na 0.3 -	Mg 0.4 0.5 0.5 - Mg 0.5 0.4	Al 3.0 2.1 3.1 2.5 Al 2.5 3.1	14.8 9.9 13.9 12.8 Si 10.7 15.6	1.0 6.1 4.4 1.4 P 5.4 0.8	0.8 0.6 1.6 0.4 K 0.7 0.9	Ca 2.2 8.9 7.5 1.8 Ca 8.1 2.3	Ti - 0.4 0.2 - Ti 0.2	Mn 0.8 0.6 1.2 0.6 Mn 2 0.6	Fe 8.2 5.3 11.8 6.9 n Fe 5.7 3 8.2	Zn 0.4 - 0.4 0.6 Zn 0.2 0.5	As - - 0.5 Ba - 1.4	1.5 - 1.8 - Pb 16. 23.	22.7 15.4 8.8 27.9
1 Spectrum A B C D 2 2 Spectrum dark Light gray dark	C 3.3 4.0 5.2 4.3 C 2.7 - 2.5	O 40.8 45.7 39.5 40.2 O 46.0 42.6 46.8	Na - 0.5 - 0.3 Na 0.3 - 0.6	Mg 0.4 0.5 0.5 - Mg 0.5 0.4 0.6	Al 3.0 2.1 3.1 2.5 Al 2.5 3.1 2.0	14.8 9.9 13.9 12.8 Si 10.7 15.6 8.6	1.0 6.1 4.4 1.4 P 5.4 0.8 7.6	0.8 0.6 1.6 0.4 K 0.7 0.9 0.6	Ca 2.2 8.9 7.5 1.8 Ca 8.1 2.3 11.3	Ti - 0.4 0.2 - Ti 0.2 5 0.2	Mn 0.8 0.6 1.2 0.6 Mn 2 0.6 0.8 2 0.6	Fe 8.2 5.3 11.8 6.9 n Fe 5.7 3 8.2 5 4.9	Zn 0.4 - 0.4 0.6 Zn 0.2 0.5 -	As - - 0.5 Ba - 1.4	1.5 - 1.8 - Pb 16. 23. 13.	22.7 15.4 8.8 27.9
1 Spectrum A B C D 2 Spectrum dark Light gray	C 3.3 4.0 5.2 4.3 C 2.7 -	O 40.8 45.7 39.5 40.2 O 46.0 42.6 46.8	Na - 0.5 - 0.3 Na 0.3 -	Mg 0.4 0.5 0.5 - Mg 0.5 0.4	Al 3.0 2.1 3.1 2.5 Al 2.5 3.1	14.8 9.9 13.9 12.8 Si 10.7 15.6	1.0 6.1 4.4 1.4 P 5.4 0.8 7.6 6.6	0.8 0.6 1.6 0.4 K 0.7 0.9	Ca 2.2 8.9 7.5 1.8 Ca 8.1 2.3	Ti - 0.4 0.2 - Ti 0.2 5 0.2	Mn 0.8 0.6 1.2 0.6 Mn 2 0.6 0.8 2 0.5 0.4	Fe 8.2 5.3 11.8 6.9 n Fe 5.7 3 8.2 5 4.9 4 4.8	Zn 0.4 - 0.4 0.6 Zn 0.2 0.5 -	As - - 0.5 Ba - 1.4 - 0.8	1.5 - 1.8 - Pb 16. 23. 13.	22.7 15.4 8.8 27.9

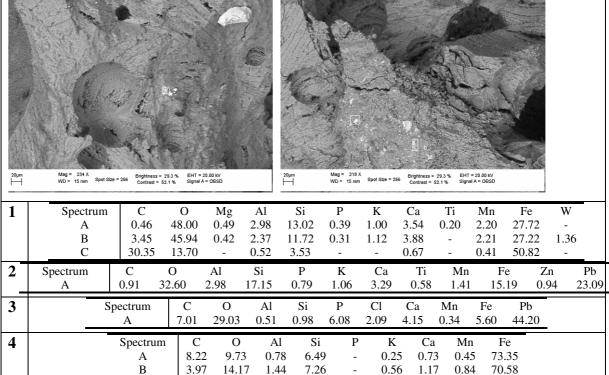






Macroscopic observation: brown-reddish external surface, the fracture area shows a black and highly porous surface **SEM-EDS analysis:** the slag is totally composed by a crystallization of fayalite, where microparticles of Pb are easily found. Fe microparticles indicate a very strong reducing atmosphere in the furnace.





0.37

1.89

2.60

1.00

52.07

5.23

23.38

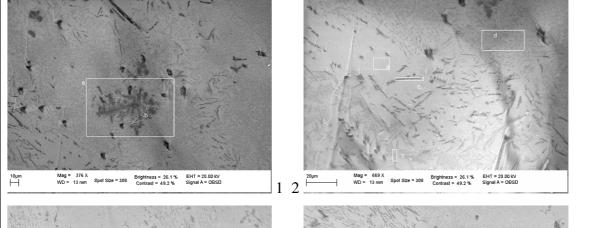
3.05

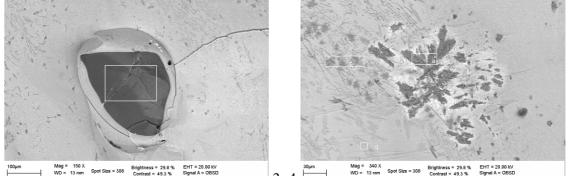
10.41

С

Macroscopical observation: the slag is black and highly vitrified.

SEM-EDS analysis: lead silicate matrix and fayalite with microparticles of metallic lead. The dashes-shaped structure seems to be calcium phosphates. Quartz hadn't react during the pyro-metallurgical cycle and remained unaltered, surrounded by glassy phase.





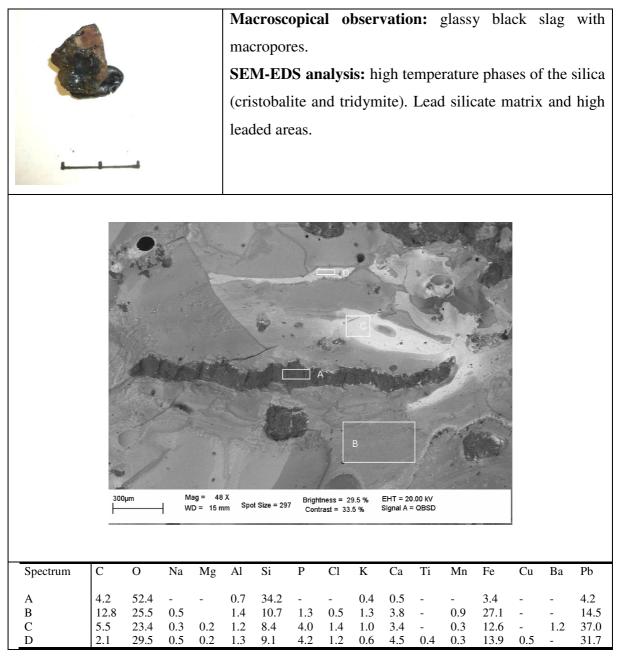
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		5	т	Management				No. of Concession, Name			

1	Spectrum A	C 2.8	0 8 41.4	Mg 49 0.3	·	Si 14.4	P 1 1.8	K 30 0.9	Ca 01 3.7		Mn 2 0.88	Fe 3 9.70	Zn 0.33	Pb 3 20.44
	В	8.4	2 32.2	25 0.4	1 2.65	14.5	0 1.6	50 1.0	9 4.0	0.4	5 0.94	4 11.7	1 0.34	4 21.64
2	Spectrum A B C D Spectrum	Mg 0.71 0.90 - 0.70	Al 5.99 5.39 5.52 5.67	Si 29.95 28.81 28.83 29.35	P 1.20 3.49 3.31 2.94 Fe	K 1.41 1.41 1.69 1.69	Ca 3.50 5.33 5.81 5.52	Ti 0.59 - -	Mn 1.05 1.07 0.98 1.27	Fe 9.55 8.42 8.48 8.71	Zn 0.22 0.11 -0.16 -0.06	Ba - 1.73 2.60 -	Pb 45.84 43.35 42.94 44.22	
	A	2.9	-			.4								
4	Spectrum	Na	Mg	Al	Si	Р	K	Ca	Ti	Mn	Fe	Zn	Ba	Pb
	A	0.65	0.86	7.64	32.43	2.52	3.76	7.01	-	1.94	18.87	0.67	5.27	18.38
	В	-	-	1.01	6.89	7.05	-	15.61	-	0.59	6.24	-	-	62.62
	С	0.63	0.92	4.68	25.36	6.44	2.04	11.92	0.71	1.21	14.40	0.74	-	30.95
	D	2.28	0.41	5.56	28.14	4.64	2.14	9.33	0.60	0.81	8.69	-	-	37.38

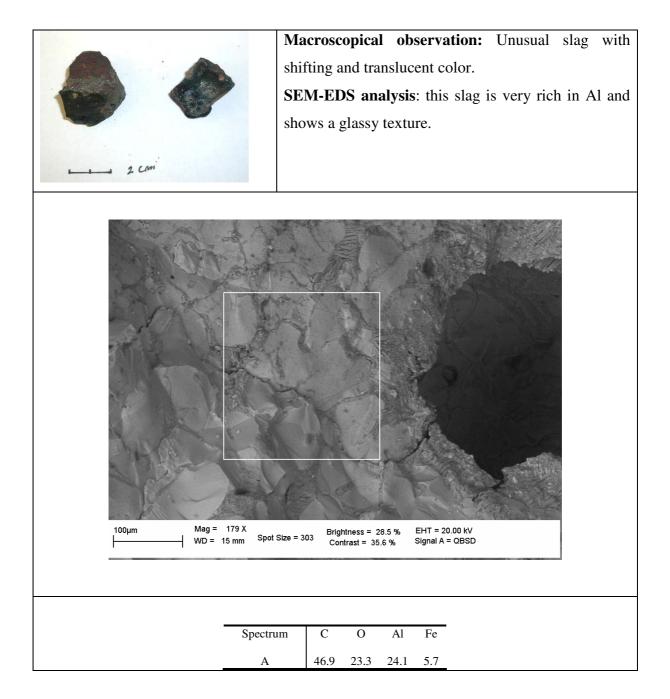
Diag	5.0															
	Macroscopic observation: the slag is highly vitrified and nextensively porous. SEM-EDS analysis: lead silicate matrix and large globule lead. Silver microparticles are detected.															
100µm	Mag = 118X WD = 15mm Spa	1 Size = 237 Eng	6 6 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	EhT = 20. Signal A = 1	00 KV GBSD			00µm	Mg = 67 WD = 15 m	A A] = 297 Brig Co	phiness = 29.3 Intras = 32.9	B % Eht = 20.6 Signal A = 1			
1	Spectrum	Mg .	Al S	Si I	Р	K	Ca	Mn	Fe	Cu	As	Ag	Sb	Ba	Tl	Pb
	A B C		3.1 2	1.7	9.6		- 4.5 6.9	- 0.5 -	0.6 20.0 7.6	1.9 - -	1.2 - -	1.0 - -	12.7 - -	- 2.2 -	1.8 - -	79.5 40.4 69.1
2	Spectrum	C	0	Mg	Al	Si	Р	Cl	K	Ca	Ti	Mn	Fe	Zn	Pb	
	A B C	11.8 6.0 3.6	49.8 28.1 36.8	- 0.2 0.5	2.1 0.9 2.3	11.9 5.4 17.7	5.5	5 0.9	0.4 0.2 0.9	1.3 4.7 2.2	- 0.3	- - 0.4	7.5 5.4 16.8	- 0.8 0.5	14.0 41.8 16.7	

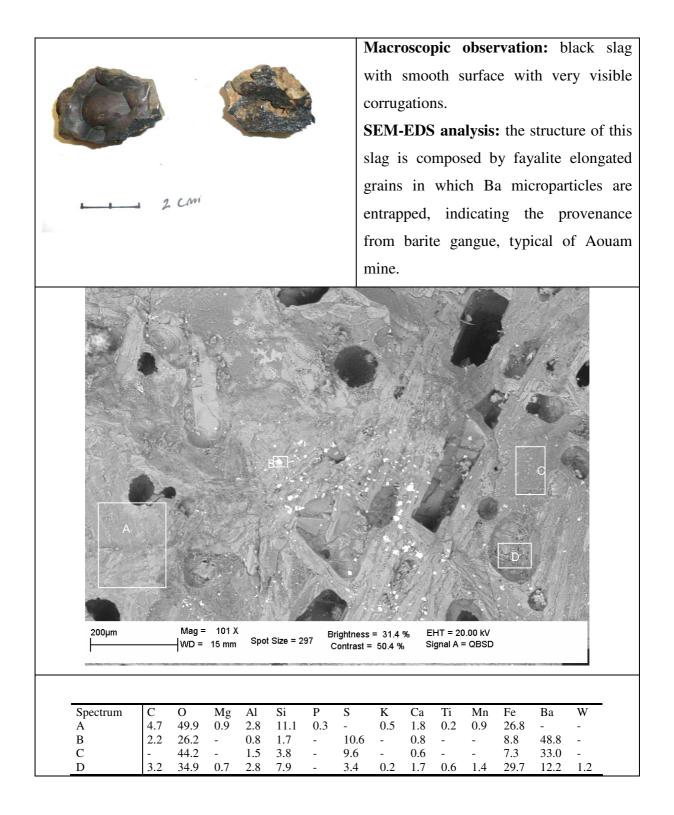
Slag B



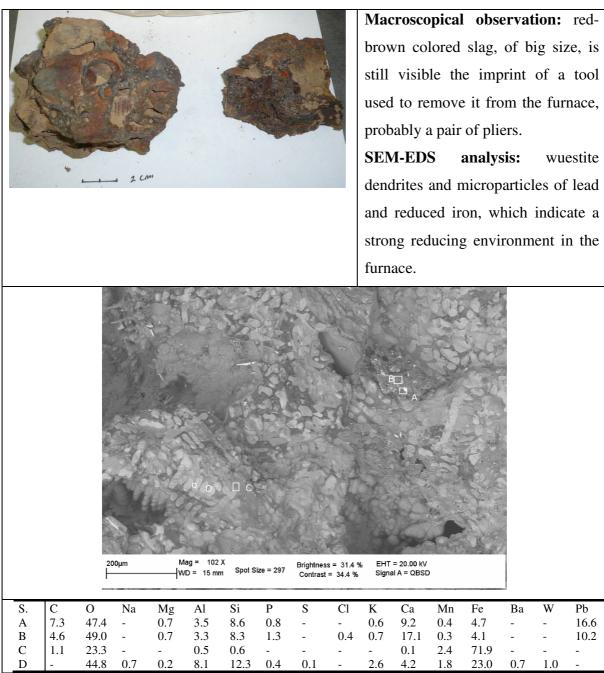












Macroscopical observation: this slag is rusty-red colored, also on the fracture surface (in the inside). SEM-EDS analysis: Iron oxides and iron microparticles (n.3). Large crystals of quartz are detected, which could suggest an intentional addition during pyro-metallurgical process or the presence of quartz in the embedding rock, or incorporation of fragments of furnace hearth. Beside these big, unreacted crystals, the matrix is composed of iron oxides, which have been identified with the aim of XRD analyses Mag = 51 X WD = 15 mm ess = 52.5 % ast = 50.8 % Brightness = 52.5 % Contrast = 50.8 % EHT = 20.00 kV Signal A = OBSE Spoi Size = 256 Brig Spot Size = 256 2 Mag = 99 X WD = 15 mm Spot Size = 256 Brightness = 52.5 % Contrast = 50.8 % EHT = 20.00 kV Signal A = OBSD 3 Fe 1 Spectrum С 0 Mg Al Si Р Κ Ca 4.67 60.42 0.20 0.39 27.10 0.50 6.72 А В 3.51 43.36 0.49 1.04 4.17 1.10 0.19 1.87 44.27 2.97 C 49.86 0.76 0.84 4.32 0.86 1.28 39.11 2 Spectrum Ca С 0 Mg Al Si Р Fe 3.99 42.90 0.49 0.31 2.14 0.65 2.24 47.28 3 Spectrum С 0 Mg Al Si Р Ca Fe White particle 6.11 40.20 0.75 0.41 2.16 0.65 0.72 49.01

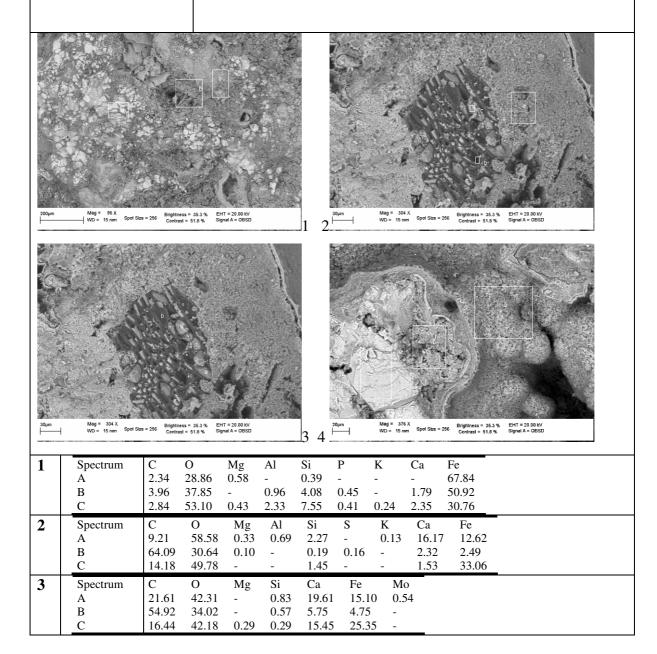
Vol 03 5606



Macroscopic observation: this slag have a rusty-red surface and inside shows black and brilliant spots.

SEM-EDS analysis: Iron microparticles and iron oxide matrix. In the 2 and 3 images fragments of carbon, entrapping iron microparticles, can be seen. The detection of a noteworthy quantity of calcite can suggest a calcite gangue of the mineral of provenance or the addition of limestone

The presence of Mg and K could suggest the addition of lowmelting compounds or the incorporation of hearth particles



4	Spectrum	С	0	Mg	Si	Р	Ca	Fe	Br
	А	-	25.61	-	2.15	0.33	1.35	70.56	-
	В	2.87	49.06	-	1.73	0.42	1.51	43.94	0.48
	С	1.90	34.81	0.62	0.50	-	5.09	57.08	-

Vol 034198

			24		159		Μ	acros	copic	obser	vation	: the	slag	is	
03					The of		re	d-brov	vn in t	the ou	tside b	ut the	e fractur	re	
	6 Section						su	surface shows a blackish colour.							
					2	SI	SEM-EDS analysis : this slag is								
						co	mpose	ed mo	ostly	by irc	on ox	kide an	nd		
	-	-1				ca	lcite a	nd in 1	the im	age n.1	, in tl	he area	E		
							al	large s	ilica ir	nclusio	on is ag	ain de	etected.		
						A	gain, a	s in th	ne prev	vious s	lag, f	ragmen	ts		
						of	carbo	n can l	be dete	ected.					
	00µm Mag 98 X ₩20 15 mm \$9	o of Size = 337 Efficiency of Size = 337	Annes = 33.8 % End	()))))))))))))))))))	And 2 Jul 2013	2	200m Ma	ā = 62 X = 55 m Spel 1	b Stee = 337 Briefer	Pets = 33.8% EHT stat = 50.9% Sens	= 20.00 kY (A= OBS) Du	ne 2 Jul 2013			
1	Spectrum A	C 4.2	O 48.6	Na -	Mg 0.5	Al -	Si 0.8	Р -	S -	Ca 2.1	Fe 43.2	Co 0.1	Br 0.5		
	B C	12.6 9.1	51.3 54.1	- 0.4	0.8 0.9	0.3 0.4	0.6 0.8	0.2 0.2	0.3 0.1	19.3 7.2	14.6 26.8	- -	-		
2	D Spectrum	4.2 C	55.2 O	- Mg	- Al	0.3 Si	37.2 P	- Ca	- Fe	0.8	2.3	-	-		
	A B	25.7 31.1	33.4 35.7	0.8 0.3	0.6 0.3	0.6 0.6		11.1 8.4	28.4 23.6						
	Б С D	53.9 60.8	31.2 20.3	0.3 0.6 0.3	0.3 0.3 0.3	0.0 0.2 0.4	0.1	8.4 8.6 3.7	23.0 5.1 14.2						
1	U	00.0	20.3	0.3	0.3	0.4		5.1	14.4	_					

XRD analysis

XRD analysis have been conducted on six slags, grounded with an agate mortar, both from Aouam and Volubilis, in order to confirm some results obtained with SEM-EDS. The samples underwent an XRD analysis using an X rays diffractometer Miniflex Rigaku equipped with a Cobalt tube, operative conditions: voltage of 30 kV and an electric current of 15 mA. Counting time is of about 3600 seconds to allow a good statistic.

The following table shows the results summarize the results for slag n.1, 2 and 3 from Aouam and for the whole set of slags from Volubilis, Vol 02 4824, Vol 034198 and Vol 03 5606 (spectra in figs. 10-15):

Sample N.	Quartz	Calcite	Goethite	Hematite	Magnetite/ Maghemite	Fayalite	Other compounds
Vol 03 4198	±		+++	+++	+	±	
Vol 02 4824	+	+++	+	++	+	<u>+</u>	Wustite
Vol 035606	+	++	++	++	<u>+</u>	+	Lepidocrocit e Wustite
Aouam 1	+++						
Aouam 2	+++						
Aouam 3					<u>+</u>	+++	

Legend: +++ abundant; ++ moderately abundant; + low abundance; + scarce/trace level

SEM-EDS and XRD results are matching well, in fact Volubilis slags are composed of different iron oxides, connecting this kind of slags to steps of iron pyro-metallurgy. No fayalite or a very scarce presence of them is underlined by both SEM and XRD on Volubilis slags. For the Aouam slags glassy matrix causes problem in the XRD instrumental response, that is not able to distinguish vitreous phases in the sample. On the other hand, this lack of response confirms the high presence of amorphous silicates of lead. In Aouam 3 the XRD detect a huge quantity of fayalite, confirmed by the SEM-EDS analysis. Fig.10: XRD spectrum Aouam 1 (q: quartz); other compounds found in SEM-EDS analysis are not detected with XRD because of amorphous nature of lead silicate matrix and cristobalite and trydimite.

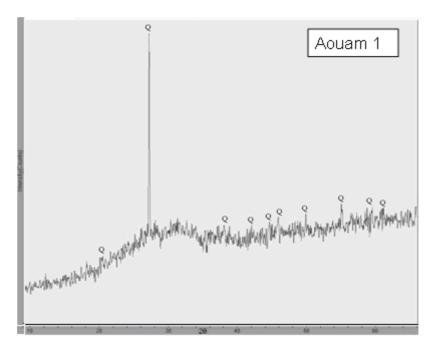


Fig.11: XRD spectrum Aouam 2 (q: quartz); other compounds found in SEM-EDS analysis are not detected with XRD because of amorphous nature of lead silicate matrix and cristobalite and trydimite.

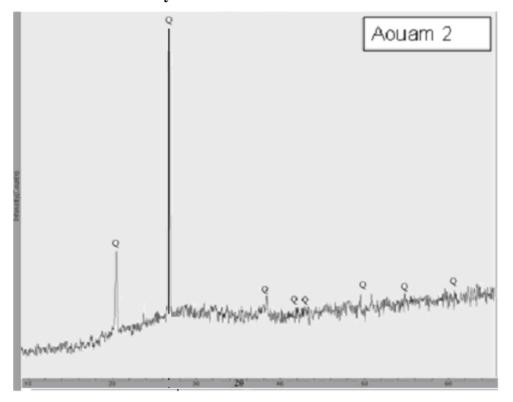


Fig.12: XRD spectrum Aouam 3 (fay: fayalite, ma: maghemite)

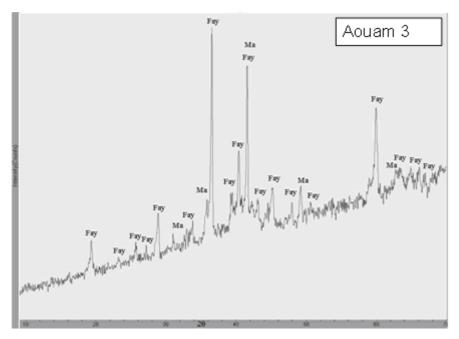


Fig.13 XRD spectrum Volubilis n. 4198 (Q: quartz, Goe – goethite, Hem – hematite, Fay – fayalite, Ma - maghemite

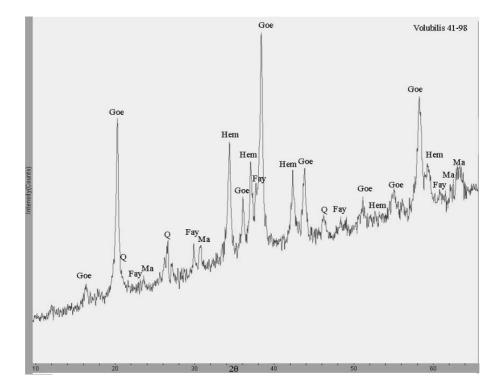


Fig.14: XRD spectrum Volubilis 48-24 (Q- quartz Ca – calcite Goe – goethyte, He – Hematite, Fay – fayalite, Wu – wustite)

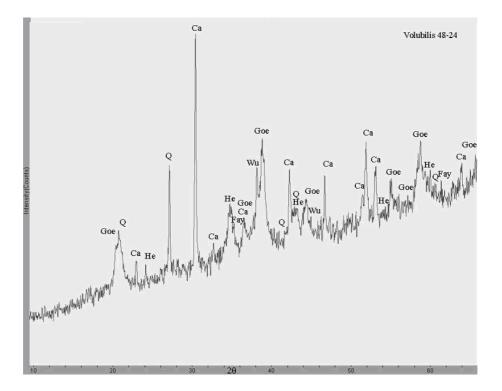
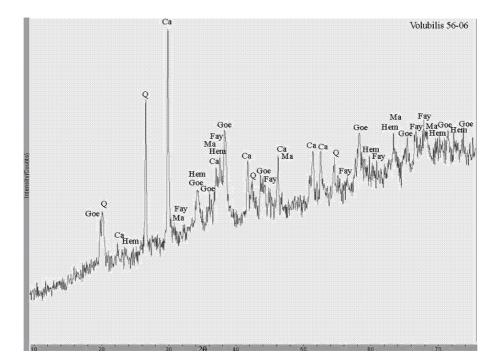


Fig. 15: XRD spectrum Volubilis 56-06 (q: quartz, Ca – calcite, Goe – goethite, Hem – hematite, Fay – fayalite, Ma - maghemite



5.5.1 Results on the slags

The introductory paragraph on slag production provides some basic parameters for the interpretation of analytical results obtained from Volubilis samples. Currently, given the small number of samples, and the necessity to obtain other evidence of arcaheological context (presence of crucible, tuyeres, minerals etc...) we can suggest some possible interpretations. From the evidences that emerged as result of SEM-EDS and XRD analysis it is established that these slags are belonging to two different types, from different metallurgical process and from different minerals.

Aouam slags were produced during the smelting of argentiferous galena extracted from the mine, as further confirmed by the presence in many samples with high level of barium, coming from barite gangue of which is composed the embedding rocks in this area. The big dimension and the composition suggest that the slags were coming from a furnace and not from crucible operations. Furthermore another information that can be inferred form the analysis is the high reducing atmosphere of the furnace. The presence of pyrite and chalcopyrite in the gangue, as in the case of Aouam, favored a slagging of the mineral without the need for addition of fluxing agents. The presence however of grains of silica, which remain unaltered during the pyrometallurgic process, indicates the addition of this agent to prevent it from melting, before the metal separates entirely from the slag (probably free-silica slag type). The presence of bone fragments however may indicate a deliberate addition of bone ash in order to further lower the point of slagging.

With regard to the possibility that cupellation were executed on the site, there are various clues in this direction (such as the presence of silver particles and ash in some slag may indicate that the execution of the process in the cupels produced with the animal ash-bone) but they need to be further confirmed by other prospection in this area.

Volubilis slags, on the other side, are different in composition and size. The composition, quite unusual, could link this type of waste product to a metallurgical operation in the pyrometallurgical cycle of iron processing.

Volubilis slags are characterized by the presence of iron oxides such as hematite, magnetite, maghemite, wustite, iron hydroxides like goethite and lepidocrocite, iron silicate like fayalite (few), whose presence could suggets a connection of this type of waste product to pyrometallurgical or mechanical operation of iron (roasting and forging phase). Also the

macroscopic appearance, with the external surface of ferrous-red color and the fracture surface of a darker color and with slight metallic luster, could suggests this hypothesis (Ingo et al. 1995). Is difficult that these slag belong to a smelting process since the production of fayalite is more scarce than the presence of oxides and idroxides. The presence of alkali, alkaline earth metals and phosphorus (for example by the addition of ashes or ash of bones), may indicate the addition of low melting fluxes, which allowed to conduct the pyrometallurgic process at lower temperatures (up to 150 $^{\circ}$ C in less).

The presence of metallic iron microparticles also seems to indicate a pyrometallurgical operation starting from iron ore. The iron particles allow to suggest this hypothesis. Indeed, as explained in the introductory paragraph, slag rich in iron oxides may also be produced during the cycle of transformation of the minerals of copper or lead, in order to remove the component of the silicate mineral. In particular, the presence of oxides and hydroxides of iron more than fayalite, could also suggest metallurgical operations of non ferrous ore carried out in crucibles with non-reducing atmospheres. Since we didn't find any copper or lead metallic micro-particles are we are inclined more to consider a process related to iron - based minerals. As the wüstite, present in two of the analyzed slag, is stable above 560 $^{\circ}$ C, it is possible to imagine that the cooling has been fast in these two cases. If the cooling is slow the wüstite decomposes into iron and magnetite (this maybe could explain the absence of it in the first slag, n. 034198).

In conclusions it could be hypothesized that the three slags could be produced during: roasting of the iron ore or during forging phase. Smelting of iron ore is less probable because of a lesser presence of fayalite or other silicate phases. The presence of charcoal suggests that these fragments are not simply ores but came from a metallurgical operation of processing.

These considerations, observed from a generic and broader point of view, and the small sizes and quantity of slags found here, may suggest that the big-scale smelting operations of large quantities of materials were carried out in the vicinity of the mine, while smaller scale operations, as the operations of processing, refining, forging or production of objects, but also of smelting of minor amounts of minerals were also carried out inside urban centers.

The presence of calcite and quartz grains in the slag of Volubilis could give an indication of provenance for the mineral worked here. To state with certainty, as it will be exposed subsequently, which was the supplying area of Volubilis it is necessary to proceed with isotopic analyzes of both slags and metal products from Volubilis.

Both types of slag are consistent with production techniques that were applied during Bronze Age and Iron Age. Is therefore obtained a *terminus post quem* for the production of these slags for Aouam slags coming from superficial survey. Otherwise it is important to take into account that metallurgical production didn't evolve much from this period to the later periods (until Middle Age), so we will need to perform additional exploration in these areas, to obtain a *terminus ante quem* and to produce results related to a narrower period. To state with certainty which was the supplying area of Volubilis, it is necessary to proceed with isotopic analyzes of both slags and metal products from Volubilis; and some firsts interesting results are shown in the dedicated paragraph.

5.6 Metal artefacts from Volubilis archaeological stratigraphy

Form Volubilis archaeological settlement a considerable number of metal artefacts has been discovered during archaeological excavation. Some of these artefacts have been analysed in a way to understand which was the possible connection between the artefacts and the slags. In the table below are listed all the metal specimens under study, followed by the description given by the archaeologists who collected them:

- Vol Y: lead fragment;
- Vol X: lead fragment;
- Vol 05 1051: lead fragment;
- Vol 03 5866: lead fragment;
- Vol 05 6719: lead fragment;
- Vol 03 3182: lead fragment;
- Vol 05 1054: copper alloy fragment;
- Vol 05 7085: copper alloy fragment;
- Vol 03 4197: copper alloy fragment;
- Vol 02 4404: iron fragment;
- Vol 02 3168: iron fragment.

In the table below are summarized the stratigraphic information about the specimens under study. Furthermore in the column named *object* is also described the artefact to which the sample was part. After having carried out the analyses on the findings, the dating has been

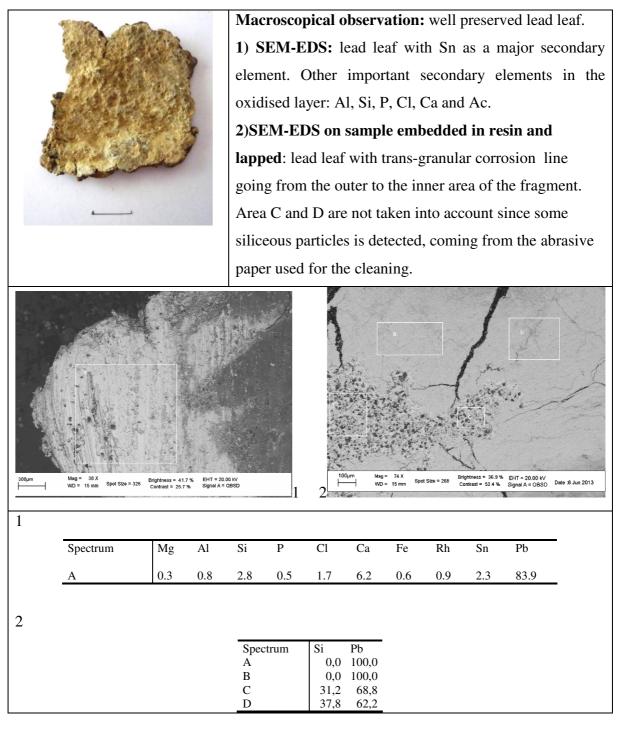
N°	Object	Stratigraphic unit	Localization	Cronology
Vol.02.3168	Sickle	00	Ch. 2	Before VIII century
Vol.02.4404	Ring	01	Ch. 1- Esp.1	IV century
Vol.03.3182	Plaque	33	Ch. 3-P. 3-4	150-250
Vol.03.4197	Plaque	46	Ch. 2- Esp.1	II century
Vol.03.5866		07	Ch. 4	IV century
Vol.05.1051		62	Ch. 2- Esp.3	150-250
Vol.05.1054	Plaque	62	Ch. 2- Esp.3	150-250
Vol.05.7085	Plaque	98	Ch. 1- Esp.6	III century
Vol 05.6719				150-250

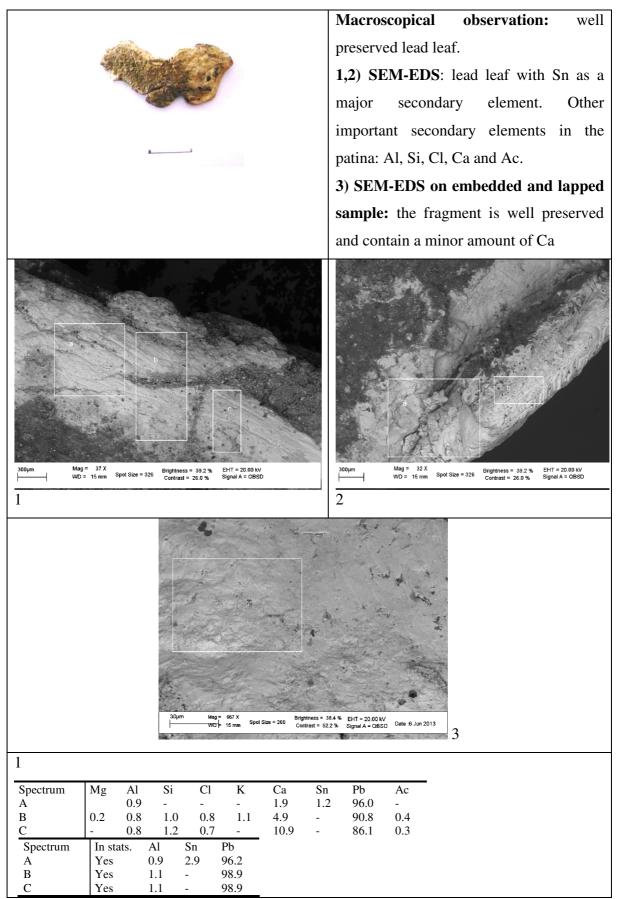
defined as belonging to the Roman and post-Roman occupation of Volubilis, then posterior to the Punic settlement. However it has been decided to report the analytical results because, as also happens in the Iberian Peninsula, Roman settlements replaced the Punic ones as a result of the conquests, but the populations maintained, often for centuries, a cultural continuity with their Punic origins, sometimes also in the manufacturing. In this light, the preliminary study of these artefacts can give also interesting information on previous Punic occupational phase. As the collaborative project is currently in its preliminar phase, we are proceeding with the research and the demand for manufactured goods of certainly Punic origin, both from the site of Volubilis (pre-roman occupation) and the burial mounds of Tayadirt (Lambert 1968).

All the artefacts have been analysed on a cleaned area to obtain images and analytical tables which correspond to the original alloy, without taking into consideration the oxides patina.

The major part of the samples have been also embedded in resin and lapped to provide as much as possible a flat surface. On the copper alloys an etching attack with a ferric chloride solution (in ethanol and hydrochloric acid) has been performed to indentify the phases in the alloy. That's way in some of the analysis Cl shows very high percentage (an overestimation). When the sample analysed sample still contain some amount of external patina the table contains all the detected element. When the sample has been highly cleaned or embedded in resin and then lapped the results has been normalized reoving C and O. All EDS results in tables are expressed as weight %.

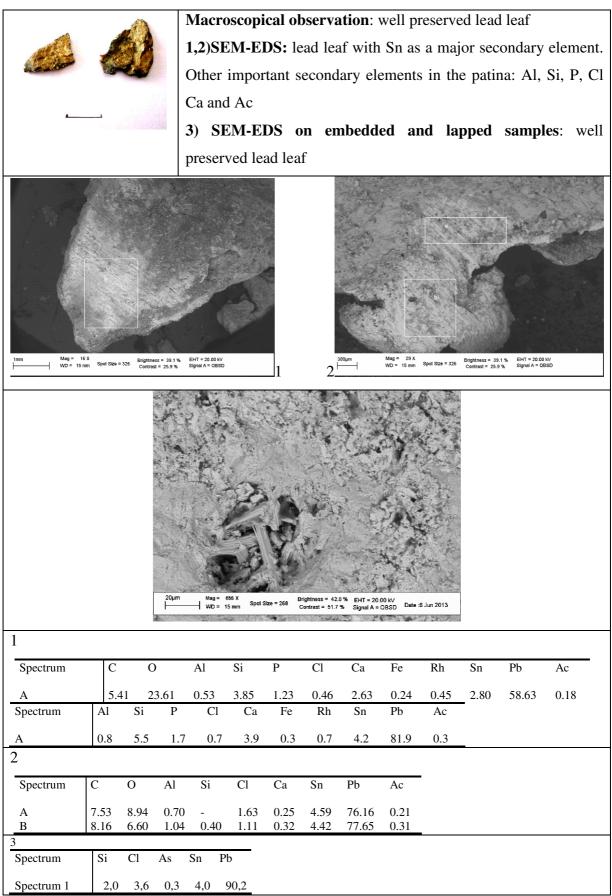






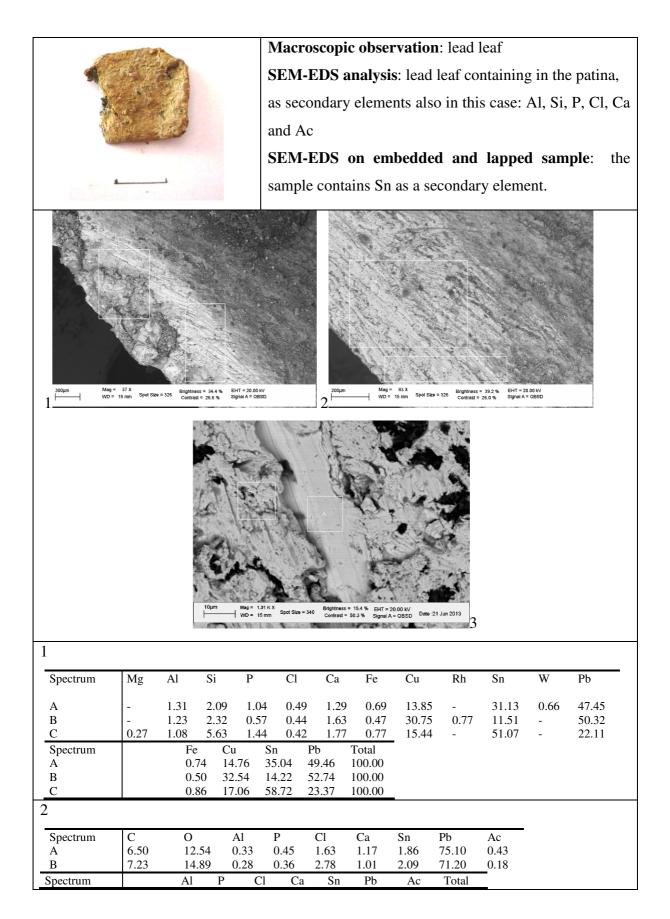
Spectrum A B	C 9.70 16.03		.91	Na - 0.28	Al 0.22 0.35	Si - 0.81	P 0.27	Cl 0.73 0.94	Ca 1.76 1.22	Rh - 0.59	Sn 0.93 -	Pb 64.09 58.39	Ac 0.39 0.35
0	10.02		.01	0.20	0.00	0.01		0.71	1.22	0.07		50.57	0.00
Spectrum A	Na -	Al 0.34	Si -	P 0.40	Cl 1.28	Ca 2.82	Rh -	Sn 1.50	Pb 93.01	Ac 0.65			
В	0.49	0.60	1.34	-	1.83	2.15	1.13	-	91.83	0.64			
Spectrum A B	Al 0.35 0.74	Sn 4.03	Pb 95.62 99.26										
_			,,										
Spectrum	Ca	Pb 98,9	-										

Vol 03 5866



Vol 056719

	Macroscopical observation: well preserved
	lead leaf
and the second se	SEM-EDS: lead leaf with Sn as a major
	secondary element in the patina.
	Other important secondary elements in the
	patina: Ca and Ac
Juni Mag = 67 X WD = 15 mm Septilize = 227 BigHnines = 23.9 K Central = 31.1 K BigHnines = 23.0 K Signal A = 0BSD 1	Image: Mage: YMX With the mage state
Spectrum C O Ca Sn Pb Ac	
A 7.0 13.1 0.5 1.4 77.8 0.2	
Spectrum Ca Sn Pb Ac	
A 0.5 1.7 97.37 0.2 2	
Spectrum Cl Br Pb	
A 100.0	
B 6,7 1,5 91,8	



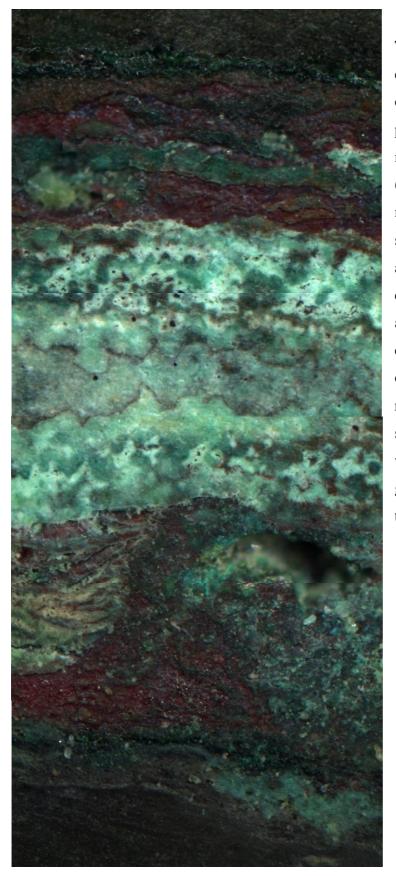
A B		0.4 0.3			23 1.5 00 1.3		92.26 90.67		100. 100.				
Spectrum		C		0	Al	Si	Р	Ca	Fe	Sn	Pb	Ac	Th
А		6	.28	12.68	1.50	0.91	0.55	1.78	0.26	1.72	74.16	0.16	-
В		6	.98	11.38	2.10	-	0.44	1.24	-	1.72	75.75	0.29	0.09
3													
6													
-													
Spectrum	С	0	Sn	Pb	_								
	C -	0	Sn 2.39	Pb 97.61	_								

	Macroscopic observation: copper alloy leaf.									
And the second s	SEM-EDS analysis: the sample is completely									
	oxidized. After the etching also micro-particles of									
	silver are detected. Also from this analysis the									
	sample seems to be composed only by layered									
	strata of oxidized compounds.									
Wight Wight 17.1 % Big Hears = 3.4 % Big Hears = 7.3 %	SPEX Tope Else + 32 BigHeat + 26 KB BigHeat + 26 KB SPEX Tope Else + 32 BigHeat + 26 KB BigHeat + 26 KB									
1										
1 0	Cl Ca Fe Cu Sn Pb 0.49 1.29 0.69 13.85 31.13 47.45									
В - 1.23 2.32 0.57 0	$\begin{array}{cccccccccccccccccccccccccccccccccccc$									
Spectrum Fe Cu Sn	Pb									
A 0.74 14.76 35.04 B 0.50 32.54 14.22	2 52.74									
<u>C</u> 0.86 17.06 58.72	2 23.37									
2										
	35 0.50 0.45 0.29 21.43 31.76 9.81									
B 4.79 31.16 0.44 1.69 0.	32 0.41 0.60 0.26 18.59 32.23 9.52									
Spectrum Fe Cu	Sn Pb									
A 0.44 31.60 B 0.42 28.43	6 51.84 16.06									
3										
Spectrum C O Al Si										
A 4.73 31.55 0.30 1.	92 0.54 0.37 0.77 0.65 16.89 30.82 7.66 29 0.53 0.59 0.57 0.63 17.74 34.28 7.79									
C 8.15 22.53 0.22 - D 3.14 35.60 0.28 1.	1.10 - 12.52 6.06 49.43 47 0.47 0.24 0.53 0.42 15.92 32.30 9.64									
Spectrum Fe Cu Sn P B 1.11 27.74 57.07 1	b 4.07									
A 1.01 27.25 58.26 1	3.48 2.26									
	6.94									

Vol 05 1054

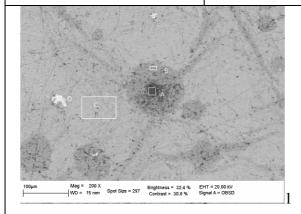
Analysis on embedded and lapped sample

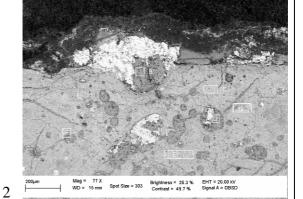
1 1 1 1 1 1 1 1 1 1 1 1 1 1	Spot Size = 30 $Bighines = 24.6 % Bighal A = 085D$ 2 $Diffunction = 100 M Bighal = 04.5 % Bighal A = 085D$ $Diffunction = 100 M Bighal = 0.085 Bighal A =$
Spectrum	C N O Mg Al Si Cl Fe Cu Ag Sn Pb
A B	5.4 - 23.2 - - 0.5 6.5 0.7 6.1 1.0 47.0 9.6 10.9 -1.4 22.2 - - 0.9 5.2 0.5 5.7 3.4 42.3 10.4
C B	5.9 - 21.5 - 0.4 9.9 1.3 10.0 - 42.6 8.3
D	4.8 - 40.8 6.4 2.6 4.2 6.1 2.4 5.2 - 22.0 5.5
Spectrum	Fe Cu Ag Sn Pb
A	1.0 8.7 1.5 71.4 17.4
B	0.7 8.3 5.4 66.8 18.8
C D	2.0 14.8 - 66.6 16.6 6.5 13.5 - 62.0 18.0
2	0.5 15.5 - 02.0 10.0
Spectrum	C N O Si Cl Fe Cu As Sn Pb
A	17.3 - 4.8 1.0 19.3 0.4 2.2 - 2.6 52.3
B	15.2 - 3.1 2.3 3.8 0.3 70.4 0.0 2.0 2.8
C D	6.1 -0.7 24.7 0.8 7.7 1.0 3.6 - 43.4 13.5 10.2 - 2.7 0.7 1.5 0.2 79.8 - 3.2 2.0
E D	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$
F	9.2 -2.2 17.9 7.7 7.3 1.1 14.0 - 31.4 12.7
G	7.9 - 14.5 0.9 6.2 0.5 37.5 - 24.7 7.8
Н	11.4 - 9.4 - 14.5 0.3 4.5 - 3.3 56.4
Spectrum	Fe Cu Sn Pb
A	0.6 3.2 4.6 91.6
B C	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
D	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
Е	1.3 12.4 60.8 25.5
F	1.8 22.0 52.9 23.3
G	0.7 50.6 35.2 13.4
Н	0.5 5.9 5.2 88.4



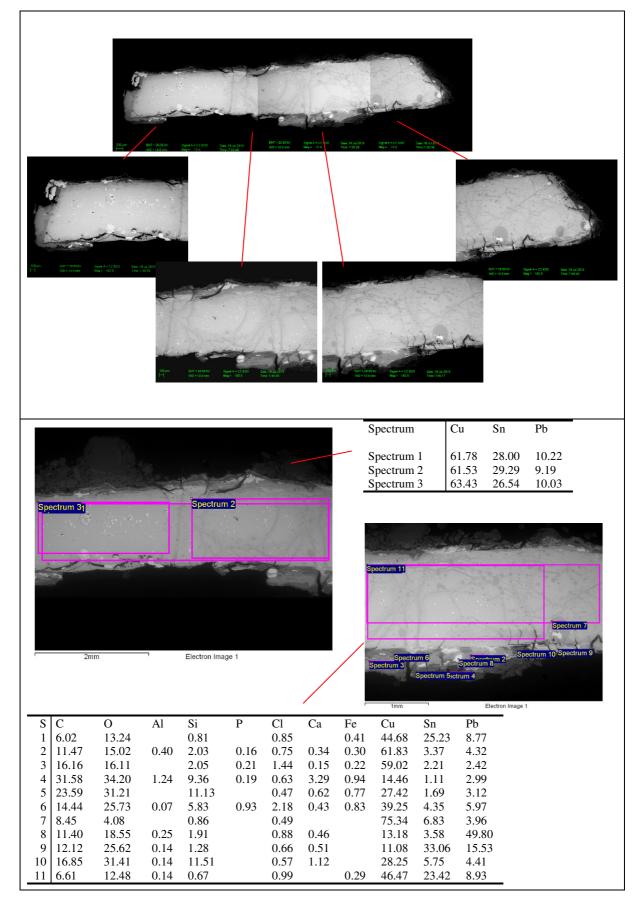
То recognize easily the composition of the layered oxidised compound some picture on metallographic microscope have been taken (50X), and composed to form a mosaic of the entire sample, showing cuprite strata (red) and a quartz grain. The green compound could be malachite, atacamite, paratacamite or copper sulphate. The presence of high amount of lead and tin revealed by SEM EDS could suggest the presence of the white cerrusite and the whitegray areas are explainable with the presence of cassiterite

Macroscopical observation: copper alloy leaf **SEM-EDS on embedded and lapped sample**: bronze ternary alloy, with a high presence of lead and tin. In the images globule of α phase with a β coating phase (peritectic reaction). The matrix is highly tinned (big grains of δ phase). The absence of dendrites suggest that the leaf was produced from a cycle of annealing and hammering, till the obtaining of the right thickness

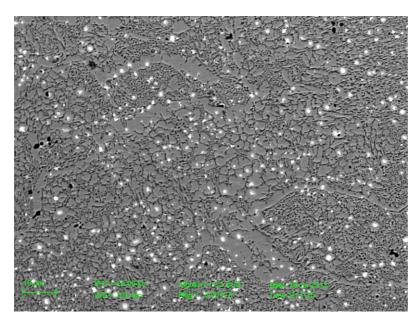




1											
Spectrum	С	0	S	i Cl		Fe	Cu	Sn	Pb		
Â	12.4	3.2	1	.0 0.2	2	-	79.6	2.4	1.2		
В	30.7	11.0) 2	.9 0.4	1	-	39.9	11.1	4.0		
С	6.7	16.8	3 0	.6 1.	1	0.4	33.3	31.7	9.4		
D	14.9	2.4	-	19	.2	-	5.9	2.8	54.8		
Spectrum	Cu		Sn		Pb)					
А		95,7		2,			1,4				
В		72,5		20,			7,3				
С		44,8		42,	6	1	2,6				
D		9,3		4,4	4	8	36,3				
2											
Spectrum	С	Ν	1	0	Si	Cl	Fe		Sn	Tl	Pb
А	17.			4.8	1.0	19.			2.6		52.3
В	15.			3.1	2.3	3.8					2.8
C	6.1		0.7	24.7	0.8	7.7			43.4		13.5
D	10.			2.7	0.7	1.5					2.0
E	7.0			23.1	0.6	7.6			38.4	1.0	13.9
F	9.2		2.2	17.9	7.7	7.3				1.0	12.7
G	7.9			14.5	0.9	6.2					7.8
H	11.		C	9.4	- D'	14.	5 0.3	4.5	3.3		56.4
Spectrum			Cu	Sn	Pb						
A			3.2	4.6	91						
B C			92.1 5.2	2.7 68.5	4.7 24						
D			5.2 93.1	08.5 3.8	24						
D E			95.1 12.4	5.8 60.8	2.8 25						
F			22.0	52.9	23						
г G			50.6	35.2	13						
H			5.9	5.2	88						
11	,		5.7	5.2	00	. r					

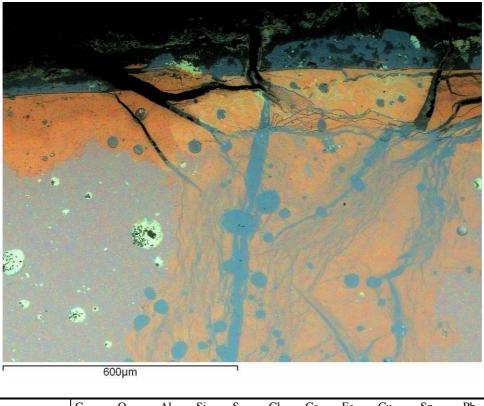


Vol 03 4197

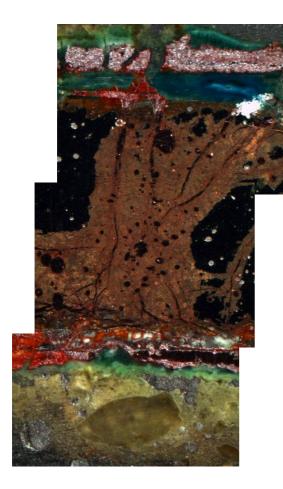


The picture below is a Backscattered electron image, which shows an high amount of small lead globules in a Cu-Sn matrix. The structure, highlighted by the use of the etching solution, is finely structured, and consists of dendrites which are closing to fine polygonal grains

The EDS elemental map below (copper: blu; tin: red; lead: light green) shows copper enrichments of circular shape. Each globules of copper is connected to the others from lines of enriched, almost pure, copper. These lines follow the trend of corrosion, so they can be considered areas impoverished of tin and lead.

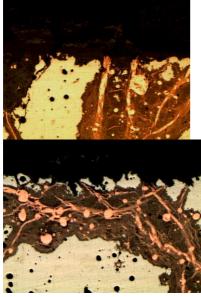


Spectrum	C	0	Al	Si	S	Cl	Ca	Fe	Cu	Sn	Pb
Sum Spectrum	13.43	15.02	0.12	2.19	0.06	0.86	0.32	0.36	39.93	19.32	8.39



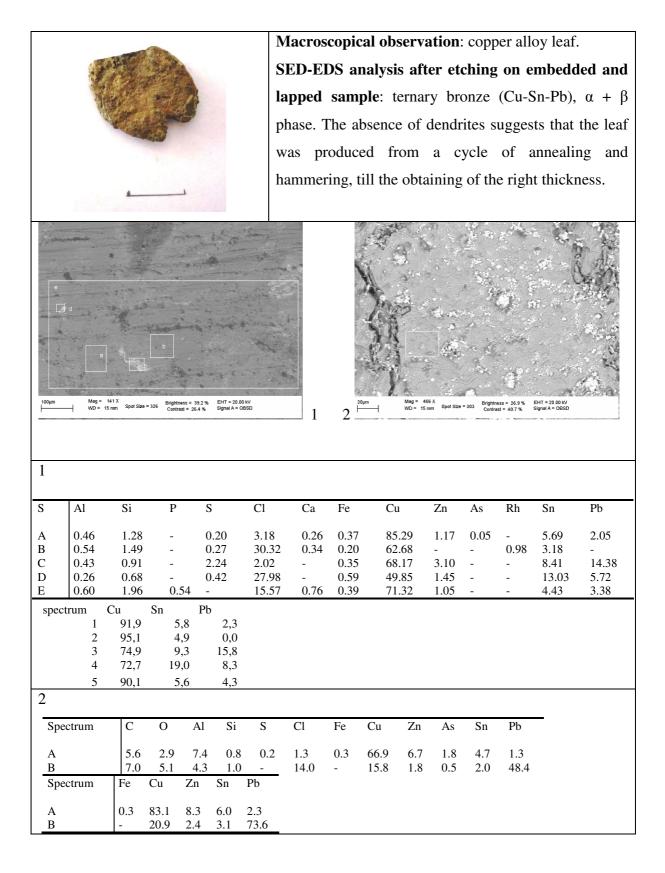
The picture on the left is a mosaic of metallographic optical microscope images token at 50X, covering almost the whole tickness of the sample. The patina, as confirmed also by SEM-EDS, is made of cuprite layers (red) and a quartz grain while the green compounds are suitable to be malachite, atacamite. paratacamite or copper sulphate(even if an absence of elevated levels of S in the SEM-EDS analis 2/4, makes this last observation to be unlikely), while the white dusty compound is accordant to the presence of cerrusite. The metallographic images are made on dark field, to better show patina, so in this mosaic the metal areas are rendered in black. In this way is interesting to notice the copper enrichments, surrounded by the developing of

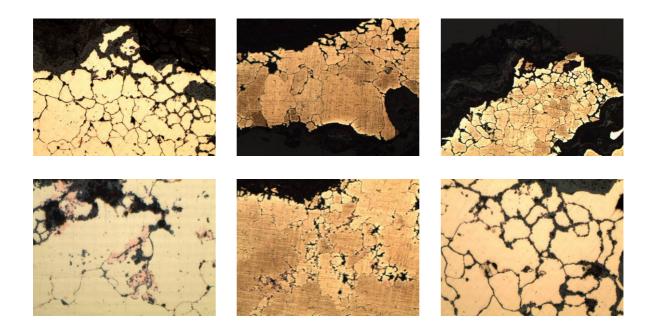
an high corrosion. The metallographic pictures below (50X and 100X), again show in high detail, the structures of copper enrichments.



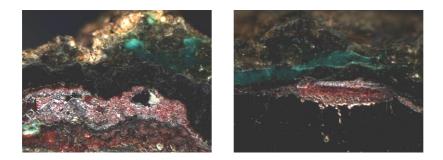






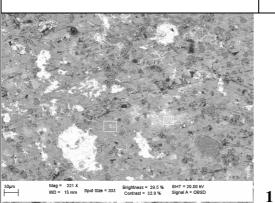


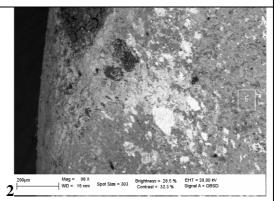
To better understand the structure of that sample, it was decided to carry out a deepening of the sample using metallographic microscope. The following picture, performed at 50X and 100X on the sample embedded in resin and treated with FeCl₃ etching solution, shows an intergranular corrosion along the grains borders and lead globules (dark gray) and copper sulphur (light gray). The structure is polygonal and the grains are of different dimensions. That structure is the result of a thermal homogenization, an annealing, which makes the dendritic structure disappear. In the second picture, taken with a 100X magnification also shows pink areas of copper enrichment along the corrosion lines.



Some pictures with metallographic microscope have been made of the patina: cuprite strata (red) and a quartz grain is easily recognized. The green compounds could be malachite, atacamite, paratacamite or copper sulphate.

Macroscopical observation: copper alloy leaf, with the sign of the use of a saw on the surface. **SEM-EDS analysis**: ternary bronze alloy with big size globules of lead. $\alpha + \delta$ obtained by the cycle of hammering and annealing to obtain the bronze leaf. After the etching, the globules of lead are much more evident

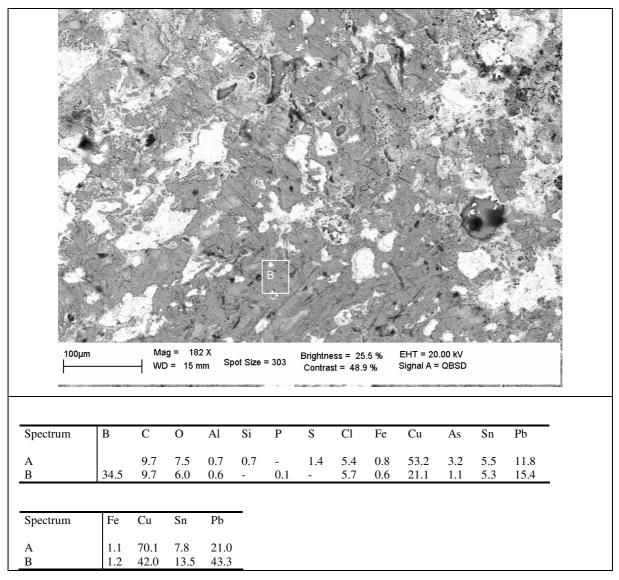




1														
Spectrum	С	0	Al S	Si S	Cl	Ca	ı Fe	Cu	As	Sn	Pb	_		
А	6.5	15.5	0.7 -	-	14.	9 -	1.4	6.3	0.5	19.1	35.6			
В	9.7	6.6	1.1 2	2.4 6.1	3.5	0.2	2 0.9	59.9	3.2	6.5	-			
С	8.2	12.2	0.3 0).4 -	5.2	-	0.8	13.1	0.8	11.1	13.4			
												_		
Spectrum	Fe	Cu	Sn	Pb	-									
Spectrum	10	Cu	511	10										
А	1.9		29.6	59.9										
В	1.2	77.5	9.4	11.9										
С	1.7	27.9	30.1	40.3										
2														
Spectrum	В	С	0	Al	Si	Р	S	Cl	Ca	Fe	Cu	As	Sn	Pb
А	-	8.9	12.3	1.4	-	-	-	13.4	-	0.6	4.8	0.5	4.8	53.2
В	-	9.5	6.0	1.3	0.7	0.2	0.2	4.1	0.2	0.9	63.8	3.8	6.0	3.3
С	39.6		8.9	0.9	0.2	-	-	5.0	-	0.5	12.9	1.5	9.8	11.0
Spectrum	Fe	Cu	Sn	Pb	-									
А	0.9	6.5	7.7	84.9										
В	1.2	84.1	8.5	6.1										
С	1.3	30.9	29.9	38.0										

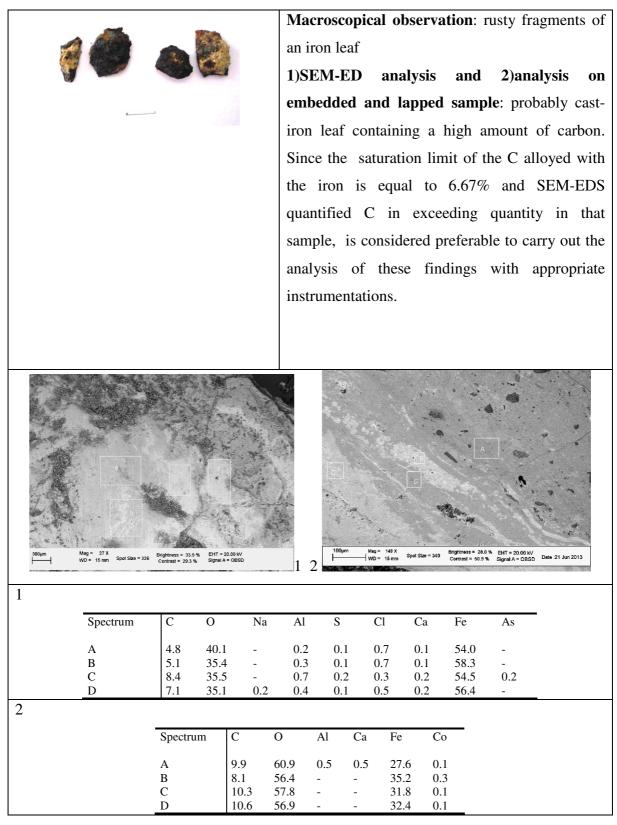
Vol 03 3182





Vol 02 4404

				Macrosco iron leafs. SEM-ED of oxidize	S anal	ysis : i	ron lea	f with r	esidual		
Spectrum	→ wD= O Mg		si P		nal A = QBSD K	Ca	Fe	Cu			
A B C	25.3 0.3 30.7 - 30.1 -	2.3 1.9		.2 3.3 2.1 1.0	R 0.6 0.2 -	1.2 4.1 0.7	60.5 58.2 66.7	0.6 - -			



Mass Spectrometry analysis on lead fragments from Volubilis

A preliminary provenance test has been carried out with TIMS instrumentation (Thermal Ionization Mass Spectrometry) instruments of IGG (Institute of Geoscience and Georesources) institute of CNR, on samples taken from leaf n. 051051. Isotopic abundance has been measured on three different isotopic ratios: Pb206/Pb204, Pb 207/Pb204 and Pb208/Pb204.The data reported below are already corrected in reference to the international standard 981

 $Pb206/Pb204 = 18,2815 \pm 0,0006$

 $Pb207/Pb204 = 15,6203 \pm 0,0006$

 $Pb208/Pb204 = 38,2962 \pm 0,0016$

These data are connectable with a galena extracted from the Earth's crust during Middle Paleozoic Era. These preliminary results have been compared with isotopic database containing data from the major ore basins exploited. This database doesn't contain data from Northern Africa, therefore it was decided to perform the confrontations and mapping to see if there is a possibility that the lead leaf comes from basins outside Morocco. These data, plotted in diagrams 207/204 vs 206/204 and 208/204 vs 206/204 overlap the range of variation of lead ores of south-central Spain (Linares area) with a partial overlapping with the field of lead ores of Sardinia (Fluminese-Arburese area). Considering this preliminary data it could be argued that the raw ore used for leafs production comes from Spain, using a database that contains no African data. Since the isotopic fingerprint is connected with the petrogenesis of the basin, and the same isotopic ratio is sometimes shared by ores from very far provenance (considering the variation field), attempting a more detailed literature research on lead isotopic ratios for Moroccan basins (Bouabdellah et al., 2009), comparable data have been found, suggesting also a possible local provenance: in fact these isotopic ratios are peculiar of a petro-genetic process which took place during Palaeozoic era in the system mantle / crust. This first observation has been done on data from minerals that probably had not been exploited in antiquity (mixed sulphides).

Secondly a more interesting comparison with Volubilis isotopic data has been performed on the isotopic data of Aouam-Tighza basin found in the research carried on by Boushaba & Michard (2011). The isotopic characteristics of the Pb, Ag, Zn veins are characterized by a comparable ratio for 206 Pb/ 204 Pb = 18,25 and 207 Pb/ 204 Pb = 15,68. Differences in the value can be interpreted with the variation field of isotopic fingerprint. Because of its Volubilis proximity to the mining site of Aouam, it is suggested that the exploited basin was precisely the one of Aouam. This would also explain the motivation to install a Punic site, later occupied by Romans in an area so far away from the sea, whose reaching was also impervious.

To strengthen this hypotesis of provenance our equipe needs to continue with the collection of Moroccans isotopic data to get a reference database that could point to a sure local provenance.

This result has therefore highlighted the need for further isotopic analysis, to obtain less ambiguous data, performing measurements of the isotopic ratios of other elements (Nd, Sr, S) and trace element analysis (PIXE) in order to properly characterize the finds from Volubilis. This step of research has therefore provided the guidelines indicating the analytical path that will be followed in the future. Will thus be necessary to characterize each mining areas in Morocco, in order to produce a database to implement the existing ones. The lack of data on Morocco and North Africa in general, therefore, generated probably a wrong interpretation of provenance of many of the artefacts in the Mediterranean basin for which have not been taken into account these important mining areas in Africa.

5.7 Discussion of the results on the artefacts

The analytical characterization of the artefacts shows some interesting results, throwing light on some unclear aspect on the metallurgical attitude of this neglected African area.

First of all, the artefacts seems to be composed of:

Vol Y, Vol X, Vol 03 5866, Vol 05 6719, Vol 05 1054: lead leafs;

Vol 03 4197, Vol 05 7085, Vol 03 3182, Vol 05 1051: ternary bronze alloy leafs;

Vol 02 4404, Vol 02 3168: iron leafs.

On the lead artefacts, one interesting result comes from the detection of the secondary elements in patinas. In the four laminas a major secondary presence of Sn and a constant presence in the oxidized layer of Al, Si, P, Cl, Ca and Ac has been detected, giving the possibility to formulate hypothesis on the ore provenance. On the presence of actinium was performed interpretive hypothesis. First of all Actinium is a product of Uranium fractioning, and this element is present both in sandstone, granite and in phosphorite deposits, extensively present in Morocco. The presence of P, as a constant secondary element in the alloy, could be hypothesized as the exploitation of a mine containing

phosphate mineralization (Morocco is one of the first producer), such as for example the multi-metallic mineralization of Marrakech where, beside galena, blend, copper, oligist iron etc. is beared and accompanied by phosphate layers. This statement should be taken just as hypothesis since is also possible that the constant presence of this elements is to be connected with the earth of burial.

In any case the presence of pyrometallurgical process inside the settlement can confirm the use to install inside towns small metallurgical workshop for conduction of small scale processes, leaving the processes on a larger scale in the vicinity of the mine.

Iron, bronze and lead furthermore suggest the presence in this ancient city of diversified activities with the use of different types of metals. The presence of so many fragments of tools also suggests that semi-finished products were produced here, to be traded from the naval harbors on the coastline. In this regard is possible to propose that a small scale workshop of metal forgery was present inside the settlement.

Since the work in collaboration with Equipe de Geoexploration of Meknes Universitè Moulay Ismail and with ENSAP (Institut National des Sciences de l'Archéologie et du Patrimoine) should be extended for other five years, the work will go on also under the suggestion given by this SEM-EDS analysis and by the illuminating results obtained with mass spectrometry. Archaeological excavations have been promoted in the mentioned areas, given the good results during the prospecting of 2012. An archaeological excavation is going to be organized (in case there will still be the right conditions) in Aouam mine under this bilateral agreement during 2014, in such a way to obtain samples connected to a precise stratigraphy and with a much more sure archaeological context.

In conclusion it can be stated that Morocco important role in Circle in the Strait of Gibraltar metallurgical production, as one of the richest areas of metal ores, also exploited in ancient times. Surely we need to carry out future archaeological archaeometric research to have more precise information on the location of mines and type of metallurgical operation .

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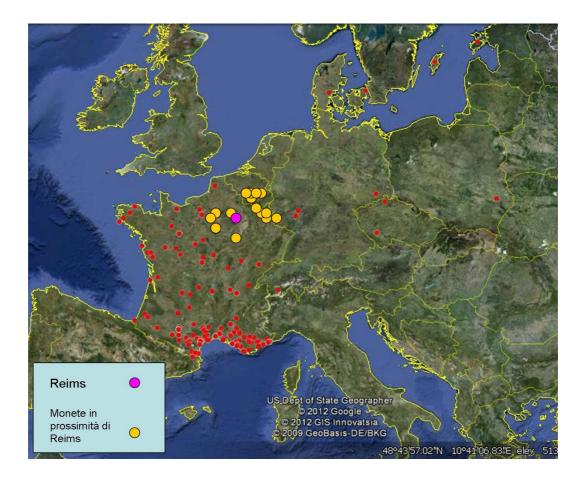
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CHAPTER VI





Google Earth map on punic and neopunic coins in France and Belgium

Punic and Neo-Punic coins in France and Belgium

Collaboration agreement with CRESTIC and GEGENAA institutes - Université de Reims Champagne-Ardenne

Highlights

The research on Punic bronze coin production was developed starting from the localization of numismatic findings to highlight their presence in foreign areas. The study regards the localization patterns of Punic and neo-Punic coinage recoveries in France and Belgium and other north-western areas, through the application of geographic information systems (GIS). The research starts from the collection of a database of coinage findings from literature, accompanied by information on archaeological context and the dating of coin types. Also compositional information of the coins are inserted into the database (where they are available). The conversion of the database in geodatabase has allowed us to offer some numismatic and archaeological interpretations on this large number of coins, also in relation to the presence of rivers, archaeological sites, naval and land routes and mines. In fact it was decided to study, also in this case, the phenomenon of the presence of these allochthonous objects in areas faraway from the motherland and from production centres. For this reason it was decided to study the numerous coinage discoveries in France and Belgium, and with a lesser extent some areas of northern Europe and UK, in order to understand which routes led so far these objects. The research was carried out in this case with the only use of the GIS system in order to achieve interpretative assumptions on these findings. This will make it possible to draw more substantiated routes of penetration which can be monitored by future archaeological surveys and prospections

6.1 Introduction

The study of the Phoenician and Punic coins always suffers from lack of "visibility" due in large part to the reluctance to recognize in the Phoenician and Punic cultural identity, something autonomous and not provincial, with respect to Greek and Roman world. In particular, for the Phoenician West areas many difficulties is encountered in admitting the role of Carthage as a hegemonic power in the Mediterranean. This attitude is reflected in the still widespread practice of cataloging the Punic and neo-Punic coins between the Greek ones. This common misleading interpretation required a deepening of the Punic numismatic issue, since a widespread diffusion of those coins, also outside the emitting areas, is to be noticed. From December 2011, a collaboration between CReSTIC (Centre de Recherche en Sciences et Technologies de l'Information et de la Communication)/GEGENA2(Groupe d'Etude sur les Géomatériaux et les Environnements Naturels Anthropiques et Archéologiques) of Universitè de Reims Champagne Ardenne and ISCIMA-CNR Italy has been activated on a common project entitled: *Spatial Analysis of the Punic and neo-Punic*

coins found in Continental France and Belgium with the aim of Geographic Information System (n.prot. 0000023ISCIMA/CNR). The project consists in the setting up of a digital catalogue of the information about Punic and neo-Punic numismatic findings in a GIS environment, to permit the spatial analysis and augmentation of the collected data.

Punic and neo-punic coins(Acquaro et al., 1992; Acquaro & Manfredi 1992, Manfredi, 1995; Viola, 2010) have been found in France and Belgium in such a large number (Bar, 1985, 1991, Doyen, 2011; Feugere & Py, 2011; Fischer, 1978) to induce this fundamental question: when have these coins appeared in the area and why these coins have circulated on the French territory? (Fischer, 1978). Object of the study is to define the historical dynamics that led to the presence of Punic and neo-Punic coins in France and, more generally, in the continental Europe, with particular attention to the area in the Champagne-Ardenne. The Punic and neo-Punic coins in France, considering also the multiple findings, are 383 while in Belgium there are 17 findings (the catalogued coins comes mainly from Feugere & Py, 2011; Fischer, 1978), minted in various areas like Sicily, Sardinia, Carthage, Ibiza, Numidia, Mauritania etc..., of which has been performed the digital cataloguing and georeferencing with GIS system, that will be in the near future accessible on the GEGENA2 website. The cataloging of coins is completed with tabs about the archaeological contexts of discovery, and detailed information on the type of coins. Each one has been also cataloged and accompanied with an image, in order to permit an easy comparison of the coin types. The insertion of more than 300 coins in the geodatabase has therefore allowed to perform a spatial analysis of this phenomenon. Reims, home of the partners institutions, is located where once stood the capital of Belgic Gaul, Durocortorum, area of greatest interest for this study because, as will be seen below, the history of these coins goes up to Caesar and his war campaigns.

All the information relating to Punic and neo-Punic numismatics found in literature, such as anatomy of the coin, the ponderal systems, metal alloys etc... have been discussed in detail in Appendix C (Bar, 1991;Barello, 2008; Finetti, 1987; Manfredi, 1990).

In this chapter, the spatial analysis of Punic coins, in French, Belgian and North European areas will be also discussed (Milne, 1948), with the main objectives to understand, through the study of archaeological contexts, the historical dynamics that led to their presence in so far away areas from the center of the Punic culture.

This first brief bibliographic review showed that the problems related to the distribution of Punic coins on French area and in general in continental Europe, are many and not easy to understand from both the geographically and chronologically point of view. The phenomenon has been variously interpreted as being due to: 1) scarcity in some regions of the divisional numeraire which followed the Roman conquest; 2) as a means of hoarding; 3) as votive offerings. The most accredited hypothesis is that the Punic coins have been conveyed in these areas by the presence of the Roman soldiers. Other 782 Punic coins are found in other areas letting consider also other more complex reasons for this huge presence (289 are from Croatia, 4 from Bosnia, 9 Germany, 3 Denmark, 104 UK, 363 Monaco, 1 Slovenia, 4 Luxembourg, 1Sweden, 3 Czech Republic, 1 Poland). If those hypothesis can explain the spread of the Numidian and the autonomous cities coins (from North Africa) outside their territory, in Europe up to the North Sea, more problematic it seems the incidence in this vast and varied territory of the oldest Carthaginian emissions. All the interpretative considerations will be explained throughout in the next paragraphs. This georeferencing work is a preparatory phase, organized in the view of the archaeometric study on coins museum collections preserved in France.

6.2 Geodatabase

All information obtained from the bibliographic study were entered into a database whose structure is illustrated in the two tables, the first for the historical data and the latter for the material characteristics:

Locality	Archaeological	Place of	Classification	Obverse/	Mint	Context	Bibliog.
District	Context	conservation	from	Reverse	Datin	dating	
Lat/long			numismatic		g		
			literature				
Location of discovery of the coin and geographic coordinates	Occasional discovery, form surface surveys or discovery in archeological context	Private or public collection (Museum foundation etc)	Classification according to syllogoi and specialized literature	Descripti on of the stylistic and epigraph ic features of the	Place and date of minti ng of the coin	Chrono logy of context	All referenc es on the coin or context
				coin			

Conservation status	Production and minting techniques	Diameter	Weight	Metal	Link to the photos
Description of the appearance of the coin (the presence of degradation products, peculiarities, etc.)	Minting or melting Casted flan or cutting off metal tube	Diameter in mm	weight in g	Gold, copper, bronze, electro, potin, billon	All the coins are accompanied by a photo

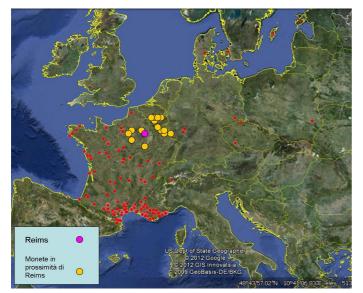


Fig. 1: Google Earth map of all the Punic and Neo Punic Coins in Northern Europe

Each coin is connected to a picture accessible from the map itself, so as to allow a comparison between different In addition types. to the archaeological/historical data, the database fields are set up also for archaeometric data acquisition on individual coin, as well as to allow comparison of compositional data from already done researchs. The database is therefore set out to contain the results of chemical and physical

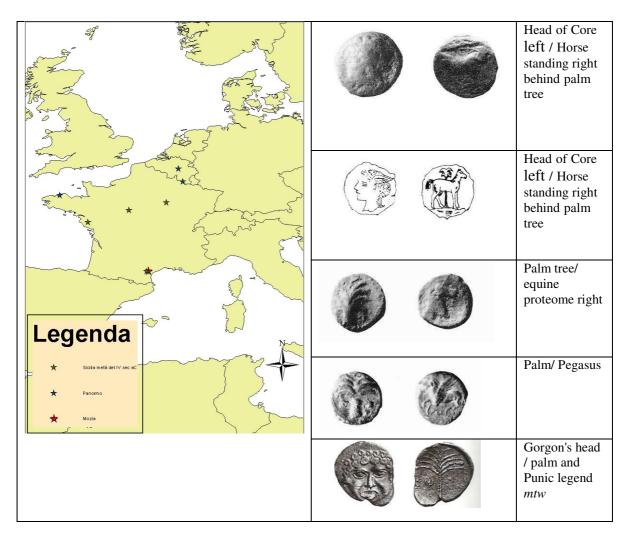
analyzes on each coin which will be possible to analyse in partnership with the Université de Reims Champagne-Ardenne. In particular, in the future developments in agreement with GEGENA2, it will be probably possible to use portable instrumentation for XRF measurements that will yield information on the elemental composition of the coins, in a totally non-invasive and non-destructive at the museums or private collections where the same are kept.

This picture (fig. 1) shows all the numismatic finding areas, in pink the city of Reims is shown as point of reference and the yellow dots are the surrounding finding areas. In fact this research puts a particular attention to the area nearby Champagne-Ardenne In this picture are shown, besides the French and Belgian areas, also other northern areas in Europe (Poland, Czech Republic, Estonia, Denmark, Sweden, Germany, Luxembourg and Britain) whose inclusion is due to the great interest for these findings, and because these will help to clarify some issues for the French and Belgian coins. As explained in the Appendix C, the discussion will be separated for these three classes of coinage: the coinage of the autonomous cities minted before power growth of Carthage, the coinage form cities and province of Carthage and the neo-punic coinage, belonging to the autonomous cities and kingdoms after the end of Carthage. Only taking into account this chronological arrangement the developments and understanding of historical, political and economic features related to it can be fully understood.

One of the developments of the work will be to transform the table into a database system, by inserting the values of the uncertainty related to each field. By superimposing these two layers in the GIS, it will be possible to produce simulations thematic maps, useful to suggest where may be other coins which is not yet found. This new structuring of the database is at run time by Universitè de Reims equipe, in order to statistically analyze the data, to build a model of localization, and create a map of forecast on possible future discoveries in considered areas, with particular reference to Reim.

Currently this common research is at this step: after a convertion in geodatabases, through the use of GIS system, thematic maps on punic coins presence has been prepared. Once the database is entered into the GIS, has been possible to perform a real analysis of the spatial phenomenon under study, using the various exploratory possibilities the system offers.

The discussion continues by taking into account the coins in chronological order of minting. Each mint and period will be handled separately by the elaboration of a thematic map and the insertion of pictures or drawings of the most important type of coins (the picture are taken from Feugere & Py, 2011; Fischer, 1978).



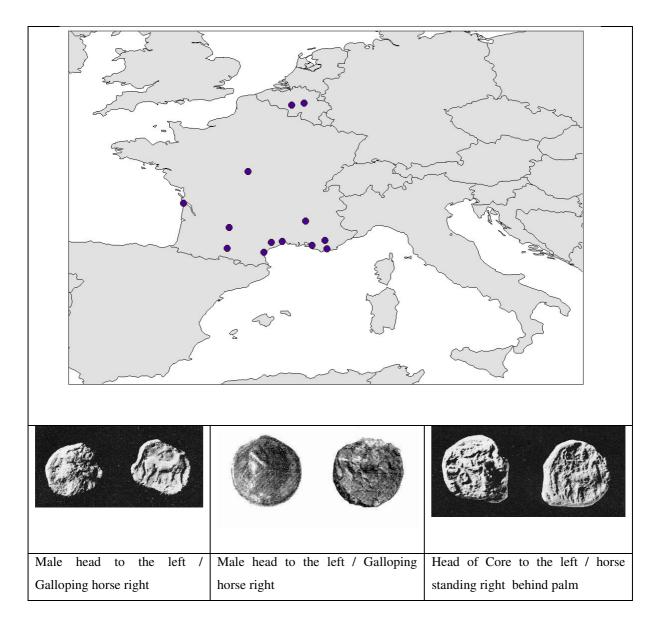
Sicilian Mint. The end of V- second half of IV century BC

Particularly important is the presence in Gaul of the oldest coins from Phoenician-founded cities of Sicily, which emissions is dated from the late V century BC – mid IV century BC, certified in eight contexts, including those of Olonzac and Mailhac and documenting a chronological context of the late VI century BC. The presence of a bronze coin of the second half of the V century BC, belonging to the type of Mothya mint, *gorgoneion / palm* with the letter *bet* under the belly, found in the territory of Mailhac in a context that goes from the end of the IV to the II cen. BC, and another one belonging to Sicilian mint, found in the oppidum of Mourrel-Ferrat in a context of the late IV-early III century BC, are to be noted. The two coins documented both on the southern coast of France, suggest the existence of possible relationships between the Phoenician cities of Sicily and southern Gaul during pre-Hannibalian period, even if their attestation does not seem to be conveyed

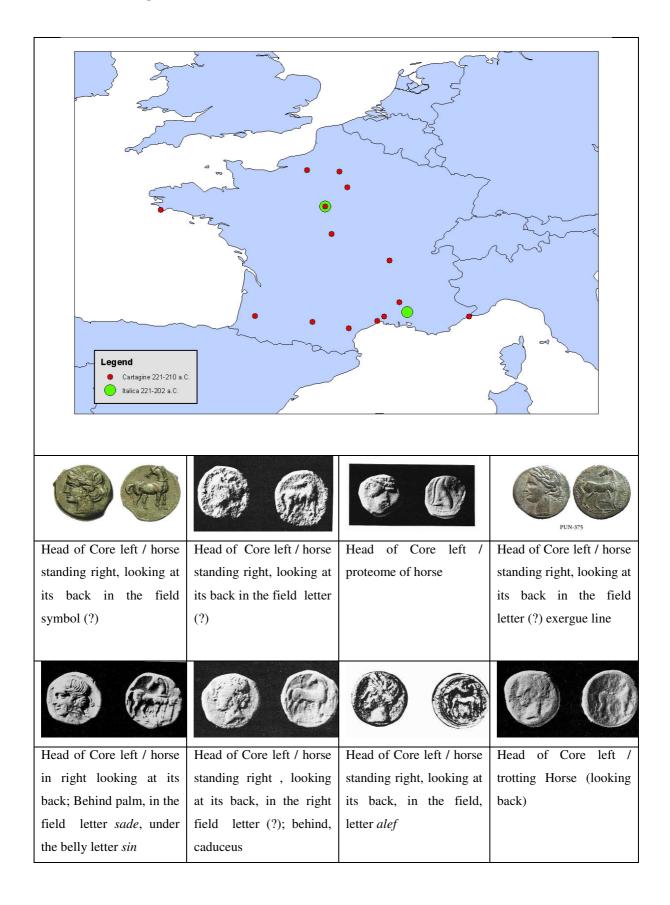
or relatable to the circulation of Greek coins coming from Sicily and from southern Italy since these latter are not documented in the two sites. More likely, their pertinence seems to be connected to a flow from the Iberian Peninsula, as suggested by the presence in Mailhac of later Iberian coins of Emporion, Bolskan, Kelse, Obulco and Ebusus. In Wasseiges in Belgian Gaul, it is also documented a bronze of the type *galloping horse / protome of human headed bull and legend 5*y*5* relevant to the mint of Panormo, dated to after 409 BC, which still shows the most ancient coinage production of the Sicilian cities of Phoenician tradition.

The autonomous cities of Solunto, Mozia, Panormo were the ones where the Punic or Punic - Greek coinage began at the end of the V century BC, and these emissions immediately show to be highly integrated into the Greek monetary and economic system in the island. The comparative study the presence of these types provides valuable information for the reconstruction of the network of business relationships, established by the individual Phoenician cities. From this examination, the range of exchange attributable to Panormo seems particularly extended, so as to assume a leading role despite the other Phoenician cities, and with the advent of Carthage influence in the island, its possible appointment as *qrthdst* "new capital" of Punic Sicily. The two finding in the south of France are connected with commerce of Sicily with Marseilles and the other coastal cities, while those found in most inland areas indicate more distant commerce and trade, which included terrestrial and maritime journeys. As will be explained fully at the end of the chapter, those coins could be connected with the presence of exploitable mines in Bretagne.

Carthage. First half of the IV century BC



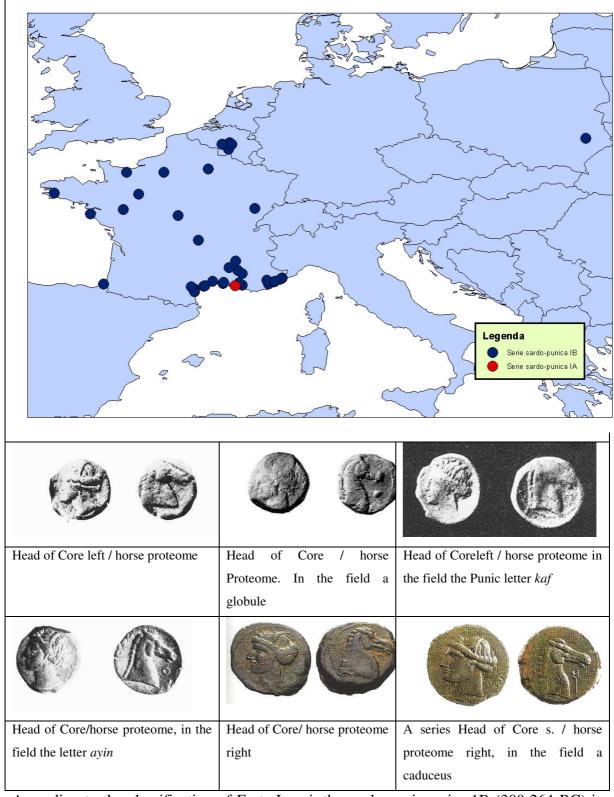
The coins of male head / galloping horse, head of Core / horse behind palm, have a large spread in the Mediterranean, and were probably produced not only in Carthage, but from many more mints under the direct control of the capital; although it is conceivable the presence of sub-mints, even if these are difficult to be distinguished. The coins with the galloping horse at the reverse are widespread in Sicily and Sardinia. From Belgian Gaul come also some coins of the mint of Carthage from this period, type *male head / galloping horse* and *head of Core / horse behind palm tree*, found in the territory of Turre-Liberchies, in Wasseiges and doubtfully at Flavion.



Mint of Carthage 221-210 BC and / or Italian Peninsula from 221 to 202 BC

The coins belonging to the Italic and the Carthage mint of 221-201 BC are associated with the presence of Hannibal in these areas as a result of the Second Punic War (Fischer,1978). Hannibal departed from Carthage to conquest the Roman Empire capital, Roma, and to cover this huge distance passed through the Algeria, then crossed the Mediterranean Sea and arrived in Cartagena. Thus he trespassed the France and reached Italia from Apennines, and trough Capua he finally reached Rome. These Punic coins dating from the III-II century BC followed Hannibal and his troops marching from Spain, crossing the Pyrenees, Provence and the Alps. This is the reason that leads these coins in these distant lands. With regards to the coins minted in Italy in this period, it is known that the troops used to emitt brand new coins in mobile mints, and also in the path of Hannibal in Italy these special coins were produced for daily transitions of the troops.

Carthaginian coins of the same period are also present in Narbonensis Gaul in Lattes, Aigues-Vives, Bollène, L'Isle-sur-la Sourg, Vieille-Toulouse.

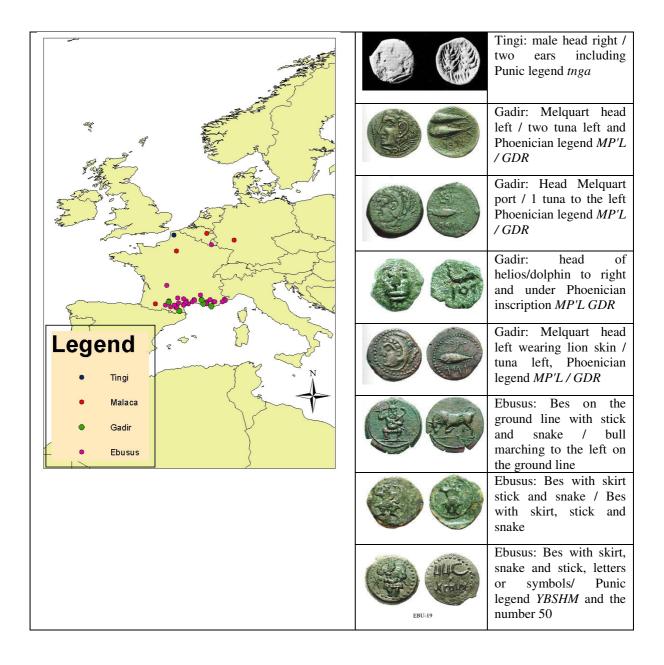


Sardinian - Punic coin series 1A and 1B (300-264 BC)

According to the classification of Forte Leoni, the sardo-punic series 1B (300-264 BC) is much older than the1A (264-241) which is bigger; the order of these two series is explained

in the light of the different dimensions. Punic coins with the horse proteome on the reverse are particularly present in southern Gaul and position themselves in particular in the areas of movement of maritime traffic, such as the ports of Monaco, Agde and Marseilles. Beside this, the 1B series has a very wide distribution (corresponding to the much older series of galloping horse) in the Mediterranean circulation, which seems to indicate that for this emission existed more than one active mint, the main was Carthage, but this kind of coin was emitted also in Sardinia in the sub-mints. This currency was of common use as the *galloping horse* type.

Series of neo-Punic coins minted from Tingi, Gadir, Malaca, Ebusus (II-I century. BC)



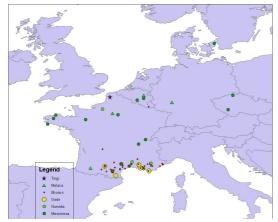
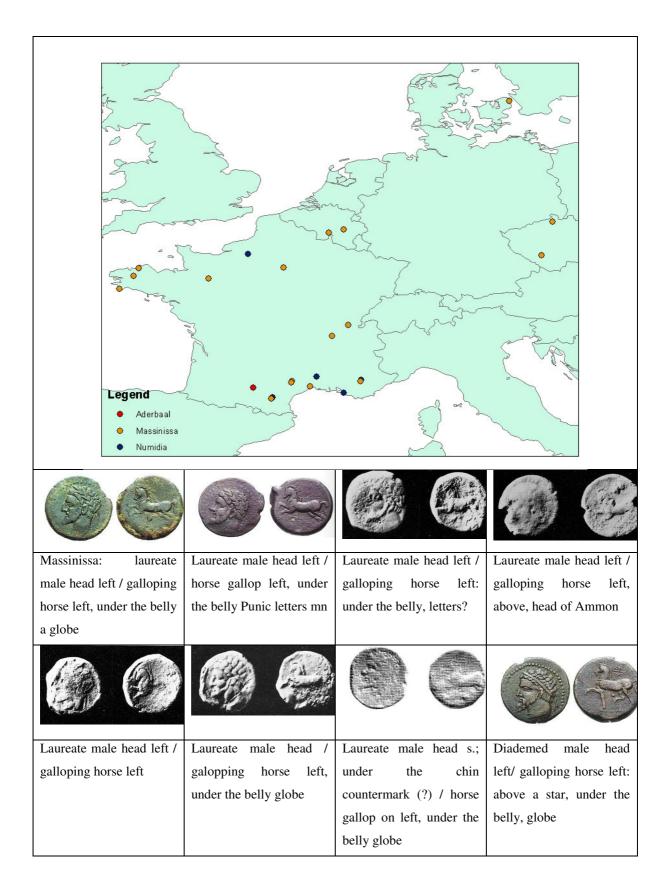


Fig.2: numidian and authonomous cities coins

After the fall of Carthage all the authonomous cities reappeared and restarted to produce coinage on their own. This is why the name of the emitting city on the faces of the coins started again to appear: all the coins from the cities of the coast of Algeria, Morocco and those of those of Spain are

epigraphic coins. This trend appeared from the III century BC, before the fall of Chartage and went on also during the I century BC. These coins circulated together with the Numidian coins (fig.2), because Numidian population recognize their value and use them to pay for mercenaries. The mercenaries arrived in the West with Hannibal and Barcides troops. The Ebusus coins collocated all in the south because Ibiza was the contact point between Africa and Europe. Numidian neo-Punic Coins: Massinissa, Aderbaal and Successors (203-148 BC)

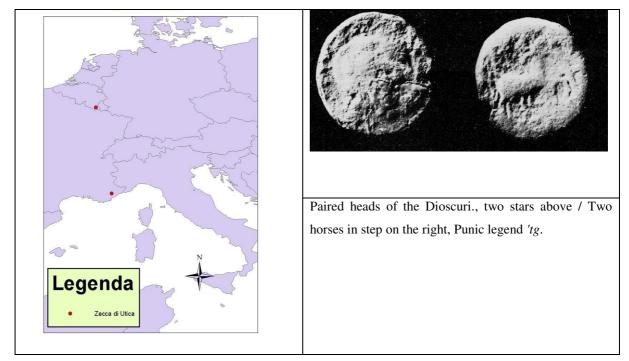


Numidian coins reached French and Belgian territory probably following two distinctive phenomenon:

-the massiles troops marching in favor of Carthage during the Second Punic War,

- under the reign of Massinissa (206 BC) emphasize the changed relations with Carthage in favor of the Romans.

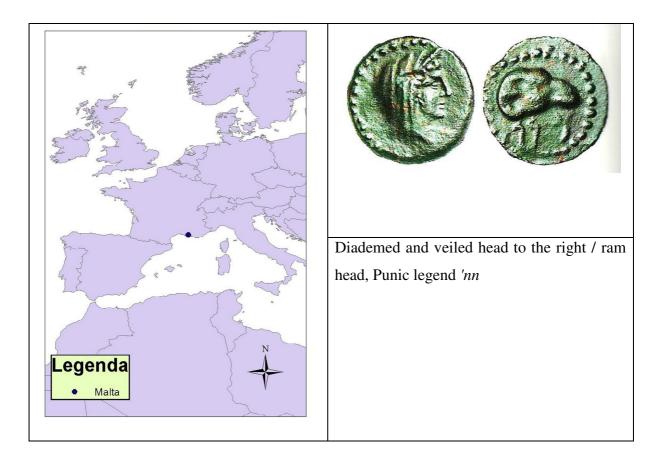
From the Reign of Massinissa and the Reign of his successor Micipsa, Numidia remains in favor of the Romans and abetting them with their mercenaries during the conquest of Gaul; the situation does not change during the reigns of Juba I and Juba II. The most recent coins instead reached Gaul after Caesar's mercenaries from Numidia, Mauretania, Cadiz and Baleares.



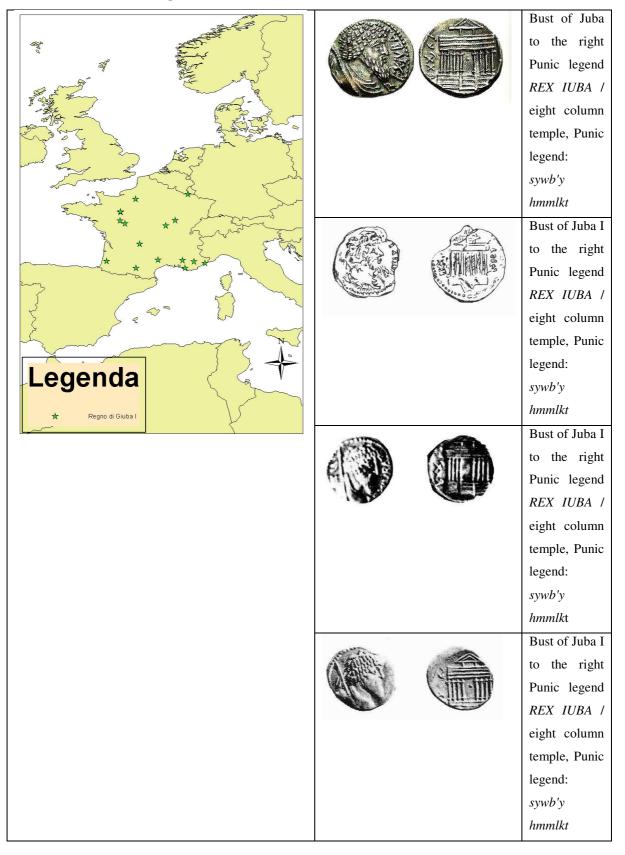
Coins from Utica, II-I century BC

The findings of this kind of coinage are very few, and it is connectable with the commercial trade with Monaco harbor. Beside this, it is quite difficult to explain the presence of one of this coins in Belgium, although the more logical reason seems to be the connection with the presence of soldiers from Africa.

Coins from Malta (I century BC)



This coins are part of the Marseille treasure, here present because of the commercial connection with the Marseille harbor area.



Mint of Numidia, Kingdom of Juba I: 60-46 BC

These coins testify a particular period in France, where lack of money made it possible the use of any foreign currency as current coinage. All the emissions that are multiples or submultiples of the Gallic Denarius were accepted. In fact, these coins were accepted to fill the shortage of currency that followed the Caesar conquest of Gaul and were accepted in the case the measure corresponded to the purchasing value. The weight of denarius was 1.90 grams and coins of Juba I were a multiple: this is the reason why the coins of Juba I are found in large number in these areas. The Punic bronzes were perfect for the movement in Gaul because the coin had a familiar appearance for the Gallic population, having coined on the obverse a male or female face and the reverse a horse, as the denarius. Numidian coins had the same characteristics. The Punic bronze coins were used as currency in a foreign country for daily commercial transactions, these emission were hoarded by the legionnaires when they changed garrison, while the silver example circulate less and are mostly found in the context of treasure. The African currencies are used until the end of the War of Gaul and to the reorganization of the system of coinage took place during the reign of Augustus between 23 and 25 BC.

6.3 Historical Conclusion

The insertion of more than 300 coins in the geodatabase has therefore allowed to perform a spatial analysis of this phenomenon (fig.6). The following are the possible conclusion reached for this research using GIS:

-the most ancient coins, belonging to the first half of the fourth century, are to be interpreted with the trade of the Carthaginians. The presence of these coins is to be interpreted, therefore, with the displacement of trade routes.

-Punic and neopunic coins dating from the III-II century. B.C. followed Hannibal and his troops marched from Spain, crossing the Pyrenees, Provence and the Alps to reach Italy. -the most recent coins reached Gaul along with Caesar's mercenaries from Numidia, Mauretania, Cadiz and Balearic Islands.

The coins connectable to trade are positioned in the coastal areas and follow the ways of penetration to the inland, while the coins connected to the presence of foreign legions in France have a wide-ranging distribution. The presence of Numidian coins in France attests the presence of Maxiles troops who marched in favour of Carthage during the Second Punic War, but also from the reign of Massinissa (206 BC) onwards, explaining the changed relations with Carthage in favour of Romans. This is the reason why the Punic, Numidian and Mauritanian coins are mixed in France between the Punic Wars and Augustus.

So, paradoxically, the African currencies provides an insight into the Gaul in a particularly turbulent period in its history (Fischer, 1978). In conclusion this spatial analysis, due to the large number of coins and the extreme variety of the types, has been addressed in an exhaustive way with the use of GIS that will permit an easy fruition even to the users of this open-source database.

6.4 Alternative interpretation on the basis of rivers localization and tin, copper and amber route

As a result of the digitization of the rivers in Europe and the study of the relationship between the numismatic findings and the latter, it has been possible to note that the majority of the Punic and neo-Punic coins have been found in conjunction with the course of major navigable rivers (fig. 3). Operating a spatial selection of the numismatic finds closest to rivers (fig.4) it is noted that almost all the coins are highlighted in the map

Fig.3: GIS thematic map with punic and neo-punic coins (red dots) and main rivers

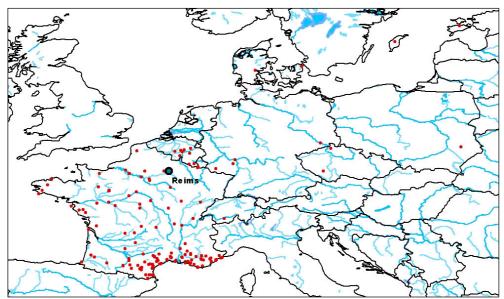
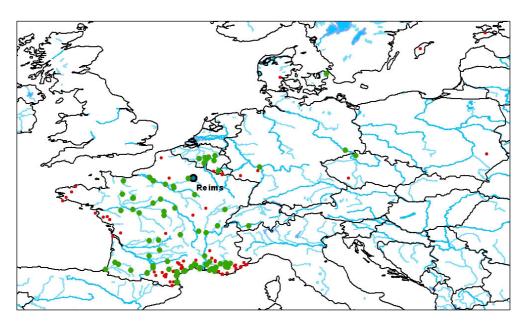


Fig.4: spatial selection of the Punic coins found near to the major rivers (green dots).



aside those already close to the coastline. Then it can be assumed that these coins could be connected with the atlantic route that carried the Punic vessels to reach the coastal lines of Northern France and Belgium and from here to follow the rivers path to reach the inland. As indicated by M. Bar other interesting findings of coins have been made in the western regions of France and particularly in Bretagne (Bar 1991), areas of great interest as they can be interpreted as evidence the frequency of an Atlantic route. In this regard, the overlap of the Punic finds to the courses of the rivers in France, reveals a remarkable and very interesting concentration of coins along the waterways from the coast to the continental areas (fig. 7). In particular, with regard to the region of Reims, we observe the presence of

Punic coins along the Seine and the Marne, reflecting a possible way of penetration by the Atlantic route, which is also observed in the recent study by J.-M. Doyen, who hypothesizes that path as a vehicle for dissemination of Ebusus coins in Belgian Gaul (Doyen, 2011). The Atlantic region in the north and south of the Strait of Gibraltar fell within an areas of influence of great strategic importance for Phoenicians (Botto, 2011; Manfredi, 2003). The frontier with the greek and roman world spread in the north and west, through the Mediterranean to Sicily, arriving to the Pillars of Hercules (Manfredi, 2003). Within this border, strongly characterized by ideological point of view, there are Punic areas which were politically and economically important and protected, off-limits for the Romans by the first Treaty with Rome (Polybius III, 22-23). This border does not have a defined path in its entirety length, but it clearly delineates the areas of influence. The oldest evidence of the knowledge of the Atlantic route by the Phoenicians is derived from the story of the journey made in the V century BC by the Carthaginian Imilcone that, according to the testimony of Pliny and Avieno, heading north, he completed a four-month expedition along the Atlantic coast and along the oceanic coasts of France, Britain, and perhaps he reached the Cassiterides (gr. Κασσιτερίδες), a name given to the islands of south-western British and Cornwall, because of the presence of tin deposits (gr. κασσίτερος)(Manfredi, 2003). In this respect interesting is the account of Strabo (Geography, III, 5.11) according to which a Punic shipowner, departed from Cadiz towards the north having noticed being followed by some Roman spy ships, preferred to let his ship aground on the rocks rather than lead his pursuers on that route. Having survived he returned to Carthage and was awarded with an entire refunds of the lost cargo (Manfredi, 2003). We do not know which goods he was carrying, but we can certainly speculate that the secrecy of the route was linked to the trade of metals that was one of the primary economic needs that determined, from the beginning of the VIII century. BC, the Phoenician frequency of the Iberian Peninsula with Cadiz, a pole of attraction and radiation in the management of the commercial network in the Far West, that goes from the estuary of the Tagus to the valley of Guadalquivir, Lixus and Mogador in Atlantic Morocco to Extremadura and the most western part of the Meseta (Guerrero Ayuso, 2008; Meds, 2008). The site on which stands the ancient Gades is, in fact, strategic and situated at the confluence of three pre-existing major commercial channels in the same Phoenician foundation: the Atlantic, the Mediterranean, the tartessian circuit (Manfredi, 2006). The Atlantic route inevitably touched Portugal where the most recent archaeological investigations have documented the presence of Phoenician settlements along the coast (Arruda, 2011) and the north of Spain. Among the findings of great interest is mentioned in particular the discovery of coins in Cadiz, Abdera and Ebusus (CALLEGARIN, 1999; MORA SERRANO, 2012; MANFREDI 2012; GONZÁLEZ-RUIBAL 2004, Ayán Vila *et alii* 2008,) in the port of Bares in Galicia. The area near this location is called Punta da Muller Marina which, according to A. González-Ruibal, could be identified, as already suggested by E. Aubet, with the *Capo Venere* mentioned in the far north of Iberia in the circumnavigation of Imilcone (Avieno, Ora Maritima, 158)(Gonzalez Ruibal 2006). Off the Spanish coast the route continued along the French coast, marked by the findings of Punic coins (Sardo-Punic type, Carthage of the IV century BC, Panormo and other ancient type) in Bidart (Pyrénées Atlantic side), Soulac (Gironde), Les Sables d'Olonne and the Ile de Noirmoutier (Vendée), Quimper and Penmarc'h (South Finistère), Ploulec'h (Côtes-du-Nord)(Fischer 1978, Doyen, 2011).

In this research perspective are of particular relevance also Punic and neo-Punic coins found on the southern coast of Britain (Doyen, 2011), and for this reason have been included in our database. At the time and based solely on published material, the samples counted on British territory are 104 (Mylne, 1948; Doyen, 2011). This aspect of the research is to be deepened but it seems interesting to repeat the data reported by JM Doyen on the discovery of Navan Fort in Ireland, in a context of the end of Hallstatt, of a vervet monkey skeleton (Macaque sylvanus) originating in the area of the Strait of Gibraltar which was for the researcher a clear indication of a "Phoenician" provenance of the animal. Those information suggest strongly a frequentation of this northern areas.

On this regard is important to note that both France and the British Isles are rich of numerous copper and tin ore deposits, some of these exploited in ancient times, suggesting a possible Punic interest in frequenting those areas (fig.5 and 6). In Provence, Vosges, in Lyon, in the Massif Central, in Armorica, in the Pyrenees (in this last are, in the Seronias-Ariege- lots of evidence of ancient pyrometallurgical operations have been found) in Languedoc (in the district of Cambrieres- Clermont l'Herault where the exploitations is attested from the Calcolitic times till the Gaul-Roman times) are localized mineral basins that could have interested the ancient Punic merchants, carrying them in those areas. In Bretagne there are also the same tin mineralization found in Cornwall (Giardino, 2002)., whose cultivation is similarly easy to do (alluvial deposits). Is interesting to note that is here (Ploulec'h, Penmarch, Quimper) that some punic coins of the ancient types (Sardo-

Punic type- 300-264 BC; Carthage 221-210 BC, Numidia 203-205, Panormo) are reported in the database (Fig.6).

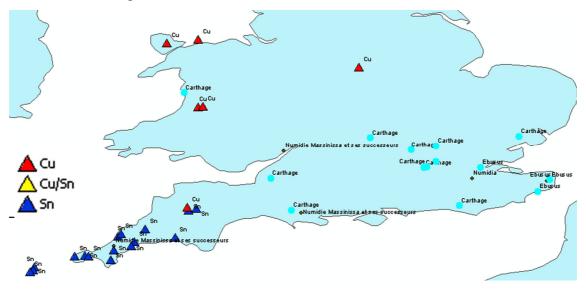


Fig. 5: ore deposits in United Kingdom, in the areas affected by the presence of punic coins.

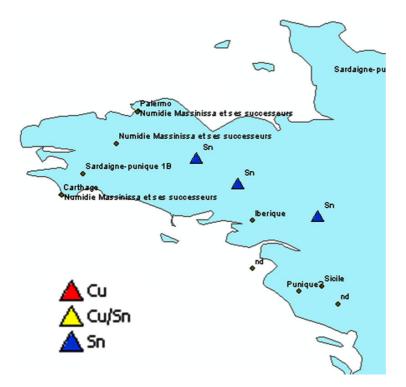


Fig. 6: coins and tin mines in Bretagne

So is possible to suggest that the frequentation of this area is to be connected with tin basins. In France, in the ancient cave of Ploermel, in Morbilhan, a huge vein of cassiterite has been exploited in roman times. Other veins are present in the Limousin area (Creuse, Haute Vienne and Charente) and in Bourbonnais in the district of Allier. This observation could partially explain the presence of foreigner coinages, especially the punic ones, since Punics used to be one of the most pushing elements in the exploitation of mines. In Cornwall a similar articulated situation is found: copper basins are present in Devonshire, Derby, Cumberland and Galles. Also in Ireland in Mont Saint Gabriel (Cork) there are some prehistoric wells, while the tin mineralizations are, beside the already mentioned Cornwall, also in Devon shire and in the Scilly islands (Giardino 2002). The tin was present also in Germany and Boemia, and Copper was widely present in central Europe (Germany, Austria, Balcan areas, Eastern Serbia, Bulgaria). Those basins are reported here to suggest other possible interpretations of these coinage findings, connecting the localization of this rich areas to the presence of Punic merchants.

Also the Baltic Amber trade, along the oceanic routes and along the river Rhine-Rhone-Saone and the course of the Danube river, can justify the presence of Punic coins in so remote areas (Poland, Estonia, Germany and the UK) as it was in the pre-protohistoric period the path for supply of this precious fossil resin. The terrestrial amber routes follows a long path which from Fratta Polesine (Veneto region, Italy) reached St Petersburg (fig.5). The findings of coins towards the Baltic region and Eastern continental Europe seem to indicate some interest, though probably indirectly, to the way of amber (fig.7). The discoveries of amber in the North African contexts and Carthage itself, dating back at least

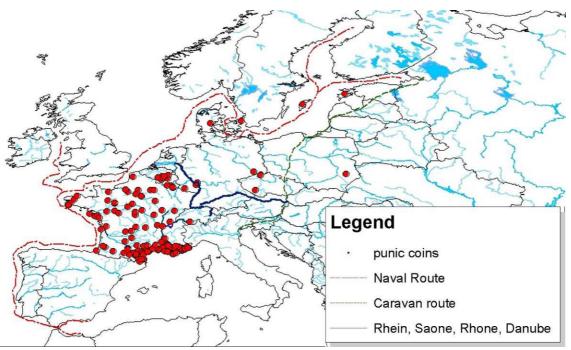


Fig.7: punic coins and terrestrial amber path (green line), river axis Rhein-Saone-Rhone-Danube and Atlantic route (red line)

to the VII century BC, have been interpreted as products of trade with the Etruscans (Fariselli, 2000). In any case, the coins found in the contexts in eastern Germany, Poland, Estonia, belong to the Punic type dating between the half of IV century. BC and 264 BC (Mielczarek, 1989).

From these excursus is thus revealed a complex and varied problem related to the distribution of Punic coins on French soil and in general in the continental Europe and the islands of the North. If a significant amount of Punic coins has been found in the south of France, those are clearly attributable to Mediterranean commercial circuits, but more difficult to understand is the dispersion of the specimens in the Atlantic and continental areas of France and in particular their presence in the region of Reims and Belgium. Because of the close relationship of these findings and these alternative routes, it is proposed, as the final conclusion, a connection with the exploitation of mineral resources and with the location of amber route (Fig.7). The rivers, especially the Danube and the axis Rhein-Saone- Rhone, shown in fig.7, could have served as connection between the Baltic areas and oceanic route, with the continental Europe and the amber route.

6.6 Some observations on simultaneous presence of different types of coins in the same archaeological context

The GIS system proved to be an excellent tool also to understand thoroughly the presence of different coin species in the same archaeological context. all the different types of context are listed in the table (only for the areas of findings of multiple coins) and the frequency of presence.

Type of context	f.	Location		
Sardinian-punic coins		Allones, Besancon, Maureuil sur Arnon,		
		Namur, Vallaurisse, Vieux		
Ebusus	4	Ambrussum, Mont Laures, Tauroeis,		
		Tolouse		
Gadir	1	Chaors		
Punic	1	Elne		
Massinissa	3	Lyon, Stradonice, Riez		

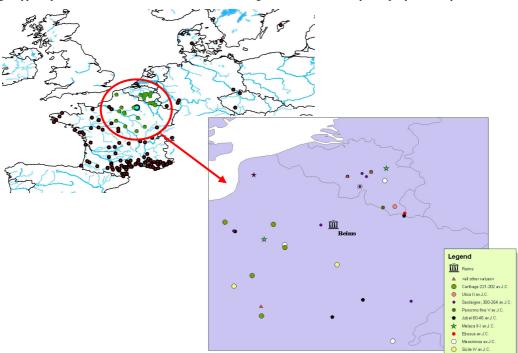
Punic	2	Coustouges, Terrats
Juba I	2	Le Mans, Lissac
Sardinian-Punic+ Ebusus	2	Agde, Narbonne
Sardinian- Punic+Ebusus+Juba I	1	Beaulieu
Sardinian-punic+ Carthage + Ebusus	1	Bollene
Ebusus+ Mozia+ Carthage + Sardinian-	1	Mailhac
Punic		
Sardinian-Punic +Ebusus+Malta+Gadir	1	Saint Pierre le Martigues
Ebusus+Punic+Carthage	1	Cazeres
Sardo-punic + Carthage + Punic	1	Marseille
Sardo-punic + Carthage	2	Rosas, Sigean
Ebusus+ Numidian+ Massinissa+ Chartage+Gadir	1	Vieille Toulouse
Massinissa+ Sardinian-Punic +Sicily	1	Flavion
Sardinian-Punic + Carthage+ Sicily	1	Luttre
Tingi+Malaca	1	Bavai
Sicily + Carthage	1	Plan de Joux
Sardinian-Punic + iberian	1	Monaco
Carthage+ Massinissa	1	Pen March
Carthage+ Numidian	1	Pitres
Panormo+ Massinissa	1	Ploul'ech
Malaca+ Punic	1	Saint lerez
Sicily + Carthage + Panormo (ancient coins)	1	Wasseiges

The most frequent mixed context contain Ebusus + Sardinian-Punic coins and the most frequent multiple findings of the same type of coins are again that of the Sardinian-Punic and Ebusus type. Also the Carthaginian coinages are often found in multiple context, indicating that these three type of emission were not only very popular, but also very easily accepted even in foreign areas.

6.7. Going forward with the research in the future

The collaboration with the institutes GEGENAA and CRESTIC the University of Reims Champagne-Ardenne will continue not only for the proposed online publication of the database, but is already developing a more complex system that describes this phenomenon and allows to make hypothesis on coinage distribution models (also to predict possible future findings). In fact, the planning of further research on Punic and neo-Punic coins in Europe will follow this path: the creation of a geodatabase in GIS environment that allows to predict the presence of additional numismatic finds, not yet been discovered, right on the basis of: the uncertainties of database data and the study of naval and land routes that led to the presence of coins currently known. We will proceed with the assignment of quality attributes to the information currently listed in the database (information of context, dating, type, composition, type of discovery). In this way it will be possible to assign a different weight to the discoveries accompanied by archaeological information, and to the unsure finds, out of archeological context. This will make it possible to draw more substantiated routes of penetration which can be monitored by archaeological surveys and prospection. Also in the near future it will be started an archeometric session with analytical non-destructive methods (XRF and SEM-EDS) on the coins held in the collections and museums closer to Reims (fig.8), with the intent to extend the analysis to those more distant and out of the French and Belgian territories.

Fig.8: type of punic coins found near Reims in France and Belgium, selected with an spatial query on GIS system.

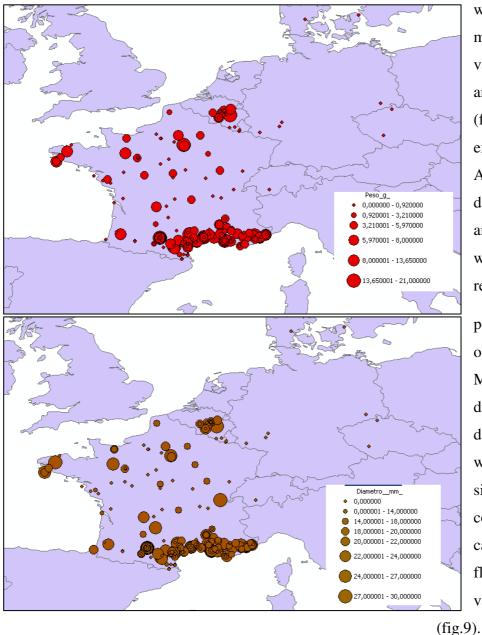


The mission of June 2013 was organized precisely to enable the Italian team to study the punic coins preserved in the collections and museums in the neighboring city near Reims, in order to lay the foundations for the beginning of the archaeometrical sessions and for the

selection of coins on which carry out compositional analyses. The proposal of archaeometric research is actually suggested for the study of punic coins in Charleville, Namur and Bruxelless.

6.7 Some Observation on Weight and Diameter values

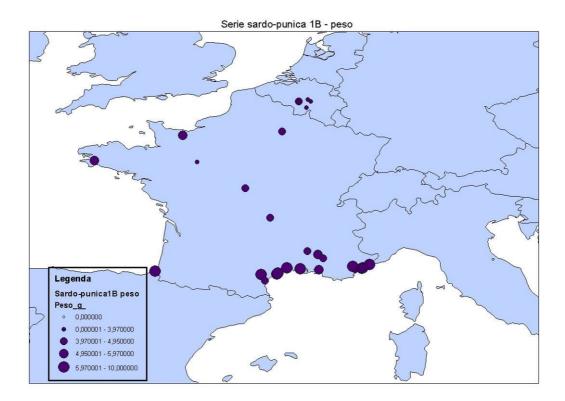
Another possibility of using GIS system is the application of symbols whose dimensional gradient allows the display of different values of selected characteristics. In the figures, it



decided to was different map values of weight and diameter (fig.9). As explained in the Appendix С different weights and diameters were used in relation to the ponderal system of reference. Mapping the different values of diameter and weight of each single kind of coin, also in this case, very high fluctuation in values is detected

Fig.9: weight and diameter gradients for Punic coins

Fig.10: weight distribution of Sardinian-Punic coins



It was assumed that even in the same type of emission of coinage existed subspecies, produced in different mints or periods. This led to the idea of analytically studying a same series of coin in order to verify the presence of these subgroups, as it will be deeply explained in the next chapter.

It was decided to proceed in this numismatic research, with the study of a type of Carthaginian coin: *male head / galloping horse*, which it was seen, from both the thematic maps and in the multiple discoveries, to be a very common discovery both in France and Belgium, but also in Italy (Sicily and Sardinia) and other areas of the Mediterranean, in order to understand if there were different mints that provided this type of coins, spread over this vast territory. Given the vastness of the presence of the *male head/galloping horse* type, it is almost impossible to imagine it to be produced only in Carthage, which was probably the issuing authority. The ultimate aim of the work that follows is also to understand, since the fluctuation of the values of weight and diameter for the same kind of coins, the possibility of existence of sub-types coined on different ponderal systems.

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CHAPTER VII

ARCHAEOMETRIC STUDY OF PUNIC COINS FOR THE INDIVIDUATION OF SUB-GROUP SPECIES

Type: male head/ galloping horse



SEM-EDS analyses on sixty coins (type: male head/ galloping or rearing horse) belonging to M. Viola's Collection.

Highlights

The study of Punic coins continues with an in-depth analysis of a particular species of coinage (male head/ galloping horse) in order to discover the presence of subspecies of coins not emitted by the issuing authority but rather by minor mints, located in strategic areas. This part of the research was carried out by performing the study of all the measurable characteristics of the coin such as the edges, the diameters, weights and compositions (SEM-EDS).

7.1 Introduction

A new procedure, different from the one employed for the georeferencing work applied on Punic coins found in France and Belgium, has been used for another part of the research, applied specifically on a particular type of Punic coins with obverse: *male head* and reverse: *galloping or rearing horse*, classified at number 126 of *Corpus Nummorum Punicorum* (Viola, 2010). The importance of the series with the galloping or rearing horse on the reverse, dated to the middle of the IV century BC, is in the emerging possibility that various mints have contributed to their issue. For this type, together with the series with *horse standing behind palm* on reverse, immensely popular throughout the Mediterranean basin, it can be supported the hypothesis that these two emissions represented the role of the basic numeral in the Punic coinage of Western areas. The intrinsic value of this series of coins is minor, but seems that the coinage had been the first in order of time to be issued under Carthage direct control.

This consideration brings to the indication that, for the galloping and rearing horse type, the emitting authority was Carthage and the chronology can be set ranging from the late IV and early III century BC (Acquaro & Manfredi 1992, Candellieri, 1989). The indication of the emission is unsure, because for this series it was also hypothesized a Sicilian provenance. In addition to these two suggested mints, it is possible to hypothesize the presence of other sub-mints in the most strategic commercial zones, that might have contributed to their emission.

The archaeometric investigation is devoted to try to carry out significative indications for the existence of different series inside the same obverse/reverse type, by considering physical factors such as metal composition, shape, dimensions, by allowing to distinguish different production centres. A previous study was carried out on the same kind of coin by the numismatic Candellieri (1989) who measured weights and studied the edges of coins and managed to distinguish between different emissions using that features. The collector and numismatic Mauro Viola has been involved in this part of investigation, by submitting 54 coins (two of them are shown in fig.1) of his collection to non-destructive analysis. *Collezione Numismatica Viola* is one of the largest and most varied collection of this type, in fact its creation began in the second half of the XIX century in Palermo by Ernesto Viola and the original nucleus consisted of some fragments of *aes rude*, 216 Roman coins, 93 Greek, 39 Punic and some medieval and contemporary specimens. The original 39 Punic coins are only 10% of the current consistency of the collection, enhanced drawing on the antiquities market.

The instrumental investigations have been performed on the already restored coins through a micro-mechanical cleaning, performed with the aid of a microscope by the owner himself. This preliminary work is necessary for the elimination of potting soil and thick layers of metal oxides, which would prevent the correct reading of features such as the execution of the edge and the faces but also the presence or absence of the exergue.

These 54 coins have been studied both from the compositional point of view (SEM-EDS) but also from the dimensional and ponderal aspect, measuring the weight, diameter and the characteristics of the faces and edges to obtain information about tecniques of production (Bar, 1991;Barello, 2008; Finetti,1987; Manfredi, 1990). In fact, since the coins showed also very different obverses/reverses between each other, also the morphological appearance and characteristics have been taken into consideration. Crossing the results obtained on the 54 coins, it was possible to perform the so-called study of the *coinage links* (in Italian language *studio dei legami di conio*) which allowed the identification of a subseries in the *male head/galopping-rearing horse* coins.



Fig.1: pictures in real size of two of the examined Punic coins (M.Viola)

7.2 Representation of the obverse/reverse

As previously stated, the almost sixty coins showed sometimes very different representation of the male head and the horse. This discrepancy can be explained starting from the production techniques of the dies. In fact, anvil and hammer dies were individually handmade, produced by craftsmen for engraving, and obviously this operation cannot be defined as "standardized" but, from the presence of same details, it is possible to derive indications of emission from different mints.

In other words each local mint produced itself new dies every time it was necessary because of their natural wear or for a breakage or owing to the necessity to pay for soldiers in a military campaign. Inevitably coins had somewhat fluctuating aspects and the type of coins were only recognized thanks to the familiar images impressed. In this regards two coins with different features of obverse/reverse can come equally from the same mint (but from different dies) or by different mints and obviously by different dies. Instead it is possible to state with certainty that two coins with the same obverse/reverse were produced by the beating of the same authority and from the same mint. In order to define the features of the coins obverse/reverse it was decided to proceed with the following lineup: high resolution photos with dimensional ruler; handmade micro-relief; use of the software ImageJ to perform precision measurements on the pictures like the size and the angles of the items; use of the software Adobe Illustrator for the execution of a digital micro-relief of the photos. Since the obverse/reverse figures depend also strongly on the beating direction and strengt it was impossible to find groups on the basis of these characteristics and this part of the chapter is reported just as an attempted step. A singularity appears for the coin number 594: it is a coin of the same species but of significantly lower module and weight, perhaps it is the lower nominal of the series "galloping horse", whose sphere of circulation is restricted to the metropolitan territory of Carthage and is rarely witnessed in the more generally Punic field (Candellieri, 1989).

Beyond this particular coin, others have similar but not identical obverse and reverse, partly because the result is also due to the force exerted on the hammer die.

In a study performed by Acquaro, Manfredi and Viola himself (Acquaro & Manfredi 1992), the coins were distinguished within the "rearing horse on the reverse" group and the "galloping horse on the reverse" group, although the authors themselves have defined it more as a formal than a real distinction. In fact, the difference in the inclination of the paws

is sometimes slight. However, for a completeness of the discussion, the distinction is reported below:

Galloping horse: 238, 318, 320, 339, 392, 410, 413, 414, 446, 554, 593, 631, 26, 240, 241,319, 322, 323, 393, 396, 397, 398, 400, 401, 402, 403, 404, 405,, 407, 408, 411, 549, 552, 553, 590, 591, 592., 594, 595, 596, 598, 608, 632, 633, 634, 641 **Rearing horse:** 239, 394, 395, 399, 412, 597

7.3 Production of the flan and rounded or truncated edge



Among the different possibility of classification of the coins, it is significative their division in relation to the edge (Candellieri, 1989): in fact, from a careful observation of the 54 samples it is possible to identify a type with rounded edges and another one with a truncated edge. The rounded edge flan were produced using a casting technique known as "*en chapelet*". This method consists of digging a series of niches, linked together by a channel, in two specular stone slabs, firmly joined together, which serve as a matrix for the molten alloy. The result is a series of flan of spherical shape subsequently used to produce the coins. This method produced flan that, on

Fig.2: rounded large numbers and facing a continuous production activities, can be and truncated edge coins considered rather standardized. The characteristic that allows the recognition of this technique is, in addition to the rounded edge, the presence of the tang of fusion, evidence of the presence of the channel in the melting matrix. The truncated coins, whose appearance is trapezoidal, testifies the subsequent use of pouring the molten metal in conical moulds which allow easy extraction by simply turning the valve upside down (fig.2). This new system made it possible to obtain more standardized diameters and thicknesses. Production technique for truncated coins, whose appearance is trapezoidal, is also interpreted (Bar, 1991) with the subsequent use to cut the flan from a metal bar. This new system made it possible to obtain standardized diameters and thicknesses.

Specificately the analysis of the edge made possible to divide the of the 60 coins into two groups:

Truncated edge: 238, 318, 320, 339, 410, 413, 414, 446, 554, 593, 631

Rounded edge: 26, 239, 240, 241,319, 322, 323, 393, 394, 395, 396, 397, 398, 399, 400, 401, 402, 403, 404, 405,, 407, 408, 411, 412, 549, 552, 553, 590, 591, 592., 594, 595, 596, 597, 598, 608, 632, 633, 634, 641

Crossing the groupings of rearing or galloping horse, and truncated edge and rounded edge, is possible to see that the data do not cross for these categories.

7.4 Weight

Each coin has been weighted with a balance with a sensibility of 0.01 g.

The complete catalogue (n.1) of the weight, the edge and the reverse is as follow (R= rounded edge; T: truncated edge coins):

Catalogue n.1

	Weigh	edge	reverse	n.	weight	edge	reverse	n.	weight	edge	reverse
n	t										
26	5.04	R	galloping	394	5.49	R	rearing	403	4.66	R	galloping
239	9.15	R	rearing	395	4.61	R	rearing	404	4.70	R	galloping
240	4.38	R	galloping	396	4.75	R	galloping	405	4.55	R	galloping
241	5.68	R	galloping	397	5.34	R	galloping	407	6.15	R	galloping
319	4.11	R	galloping	398	3.88	R	galloping	408	4.06	R	galloping
322	5.92	R	galloping	399	5.25	R	rearing	409	4.77	R	galloping
323	4.89	R	galloping	400	4.60	R	galloping	411	4.84	R	galloping
392	5.33	R	galloping	401	5.19	R	galloping	412	5.92	R	rearing
393	4.04	R	galloping	402	4.30	R	galloping	549	5.10	R	galloping

n.	weight	edge	reverse	n.	weight	edge	reverse	n.	weight	edge	reverse
552	5.09	R	galloping	598	3.49	R	galloping	320	2.96	Т	galloping
553	4.61	R	galloping	608	5.56	R	galloping	410	4.47	Т	galloping
590	5.96	R	galloping	632	5.40	R	galloping	413	4.21	Т	galloping
591	3.60	R	galloping	633	7.23	R	galloping	414	3.21	Т	galloping
592	7.06	R	galloping	634	5.08	R	galloping	446	3.18	Т	galloping
594	1.59	R	galloping	641	5.32	R	galloping	554	3.00	Т	galloping
595	6.54	R	galloping	339	3.18	Т	galloping	593	3.46	Т	galloping
596	4.49	R	galloping	238	3.91	Т	galloping	631	3.84	Т	galloping
597	5.33	R	rearing	318	2.52	Т					

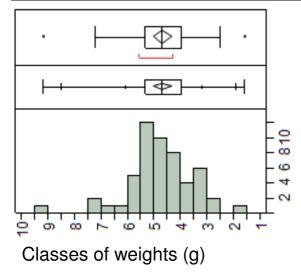


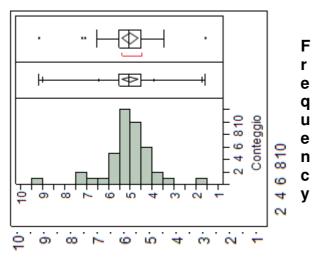
Fig. 3: weight distribution of the 54 coins

- **F** The statistical parameters of the weight
- r distribution are (fig.3):
- **q** Minimum weight: 1.59 g.
- U Maximum weight: 9.15 g.
- e Extension of the series: 8.44 g.
- n c Average value: 4.73 g
- y Median: 4.70 g
 - Standard deviation: 1.26 g

The diagram in the upper part of the frequency distribution are Box Whisker plots, whose reading method has been explained in chapter II.

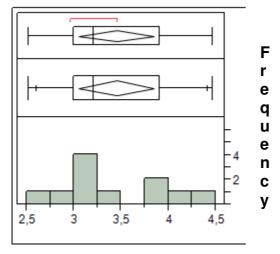
This distribution is very wide as described by the value of standard deviation.

From a thorough study of the data emerged that the weight values for truncated edges and the globular are different. Since the histogram graph seems to have a bimodal distribution with two different peaks, one around 5 grams and the other around 3 grams, the distribution were splitted into two groups, separating the lighter coins from the heaviers. Since the values of weight of about 3 grams are possessed especially by all the coins with the truncated edge, the values were separate into two different distributions for globular and



Classes of weights (g)

Fig. 4: weight distribution for rounded edge coins



Classes of weights (g)

Fig. 5: weight distribution for truncated edge

truncated edge coins.

Rounded edge coins (fig.4):

Minimum weight: 1.59 g. Maximum weight: 9.15 g. Extension of the series: 8.44 g. Average value: 5.07 g Median: 5.06 g Standard deviation: 1.17 g

Truncated edge coins (fig.5): Minimum weight: 2.52 g. Maximum weight: 4.47 g. Extension of the series: 2.5 g. Average value: 3.44 g Median: 3.21 g Standard deviation: 0.59 g

As can be seen from the separation of the two distributions, truncated coins have a lower weight (as seen also by Candellieri, 1989 using Student Test) and a much lower variability, indicating a more standardized production.

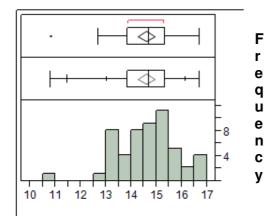
7.5 Diameter

Catalogue n.2

The diameter has been measured by the means of Image J software, by the confrontation with the dimensional ruler. Each measurement in the table below is expressed in mm

n.	Diam.	edge	reverse	n.	Diam.	Edge	reverse	n.	Diam.	edge	reverse
26	14.1	R	galloping	394	15.2	R	rearing	403	14.5	R	galloping
239	16.7	R	rearing	395	13.3	R	rearing	404	13.3	R	galloping
240	14.4	R	galloping	396	14.7	R	galloping	405	15.3	R	galloping
241	15.6	R	galloping	397	14.0	R	galloping	407	13.7	R	galloping
319	13.0	R	galloping	398	13.0	R	galloping	408	13.1	R	galloping
322	16.2	R	galloping	399	13.0	R	rearing	409	16.7	R	galloping
323	14.3	R	galloping	400	13.8	R	galoppo	411	13.4	R	galloping
392	15.9	R	galloping	401	15.7	R	galoppo	412	14.1	R	rearing
393	14.0	R	galloping	402	12.7	R	galoppo	549	16.6	R	galloping

n.	Diam.	edge	reverse	n.	Diam.	edge	reverse	inv	Diam.	edge	reverse
552	14.2	R	galloping	598	14.6	R	galloping	320	15.3	Т	galloping
553	16.1	R	galloping	608	15.8	R	galloping	410	14.7	Т	galloping
590	14.8	R	galloping	632	13.5	R	galloping	413	15.4	Т	galloping
591	13.4	R	galloping	633	14.8	R	galloping	414	15.0	Т	galloping
592	14.7	R	galloping	634	15.0	R	galloping	446	14.0	Т	galloping
594	10.8	R	galloping	641	15.2	R	galloping	554	15.0	Т	galloping
595	15.5	R	galloping	339	14.6	Т	galloping	593	13.9	Т	galloping
596	15.3	R	galloping	238	15.2	Т	galloping	631	15.4	Т	galloping
597	14.8	R	rearing	318	16.7	Т					



Classes of diameters (mm) Fig. 6: diamater distribution for the 54 coins As previously done with the weight measurement, the frequency distribution has been calculated also for the diameter values (fig.6).

Minimum diameter: 10.8 mm Maximum diameter: 16.7 mm

Extension of the series: 5.9 mm

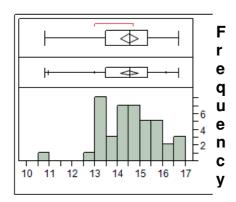
Average value: 14.6 mm

Median: 14,7 mm

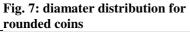
Standard deviation: 1.17 mm

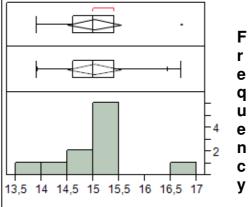
The values of coins diameter are quite variable, and this variability is an indication of how much the process of minting was not easy to control. In this case it was also decided to separate the two distributions for the truncated and rounded edge types, to see how the diameters behave.

Below, the distribution for the rounded coins is shown, and on the right side of the diagram is enumerated the statistical factors (fig.7).



Classes of diameters (mm)





Classes of diameters (mm) Fig. 8: dimeter distribution for truncated coins Minimum diameter:10.8 mm

- Maximum diameter: 16.7 mm
- Extension of the series: 5.9 mm
- Average value: 14.49 mm
- Median: 14.55 mm
- Standard deviation: 1.24 mm

The distribution is very wide and characterized by highly variable values.

The distribution of the truncated edge coins (fig.8) shows a narrower range of diameter, with a lower
standard deviation and more than the half of the population centered in the range of 15-15.5 mm.
Minimum diameter: 13.9 mm
Maximum diameter: 16.7 mm

- Extension of the series: 2.8 mm
- Average value: 15.02 mm

Median: 15.00 mm

Standard deviation: 0.76 mm

In conclusion, from the study of the values of weight (Candellieri, 1989) and diameter it has

been possible to identify two sub-species, which do not take into account the presence of the galloping or rearing horse, more classical classification, but rather the production techniques of flan. The technique of conical moulds allowed the production of a more standardized series of coins from the weight and size point of view. The discussion now proceeds with the study of composition of the coins, in order to see if this subdivision is also encountered in the type of alloy.

7.6 Composition

At this point of the work all the coins have been analyzed with SEM-EDS, on low magnification areas, in order to obtain the averaged data of the compositions. Since it was not possible to clean the coins at all, it must be considered an increase in the values of tin and lead, which predominantly oxidize and form the patina, according to the phenomenon of surface enrichment. It must however be considered the fact that the coins have all been restored before the execution of the analysis, minimizing the influence of this presence to the compositional data. In any case, the data (reported in the table) are taken into account only for a qualitative distinction of the coins in compositional groups. The values expressed for the major elements are shown in the table below (all reults are in weight %):

Catalogue n.3

n.inv	Cu	Sn	Pb	n.inv	Cu	Sn	Pb	n.inv	Cu	Sn	Pb
26	90.5	9.5	0.0	323	43.9	18.4	37.7	398	77.1	22.9	0.0
<mark>238</mark>	<mark>32.5</mark>	<mark>63.5</mark>	<mark>4.1</mark>	<mark>339</mark>	<mark>74.0</mark>	<mark>25.9</mark>	<mark>0.0</mark>	399	73.5	26.5	0.0
239	61.3	38.5	0.1	392	80.8	19.2	0.0	400	68.9	31.1	0.0
240	58.5	10.9	30.6	393	41.7	48.7	9.7	401	87.4	12.6	0.0
241	96.3	3.7	0.0	394	46.7	16.7	36.6	402	13.9	15.6	70.5
<mark>318</mark>	<mark>80.9</mark>	<mark>19.0</mark>	<mark>0.0</mark>	395	48.2	51.8	0.0	403	78.6	21.4	0.0
319	48.5	43.8	7.7	396	100.0	0.0	0.0	404	80.9	7.8	12.6
<mark>320</mark>	<mark>52.5</mark>	<mark>47.4</mark>	<mark>0.0</mark>	397	45.1	47.9	6.9	405	24.5	17.5	58.0
322	48.8	10.2	40.9	323	43.9	18.4	37.7	407	86.1	13.9	0.0

n.inv	Cu	Sn	Pb	n.inv	Cu	Sn	Pb	n.inv	Cu	Sn	Pb
408	44.1	47.9	8.1	552	78.0	21.9	0.0	596	82.3	4.8	12.9
409	76.9	23.0	0.0	553	59.7	30.9	9.4	597	59.4	40.6	0.0
<mark>410</mark>	<mark>59.9</mark>	<mark>40.4</mark>	<mark>0.0</mark>	<mark>554</mark>	<mark>63.9</mark>	<mark>20.3</mark>	<mark>15.8</mark>	598	80.4	19.6	0.0
411	60.9	39.1	0.0	590	78.2	21.8	0.0	608	87.3	12.7	0.0
412	34.4	58.2	7.4	591	94.1	5.9	0.0	<mark>631</mark>	<mark>82.5</mark>	<mark>17.5</mark>	<mark>0.0</mark>
<mark>413</mark>	<mark>82.5</mark>	<mark>17.5</mark>	<mark>0.0</mark>	592	86.0	13.9	0.0	632	86.9	13.1	0.0
<mark>414</mark>	<mark>53.9</mark>	<mark>43.7</mark>	<mark>2.3</mark>	<mark>593</mark>	<mark>81.4</mark>	<mark>18.6</mark>	<mark>0.0</mark>	633	83.1	16.9	0.0
<mark>446</mark>	<mark>53.8</mark>	<mark>46.2</mark>	<mark>0.0</mark>	594	64.4	35.6	0.0	634	90.8	6.6	2.6
549	60.7	25.2	14.0	595	88.5	10.5	1.0	641	76.3	21.5	2.2

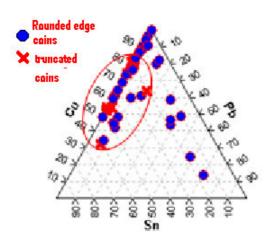


Fig.9: ternary diagram (Cu-Sn-Pb) for punic coins

In some studies carried out on similar archaeological bronze (Figueredo *et al.* 2007) it was found that the analyses performed on the uncleaned artefacts may have levels of lead of up to four times higher than the real value of the item. Moreover, in the case of cleaned areas of bronze study, it incurs the contrary result, in which lead is perceived as less than the real amount that was in the original alloy. Despite these considerations, and taking into account the intention of the work, that is to qualitatively discern groups of different alloys (binary or ternary bronze), it was decided to consider as binary bronze alloy only the samples for which the SEM-EDS analysis results to have a content of lead <2-3%. From the elemental data on the coins (fig.9), two broad groups can be distinguished, belonging to binary and ternary alloy. All the made considerations lead to observe how globular and truncated coins behave, and also in this last case, it is possible to easily separate these two types of coins. All the data underlined in yellow are coming from the truncated edge coins and, as it can be easily observed, these seem to be, for more than a 90% of the entire population, composed of binary bronze Cu-Sn. The rounded edge coins, on the contrary, seem to be composed alternately by binary and ternary bronze suggesting, also in this case, that the production of this type of coins was less standardized even from the compositional point of view. The compositional data were also plotted by the statistical method of multivariate analysis PCA (fig.10) in order to further verify the compositional trend of the rounded and truncated coins.

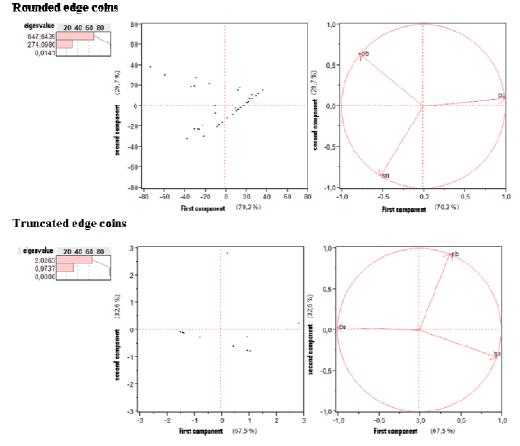


Fig. 10: PCA plot for rounded and truncated coins

The application of this method is very useful to get in order to obtain the description of compositions and the division into macro-groups of coins. In fact, since the system is constituted by more than three components (three numerical components of composition ie Cu, Sn, Pb and a nominal variable ie the type of edge), this technique can be used to find connections in multicomponent systems. The plot in the figure 10 (the plot is elaborated on convariance matrix since only the major elements are included) have the advantage of being easily readable, thanks to the location of the original variables (Cu, Sn, Pb) in the graph next to each plot. The cloud of points will have a trend that is positioned towards the element that characterizes it most. So also in this case it is possible to see how much the rounded coins compositional values are distributed both toward the tin than towards the lead (as it is supposed to be a non-standardized production), while for the truncated coins there is a trend towards the tin only (the composition is considered more controlled). In conclusion both from the PCA plot and from the triplot in fig. 3 is possible to notice a partial overlapping of the rounded edge and truncated edge coins compositions for the binary bronze alloys areas of the plots, as the rounded coins are placed on broad compositional range, covering also the narrower range of truncated coins, while almost only for the rounded coins the ternary composition have been detected.

7.7 Conclusion

The rounded edge coins have very wide range of composition, weight and diameter: the average weight is 5.07 g with a standard deviation equal to 0.6 and the average diameter is 1.47 cm with a standard deviation of 0.12, while the composition is both binary and ternary bronze.

These are typical characteristics of a not standardized production. On the contrary the truncated edge coins have very narrow range of composition, weight and diameter: these coins are composed of binary bronze Cu-Sn, the average weight is 3.4 g. with a standard deviation of 0.4 g, while the diameter is 1.5 cm and the standard deviation is 0.07 mm. In this case the production is controlled and standardized. Therefore, as the coins produced by using conical moulds were also more compositionally homogeneous, it is possible that this series of coins was more recent and based on improvements in the production system. Since the truncated edge coins are lighter than the rounded edge ones, it is possible to assume that this was a separate emission, probably minted during a period of devaluation of the coins.

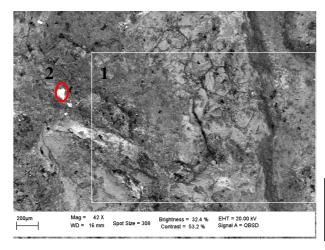
Is to be held in high regard, however, that the 5 g coins were produced according to the monetary Punic foot, while 3g followed a fraction of the Italic foot. Then this may indicate a change of weight system, that can give clues to influence changes in the period of coinage. Therefore, in conclusion, it can be said that this type of widespread type of coin underwent a change in its emission, thus indicating a passage of weight unit, which could suggest the presence of more than one mint under the direct control of Carthage. This observation also comes from the widespread geographical diffusion of this coinage type, which could be interpreted with the presence of other sub-mints, located in strategic areas.

On the other hand, starting from the fact that the truncated edge coins don't contain lead in the alloy, it is obvious to expect that these subgroup weighed less and therefore, especially considering that the diameter instead had not changed, it can be stated that this modification in composition was due to a technological improvement, because of the use of this new kind of moulds. In fact the presence of lead would have caused an increase in the fragility of the bar, making it much more difficult to hammer the flans.

From this last point of view, the truncated edge coins were produced by a more skilled procedure of emission, that can be considered as more recent.

In general conclusion, studying all the measurable characteristics of a numerous series of coins, it is possible to distinguish different subspecies, which are the ones that give the most interesting information, that can be used and interpreted in the light of archaeology, history and numismatics.

Particular details revealed during the study:



From EDS analysis and from backscattered images observation, two particular coins emerged, one for its particular superficial composition, the second for a peculiar pattern on its obverse.

S	Fe	Cu	Ag	Sn	Pb
1	1.8	66.5	nd	28.3	3.4
2	nd	5.1	94.9	nd	nd

Fig. 11 and tab. 1: SEM-EDS analysis of coin n. 533

Coin n. 553, (shown in the picture BSE

images, EDS table), shows a 100 micron particle of metallic silver, whose presence has been detected sporadically on the whole extension of the coin. These microparticles may suggest the presence of a silver coating, probably produced for depletion silvering, which was probably removed during mechanical cleaning by the restorer. This observation might suggest the presence of a higher value numeral which, together with the lower module currency represented by the coin n. 594 mentioned previously, might indicate further sub-type of the male head/galloping horse species.

The next BSE photos show a fabric texture imprinted on the patina of the coin n. 497; this is a curious example, suggesting that this coin had been kept in a tissue bag that was than destroyed during burial, while leaving its visible trace (fig.12). The burial of hoards of coins was very common saving way, whose archaeological discovery is then of great interest for numismatists studies, as explained in the previous chapter on French and Belgian Punic coins.

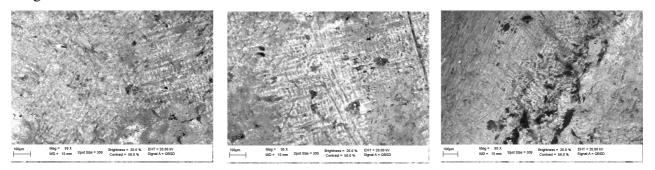


Fig. 12: SEM-EDS analysis of coin n. 497

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Final Conclusions

The thesis is configured as a setting out of analytical and research protocol applied to as many as possible classes of bronze objects, arranged as a collection of contributions to the study of ancient bronze metallurgy in areas influenced by Phoenician and Punic presence, aimed to interpret a wide range of metal production steps and their evolution between the Late Bronze Age and Iron Age.

In addition to the achievement of results from each case study, explained in each chapter, it was possible to validate the analytical protocol, which has shown to have good flexibility and adaptability, allowing to observe the problem from different points of view. The rapidity of response to archaeological problems makes the procedure suitable to be applied to similar field of research: for this reason it is ultimately proposed as an archaeological research tool.

The protocol has been internationally accepted and promoted during the sessions of collaborations with foreign partners.

In the detail of each single contribution, the conclusion, fully explained separately in each chapter, can be summarized as follows:

Votive artefacts from Portuguese area: using a process of analysis/comparison of analytical data, it was possible to support hypothesis and suggestions about provenance and chronology for the collection in the Museum of Evora, whose origin was lost. 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 is considered as influenced by Phoenicians. This kind of ternary alloy was observed in similar studies for the Portuguese area (P. Valerio and E. Figuereido) in connection with Phoenician presence. Furthermore it can also be stated that the type of metallurgy was already distinguished in relation to the type of artefact. In fact lead, often present in artefacts of decorative adhibition, is not present in the tools or in the weapons, whose liquation could make the alloy less resistant. 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, who probably acquired knowledge from the Phoenicians metalworkers.

Bronze pear-shaped jar from Alcacer do Sal: this jar seems to be a particular production in its kind, since the presence of a welding with brazing alloy has been detected,

while other similar jars do not show it commonly. Also in this case it would seem that the site of Alcacer do Sal was a highly frequented area in which the metal work, characterized by a blend of local and non-native technical skills, created a very complex bronze production.

Sardinian votive bronze statuettes and artefacts: being highly developed even during the Iron Age, Sardinian bronze production interfaced itself in a slightly different way with the Phoenician technology transfer. In fact, ternary alloy seems to be used also for production of typically local *bronzetti nuragici*. A good connection between the type of alloy and mining areas has been instead found, which exploitation was highly favored by the huge demand of Pb, Sn and Cu, fueled from the Phoenicians.

Sardinian ingots: the presence of secondary type of ingots (Cu-Sn-Pb) proves that the addition of lead was already planned since the preliminary stage of the production of semi-finished objects.

Sardinian tools: also for Sardinian bronze production, only votive or decorative artefacts show highly leaded bronzes, testifying a differentiation and a control of the alloy phases.

Moroccan ancient mines: Morocco is not commonly included among the exporters of lead and minerals in the Iron Age, so its role in commercial trade in the Circle of the Strait of Gibraltar is not generally recognized. However, since its coastline is studded with Punic sites, and mines appear to have been exploited in ancient times, it can be assumed that Morocco was one of the supply point for minerals and half-finished materials. Through the application of GIS system it has been possible to localize interesting sites which suggest a Punic frequentation. Especially in Aouam mine, the superficial prospection has allowed the collection of slags and one tuyeres fragment, whose analytical study defined as smelting slags of argentiferous galena. Since all the samples came from superficial surveys we need to proceed in the future with a archaeological excavation to obtain samples from precise stratigraphic layers, allowing a more precise chronology of the findings (in the case we will still have possibility to go on in this study. In any case, currently the necessary concessions have been given for archaeological digging from the manager of the mine).

Volubilis artefacts: in the stratigraphic phases of Volubilis (almost from Roman period) fragments of bronze and iron artefacts have been found, along with the presence of small slags coming from metallurgical operations on iron ores, attesting the presence of metallurgical workshop in the city. The Pb isotopes fingerprint of one lead leaf, firstly

attributed to an Iberian basin, proves a more convincing connection with Moroccan basins, precisely with the argentiferous galena mine of Aouam, emphasizing even more the need to perform an archaeological excavation of this area.

French and Belgian Punic coins, and Punic coins from private collections: this part of the research shows how important it is to apply georeferencing and statistical means to numismatics. In fact, a study on a large number of coins allowed the identification of subspecies, which can give very useful information on the issuing authorities. Moreover, the presence of these coins in a wide area of central and northern Europe has also allowed us to study the mechanics that led to these findings, so far away from their motherland.

This research also highlighted the need to integrate the archaeological and archaeometric work with a different approach, unifying the study of each single case study in a more general vision. Beside this, it has been identified also the need to make common habit of inserting the GPS coordinates of the sites of archaeological settlements.

This analytical research applied to the Phoenician-Punic bronze production is far from being fully completed. Since the doctoral thesis has allowed us to activate two bilateral agreements with CNR and CNRST (Morocco) and FCT (Portugal), hopefully the research will proceed in the forthcoming years with the isotopic study of all the artefacts from these areas and with the study of other types of materials.

Another desired perspective for future development is to study artefacts from Lebanon and Palestine, other geographical area that currently has been interested by few archaeometallurgical research. This will make it possible to close the circle of the Mediterranean Basin and get an overview of the transfer of technology between local and Phoenician-Punic culture.

Appendix A

From the extraction of ores to the production of artefacts: a brief *excursus* on pyrometallurgical and mechanical operations

In order to clarify some recurrent points and arguments of this thesis, a brief excursus of the pyrometallurgic cycle and some basic mechanical operations will be discussed in this appendix, starting from the extraction of the ores, going through the discharge of gangue and production of slag, till the alloying and the modelling of the object (Giardino, 2002; Hauptman, 2007; Rothemberg, 1990). In this way the Appendix provides a sort of glossary to refer to during the reading of the thesis.

1) Extraction of the ore

Metals are generally scarce on the earth crust: thus the deposits where metal ores have naturally accumulated have had a great importance throughout the times. In the epigenetic deposits the mineralization has often the form of veins, tabular or lenses shaped deposits, which had produced in the cavities or fractures of the rocks, while the singenetic ones are often connected with erosive phenomenon producing alluvial reservoirs. The ancient extractive activities were focalised on the superficial deposits, easier to be exploited with the technologies available for that time. The ores are constituted by four different components: the sterile inclusions (the embedding rocks), the gangue (non metallic minerals such as quartz, calcite, barite, fluorite), other metal ores (like the zinc sulphur), and the exploited ore. The deposits could be cultivated using both open pit or galleries. The oldest techniques for the exploitation of metalliferous deposits are based on the experiences of Neolithic flint mining, whose deposits were achieved through pits or tunnels. An area was chosen through careful observation, in such a way to find products of alteration of the mineral or through the observation of the vegetation. If the place looked promising were carried out surveys of prospection. Once found the deposit, the rock could be weakened by fires (firesetting). Before being sent to the smelting furnaces, the mineral needed to be enriched before, by mechanically removing as much as possible the gangue. Minerals were crushed and the obtained pieces were hand selected, taking into account the colour and weight. The selected pieces were then grounded and the powder was subjected to washing, separating the metal from the rocks using their difference in specific weight. Grinded mineral reach very small dimensions, often millimetre size (Giardino 2002).

2) Extraction of metal from the ore

To efficiently extract the metal from the ore, it was necessary to have a furnace in which could be obtained, by appropriate supply of air, the sufficient temperature to reach the melting point. The fuel was made from charcoal that also provided a reducing atmosphere.

The procedure can be summarized in that way:

Ore + heat + reducing agent = metal + slag + gas

This are the general lines of the pyro-metallurgical procedures; now the extraction will be explained in detail for copper and tin (while lead is fully explained in Chapter V) starting both from oxides and sulphurs.

2.1) Copper Oxides

The following minerals are defined as oxides, in modern terms:

- malachite: CuCO₃*Cu(OH)₂
- azurite: 2 CuCO₃*CU(OH)₂
- cuprite: Cu₂O
- tenorite: CuO
- chrysocolla CuSiO₃*2H₂O
- antlerite: Cu₃SO₄*(OH)₄

Upon heating to relatively low temperature (400-500°C), malachite and azurite are converted into oxide CuO:

 $CuCO_3*Cu(OH)_2 => 2 CuO + CO_2 + H_2O$

Carbon monoxide is the main reducing agent(Lo Schiavo et al., 2005).

 $C+1/2O_2 =>CO$ $C+O_2 => CO_2$

 $CO_2 + C \Longrightarrow 2 CO$

 $CuO + CO => Cu + CO_2$

2.2) Copper sulphides

The most common copper ores are sulphides. Oxide ores are the results of weathering by atmospheric agents of sulphide veins exposed at the surface. Thus oxides and sulphide ores are usually bordering of the same orebodies. The most common minerals are:

- chalcocite Cu₂S
- covellite CuS

- bornite: Cu₅FeS₄

- chalcopyrite: CuFeS₂

The most common sulphide is chalcopyrite, where copper is accompanied by a significant amount of iron and sulphur, which complicate the extraction process. The metallurgical treatment of chalcopyrite was carried out in two stages:

- ore roasting on a open fire

- smelting of the oxides discussed in the previous point 2.1.

Nowadays the flowsheet for metallurgical processing of chalcopyrite differs, yielding an intermediate product known as matte, a Cu rich copper-iron sulphide.

The main reactions taking place in the roasting process can be schematized as follows:

 $CuFeS_2 + 3 O_2 => CuO + FeO + 2 SO_2$

The oxide is then reduced following the previous reaction.

The iron oxide shows a chemical affinity for silica and the combination product is a fluid slag that flows to the bottom of the furnace. The product of the roasting process can then be fed to the smelting furnace, where the iron oxides combine preferably with the silica, to form the main constituent of the slag; the remaining copper sulphide may react with the copper oxide producing metallic copper and sulphur dioxide. In other words, in order to separate the gangue from the metal, this waste material need to be combined to form a molten slag. If the mineral is rich in gangue, so much that the iron oxide already present in chalcopyrite is not enough to combine with silica to produce the fluid slag, the slagging can be achieved adding a flux to the furnace. The fluxing agent was composed of iron or manganese ores and quartz rock (in the other case, if the iron oxide was to much in the gangue), bones, limestone or ash: these were all compound available in antiquity. The final result of this operation was a lowering of the melting point of the slag (Lo Schiavo *et al.*, 2005). For a complete explanation of the slagging and fluxing process refer to the Chapter V, were these process are fully rendered for iron and lead ores.

The metal obtained through these procedures usually underwent to refining operation, in order to eliminate other metals present as impurities, along with other elements such as oxygen and sulphur. This operation is called fire refining since the procedure is done in furnace using crucibles (Chapter V). Summing up, copper refining can be carried out as follows:

- the raw copper was placed in a clay or stone crucible and heated to the meting point;

- the molten copper was stirred and blow onto the surface. Granulate silica was added (it floats on the surface); the slag formed on the surface was skimmed off;

-the copper melt was then covered with a layer of charcoal and stirred with a stick of fresh wood.

Essentially the refining techniques relies on oxidation of the metals. The produced slag was then removed from the surface. Volatile components dissolve in the fumes. Part of copper oxidises during the process but it is reduced once again to metal in the final phase by charcoal addition.

2.3) Tin

Cassiterite (SnO_2) is probably the only tin-bearing mineral used by the early civilization. As cassiterite is an oxide, it can be metallurgically processed without a preliminary treatment required by sulphide ore. The chemistry of the extraction can be summarized by the simple overall reaction:

 $SnO_2 + 2CO \implies Sn + 2CO_2$

If necessary the tin can be refined carring out the following steps:

- metals with a higher melting point than the tin remain on the bottom of the bath, heated by temperatures slightly above the melting point of tin, and separated tin is then tapped from the top of the crucible;

- the metals with low melting point can be oxidised by blowing air on the bath, transforming them into slags (Lo Schiavo *et al.*, 2005).

3) Alloying

The most reliable method for the production of bronze consist in adding metallic tin to the copper melt, but bronze might also been produced by cementation or co-smelting.

Cementation was probably accomplished by spreading a layer of cassiterite mixed with carbon on top of the copper bath. Blowing air into the melt generates carbon monoxide, creating a reducing atmosphere which transforms the tin oxides into the metal.

Co-smelting was accomplished smelting copper and tin ores together in the same furnace.

4) Shaping the object

Smelted metal was placed in crucibles inside a furnace. Once liquid, the metal could follow different paths:

- be poured in matrices made of clay, stone or metal;

- undergone the lost wax procedure.

The simplest method to perform lost wax procedure, the so-called direct method, starts first of all giving shape to the object in the malleable wax. Then sticks of wax are applied on the model, so as to achieve the drains of the wax, while a casting cone was inlaid to allow the pouring of molten metal. The model is covered with clay which should reproduce the details to the negative. The form of clay is placed in the furnace, in order to allow the wax to go out from the previously created channels. The cavity thus produced can accept the molten metal dripping from the main entrance cone. To produce hollow objects, the wax was spread on a clay core and to prevent the collapse of the core during the melting of the wax, the core was fastened with metal nails.

Vases, jars and bowls were often produced by hammering a flat and very thin sheet, exploiting the metal ductility (Giardino, 2002).

These are brief outlines of the very complex pyro-metallurgical procedures which also in ancient times took place, using furnaces, crucibles, tuyeres and specialised equipment, near the orebodies and mines (for large scale operations) or inside settlements (for small scale operations). The whole production cycle was composed of delicate operations, whose proper conduct required a high degree of specialization, a fundamental clue that describes the type of skill levels and technology available in a given area. This brief appendix wants to give an idea of how metallurgical operations should have occurred during ancient times.

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Appendix B

Multivariate analysis with Principal Component Analysis (PCA) Method

PCA analysis is a technique used in the field of multivariate statistics for the simplification of the original set of data (D'Adria, 2006; Drennan, 1996; Sadocchi, 1993; Smith, 2002).

As explained previously in a brief way in the Chapter I, the aim of this method is to reduce the number of variables in a lesser number of latent variables, trough a linear transformation of the original variables that projects these in a new Cartesian system, where the variables are arranged in descending order of variance. These new directions or linear combinations are computed iteratively in such a way that the first component is characterized by a greater explained variance, the second component will reduce the residual variance at a slightly lesser extent than the first component, and so on for the third and subsequent

For *n* original variables, *n* principal components are formed as follows:

- the first principal component of the linear combination of the standardized original variables has the greatest possible variance;
- each subsequent principal component is the linear combination of the standardized original variables, that has the greatest possible variance and is uncorrelated with all the previously defined components.

Therefore, the variable with greater variance values is projected on the first axis, while the second will be put on the second axis and, in the case of three-dimensional plots, also a third axis which exits from the two-dimensional plane is to be taken into account. Considering only the principal variables, the reduction of the system complexity occurs.

In order to reduce the dimensionality of the data set, a loss of information contained in the original system takes place, but PCA method permits to control the balance between this losses and the required simplification. PCA is a way to picture the structure of the data as completely as possible, by using as few variables as possible.

JMP software offers several types of elaborations to help interpret the extracted components. Principal components can be accessed through the Multivariate platform or the Scatterplot 3D platform or through principal Components command on the analyze-Multivariate Methods menu. Matlab language of technical computing has been also applied

to a wider database of Iberian analytical data, in the occasion of a tutoring towards a student (Matlab elaboration carried on by V. Graziani) who will graduate in Science Applied on Cultural Heritage (Sapienza University of Rome).

Each principal components, in a more detailed point of view, is calculated by taking a linear combination of an eigenvector of the correlation matrix, with a standardized original variable. The eigevalues show the variance of each component.

The method

Principal components analysis, regarding *p* variables (X1,X2,Xi,..., X*p* with i= 1,2, ...,*p*) allows to detect other p variables (Y1,Y2,Yi,...,Y*p*), being each a linear combination of the original variables *p*.

The original data are considered as a matrix, where the columns represent the observations and the rows are the variables considered for the phenomenon under analysis (matrix X). the goal of PCA is to identify appropriate linear transformations of the observed variables Yi, capable of synthesizing the information inherent in the initial matrix X. From the linear combination of the formers the new matrix Y is obtained:

Y = LX

Where the i-th component of Y is:

 $Yi = l_i^T X$

which corresponds to a variance and a covariance (the variance is a measure of how far a set of numbers is spread out and the covariance is a measure of the linear relationship of two random variables):

Var (Yi)= $l_i^T \Sigma l_i$

Covariance can be expressed as below:

 $Cov(Yi, Yj) = l_i^T \Sigma l_j$

L is the linear transformation matrix, while Yi are the principal components.

The multivariate vector is such that the first element Y1 includes the possible greater variability (and more information) of the original variables and that Y2 represents the greater variability of Xi and Yp takes into account the smallest fraction of the original variance. To have a unique solution, it is necessary that the vector l must have unit norm.

To individuate the first principal component the bounded, maximum problem shown below should be solved.

 $Var(Y1) = l_1^T \Sigma l_1$ where $l_1^T l_1 = 1$

Maximizing the objective function with respect the means to find the appropriate vector of weights to be assigned to the matrix X, so that Y explain the maximum possible quote of the total variability. From Rouchè-Capelli theorem it is possible to identify a linear system that admits solutions if the matrix is singular. It follows that the problem of the maximum bound will result in a problem of eigenvalues and eigenvectors as the vector l is nothing but the eigenvector of unit norm of the matrix. The variance of the first principal component will therefore be maximized, as it is chosen for the biggest of the eigenvalues.

The criteria used for choosing the number of components are three:

1) take only those components that represent the 80-90% of the total variability;

2) follow the rule of Kaiser namely take only those components that have an eigenvalue greater than or equal to 1, or the components that have a variance greater than the average;

3) the choice of the number of components can be made through the graph of the eigenvalues or screeplot. The plot is constructed by placing on the horizontal axis the serial numbers of the eigenvalues (1,2, K, k), and on the vertical axis the eigenvalues corresponding to these. Inside the graph you choose the number of components corresponding to the point of elbow of the polyline. The number of principal components to be used will be given by the smallest eigenvalues, such that to the left the trend of the plot is strongly decreasing, while to the right the trend must be almost constant or slightly decreasing.

In the case in which the source data are defined by units of measurement not comparable with one another or from very different amplitude, the correlation matrix, and not the covariance matrix, is used. Often it may happen that the available source data are characterized by units of measure that cannot be compared with each other, or from very different amplitudes of the subsamples. In such cases it is not possible to work with the known sample, but it is necessary to standardize the random variables. The process of standardization allows to normalize the variables and thus obtain a new sample, that will have zero mean and unitary variance. For this new sample, the covariance matrix coincides with the correlation matrix of the sample of origin. The PCA is normally performed on the covariance matrix, but if the components of X are very different among themselves, can be useful to calculate also the PCA on the correlations, to see if the plot is better elaborated.

Interpretation of the principal components

The coefficient, associated to the variables in the loading plot, represents the weight that the variable Xi has in the determination of the principal component Y. The larger the value (considered as absolute value), the greater will be the weight that the values of Xi have in determining the principal component Yi. In other words the principal component Y will be better characterized by variables Xi, which correspond to larger numerical coefficients, in terms of absolute value. In this way the numerical coefficients give significance to the principal component Y. This means that, to determine the meaning of the main components, the variables Xi with which is more closely related will contribute more. Furthermore this type of analysis can also be performed graphically or: if we consider in the plane of the first two principal components a circle of unit radius, it is possible to assess the coefficient of correlation between the different X and Y. Each variable X is plotted inside the circle, with the following coordinates (Corr (Y1, Xi) Corr (Y2, Xj). In this way it is possible to have a graphical indication of which variables determine one, the other or both of the main components and which ones are positively or negatively correlated. From the geometric point of view the data matrix X is representable as p points in dimensional space R (as shown in the chapter III). The principal components identify a new coordinate system, that is likely to have on the first axis (Y) the maximum variability of the system, on the second a variance lower than the first and so on.

Some examples of the passages reported in this appendix, coming from the database collected for Portuguese bronze artefacts, are shown in pictures 1 and 2.

Type of object: "orientalizing"

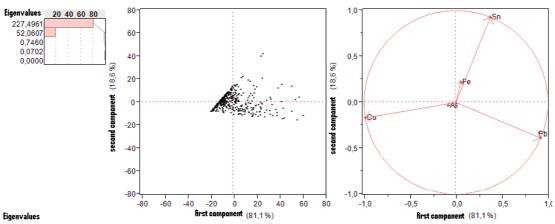
Correlations

	Fe	Cu	As	Sn	Pb
Fe	1,0000	-0,1084	-0,0391	0,1825	-0,0600
Cu	-0,1084	1,0000	0,0869	-0,5233	-0,8429
As	-0,0391	0,0869	1,0000	-0,0731	-0,0775
Sn	0,1825	-0,5233	-0,0731	1,0000	-0,0125
Ph	-0.0600	-0.8429	-0.0775	-0.0125	1 0000

Covariance matrix

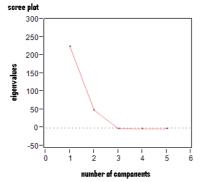
	Fe	Cu	As	Sn	Pb
Fe	0,58832	-0,98213	-0,00715	0,86564	-0,46468
Cu	-0,98213	139,50324	0,24477	-38,23192	-100,5340
As	-0,00715	0,24477	0,05684	-0,10787	-0,18660
Sn	0,86564	-38,23192	-0,10787	38,25676	-0,78261
Pb	-0,46468	-100,5340	-0,18660	-0,78261	101,96787

Plat



п.	eigenvalues	%	20 40 60 80	cumulative :	% Chi-squai	red DF	Prob/ ChiSquared
1	227,4961	81,141		81,141	17368,8	14,000	<,0001*
2	52,0607	18,568		99,709	15678,3	9,000	<,0001*
3	0,7460	0,266		99,975	11710,5	5,000	<,0001*
4	0,0702	0,025	(1) (1) (1) (1)	100,000	10519,2	2,000	<,0001*
5	0,0000	0,000		100,000	0,000	0,000	

eigenv	eigenvectors										
	1 principal	2 principal	3 principal	4 principal	5 principal						
Fe	0,00266	0,02349	-0,86173	0,23846	0,44721						
Cu	-0,77176	-0,27509	0,28317	0,22029	0,44721						
As	-0,00141	-0,00100	-0,01541	-0,89429	0,44721						
Sn	0,15338	0,79408	0,31429	0,21709	0,44721						
Pb	0,61713	-0,54148	0,27969	0,21846	0,44721						



Type of object: local

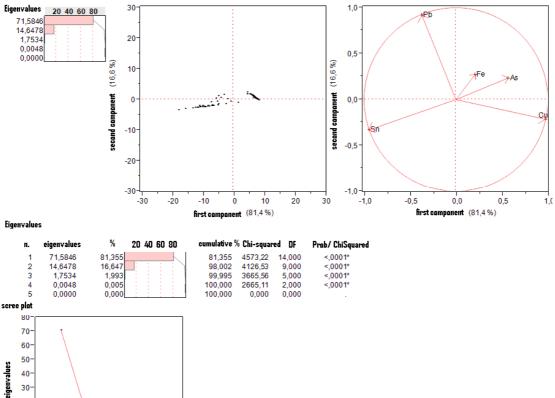
Correlations

	Fe	Cu	As	Sn	Pb
Fe	1,0000	0,1501	0,0985	-0,2877	0,1845
Cu	0,1501	1,0000	0,4471	-0,8466	-0,5465
As	0,0985	0,4471	1,0000	-0,6413	-0,1094
Sn	-0,2877	-0,8466	-0,6413	1,0000	0,0529
Ph	0 1845	-0 5465	-0 1094	0.0529	1 0000

Covariance matrix

	Fe	Cu	As	Sn	Pb
Fe	0,00440	0,06204	0,00940	-0,11540	0,03955
Cu	0,06204	38,86791	4,01272	-31,92680	-11,01587
As	0,00940	4,01272	2,07250	-5,58525	-0,50938
Sn	-0,11540	-31,92680	-5,58525	36,59377	1,03368
Pb	0,03955	-11,01587	-0,50938	1,03368	10,45202

Plat



eigenvectors

Fe Cu As Sn Pb ò

1 principal

0,00164

0,71792 0,09669

-0.67468

-0,14158

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5 principal

0,44721

0,44721 0,44721

0 44721

0,44721

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3

number of compo

ż

2 principal 0,00477

-0,34802 0,08862

-0.52021

0,77484

4 5

nents

3 principal

-0,00481

-0,33600 0,85706

-0,15995 -0,35629

6

4 principal

0,89440

-0,22488 -0,21965

-0.22046

-0,22941

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Appendix C

Punic and Neo-Punic Numismatics

The ancient coin is an exceptional source and an interesting technological indicator for the archaeometric research and for the understanding of ancient world, providing an indispensable contribution in a huge number of ways, especially from the point of view of history, politics and economics. In fact there are many points of view through which is possible to study coinage:

Appearance: metal, weight, minting techniques;

Economic aspect: the study of monetary reforms, weight systems, use of the coins, currency circulation;

Administrative aspect: a study of financial, fiscal characteristic of the emitting authority; Typological aspect: the study of coins as a historical, political and artistic document (used as a propaganda tool, a source for historical and religious information);

Archaeological aspect: dating of archaeological levels.

The Phoenician coinage (Bar, 1991; Barello, 2008; Breitenstein 1983; Finetti,1987; Manfredi, 1990; Manfredi, 1995; Viola, 2010) includes the production of coins from the independent cities of Phoenicia, who began their minting activity around the middle of the V century BC and ended in 333 BC simultaneously with the conquest of Alexander. This type of coins production was "palatial", linked to the emission of each indipendent city and for this (in order to keep visible the identity of the coin) that kind of emission was always accompanied by an inscription of the issuer city. The use of the Phoenician alphabet for the legends on Phoenician coins is attested until the second-first century BC. The Phoenician coinage, therefore, was created and developed during the Persian domination and is maintained even in the Hellenistic period, until the Roman conquest.

The mint of Carthage started to be the only issuing authority from the middle of 410 to 146 BC, because it is in this period that this settlement gained all the African inland and became a power. The coins of this period, of Carthaginian brand, in fact do not need a legend with the indication of the city so far, being the only official mint. Given the widespread use of these coins, however, it is possible that there exist other subordinate mints, located in all areas of Punic domination. Also the large span of time suggests that the numismatic Punic coinage was a transversal production, not attributable to a single political and cultural

entity. The oldest representations of the Punic coinage was developed in Sicily and was nourished by the contacts of the Greek world showing this influence, while the later types, the neo-Punic coins, keep alive the tradition of Carthage in a period and a culture that is already Romanized. Large is also the geographical area covered by the Punic numismatic: attestations relate to the most eastern coinage of Tripoli emporia, today's Libya, and the most western was that one in the south-western coast of the Iberian Peninsula, in the north and south to the High Guadalquivir, North Africa and the southern borders of today Sala in Morocco. The Punic coinage coincides with the limits of Phoenician expansion in the West. The main difference, therefore, between the Phoenician and the Punic coinage is that the first is a coinage belonging to autonomous cities that recognize the individual power of each other, while the second assumes the characteristics of an imperialistic coinage type. The neo-punic coinage started to circulate near the date of Chartage's fall, at the end of III century BC, when its power started to decrease. As soon as the autonomy reappeared, autonomous cities produced their coinage and, as a consequence of this, the legend of the city restarted to appear on the coin, indicating the emitting authority (coasts of Algeria, Morocco and those of Spain). These kind of coins begin to be produced in a period when Carthage had still power, the oldest were the silver ones of Cadiz and Iol Caesarea, belonging to the late III century BC. From the II to the I century BC all the emissions from independent cities started to increase. North Africa, after the fall of Carthage, is divided into two parts: Numidia (now Algeria and western Tunisia) and Mauritania (Morocco and part of Algeria). Massinissa, king of Numidia, plays a key role in the fall of Carthage since, with his help, the Romans manage to complete the destruction of the empire capital. The intent of Massinissa was to replace Carthage, and let the Punic culture to survive, this is why the Phoenician-Punic culture and language is maintained, and neo-Punic coins always showed the legends in Phoenician characters.

Punic numismatics, in its alternatingly autonomous and imperial character, covers a considerable range of time permitting, from this study, to obtain a number of conclusions from the historical and archaeological standpoint.

Despite this, the study of the Phoenician and Punic coins always suffers from lack of "visibility", due in large part to the reluctance to recognize the Phoenician and Punic cultural identity, saw as something autonomous and provincial with respect to the Greek and Roman world.

Anatomy of the coin

A coin is a piece of metal (usually circular and flattened) of a particular type, weight and model, issued by a competent authority. The fundamental characteristic are:

Type of coin: the classification of coins (depending on obverse/reverse on the sides of coin);

Field: the space occupied by the type;

Exergum: the lower part of the field separated from the rest of the coin by a line;

Obverse: main figuration, from technical point of view is the figure produced by the bottom die (sometimes called the *anvil die*);

Reverse: secondary figuration;

Legend: inscription;

Mint: place of emission;

Issuing authority: State or civic entity under whose authority the coins are minted.

Among the most discussed problems in the Punic numismatic stands out the identification of emitting mint. A major difficulty lies in the different approach that often occurs, depending on the cultural model of reference adopted for the interpretation. If the Punic coinage is considered analogous to the coinage of Greek tradition, the production of coins should be tied to sovereignty town and difference between issuer authority and mints should not be found. If the model is more similar to the imperialistic one, as the Persian Empire and the Roman Empire, the issuing authority and the mint emission coincide only for the capital and is further accompanied to more provincial centers of production. In the long history of the Punic coinage both cultural models are found.

This duality and different cultural reference are also found within the intrinsic characteristics of the production of Punic coins like the characteristics of the series, emissions (groups of coins with the same characteristics suh as weight, diameter, accessorial symbols, coined with the same pair of dies or similar dies) and nominal values (multiples and submultiples of a reference unit which form a series).

Other fundamental feature for a coin description are:

Series: consists of the nominal (multiples and submultiples of the unit)

Nominal: are multiples and submultiples of the unit that form a series.

Emission: group of pieces coined in a single moment, with the same metal, with the same weight and diameter, with the same types. Equal coins then, but not identical;

Metal: gold, silver, electrum, bronze, billon (silver-copper) *potin* (copper alloy plus a high content of lead or tin);

Theoretical weight: weight given to specimens, depending on the used metal or alloy; Weight: real weight of the single coin;

These elements are defined during the preparation of flan and represent the value of the currency, whose state guarantee is given by the type and legend. The nominal weight refers to the weight systems.

Complex and not easy to read is the documentation related to the Phoenician and Punic weight system: there aren't any direct or indirect sources that provide useful elements for the reconstruction of the theoretical values, which were based on the weight systems of the East and West Phoenician cities. From inscriptions it can be deduced that only the shekel (šql) was the main weight unit of the Phoenicians and Punics while scarce and of difficult interpretation is the documentation relating to the multiples and submultiples. However, it seems plausible to assume that with the term shekel were intended different measures, accompanied by units and systems designated with different names and weights. Diversification of weight emerges clearly from an examination of weight-samples found in Phoenicia. Regarding the coinage system adopted by the Phoenician cities from the middle of the V century BC, the ponder system of g 7.76 (Phoenician shekel) is prevalent, with a greater diffusion of the heavier 14.0 g, rather than the lighter of about 7.0 g.

Only Arado coins where minted constantly on micro-Asiatic ponderal system (shekel of 11.75 g), while in the early coinage of Byblos is perhaps recognizable the use of the Syrian weight of 9.4 g. The attic ponderal system of 4.36 g appears only in the IV century BC in Sidon and Tyre, during the revolt against the satraps. The near east pre-monetal weight system, micro-Asian g 5.8 and perhaps the Syrian 9.40 g, arrive in the West as early as XI century BC, following the trade routes passing through Cyprus, touching Sardinia and the Iberian Peninsula.

The Phoenician 7.76 g unit spreads later, after having affirmed all along the Syrian coast, and is part of a Western context. In the area of Punic, the two weight of oriental origin seem to integrate to form a mixed system, open to the units of the Western tradition. The most ancient Punic coinage of silver (410 BC) are coined on Euboean- attic weights, while the series minted in gold and electro are minted on the Phoenician shekel of 7.76 g. or Syrian unit of 9.40 g, perhaps recognizable also in the weights system of g 8/9 used for Spanish-Carthaginian emissions of barcide age and those of the neo-punic cities of Malaca, Gadir,

Ebusus. Since the end of the III century BC in the Iberian Peninsula the unit of g 10/11 is also attested, considered to be of Carthaginian tradition, but which could also be inspired by systems of oriental origin.

Producing and Minting Techniques

Coins can be distinguished by the technique of flan production (the metal little disc of which is constituted the coin) and the mode of impression of the faces on the coin.

The flan could be produced by pouring the molten metal into a mold, or produced for cutting a metal tube in discs. These to different procedures are distinguishable because each one leave clues on the coin: the fused flan has often a fusion tang on the border and the final shape of the coin is a disc with rounded edge or truncated edge in relation of the shape of mould cavities, while the cut flan has a cut edge and doesn't show any fusion tang.

The obverse and reverse can then be produced both by minting the flan with the two dies and pouring the fused metal in a mould, which reproduce precisely the obverse and reverse.

Fusion: The prototype was probably made of wood and was reproduced by pressure in a refractory material (clay) and then baked. Molten metal was subsequently poured inside. This was a simple and fast system of coinage, but typologically inexact and easily falsifiable. This is way gold and silver coins were never produced by fusion.

Minting: hammering a hot-melt flan, at the state of malleability, between two dies that bore carved cavity, which reproduce the reverse and obverse of the coin and legends, that after the minting appear as embossed on the coins.

One of the two dies was firmly fixed to the anvil (fixed die), the other was mobile and was used in the beating, tightened with pliers (mobile die placed over the flan): this was the so called hammer-die. The hammer-dies (reverse) were much faster destroyed than an anvildies (straight). The dies in ancient times were made of bronze or iron, but very few of these persisted from the past to our days because there were the common use to destroy the dies after the end of the emission. The coin was engraved by hand by an artist, which reproduced a model-sample of larger diameter. The art of engraving the dies was an official art, also important for historical and religious implications, because the carved issues as deities and other valuable items for the phoenician and punic culture, are represented in their official aspect. In this appendix were highlighted the most important features that will

be addressed in the numismatic study object of chapter VI and chapter VII, on the archaeometric study of privates collection of Punic coins.

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Index of abbreviations used in the text

EBA: Early Bronze Age MBA: Middle Bronze Age LBA: Late Bronze Age IA: Iron Age EIA: Early Iron Age MIA: Middle Iron Age

Index of Latitude and Longitude (in decimal degrees) of the major settlements and areas mentioned in the text

Place	Latitude	Longitude
Evora	38,638327	-7,907867
Alcacer do Sal	38,396568	-8,522873
Teti	40,098034	9,120097
Monte Sirai	39,184568	8,483319
Bonorva	40,417678	8,769135
Flumenelongu	40,559200	8,318853
Antas	39,431950	8,486023
Baunei	40,029718	9,666581
Lanusei	39,826995	9,598503
Sardara	39,615210	8,820720
Oschiri	40,720136	9,099841
Nule	40,464711	9,188898
Ossi - Sa Mandra/Sa Giua	40,677123	8,597646
Perfugas/Canopoli	40,829073	8,883719
Illorai	40,355113	9,003375
Siligo	40,577977	8,727858
Olmedo	40,648866	8,382211
Ardara località de Boes	40,622096	8,809397

Laerru	40,814069	8,825426
Giorrè/Florinas	40,650950	8,661897
Posada/Nuraghe Pizzinnu	40,634147	9,716878
Sarrok	39,076243	8,982582
Villacidro	39,459987	8,733345
Aouam	33,197328	-5,631294
Volubilis	34,074542	-5,555116

To ask for the tables collected from literature and used for elaboration of the PCA and GIS shown in the thesis please send an email to the author: angela.celauro @ tiscali.it

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