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THE ORIGIN AND USE OF COPPER AT THE CHALCOLITHIC FORTIFICATION OF LECEIA (OEIRAS, PORTUGAL)

1. Leceia - a fortified settlement of the 3rd millennium B.C.

The prehistoric settlement of Leceia is located on a brow overlooking the valley of the river Barcarena, about 3 km from the Tagus estuary (Oeiras, Lisbon; fig. 1-3). Although the site is known since the 19th century¹, systematic excavations were conducted only between 1983 and 2002, revealing a complex settlement fortified by a defence system of massive stone walls and towers². The well-preserved stratigraphy reaches from Bell Beaker contexts down to a late 4th millennium Neolithic stratum. The Copper Age occupation at the site can be divided into an Early Chalcolithic (layer 3), dating between 2800 and 2600 B.C., and a Middle Chalcolithic (layer 2), dating between 2600 and 2300/2200 B.C. The Bell Beaker complex occurred at around 2600 B.C. and seems to have been present at the site until 2000 B.C.; i.e. it went all along with the Middle Chalcolithic material assemblage and finally outlived it chronologically³. The settlement comprises a series of roundhouses with stone foundations, remnants of adobe superstructures as well as fireplaces often defined by standing plate stones. Remains of a stone pavement linking some of the stone structures were found in the south-eastern part of the site.

However, thus far little is known about the living activities at Leceia and the socio-cultural and socio-economic role played by the site in the late 4th and 3rd millennium B.C. Its elevated position, size and rich material assemblage are strong arguments for interpreting it as a 'Central Place'⁴ similar to Vila Nova de São Pedro (Azambuja)⁵ and Zambujal (Torres Vedras)⁶, probably controlling the local settlement area⁷. In this paper we present and discuss new chemical and lead isotope data of archaeometallurgical remains from the site, in order to learn more about the role the innovation of

¹ Ribeiro 1878.

² Cardoso 1994; Cardoso 1997; Cardoso 2000; Cardoso - Soares 1996.

³ Cardoso 1994; Cardoso 1997; Cardoso 2000.

⁴ Kunst 1995.

⁵ Jalhay - Paço 1945.

⁶ Sangmeister - Schubart 1981; Kunst - Uerpmann 1996.

⁷ Cardoso 2000.

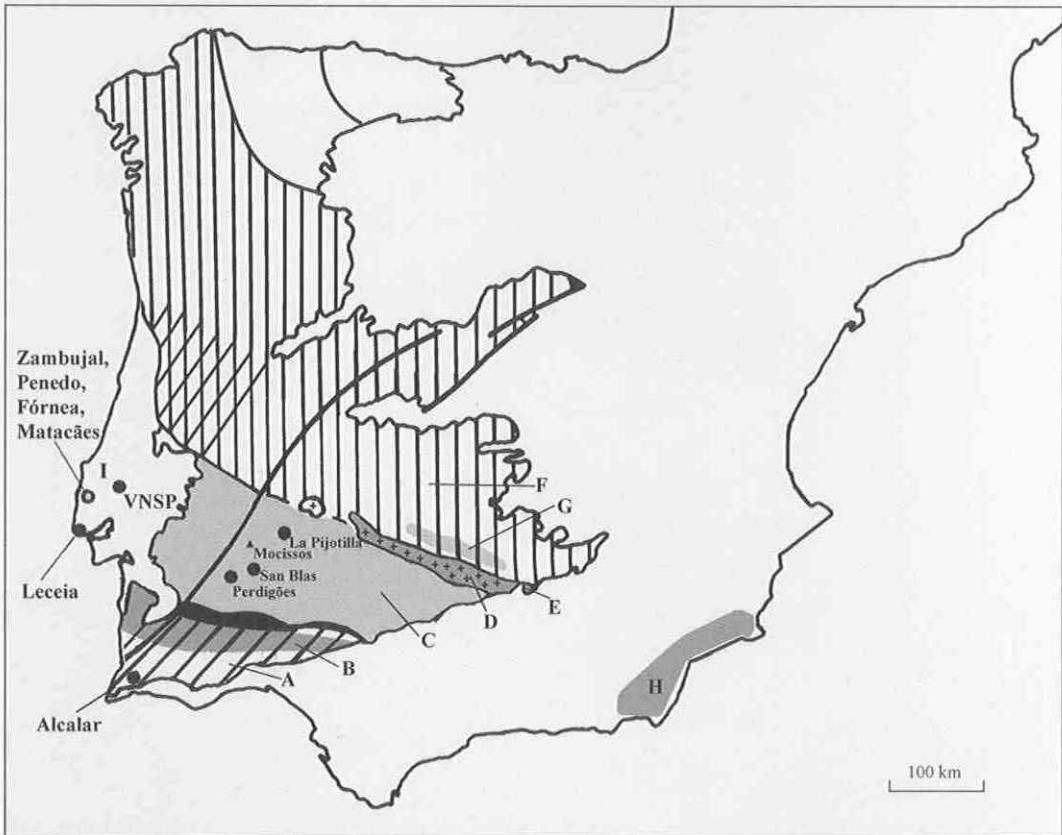


Fig. 1 The Iberian Peninsula with geological units and archaeological sites mentioned in the text. A South Portuguese Zone; B Southwest Iberian Pyrite Belt which is part of the SPZ; C Ossa Morena Zone; D Los Pedroches Batholith; E Linares - La Carolina district; F Central Iberian Zone; G Alcudia Valley; H Ore deposits of southeast Spain (Betic Cordilleras); I Postpaleozoic sediments of the Estremadura. The grey areas represent geological units that contain copper occurrences of which lead isotope data exist; these are discussed in the paper. There are small copper occurrences in the south-western part of the CIZ which have not been studied thus far (checked zone). Spots - some of the largest Chalcolithic fortifications of southwest Iberia.

copper metallurgy played at the site in terms of production practices and resource procurement.

2. *The metallurgical remains and their domestic contexts*

The excavations at Leceia revealed about 130 copper objects of small size and rudimentary shape, like awls, chisels, saw blades, fishhooks and Palmela points; the heaviest implements are tanged daggers and small axes⁸. The presence of copper prills, lumps and tormented, apparently unfinished copper artefacts - some of them may have been ingots - found in some domestic areas indicate that the metal was shaped inside the

⁸ Cardoso 1997.



Fig. 2 The Chalcolithic fortified settlement of Leceia. Aerial view.

settlement; a fragment of a crucible was discovered as well (fig. 4). All in all, the copper found at Leceia accounts for no more than 1 kg. Indications for extractive metallurgy, like smelting slags or copper ores, were not found at the site.

The stratigraphy documented at Leceia demonstrates clearly that copper metallurgy developed significantly only from the Middle Chalcolithic onwards; only a knife and three amorphous copper fragments were found in the Early Chalcolithic layer 3. Hence, it appears that the metal innovation did not trigger, but rather resulted from the 3rd millennium cultural change, which was accompanied by social differentiation and the emergence of complex settlements such as Leceia.

During the last decades, numerous chemical and metallographic analyses of the Leceia copper artefacts produced first significant insights into the 4500 year old metallurgy⁹. 22 quantitative chemical analyses (using neutron activation) have shown that the artefacts are made of arsenical copper with very low trace element concentrations. It was suggested that the arsenic most probably derived directly from the processed ores.

⁹ Gil et al. 1979; Cardoso – Fernandes 1995; Cardoso – Guerra 1997/1998.

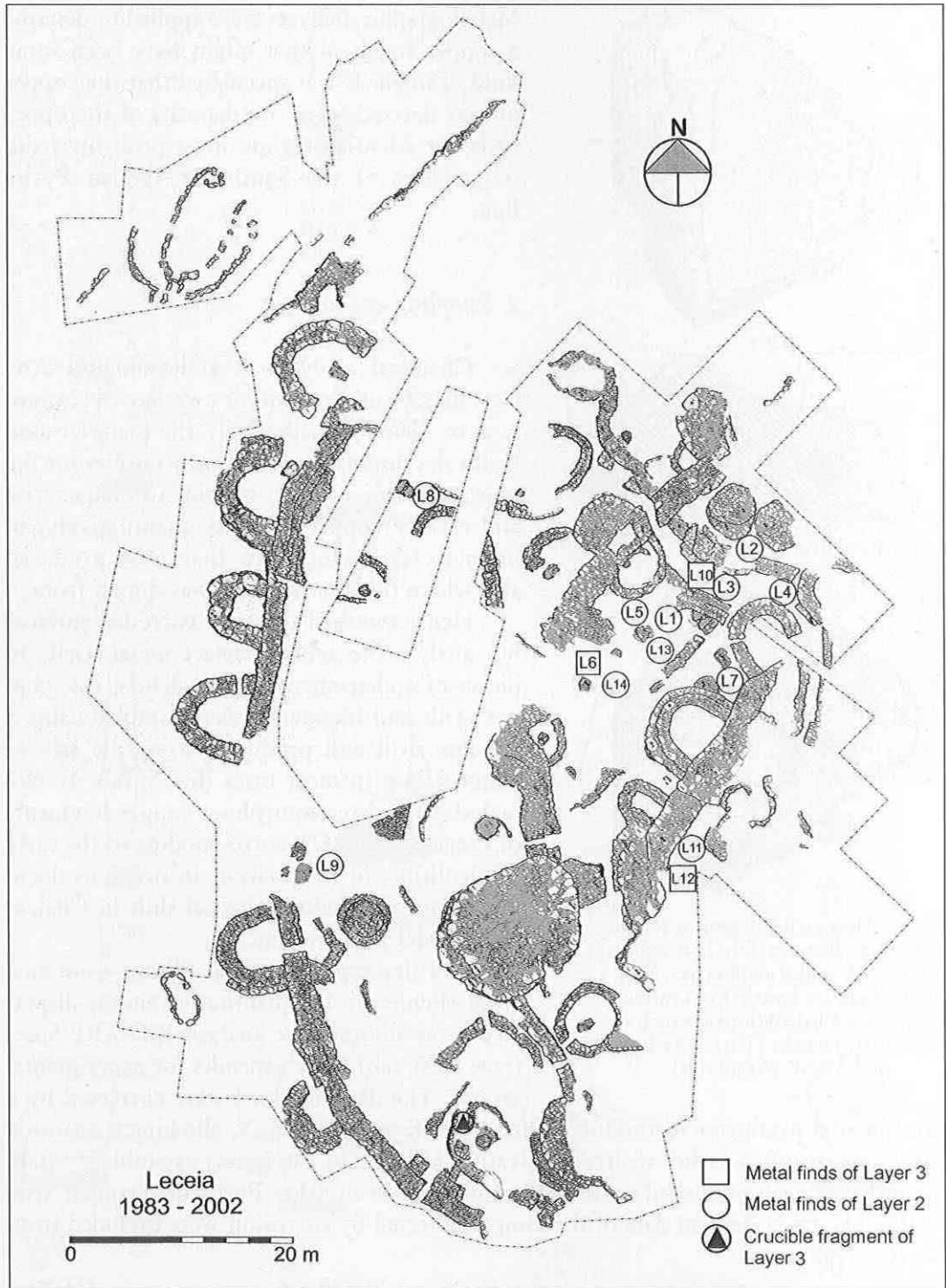


Fig. 3 Plan of the site of Leceia showing the distribution of the metal finds analysed in this paper, as well as the find spot of the only crucible fragment discovered at the site.

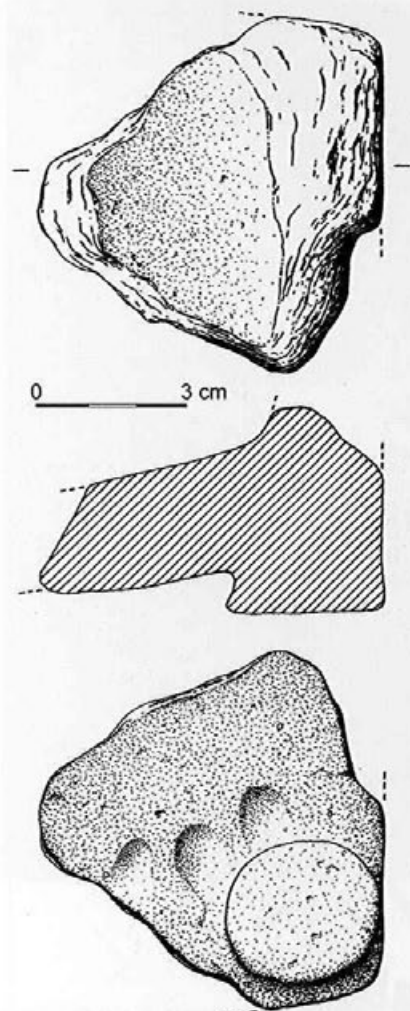


Fig. 4 The crucible fragment found at Leceia. The crucible is roughly made, thick-walled and has feet. Very similar finds are known from various Portuguese Chalcolithic sites such as Zambujal, Penedo (Torres Vedras) and VNSP (Azambuja).

a fundamental parameter method described by Lutz and Pernicka¹⁰, allowing a quantitative comparison to earlier neutron activation analyses of the Leceia assemblage¹¹ (tab. 2) and previously published optical emission analyses of other Portuguese copper artefacts¹². The trace element data of the samples affected by corrosion were excluded from the discussion.

Metallographic analyses were applied to describe a copper fragment that might have been some kind of ingot. It was speculated that the copper objects derived from ore deposits of the upper or lower Alentejo region, most probably from occurrences of the Southwest Iberian Pyrite Belt.

3. Sampling and analysis

Chemical analyses of archaeological artefacts may be undertaken for a variety of reasons, e. g. to identify qualitatively the material used or for developing conservation strategies for the object. In this project, we aim to characterise and classify copper artefacts quantitatively, in order to learn about how they were produced and where the raw material was drawn from.

Hence, we had to avoid corroded surfaces but analyse the actual artefact metal itself. 14 pieces of archaeometallurgical debris, i. e. copper prills and fragments, were sampled using a 1.5 mm drill and producing a sample size of about 0.03 g in most cases (fig. 5; tab. 1). We included the three amorphous copper fragments of Leceia stratum C3, corresponding to the early Chalcolithic, in the analysis in order to document a potential chronological shift in Chalcolithic metal procurement.

In a first step the metal fillings were analysed chemically by quantitative energy-dispersive X-ray fluorescence analysis (ED-XRF Spectrace 5000; tab. 1; see appendix for experimental set-up). The data obtained were corrected by a

¹⁰ Lutz - Pernicka 1996; Lutz - Pernicka unpublished.

¹¹ Cardoso - Guerra 1997/1998.

¹² Junghans et al. 1960; Junghans et al. 1968; Junghans et al. 1974.

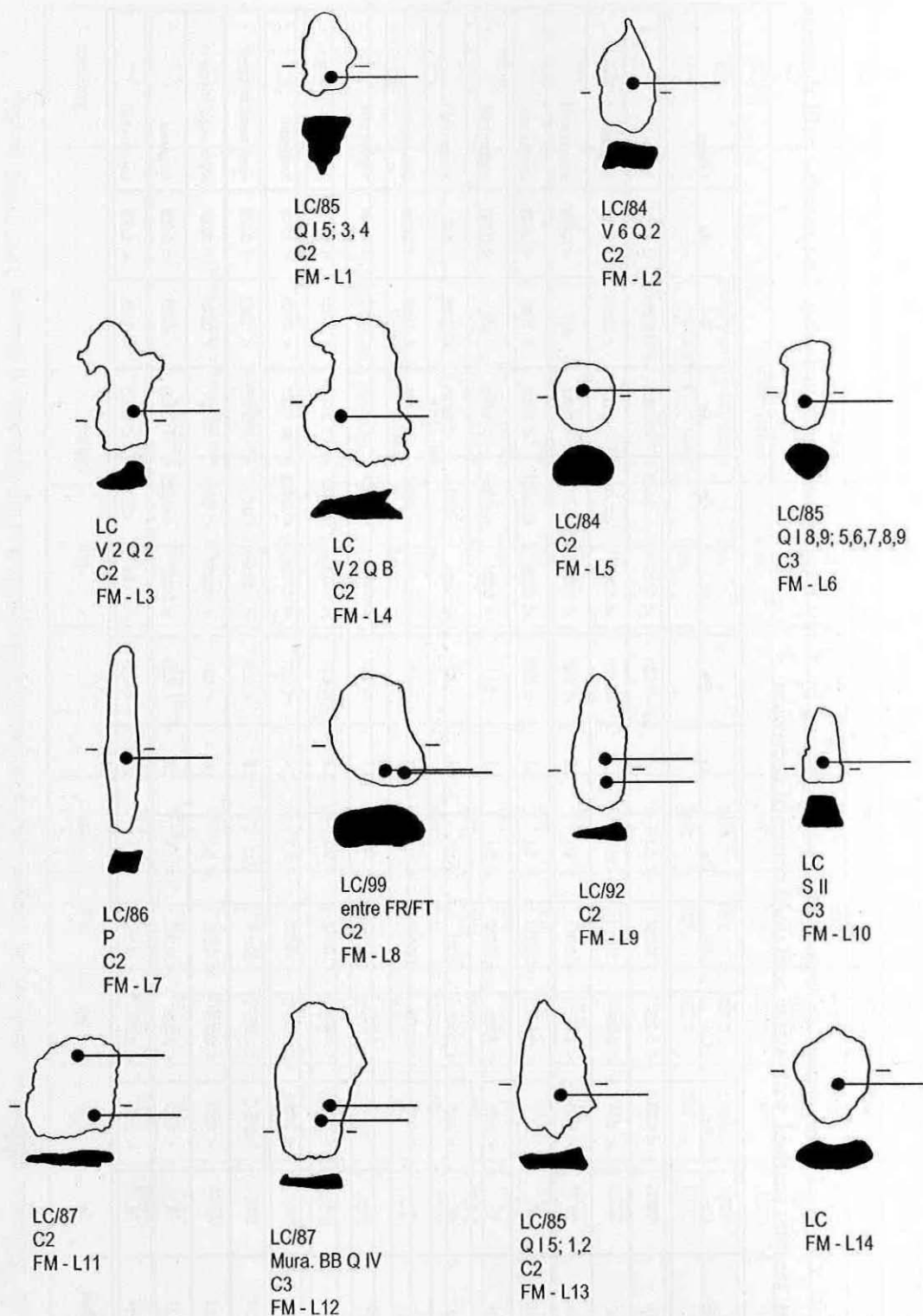


Fig. 5 The artefacts analysed in this paper. M. 1 : 1.

Tab. 1 Chemical composition of copper artefacts as detected by X-ray Fluorescence analysis (wt%). The metallic drill shavings of the samples marked with a star contained traces of corrosion.

Sample	Cu	Fe	Co	Ni	Zn	As	Pb	Bi	Ag	Sn	Sb	Tc	Object
FM-L1	98	< 0.05	< 0.005	< 0.010	< 0.1	1.8	< 0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.008	fragment
FM-L2	99	< 0.05	< 0.005	< 0.010	< 0.1	1.0	< 0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.008	fragment of a blade
FM-L3	99	< 0.05	< 0.005	< 0.010	< 0.1	0.6	< 0.01	< 0.005	0.01	< 0.005	0.01	< 0.008	copper prill
FM-L4*	98	0.1	< 0.005	< 0.010	< 0.1	1.3	< 0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.008	copper prill
FM-L5	99	< 0.05	< 0.005	< 0.010	< 0.1	1.1	0.01	< 0.005	< 0.005	< 0.005	0.01	< 0.008	copper prill
FM-L6*	98	< 0.05	< 0.005	0.02	< 0.1	1.6	0.01	< 0.005	0.01	< 0.005	< 0.005	0.02	copper prill
FM-L7	99	< 0.05	< 0.005	< 0.010	< 0.1	0.7	< 0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.008	awl fragment
FM-L8*	97	0.1	< 0.005	< 0.010	< 0.1	2.9	< 0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.008	copper prill
FM-L9	98	< 0.05	< 0.005	< 0.010	< 0.1	2.2	< 0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.008	fragment
FM-L10	98	< 0.05	< 0.005	< 0.010	< 0.1	2.4	< 0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.008	fragment
FM-L11*	97	0.2	< 0.005	0.04	0.1	2.8	< 0.01	< 0.005	0.03	< 0.005	< 0.005	< 0.008	sheet metal fragment
FM-L12	96	< 0.05	< 0.005	< 0.010	< 0.1	3.6	< 0.01	< 0.005	< 0.005	0.009	< 0.005	< 0.008	sheet metal fragment
FM-L13	98	< 0.05	< 0.005	< 0.010	< 0.1	1.8	< 0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.008	fragment
FM-L14	98	< 0.05	< 0.005	< 0.010	< 0.1	1.8	< 0.01	< 0.005	0.01	< 0.005	< 0.005	< 0.008	copper prill

Tab. 2 Neutron activation analyses of copper artefacts from Leceia published by Guerra 1997/1998 (wt%).

Sample	Ni	Sn	Sb	Ag	As		Cu	Object	Stratum
P7	0.006	0.003	0.005	0.007	1.12		98.9	chisel	C2
P8	0.003	n.d.	0.021	n.d.	0.26	Fe: 1.12	95.6	fragment	C2
P9	0.007	n.d.	0.010	n.d.	1.48		98.5	axe fragment (cutting edge)	C2
P10	0.003	n.d.	n.d.	n.d.	0.34		99.7	axe fragment	C2
P11	0.017	0.007	0.022	n.d.	3.93		96.0	knife	C3
P12	0.020	0.003	0.002	n.d.	1.54	Fe: 0.01	98.4	awl	C2
P13	0.001	0.003	0.002	n.d.	3.36		96.6	sheet metal fragment (blade)	C2
P15	n.d.	n.d.	0.035	n.d.	5.22		94.7	sheet metal fragment	C2
P16	0.01	0.01	0.011	n.d.	1.44		98.5	axe blade	C2
P17	0.012	n.d.	0.010	0.101	0.85		99.0	arrow head	C2
P18	0.022	n.d.	0.022	n.d.	2.75		97.2	knife	C2
P19	0.004	n.d.	0.014	n.d.	3.05		96.9	awl	C2
P23	0.005	n.d.	n.d.	n.d.	0.17		99.8	chisel	C2
P31	0.007	0.01	0.016	n.d.	1.86		98.1	awl	C2
P38	0.002	0.007	0.012	n.d.	5.31	Pb: 0.22	94.4	fragment of a fish hook	C2
P76	0.003	0.005	0.007	0.012	0.91		99.1	sheet metal fragment	C2
P77	0.003	0.007	0.007	0.01	0.87		99.1	scrap metal once molten	C2
P90	0.007	n.d.	0.020	n.d.	3.61		96.4	tanged dagger	C2
P91	0.007	n.d.	0.015	n.d.	3.55		96.4	tanged dagger	C2
P92	0.010	0.003	0.006	n.d.	2.19		97.8	long awl	C2
P93	0.011	0.002	0.006	n.d.	1.55		98.4	fishhook	C2
P94	0.007	0.001	0.005	0.007	1.19	Au: 0.0003	98.8	fragment (probably an ingot)	C2

Tab. 3 Lead isotope abundance ratios of Leceia copper artefacts. The 2σ error of the $^{206}\text{Pb}/^{204}\text{Pb}$ data is significantly higher than 0.1% in five cases, due to the extremely low lead content of the samples.

Sample	$^{207}\text{Pb} / ^{206}\text{Pb}$	2σ of $^{207}\text{Pb} / ^{206}\text{Pb}$ [%]	$^{208}\text{Pb} / ^{206}\text{Pb}$	2σ of $^{208}\text{Pb} / ^{206}\text{Pb}$ [%]	$^{206}\text{Pb} / ^{204}\text{Pb}$	2σ of $^{206}\text{Pb} / ^{204}\text{Pb}$ [%]	Object	Stratum
FM-L1	0.8478	< 0.1	2.0881	< 0.1	18.474	0.75	fragment	C2
FM-L2	0.8523	< 0.1	2.0880	< 0.1	18.341	0.48	fragment of a blade	C2
FM-L3	0.8577	< 0.1	2.0998	< 0.1	18.240	< 0.1	copper prill	C2
FM-L4	0.8460	< 0.1	2.0797	< 0.1	18.496	< 0.1	copper prill	C2
FM-L5	0.8568	< 0.1	2.0970	< 0.1	18.254	< 0.1	copper prill	C2
FM-L6	0.8470	< 0.1	2.0824	< 0.1	18.473	< 0.1	copper prill	C3
FM-L7	0.8566	< 0.1	2.0980	< 0.1	18.257	< 0.1	awl fragment	C2
FM-L8	0.8530	< 0.1	2.1005	< 0.1	18.303	< 0.1	copper prill	C2
FM-L9	0.8544	< 0.1	2.0914	< 0.1	18.293	< 0.1	fragment	C2
FM-L10	0.8438	< 0.1	2.0916	< 0.1	18.537	0.24	fragment	C3
FM-L11	0.8820	< 0.1	2.1263	< 0.1	17.642	< 0.1	sheet metal fragment	C2
FM-L12	0.8443	< 0.1	2.0847	< 0.1	18.589	0.64	sheet metal fragment	C3
FM-L13	0.8505	< 0.1	2.0884	< 0.1	18.421	0.34	fragment	C2
FM-L14	0.8523	< 0.1	2.0904	< 0.1	18.345	< 0.1	copper prill	surface find

In a second step, lead isotope analyses were conducted using a Multi Collector - Inductively Coupled Plasma Mass Spectrometer (MC-ICPMS Axiom, VG Elemental; tab. 3). The experimental set-up is described in the appendix.

4. *The historical and technological context of the arsenical copper from Leceia*

The analysis of Portuguese copper artefacts played a crucial part in the SAM¹³ trans-European analytical program of the 1960s and 1970s: chemical data of more than 1100 objects were obtained, the vast majority of which deriving from southern Portugal and dating to the Copper and Early Bronze Age¹⁴. The protagonists were ahead of their time: neither did computers and appropriate statistical methods exist, nor had the archaeological community had ever seen such a huge amount of artefact specific data before. As a result, the quality of the data and the point of grouping artefacts according to their chemical composition were severely questioned¹⁵. However, in the early 1980s, the accuracy of the whole SAM data set was approved by a systematic comparison of the analytical results with others, obtained using more recent analytical methods¹⁶. Unfortunately, for early metalwork from the Iberian Peninsula in particular a large scale, systematic comparison of the SAM data with results of analyses of other programs, such as the 'Proyecto de Arqueometalurgia'¹⁷ or data produced by the British Museum¹⁸, is still lacking¹⁹.

Using a graphical method suggested by Sangmeister²⁰, fig. 6-8 show the copper used at Leceia in comparison to copper found at Zambujal and Vila Nova de São Pedro (VNSP), which are the two other type sites of the Estremadurian Chalcolithic. Sangmeister proposed to plot the data of each artefact into the diagrams As/Ni, As/Bi and Sb/Ag. The data clouds were then allocated graphically to various fields defined in each diagram and each individual data point - representing one artefact - was given the respective letter representing the fields. Hence, he was able to classify each artefact according to the ratios As/Ni, As/Bi and Sb/Ag²¹. Most of the artefacts from Zambujal fell into the fields D, E and K (material group DEK), copper high in arsenic, but low in nickel, bismuth, antimony and silver. In fact, this type of arsenic-only copper²² is very typical not only for Copper Age Portugal but the entire Iberian Peninsula²³. Ac-

¹³ Studien zu den Anfängen der Metallurgie.

¹⁴ Junghans et al. 1960; Junghans et al. 1968; Junghans et al. 1974; Ferreira 1961.

¹⁵ See discussion in Pernicka 1984 and Pernicka 1990.

¹⁶ Ottaway 1982; Pernicka 1984; Pernicka 1987; Pernicka 1990.

¹⁷ Rovira et al. 1997.

¹⁸ Harrison - Craddock 1981.

¹⁹ Thus far, only small tables of analytical data of the different laboratories have been discussed comparatively, without considering in detail the biases and limits of the various methods employed (cf. Monteiro 1994; Hunt 2003).

²⁰ Sangmeister 1995.

²¹ Note that nickel, silver and bismuth should follow completely the copper during the smelting process. Arsenic and antimony on the other hand, may partly evaporate during metallurgical processes depending on the redox conditions (cf. Pernicka 1990; Pernicka 1999).

²² Needham 2003.

²³ Sangmeister 1995; Needham 2003; Krause 2003; Pernicka 1990.

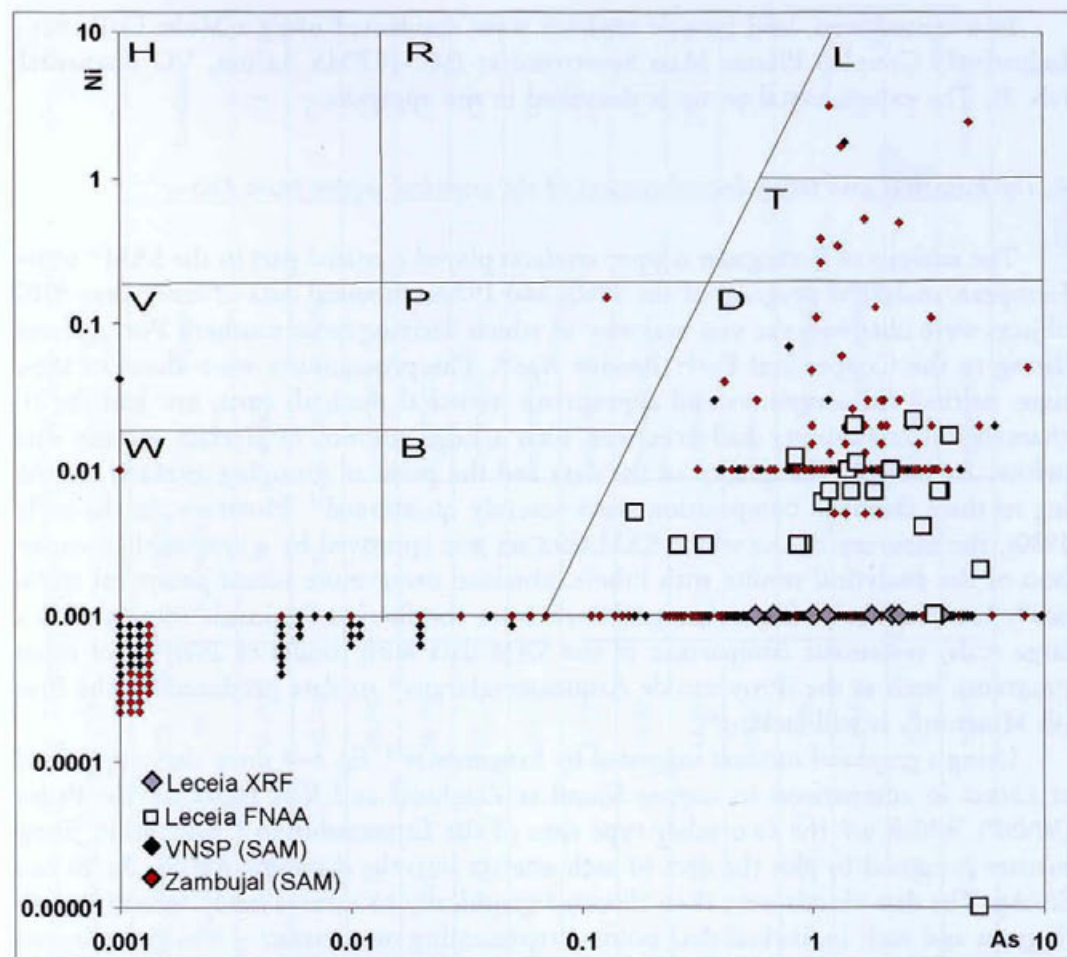


Fig. 6 As-Ni diagram of Leceia artefacts (XRF and FNAA analyses) in comparison to artefacts from VNSP and Zambujal (SAM data). The data were grouped according to a method developed by Sangmeister 1995 (see text).

According to the stratigraphy of Zambujal, this main type of copper was accompanied by pure copper (WAK), mainly occurring in Zambujal phase 1, and arsenical copper with higher impurities of silver, antimony and/or nickel (predominantly DEG), mainly occurring in the later phases of settlement occupation.

All of the Leceia artefacts analysed thus far are made of the typical DEK copper²⁴. Comparing the results chronologically to the ones from Zambujal²⁵, we face the prob-

²⁴ If the element was not detected by the analytical methods used, the artefact was given the theoretical value of the detection limit divided by 10 in order to allow a statistical treatment (cf. Pernicka 1990, 95). The SAM data was plotted using a modified version of the digital SAM data base published by Krause 2003. Non-quantitative descriptions of the SAM data as published by Junghans et al. (Junghans et al. 1960; Junghans et al. 1968; Junghans et al. 1974) such as 'trace' had been quantified by comparative analyses (Pernicka 1990, 95; Pernicka 1984; Pernicka 1987).

²⁵ A clear description of the archaeological contexts of the copper artefacts from Vila Nova de S. Pedro does not exist.

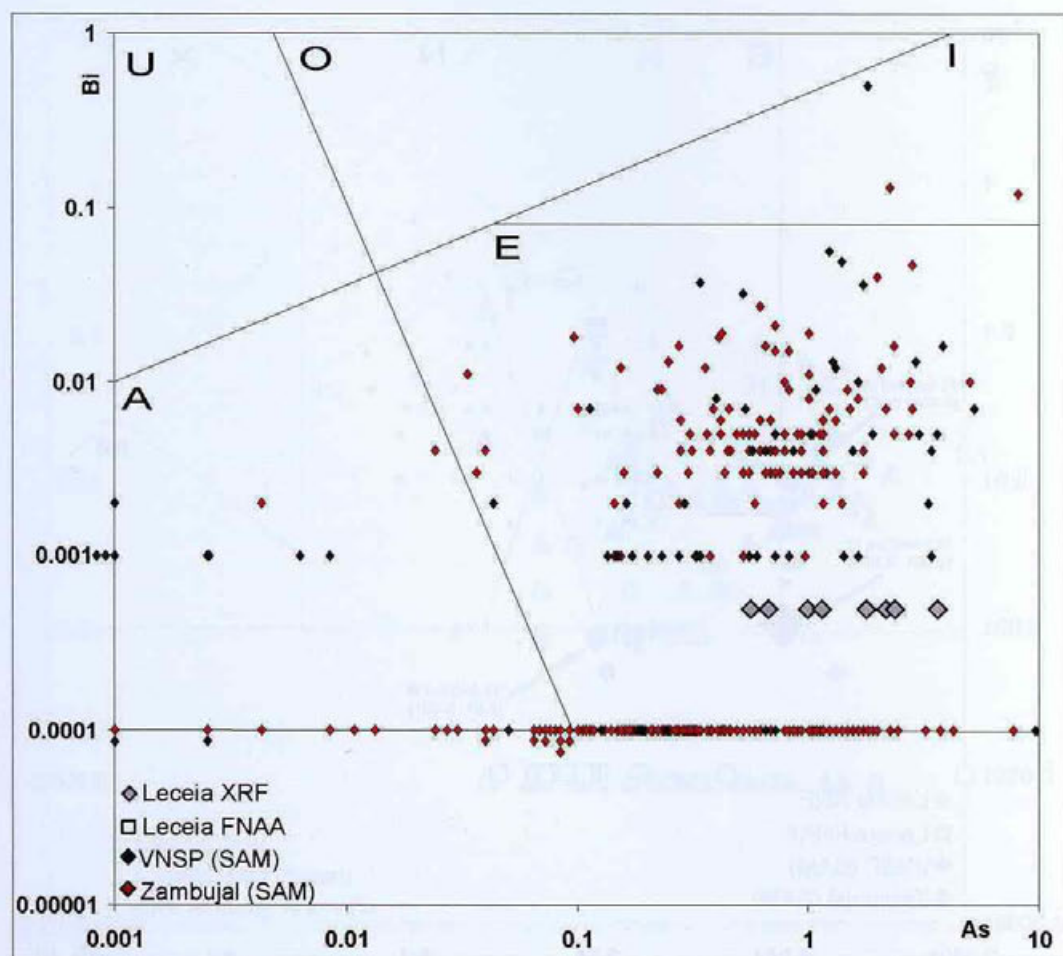


Fig. 7 As-Bi diagram of Leceia artefacts (XRF analyses) in comparison to artefacts from VNSP and Zambujal (SAM data; see text).

lem of discrepancies between the two stratigraphical sequences²⁶. Yet, it is very tempting to link the absence of pure copper (WAK) at Leceia with the absence or occurrence of only very few copper artefacts in the earliest occupation phases of the site (i. e. Late Neolithic and Early Chalcolithic). In the same manner, to link the absence of the more impure arsenical copper DEG, with the end of the occupation at Leceia dating earlier than the one at Zambujal, where the last construction phase (i. e. phase 5) dates to the Bronze Age²⁷. In the case of VNSP, the vast majority of the tin bronze artefacts were made of tin alloyed to DEG copper, hence supporting again a chronological significance of the different copper types used. In any case, we have to remember that there are far fewer quantitative analyses of artefacts from Leceia than from the other two sites²⁸.

²⁶ Sangmeister - Schubart 1981; Cardoso 1997.

²⁷ Sangmeister - Schubart 1981; see also Sangmeister 1995, 38.

²⁸ For example, there are about ten times more quantitative analyses of the Zambujal assemblage than of the Leceia one. Zambujal: 363 of 871 objects were analysed. Leceia: 32 of about 130 artefacts were analysed.

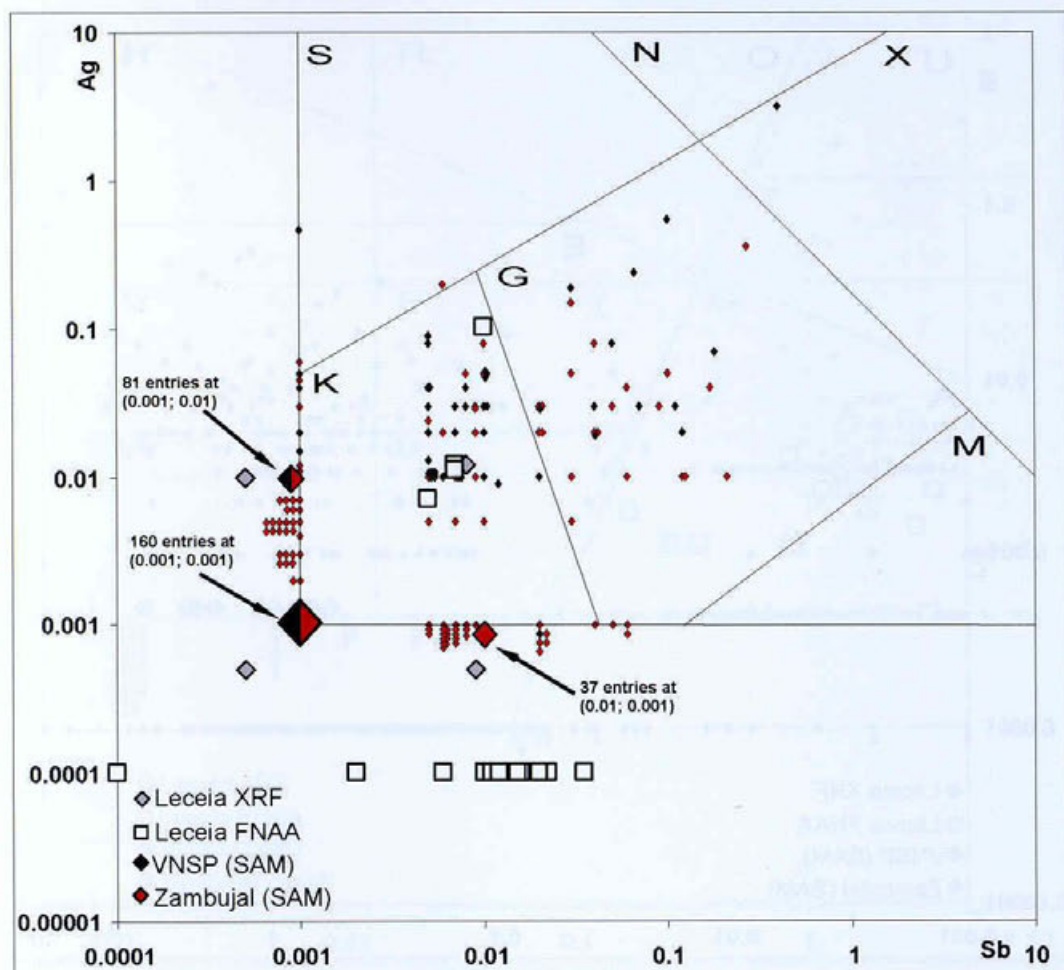


Fig. 8 Sb-Ag diagram of Leceia artefacts (XRF and FNAAs analyses) in comparison to artefacts from VNSP and Zambujal (SAM data; see text).

It is very challenging to determine the raw materials that were used to produce the DEK copper from Leceia. The extremely low amounts of impurities in the arsenical copper may indicate that the artefacts were made from native copper, which can contain up to several percent of arsenic.²⁹ A direct marker for native copper would be the detection of mercury, since the element evaporates far below the temperatures necessary to smelt copper ores³⁰. Thus far we have not been able to examine this crucial element in copper artefacts from Leceia.

In the case of Balkan and Anatolian early copper finds, Pernicka and co-workers³¹ were able to distinguish native from smelted copper by looking at the nickel and cobalt contents. Regarding the use of native copper by native Americans, Rapp and co-

²⁹ Tylecote 1987; Cardoso – Guerra 1997/1998; Pernicka 1998; Pernicka 2004.

³⁰ Kuleff – Pernicka 1995.

³¹ Pernicka et al. 1993.

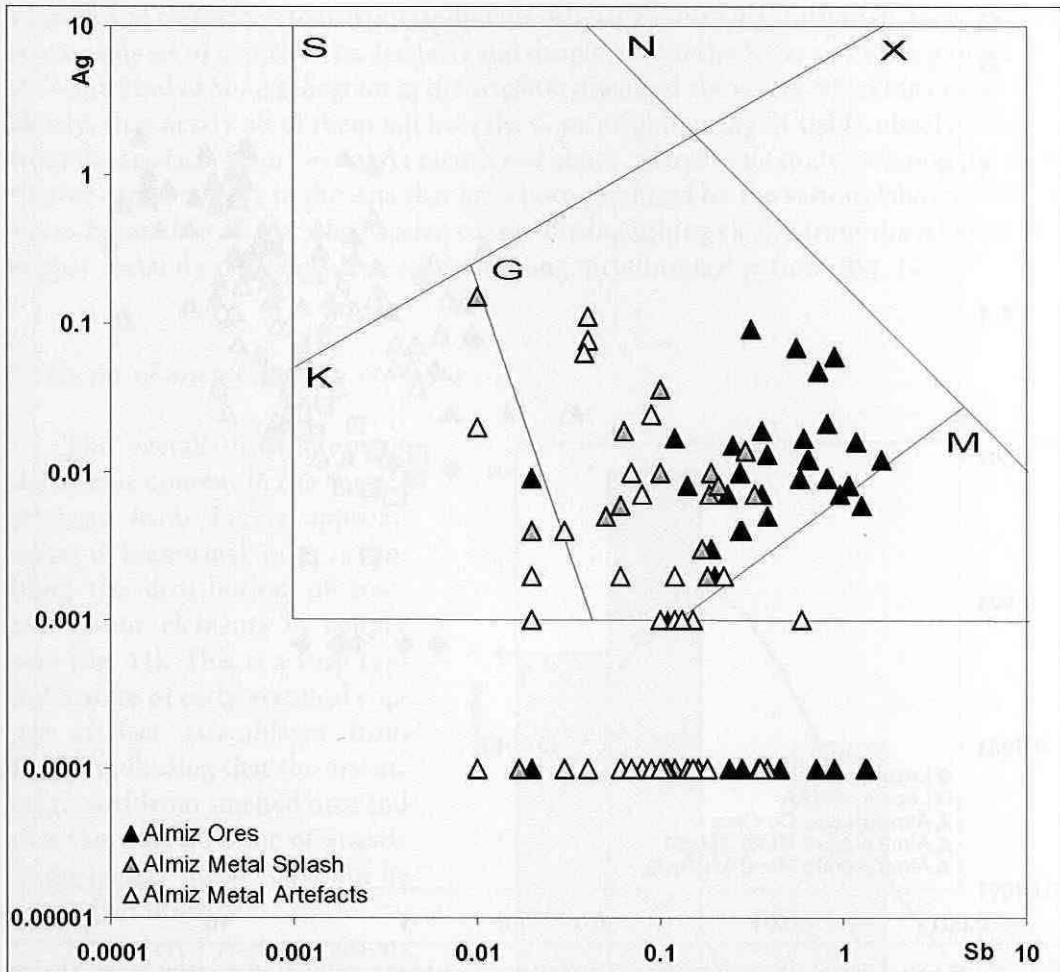


Fig. 9 Sb-Ag diagram of copper artefacts, droplets and ores from Almizaraque based on XRF data published by Rovira et al. 1997. In contrast to the artefacts from Leceia, most of the metal objects and ores fall into the G-field, say have higher amounts of Ag and Sb. Together with high As values, this is a typical fahlore signature. The ores contain somewhat more Sb, since the volatile element tends to evaporate during the smelting and subsequent casting procedures.

workers³² could even distinguish between different sources of native copper exploited, based on the statistical evaluation of trace element patterns. Unfortunately, comparative analyses of native copper from Portugal do not exist thus far³³.

Alternatively, the artefact copper may have been smelted from extremely rich, oxidic copper ores under poorly reducing conditions³⁴. Arsenic-rich copper smelting slags have been identified in the Estremadurian settlements of Zambujal³⁵ and VNSP³⁶,

³² Rapp et al. 2000.

³³ Personal communication from Dr. João Matos, Departamento de Prospecção de Minérios Metálicos, Instituto Nacional de Engenharia, Tecnologia e Inovação (INETI), Portugal.

³⁴ Craddock - Meeks 1987.

³⁵ Müller et al. in press.

³⁶ Müller - Soares 2008, this volume.

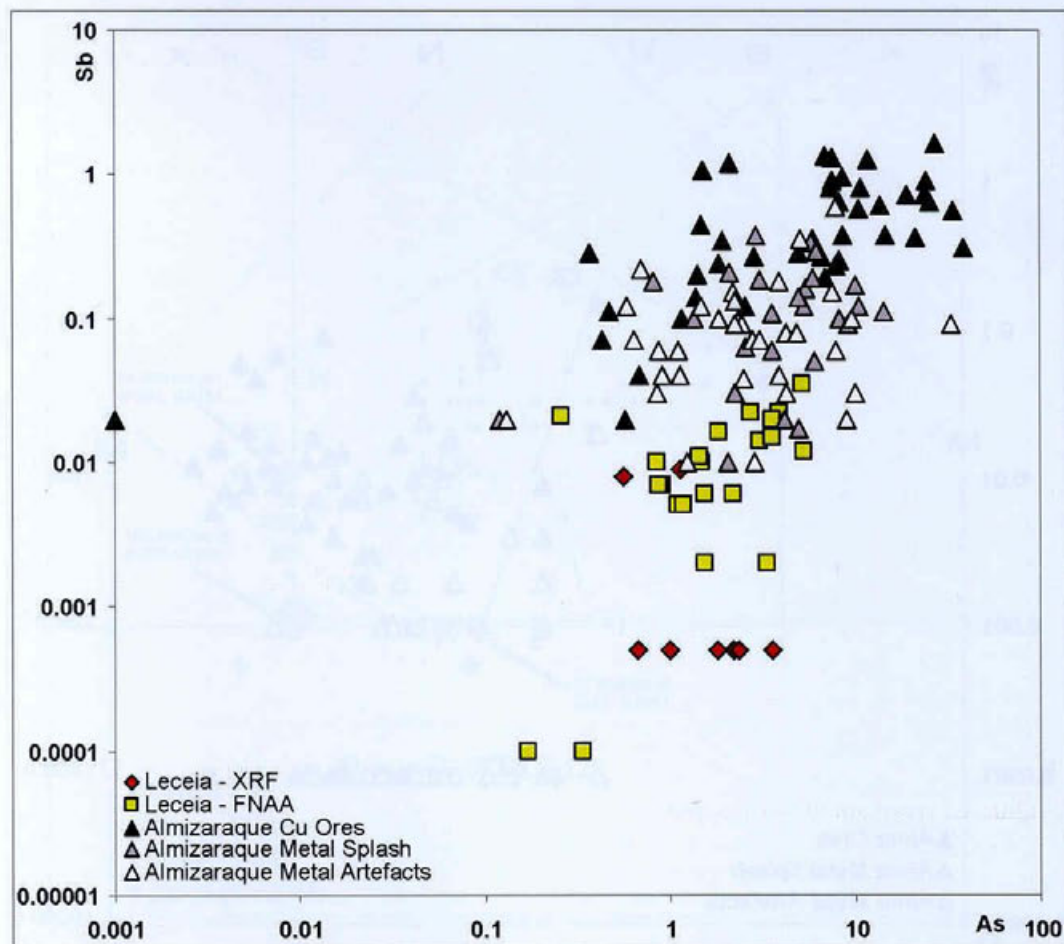


Fig. 10 As-Sb diagram of artefacts from Leceia versus archaeometallurgical remains from Almizaraque. The copper artefacts, droplets and ores from Almizaraque systematically contain higher amounts of Sb at similar contents of As. Once again, looking at the Almizaraque data cloud, one can see clearly that the volatile elements As and Sb tend to evaporate partially during the metallurgical processes.

as well as in Alentejan sites of São Pedro and Fonte Ferrenha (Redondo)³⁷, demonstrating that copper was extracted from ores in south-central Portugal in the 3rd millennium B.C. Secondary copper ore minerals associated with secondarily altered arsenopyrite may have been used to produce arsenical copper with low impurity patterns³⁸.

In contrast, arsenical copper with higher amounts of silver and antimony is very typical for copper smelted from ores of the fahlore group (mainly tennantite and tetrahedrite). Copper with this fahlore signature was used throughout Europe especially at the end of the 3rd and the dawn of the 2nd millennium B.C.³⁹ and was smelted from primary or secondarily altered fahlores at sites such as Ross Island (Ireland), Cabrières (southern

³⁷ Slags identified by Roland Müller, unpublished.

³⁸ Ixer – Patrick 2003; Cardoso – Guerra 1997/1998.

³⁹ Junghans et al. 1960; Junghans et al. 1968; Junghans et al. 1974; Pernicka 1990; Sangmeister 1995.

France) and to some extent Almizaraque (southeast Spain)⁴⁰. Quantitative X-ray fluorescence analyses of copper ores, artefacts and droplets from the latter site were plotted into the same kind of Sb/Ag diagram as the artefacts discussed above (fig. 9)⁴¹. One can see very clearly, that nearly all of them fall into the G or neighbouring M fields, clearly differing from the artefacts from Leceia. As mentioned above, a large scale study assessing the quantitative comparability of the data that have been produced by the various laboratories still has to be conducted. Yet, the Leceian copper distinguishing clearly from the Almizaraque copper certainly does reflect a real underlying metallurgical pattern (fig. 10).

5. The use of arsenical copper at Leceia

The overall distribution of the arsenic content in the copper artefacts from Leceia approximates a lognormal one, resembling the distribution of trace and minor elements in copper ores (fig. 11). This is a very typical feature of early arsenical copper artefact assemblages from Iberia, indicating that the arsenic originated from smelted ores and that the final amount of arsenic in the copper metal could not be controlled directly⁴².

However, re-examinations of the chemical analysis of early copper artefacts from Portugal conducted in the extensive SAM project have shown that the 3rd millennium B.C. metal smiths were well aware of the superior qualities of higher arsenical copper. Judging from the colour of the metal and its properties during forging, higher arsenical copper was reserved for long awls, sheet metal objects (such as saws), Palmela points and tanged daggers⁴³. In the case of

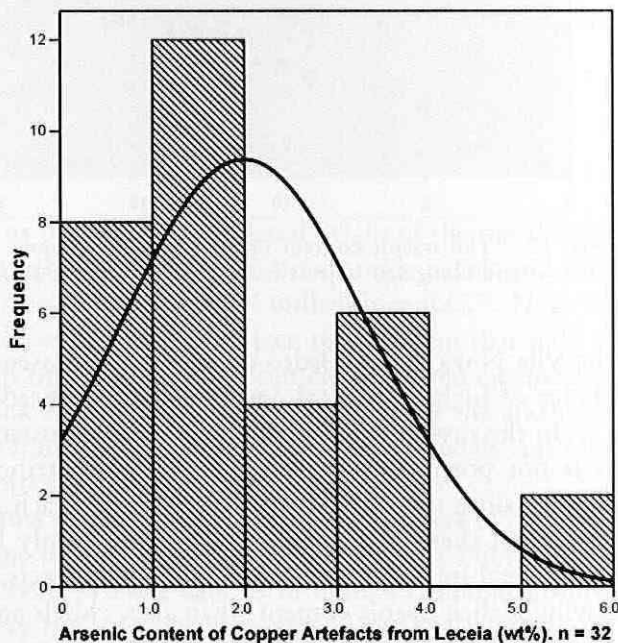


Fig. 11 Overall distribution of the arsenic content of artefacts from Leceia, based on X-ray fluorescence analyses conducted in this project as well as neutron activation analyses published by Cardoso - Guerra 1997/1998. The overall distribution approximates a lognormal one, very similar to the distribution of trace and minor elements in ores.

⁴⁰ O'Brien 2004; Prange - Ambert 2004; Müller et al. 2004.

⁴¹ The very few data of the Almizaraque material published by Rovira and co-workers (2007) that contained the non-quantitative description 'trace' were not considered in this case.

⁴² Rovira 2005; Müller et al. in press; Ferreira 1961; Pernicka 1990; Cardoso - Guerra 1997/1998.

⁴³ Müller et al. in press.

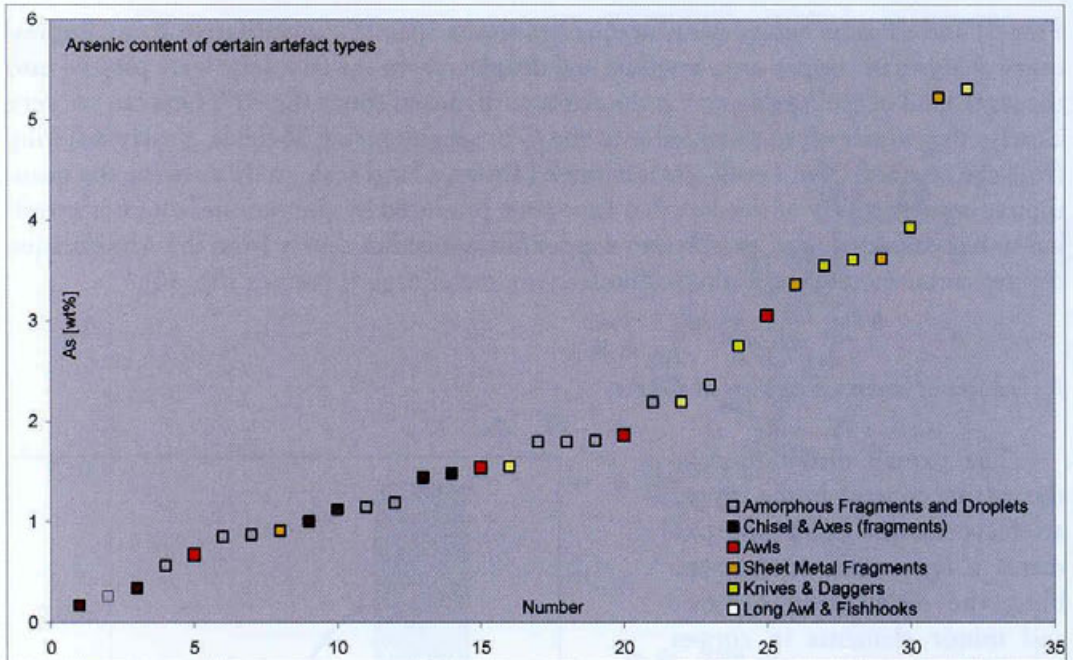


Fig. 12 The arsenic content of certain artefact types. There is a tendency of thinly hammered and elongated objects having higher arsenic contents than chisels, axes and awls.

the Vila Nova de São Pedro copper artefact assemblage, Soares could show a selective choice of higher arsenical copper for arrow heads, spear heads and daggers⁴⁴.

In the case of the Leceia copper artefact assemblage, we see a very similar pattern. It is not possible to look at the arsenic distribution patterns of individual artefact groups, since there are too few analyses for each artefact group. Nevertheless, we can show that there is a general tendency of thinly hammered artefacts, i. e. blades and sheet metal fragments, as well as elongated objects, i. e. the long awl and the fishhooks, having higher arsenic contents than axes, chisels and simple awls (fig. 12). On the other hand, a clear correlation between the arsenic content and the state of cold-working does not seem to exist: metallographic analyses of copper artefacts from Leceia, Zambujal, VNSP, and Porto Mourão have shown that awls and axes were left in a soft annealed state after manufacture, whilst chisels and the only saw thus far examined were left in a cold-worked state⁴⁵. Maybe arsenical copper was recognized not only due to its higher hardness compared to pure copper and due to its distinctive colour; but also because it can be shaped well by cold working. According to Lechtman⁴⁶, all alloys

⁴⁴ Soares 2005; Müller – Soares 2008, this volume.

⁴⁵ Soares et al. 1996; Cardoso – Fernandes 1995; Wang – Ottaway forthcoming. The hardness of a copper artefact, e. g. of a blade, depends on both the chemical composition and the state of cold-working. Alloyed copper is harder than pure copper. This difference in hardness is even greater once the artefacts are cold-worked, i. e. hammered in a cold state. Conversely, the hardness reduces once the artefact is put back into fire and cooled slowly afterwards, i. e. when it is annealed (Lechtman 1996; Rovira – Gómez 2003).

⁴⁶ Lechtman 1996.

from 0.5 to 7 weight-percent of arsenic maintain an almost constant level of ductility as the solid solubility limit is approached. They are also tougher at extreme deformations than pure copper, i.e. thin metal sheets and elongated objects would not brittle fracture as easily⁴⁷.

The question of artefact-alloy correlations have long been a major matter of dispute, regarding archaeometallurgical process reconstructions. The traditional argument is that a conscious addition of an alloying agent and successful production of alloyed copper would require a more sophisticated metallurgy⁴⁸. However alternatively, metal droplets of certain metal types may have been distinguished after the metal was extracted from arsenic bearing copper ores according to their colour and their working properties⁴⁹. In any case, only future detailed studies of south-west Iberian, 3rd millennium B.C. arsenical copper production practices including a consideration of the raw materials and slag by-products involved may allow a better understanding of this problem.

6. *The geological provenance of the copper from Leceia*

Lead isotope analyses may allow us to trace the geological origin of the metal used at Leceia and hence provide new arguments for discussing certain trade routes and patterns of exchange in south-central Portugal during the 3rd millennium B.C.⁵⁰. Next to the general limitations of the method (see appendix), we face the problem that only a small fraction of the abundant Iberian ore sources have been characterized chemically and isotopically. Furthermore, we lack a detailed understanding of how the arsenical copper was produced in Chalcolithic Portugal. As a consequence, the models and ideas developed here can only be understood as working hypotheses.

Fig. 13 shows the lead isotope ratios of 14 copper artefacts from Leceia in comparison to hitherto published lead isotope data of artefacts from VNSP, La Pijotilla and southern Iberian ore deposits. The Leceia artefact data form quite a coherent group, except for one extreme outlier, namely the sheet metal fragment FM-L11⁵¹. One may argue that this is a result of lead contamination, since the drilled sample contained some corrosion. Yet, it is quite unlikely that soils formed from the Mesozoic, i. e. geologically speaking younger, sediments of the Estremadura would provide lead to enter corrosion layers which is a couple of million years older than the copper ores that were used to smelt the actual artefact metal. Looking at the stratigraphic context of the copper, we can see a trend of artefacts originating from Leceia layer C3 (Early Chalcolithic) having smaller ²⁰⁷Pb/²⁰⁶Pb ratios than the artefacts from layer C2 (Middle Chalcolithic).

⁴⁷ Lechtmann 1996; cf. Müller et al. in press.

⁴⁸ Hook et al. 1991; Montero 1994; see discussion in Müller et al. 2004.

⁴⁹ Lechtmann 1996; Shugar 2000; Müller et al. in press.

⁵⁰ Cardoso 1999.

⁵¹ The accuracy of the analytical result was verified by conducting a repeated analysis of an aliquot of the lead sample solution using Thermal Ionization - Mass Spectrometry. Instrument used: MAT 262 (Finnigan), Institute of Mineralogy, University of Mining and Technology Freiberg (Saxony).

However, there are too few analyses to support an argument for the presence of a systematic chronological pattern.

In addition, data of copper artefacts from Vila Nova de São Pedro⁵² were included, in order to allow developing an Estremadurian, rather than a Leceian site-specific perspective on the provenance discussion. If we for instance assumed that the two sites obtained copper from the same source or source region, the Leceian lead isotope data cloud would then form part of a larger one, whose characteristic shape and location could be indicative⁵³. Indeed, some of the Leceia artefact data fall on the same lead mixing line (see appendix) as the material from Vila Nova de São Pedro (VNSP), indicating that the copper may have come from the same or similar kind of deposit; a deposit showing a strong lead isotope scatter, most probably due to a strong radiogenic overprint. Finally, the existing data of artefacts from La Pijotilla⁵⁴, a Chalcolithic ditched enclosure located in the Guadiana basin, were included for interregional comparison.

Of all Iberian copper occurrences analysed thus far, Matacães (Runa, Torres Vedras) is the one located closest to Leceia. The occurrence comprises limestones and calcareous sandstones with only very few traces of malachite⁵⁵. The material would have been unsuitable for copper smelting in the 3rd millennium B.C. Yet, strictly speaking we cannot exclude the possibility that there might have been layers at the site that were enriched in copper and which might have been dug away during the last millennia. Matacães is one of the few copper occurrences located in the Estremadura that reoccur in the discussions as potential copper sources for the metal found in the local Chalcolithic sites⁵⁶. Other occurrences mentioned are supposed to exist close to Asfamil (Rio de Mouro), Maceira and Óbidos⁵⁷. However, at none of them rich copper ores have clearly been documented and studied. In fact, a careful survey by Goldenberg and Maass⁵⁸ in the area of Matacães did not trace any signs for prehistoric mining; around Óbidos, not even the supposedly existing copper occurrence was found! The three samples from Matacães analysed so far define a very narrow field, completely set apart from both the Leceia artefact data cloud, as well as the larger data scatter of the VNSP artefacts (fig. 13).

Besides the poor Estremadurian occurrences, the nearest copper sources are located in the so-called ›Ossa Morena Zone‹, about 100 km away from Leceia and the other coastal Estremadurian sites. Tornos et al.⁵⁹ state that ›the Ossa-Morena Zone (OMZ) is the Iberian Massif geotectonic unit that displays the greatest variety of types of mineralization as well the largest number of ore deposits and showings (> 650)‹. The data plotted in fig. 13 comprise 75 analyses of 27 occurrences of various types located in the Spanish part of the Ossa Morena Zone; analyses of Portuguese deposits of the same geological zone have not yet been published. The lead isotope data of ores of ›FeO-

⁵² Müller – Soares 2008, this volume.

⁵³ Niederschlag et al. 2003, 82.

⁵⁴ Hunt 2003.

⁵⁵ Müller – Soares 2008, this volume; Goldenberg – Maass unpublished report.

⁵⁶ Sangmeister 1995; Kunst 1995; Cardoso – Guerra 1997/1998; Uerpmann – Uerpmann 2003.

⁵⁷ Sousa et al. 2004.

⁵⁸ Goldenberg – Maass unpublished report.

⁵⁹ Tornos et al. 2004, 144.

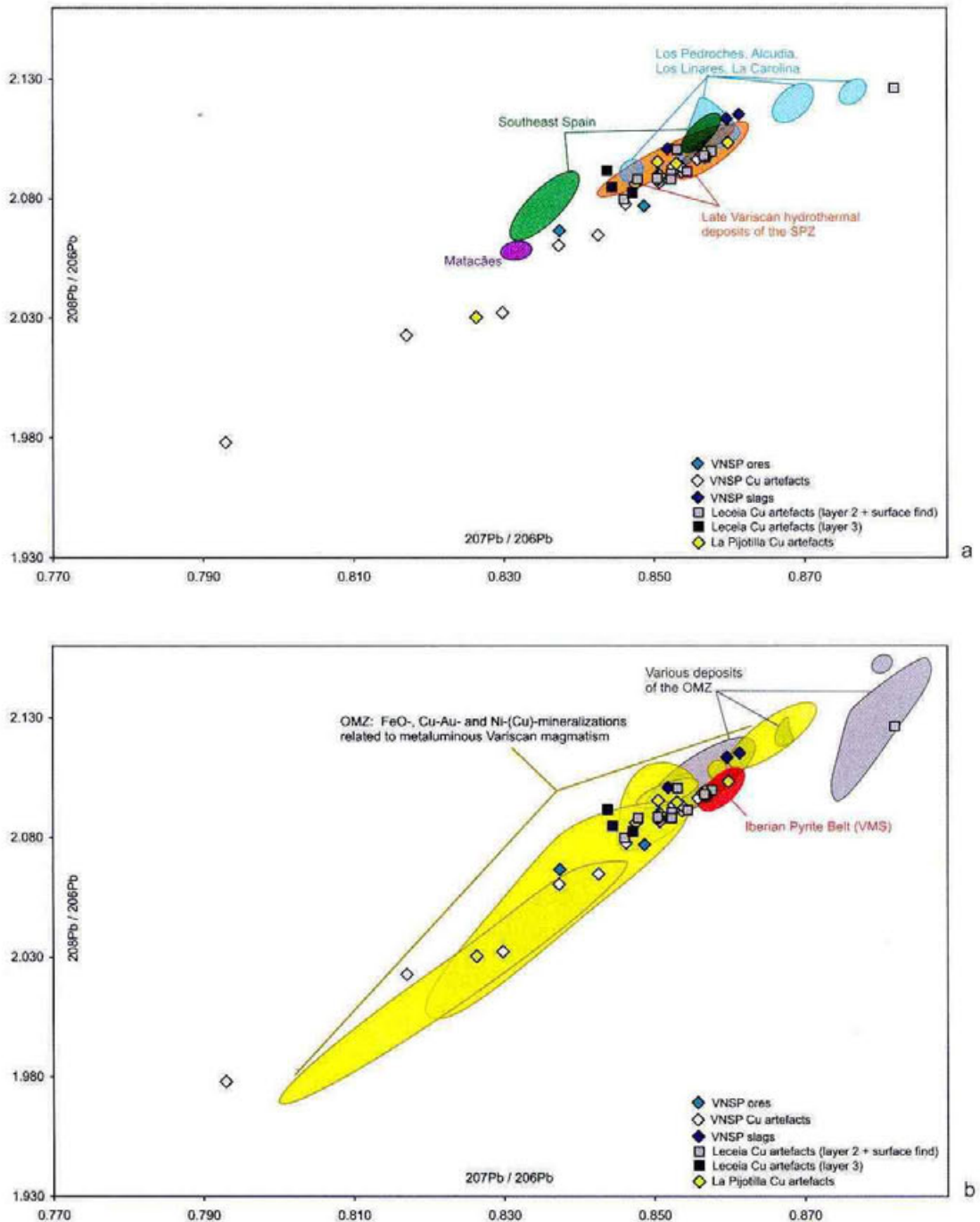


Fig. 13 Lead isotope data of Leceia copper artefacts in comparison to hitherto published data of copper artefacts, slags and ores from VNSP, La Pijotilla and of south Iberian ore deposits (see references below, in "Provenance of figures"). The isotope fields of the deposits found to be relevant in the $^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{208}\text{Pb}/^{206}\text{Pb}$ diagrams (fig. 13 a. b) are broken down to individual data points in the $^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ diagram (fig. 13 c) to allow the third dimension of the data set to be considered. a. b Analytical error d° size of the symbols; c Error cross indicates 2σ -error of 0.1%. Analyses with higher uncertainties are indicated by extended error bars.

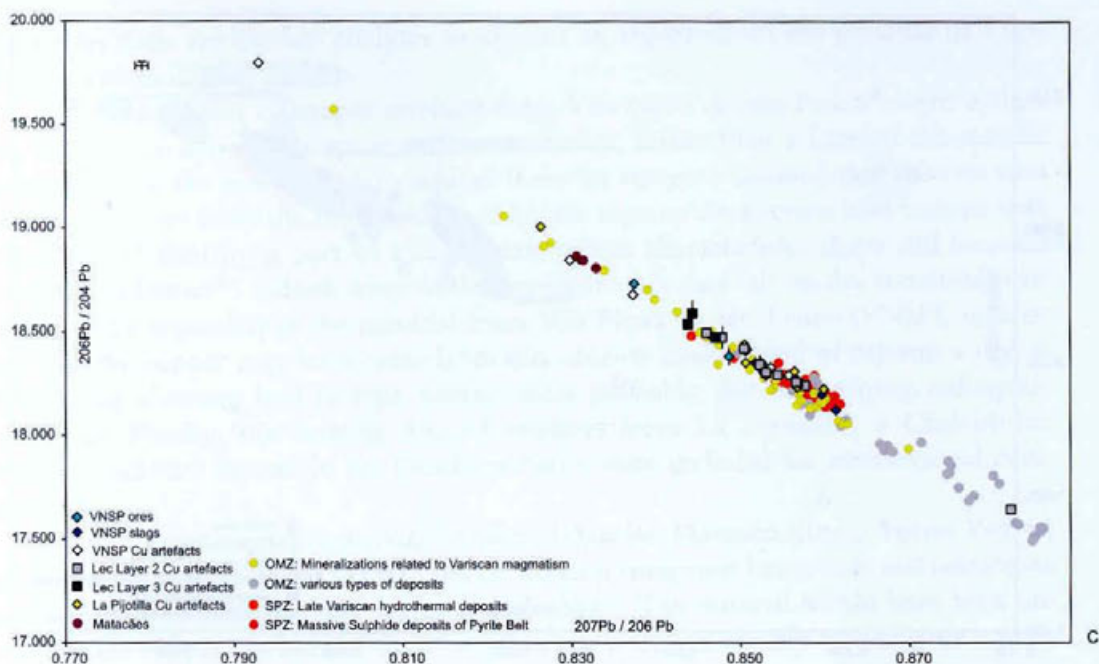


Fig. 13.

Cu-Au- and Ni-(Cu)-mineralizations related to the metaluminous Variscan magmatism (Isotope Group 5 according to Tornos and Chiaradia)⁶⁰ correlate very well both with the ones of the Leceian artefact assemblage, as well as the ones from VNSP. Remarkably, even the extreme outlier of the Leceia assemblage falls into a lead isotope field defined by mineralizations of the Ossa Morena Zone. Furthermore, the hypothesis of an exchange of copper between the Ossa Morena Zone and the Estremadura during the 3rd millennium B.C. is strongly supported by the fact that copper artefacts found at the contemporary fortified site of La Pijotilla correlate both with the lead isotope signatures of artefacts from Leceia and VNSP, as well as with ores from the Ossa Morena Zone.

At least since the Roman times, economically speaking the most famous south Iberian copper ore deposits are to be found in the lower Alentejo region associated with the volcanogenic massive sulphide mineralizations of the Iberian Pyrite Belt (IPB). The lead isotope field drawn in fig. 13 b comprises 110 analyses of primary as well as secondary ores. The small field covers isotope ratios of samples deriving from 27 of the 80 thus far known deposits and distribute over an area that is 250 km long and 25–70 km wide⁶¹. It is quite unlikely that these deposits were a major copper supplier for the Leceian community since only two of the artefacts from Leceia fall into the Iberian Pyrite Belt field. The tight field can account even less for the large lead isotope scatter of the VNSP material.

⁶⁰ Tornos – Chiaradia 2004, 977.

⁶¹ Marcoux 1998.

Next to the massive sulphide deposits, there are few other, smaller copper bearing occurrences in southern Portugal, which have not been studied much to date⁶². Lead isotope data of 19 polymetallic ores from 11 late Variscan hydrothermal deposits exist (fig. 13). Since only individual samples were analysed for some of the deposits, the data points were grouped into two fields, as suggested by their geological association⁶³. Indeed, some of the Leceian artefacts fall into the two fields. However, since the fields do not resemble individual deposits but an assemblage of individual samples from various deposits, this association would require more analyses. Furthermore, the two fields neither can account for the entire Leceian, nor for the larger Estremadurian lead isotope scatter of copper artefacts including the VNSP material.

East and northeast of the Ossa Morena Zone, in the so-called Central Iberian Zone, there are the famous ore districts of Los Pedroches, Alcudia, Los Linares and La Carolina. The deposits are associated with polymetallic Pb-Zn-mineralizations. Santos Zalduegui and co-workers⁶⁴ structure the deposits according to their lead isotope signatures into four groups based on the analysis of 135 samples of galena (fig. 13). Again, some of the copper artefacts fall into the fields defined by these deposits. Yet, the fields can neither account for the Leceian artefact data distribution, nor for the VNSP one.

Finally, southeastern Spain, i. e. eastern Andalusia and southern Murcia, is a very important region to discuss in the context of Estremadurian Chalcolithic copper artefacts. Although more recent archaeological excavations have revealed dense patterns of 3rd millennium fortified settlements in various south-Iberian regions (e. g. Guadiana and Guadalquivir basins), both the Estremadura and southeastern Spain still today are considered to be two core regions of more advanced Iberian civilizations of the 3rd millennium B.C.⁶⁵. There are several copper occurrences located in this region. They are associated with various ranges of mountains of the Betic Cordilleras. 64 samples of galena, complex sulphides and copper carbonates from these deposits have been analysed thus far (fig. 13). According to Arribas and Tosdal⁶⁶, these mineralizations can be divided into two groups. Especially the geologically younger ones, represented by the bigger southeastern isotope field located left of the smaller one, are supposed to contain rich secondary copper ores and native silver. The lead isotope compositions of these southeastern deposits clearly differ from the signatures found in the copper artefacts from Leceia and VNSP.

In conclusion, the high density of 3rd millennium B.C. fortified sites in the Estremadura are in complete contrast to the scarce copper occurrences of the region⁶⁷. Indeed, some important Estremadurian sites such as Leceia, VNSP and Penedo do Lexim show an initial phase of occupation that either lacks completely or shows only very few archaeometallurgical finds. The few analyses of ores from Matacães also indicate that – if at all – this local copper occurrence does not seem to have played a crucial role in

⁶² Dr. João Matos, personal communication.

⁶³ Marcoux – Sáez 1994.

⁶⁴ Santos Zalduegui et al. 2004.

⁶⁵ Gilman 1987; Schubart 1990; Kunst 1995.

⁶⁶ Arribas – Tosdal 1994.

⁶⁷ Sousa et al. 2004; Müller – Soares 2008, fig. 5.

Estremadurian Chalcolithic copper procurement. Hence, it is very likely that copper was imported from regions beyond the Estremadura, i. e. over distances greater than 100 km. It seems that raw copper – perhaps already in the form of ingots⁶⁸ – was exchanged, since no copper smelting, but casting remains were found at Leceia. Of all copper occurrences analysed thus far, lead isotope data of deposits of the Ossa Morena Zone show the best correlation with the lead isotope signatures of the artefacts from Leceia and VNSP. They are very similar to lead isotope signatures of artefacts from Zambujal and the neighbouring sites of Penedo and Fórnea⁶⁹. In addition, copper artefacts found at La Pijotilla – a contemporary site located in the OMZ – also show the same lead isotope signatures, supporting a model of copper mining and smelting of OMZ ores, as well as an interregional exchange of the copper produced. Indeed, field surveys conducted by Gert Goldenberg in the Ossa Morena Zone seeking prehistoric mining sites revealed a range of mines that we think were already exploited in prehistory, most probably during the Copper and/or Bronze Age (e. g. Mocissos, see fig. 1)⁷⁰.

Future analyses will have to examine critically these conclusions. Most importantly, we need more detailed mineralogical and geochemical analyses of late Variscan hydrothermal and epithermal copper deposits of the South Portuguese Zone and of Central Iberian Zone copper ore deposits just north of the Estremadura (fig. 1). The latter ones are of special interest, since they are located in a region that geographically speaking appears easily accessible for Estremadurian coastal people, due to the Tagus river connection.

The idea that the people of Leceia obtained their metal from sites in the Ossa Morena Zone is supported by the fact that the procurement of amphibolite gained more importance during the Copper Age as well⁷¹. Petrographic analyses have shown that amphibolite found at the Estremadurian sites of Leceia and Zambujal was imported over distances of 80–100 km from sources of the Ossa Morena Zone, some of them in an unworked state⁷².

7. Conclusion

The copper artefacts from Leceia analysed thus far are made of arsenic-only copper, like most of the Portuguese Chalcolithic metal artefacts. At this stage of research it is not possible to say whether the metal derived from native copper or whether it was extracted from ores. In any case, the copper from Leceia is clearly different from arsenical copper smelted from primary or secondarily altered fahlores, which were found e.g. at Almizaraque, next to other types of ores. The arsenic content of the artefacts show a natural distribution, which is very similar to the lognormal distribution of trace and minor elements in ores. Nevertheless, there is a tendency of thinly hammered ar-

⁶⁸ Cardoso – Fernandes 1995.

⁶⁹ Müller et al. in press.

⁷⁰ Müller et al. in press.

⁷¹ Cardoso 1999; Cardoso 2004.

⁷² Cardoso – Carvalhosa 1995; Uerpman – Uerpman 2003.

tefacts, i. e. blades and sheet metal fragments, as well as elongated objects, i. e. the long awl and the fishhooks, having higher arsenic contents than axes, chisels and simple awls.

According to the existing lead isotope data, it is more likely that the copper metal found at Leceia originates geologically from deposits located in the Ossa Morena Zone, than from Matacães (Estremadura), the Iberian Pyrite Belt, or ore deposits located in south-central and in southeastern Spain. This Leceian metal provenance hypothesis is strongly supported by similar lead isotope distributions of copper artefacts from Vila Nova de São Pedro, Zambujal and La Pijotilla. Furthermore, amphibolite found at Estremadurian Chalcolithic sites also seems to originate from sources located in the Ossa Morena Zone, strengthening the hypothesis of a 3rd millennium B.C. exchange route between these two regions.

It appears that during the 3rd millennium B.C., copper metal became a decisive part of an intensified production and exchange pattern in southern Portugal along with other commodities such as amphibolite, gold, flint, and probably also elaborated organic materials and food. In the case of Leceia however, the direct accessibility of copper resources do not seem to have played the crucial role for founding the complex fortified site. Metal droplets and casting remains occur in various restricted places of inhabited space indicating that copper artefact manufacture was incorporated in daily domestic life like other crafts. In order to develop further our understanding of how the new material was valued, we need to find the primary production places and need to conduct similar comparative studies with material assemblages of smaller sites which supposedly were dominated by central fortifications such as Leceia, Zambujal and Vila Nova de São Pedro.

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9. Appendix

X-ray fluorescence analysis: experimental set-up

A Spectrace 5000 instrument was used, equipped with a Rhodium target X-ray tube. Tube voltages of 35 kV and 50 kV at a current of 0.35 mA were applied. The counting time for each analysis was 1000 s. To optimize the excitation conditions, a rhodium filter was used at 35 kV and a copper filter at 50 kV tube voltages. The raw data was processed using a fundamental parameter method, described by Lutz and Pernicka⁷³. The investigation focused on metallic drill shavings instead of corroded surfaces. Accuracy and precision were ensured by measuring a BAM 227 copper standard parallel to each analysis session comprising nine analyses each. As an example, Lutz and Pernicka documented an analytical precision of $\pm 20\%$ for measuring arsenic at concentrations higher than 0.1 wt % in copper-arsenic alloys with low amounts of lead⁷⁴. Only the data of elements occurring in amounts of less than 0.1 wt % often show higher uncertainties and are less accurate⁷⁵. In the case of iron the analytical uncertainty rises significantly at concentrations below 1 wt %⁷⁶. The certified values of the BAM 227 standard were also reproduced during the analyses of this project (tab. 4). Analytical problems occurred in the measurement of selenium and gold, which is why they were excluded from data evaluation. In any case, they are not of relevance for the discussion of the results.

Tab. 4 BAM 227 standard measurements run parallel to all analysis sessions of nine measurements each. Time period: May 2004 – January 2006. The same analysis setting was used as described by Lutz – Pernicka 1996. In the case of bismuth, values just above the detection limit were detected in few cases. The silver value used to confirm the XRF measurement is an unpublished value obtained by Ivelin Kuleff using neutron activation analysis.

	Fe	Ni	Cu	Zn	As	Pb	Bi	Ag	Sn	Sb	Te
certified values	0.129	0.284	85.57	3.46	0.081	4.12	0.0088	0.019 ^a	6.01	0.160	0.0012
mean of 21 XRF analyses	0.176	0.299	86.92	2.57	0.087	4.11	< 0.005 ^a	0.018	5.49	0.136	< 0.008
rel. difference [%]	-36	-5	-2	26	-7	0.3	-	6	9	15	-
standard deviation [%]	12	10	1	9	28	4	-	18	6	7	-

Lead Isotope analysis: experimental set-up

Lead isotope analyses were undertaken using a double focusing magnetic sector based MC-ICPMS (Axiom, VG Elemental); the instrument was set-up as developed by Niederschlag and co-workers⁷⁷. The drill shavings were dissolved in ultra-high purity

⁷³ Lutz – Pernicka 1996; Lutz – Pernicka unpublished.

⁷⁴ Lutz – Pernicka 1996; Lutz – Pernicka unpublished.

⁷⁵ Compare with Lutz – Pernicka 1996; Lutz – Pernicka unpublished.

⁷⁶ Lutz – Pernicka 1996; Lutz – Pernicka unpublished.

⁷⁷ Niederschlag et al. 2003.

nitric and in hydrochloric acid and then drained over ion exchange columns filled with a cat ion-exchanger, in order to separate the lead from the copper matrix. The lead fractions of the samples – dissolved in 2 % HNO_3 – were then analysed in 3 runs, each comprising 20 single measurements. Mass fractionation was corrected by adding a standard solution to the samples, which contained a known $^{205}\text{Tl} / ^{203}\text{Tl}$ ratio. Analyses of the NIST standard SRM-981, run parallel to the sample measurements, revealed an experimental uncertainty of $\sigma = 0.01\%$ for the $^{208}\text{Pb} / ^{206}\text{Pb}$ and $^{207}\text{Pb} / ^{206}\text{Pb}$ ratios and $\sigma = 0.05\%$ for the $^{206}\text{Pb} / ^{204}\text{Pb}$ ratio. However, the analyses of the Leceia samples were somewhat less precise, due to the extremely low lead content of the copper. Regarding the $^{206}\text{Pb} / ^{204}\text{Pb}$ ratio measurements the analytical error rose higher than 0.1 % in five cases (tab. 3).

Lead Isotope and Trace Element Analyses in Archaeometallurgical Provenance Studies

Since its breakthrough in the early 1980s, lead isotope analyses have been applied in various archaeological programs to trace the provenance of early copper or bronze artefacts⁷⁸. In short, common lead is composed of the four stable isotopes ^{204}Pb , ^{206}Pb , ^{207}Pb and ^{208}Pb . The quantities of the latter three isotopes increase in the course of time, as they form the end product of radioactive decay chains of ^{238}U , ^{235}U and ^{232}Th ; ^{204}Pb is called primeval lead, because its quantity has not changed since the creation of the earth. During the formation of an ore deposit, uranium and thorium are separated for the most part from the lead, thus the formation of radiogenic lead is interrupted. In geology, this phenomenon is used for dating ore deposits⁷⁹.

Whereas during metallurgical processes, like roasting, smelting and refining, the ratio of the copper to minor and trace elements may change dramatically, the ratios of the isotopes of lead, present as a trace in nearly every prehistoric copper artefact, do not. Hence, ideally ores and copper artefacts can be related to each other. However, there are a number of limitations to the method. Firstly, several ore deposits may have the same lead isotope signature. Secondly, deposits can be very inhomogeneous, because of various cycles of mineralization. Thirdly, if the amount of lead in the ores is low, small quantities of uranium and thorium that were not fully separated during ore formation can still distort the lead isotope ratios of the deposit. This often results in a huge variation of results within one deposit. Finally, it is obvious that determining metal provenance based on lead isotope ratios becomes more complicated as soon as ores or metals from different sources are mixed during the metallurgical processes. For instance, the lead isotope composition of a copper artefact that is produced of copper from two different sources will plot onto a mixing line linking the two data points that represent the original lead isotope composition of the two separate copper sources⁸⁰. More complex mixing polygons would emerge once there are more lead sources involved (e. g. from potentially lead-rich gangue minerals, alloying agents or clays of a reactor vessel used).

⁷⁸ Gale – Stos-Gale 1981; Pernicka 1987; Pernicka 1990; Rohl – Needham 1998; Hunt 2003.

⁷⁹ Faure 1986.

⁸⁰ Pernicka 1987.

These limitations only allow to exclude, rather than to unambiguously trace down a single ore deposit as the ore source. Nevertheless, in combination with trace element analyses and other archaeological arguments, the range of possible ore sources can be narrowed down significantly in many cases⁸¹.

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Provenance of figures: Fig. 1: Map based on Quesada et. al 1990 and Müller et al. in press. – Fig. 2–5. 7. 11. 12: Authors. – Fig. 6. 8: based on XRF data of this paper and FNAA data published by Cardoso – Guerra 1997/1998. – Fig. 9. 10: based on XRF data published by Rovira et al. 1997. – Fig. 13: based on lead isotope data of this paper and data published in Arribas – Tosdal 1994; Marcoux – Sáez 1994; Stos-Gale et al. 1995; Marcoux 1998; Trincherini et al. 2001; Hunt 2003; Santos Zalduegui et al. 2003; Tornos – Chiaradia 2004; Müller – Soares 2008, this volume.

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