THE DEVELOPMENT OF SPANISH METALLURGY AND COPPER CIRCULATION IN PREHISTORIC SOUTHERN SPAIN

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1. INTRODUCTION

There is no doubt that in the Spanish Copper Age the earliest technological developments of copper smelting, and the extraction of silver and tin, were based on the mineral resources of the Iberian Peninsula. Much work has been conducted recently in Spain by scientists trying to link the trace elemental composition of prehistoric metal artefacts with the known ore sources, in particular a large amount of data on copper based metals and silver artefacts from all districts of Spain have been published recently by Rovira Llorens [1]. However, the archaeometallurgical survey of mineral resources, that needs to accompany such research, is largely limited to southern Spain, in particular the south west [2].

In the eastern Mediterranean in the last twenty years there were several projects combining archaeometallurgical surveys and chemical and lead isotope analyses of ores and Bronze Age artefacts [3 and 4]. The lead isotope compositions of ores and artefacts are used as their characteristic 'fingerprints' [5]. The analytical method used currently for lead isotope characterisation of archaeological materials and ore minerals is Thermal Ionisation Mass Spectrometry (TIMS). This technique provides highly accurate data for lead isotope (LI) compositions of lead present in ancient metals, glasses, glazes and pigments, even if lead is present only at the level of a few parts per million [6]. The LI provenance studies are based on the diversity of proportions of lead isotope atoms in ore minerals related to their geological history. This feature allows to characterise geographical sources of minerals according to the pattern of the lead isotope ratios measured in them. The lead isotope characteristics of ores do not change during their smelting, carrying the LI fingerprint of the ore into the metal.

The methodology of lead isotope provenance research is based on accumulating isotopic, geochemical and archaeometallurgical information about known ore deposits

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and ancient smelting sites, to provide the information about ancient exploitation of the deposits and their unique LI 'fingerprint'. The second step involves obtaining lead isotope and chemical analyses of metal artefacts and comparing them one by one with the datapoints obtained for ores from known localities the ores [7].

2. ARCHAEOMETALLURGY IN SPAIN

Spain, and in particular the region of Huelva, is very rich in copper, tin, silver and lead deposits. The archaeometallurgical excavations in Huelva prove that the ores were exploited since the Chalcolithic period [8 and 9]. The research into the archaeometallurgy of the south-east seems to be less advanced at present. However, archaeological and archaeometallurgical surveys have been conducted in the Vera basin, which is the locality of many Bronze Age settlements [10, 11 and 12]. On many of these sites there are excavated pieces of copper minerals [1 and 11] and therefore it is usually assumed that copper extraction was carried out in the settlements. This is a conclusion that might require a further investigation. For example, on Chalcolithic Balkan sites copper carbonates are also present in the context of the settlements, but the lead isotope analyses show that the metals found on these sites are of quite different origin than the minerals [4 and 13]. However, for example, the evidence from Almizaraque in the Vera basin seems to support the local copper extraction, because many sherds with adhering minerals have been found [11]. It would be interesting to know what was the scale of copper extraction in these settlements. How much metal was extracted in this way? Was the knowledge of copper extraction widely spread, or is it simply that the copper ores were very rich and it was easy to reduce copper carbonates to metal without the use of fluxes? In the eastern Mediterranean, even in the Early Bronze Age, the copper smelting was a highly specialised activity documented by extensive smelting sites, easy to find even today, because of the considerable amounts of slag. Some of the large slag heaps in Greece and the Near East are in the vicinity of considerable ore occurrences and show typical features of industrial, rather than habitation sites. Some of them have been extensively studied and dated to very early periods [14, 15, 16 and 17]. The slags are also abundant in Huelva [9 and 18]. A preliminary report from, what appeared to be a similar industrial site in Andalucia, was published some years ago by Rothenberg [19], but so far no more information about this or other similar sites in the south-east Spain seems available.

This might be an interesting question: if the ores in south-east Spain have been used during the Bronze Age and their smelting was confined to settlements rather than industrial sites, then the social organisation of metal extraction in this region was quite different than in other metallurgically active Mediterranean regions. It is possible that lead isotope studies of ores and metal artefacts from southern Spain could perhaps help with better understanding of the social organisation and the patterns of exploitation of local resources in the early stages of the metal using cultures.

3. THE LEAD ISOTOPE CHARACTERISATION OF SPANISH ORES

The lead isotope database for ores from southern Spain currently available is very limited and quite insufficient for a proper LI characterisation of possible Bronze Age copper sources. The lead isotope analyses for the south-east are mostly of lead ores [6 and 20] and there is no systematic and sufficient lead isotope characterisation of any ancient copper smelting or mining site. For the south-west there are nearly 100 lead isotope analyses of copper ores and slags obtained some years ago by Dr. Marcos Hunt in Oxford (mostly unpublished), but these data show quite large range of isotopic ratios and it is clear that much more systematic analytical work is needed to understand the LI patterns characteristic of copper from this region (see figure 1). However, the current LI data for Spanish ores indicate that the pattern for the south-east is confined mostly to the $^{207}Pb/^{206}Pb$ ratios below 0,84, while the copper ores from south-western Spain show the values for $^{207}Pb/^{206}Pb$ LI ratios mostly between 0,84-0,863 [6 and 20]. It is known that the ores in Sierra Alhamilla have LI ratios similar to the ores from the south-west, but these occurrences are Pb-Zn mineralisations therefore they are not relevant to the question of early copper extraction. Also it seems that

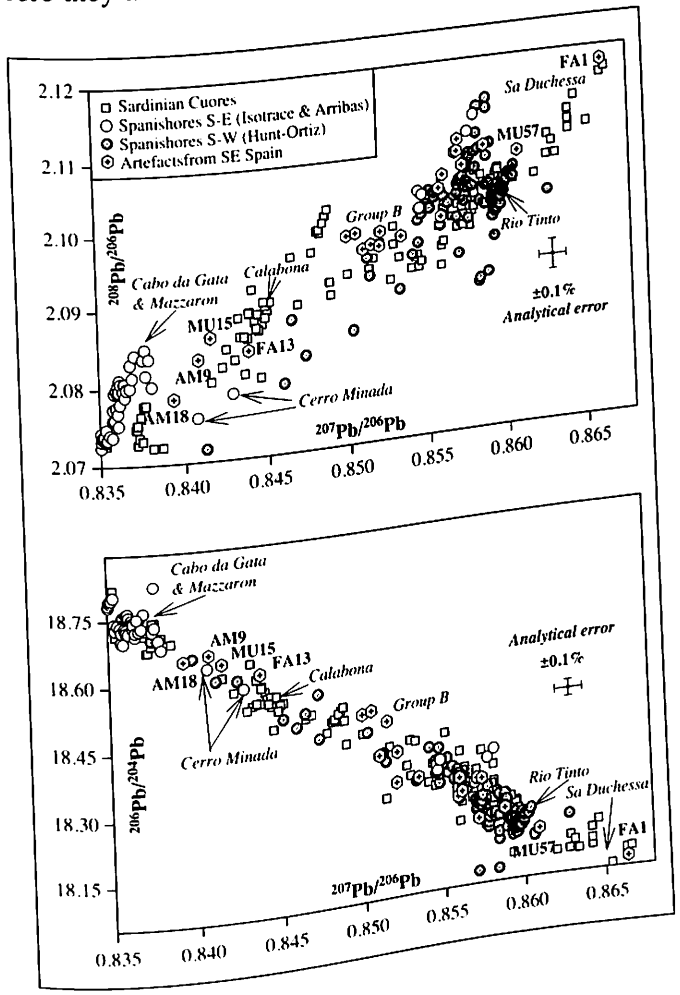

Figure 1. Lead isotope compositions of Spanish ores compared will copper ores from Sardinia and Bronze Age metal artefacts from SE Spain.

certain similarity exist between ores from some of the ore deposits on Sardinia and the ores from Huelva [6]. On the whole though, the available lead isotope data for Spanish ores shows that most of the analysed mineral samples have quite distinctive lead isotope ratios for different mines and also the differ from other Mediterranean ores. However, not all deposits are represented amongst the existing lead isotope data. Also, the number of samples of ores from each specific occurrence analysed so far is too small.

This situation resembles the early years of lead isotope studies in the eastern Mediterranean: for many years there were only 38 data points available for copper ores from Cyprus and the extend and shape of the so called 'Cypriot field' was open to much speculation. Currently we have a database of over 600 data on Cypriot ores and ancient copper slags from this island and the characterisation of 'Cypriot' lead isotope ratios is far more accurate, and significantly different. The comparative lead isotope analyses of copper-based artefacts from Cypriot archaeological sites allow identifications of the chronological and spatial pattern of ore exploitation. Also, the lead isotope analyses demonstrated that copper for large 'oxhide' shaped ingots was obtained from only one or two mines in north-west of the Troodos Mountains [7].

4. THE LEAD ISOTOPE ANALYSES OF COPPER-BASED ARTEFACTS FROM SPANISH BRONZE AGE SITES

Some years ago we have undertaken a pilot project on lead isotope analyses of the copper-based artefacts from the Bronze Age sites in south-east Spain. The focus of this study was the provenancing of copper used for the production of artefacts excavated on the site of Gatas (Almeria). This study will be published elsewhere (Chapman and Stos-Gale, 2000). In this paper I would like to present 21 analyses of copper based artefacts and two silver rings from several other Bronze Age sites in the south-east Spain. The brief description of the copper based artefacts is listed in table 1 together with the ED XRF data for their major elemental composition. The analyses were made on samples drilled from the artefacts (about 20 mg of metal). All these elemental analyses are semiquantitative with the detection limits for all major metals around 0,2% and the analytical error of about 10% of the element. Some of these artefacts have been also analysed by Rovira Llorens *et al.* [1]. Small discrepancies between the data published here (table 1) and these published by Rovira Llorens *et al.* [1] are most likely due to the inhomogeneity of arsenic in the analysed samples. The analyses of Rovira Llorens *et al.* are perhaps a more accurate representation of the composition of a given artefact, but the analyses presented here represent the metal compositions of samples for which the lead isotope analyses were done. Lead in all these artefacts almost certainly originates from the copper minerals used for extraction of copper metal. Only one artefact, a small bronze bar from Fuente Alamo (FA6), contains lead in quantity above 1%, but even that is not unusual for copper smelted from multimetallic ores and cannot be regarded as a deliberate addition of lead. Two other artefacts in this table c[ontain high amounts of tin, an awl (or perhaps another small bar) from Cabezo Negro (MU50)

Table 1. Metal composition of copper-based metal artefacts from South-East Spain.

Number	*Museum Number*	*Site*	*Description*	*Date* **	*% As*	*% Sn*	*% Fe*	*%Cu*	*% Pb*
AM 15	Almería 13763	El Argar	Small axe	Argaric	2,3	<0.2	0,2	97,5	<0.1
AM 18	Almería 14014	El Argar	Sword, five rivets	Argaric	2,8	<0.2	0,2	97,0	<0.1
AM 20	Almería 14025	El Argar	Dagger, four rivets	Argaric	1,4	5,7	<0.2	92,9	<0.1
AM 9	Almería No Inv. No.	Terrera Ventura	Copper droplet	Argaric	2,8	<0.2	0,2	97,0	<0.1
BA 1	Almería 52557	El Barranquete	Awl	Argaric	0,9	<0.2	<0.2	98,5	<0.1
BA 2	Almería 52505	El Barranquete	Bracelet	Argaric	1,8	<0.2	0,2	98,3	<0.1
BA 3	Almería 52506	El Barranquete	Small Dagger	Argaric	0,9	<0.2	0,2	98,9	<0.1
BA 4	Almería 52507	El Barranquete	Small Dagger	Argaric	1,0	<0.2	0,2	98,5	0,2
FA 1	Almería FA 600/5	Fuente Álamo	Dagger, two rivets	Argaric	1,4	<0.2	<0.2	98,5	<0.1
FA 12	Almería 1494/6	Fuente Álamo	Dagger, four rivets	Argaric	1,2	<0.2	0,2	98,7	<0.1
FA 13	Almería 1432/5	Fuente Álamo	Axe	Argaric	1,2	1,1	0,2	97,5	<0.1
FA 15	Almería FA1477/1	Fuente Álamo	Dagger, three rivets	Argaric	0,6	<0.2	0,2	98,9	<0.1
FA 6	Almería FA 74	Fuente Álamo	Bronze bar	Argaric	0,3	20,7	0,3	76,7	1,8
MU 15	Murcia 576	La Bastida	Dagger, three rivets	Argaric	1,1	0,9	0,2	97,2	0,6
MU 21	Murcia No Inv. No.	Rincón de Almendricos	Dagger	Argaric	1,7	<0.2	<0.2	98,3	<0.1
MU 32	Murcia No Inv. No.	Monteagudo	Dagger	Argaric	4,1	<0.2	0,2	95,8	<0.1
MU 48	Murcia CN-72-I-II-29	Cabezo Negro	Fragments	Argaric	1,1	1,2	0,2	97,5	<0.1
MU 49	Murcia	Cabezo Negro	Small knife	Argaric	1,4	0,5	0,2	97,9	<0.1
MU 50	Murcia 29-X-77	Cabezo Negro	Small Awl	Argaric	1,2	18,6	<0.2	80,0	0,2
MU 57	Murcia No Inv. No.	Cantera de Murviedro	Bracelet	? Copper Age	0,4	13,2	<0.2	86,4	<0.1
MU 59	Murcia 354	Cantera de Murviedro	Small saw	? Copper Age	2,2	<0.2	<0.2	97,2	<0.1

Table 2. Lead isotope analyses of metal artefacts from South-East Spain.

Number	Museum No.	Region	Site	LI Group	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{206}Pb/^{204}Pb$
BA 1	Almería 52557	Níjar	El Barranquete	0	1,82026	0,74369	21,285
MU 59	Murcia 354	Lorca	Cantera de Murviedro	0	2,05401	0,83048	18,921
AM 18	Almería 14014	Antas	El Argar	A	2,07842	0,83935	18,649
AM 9	Almería No Inv. No.	Tabernas	Terrera Ventura	A	2,08330	0,84086	18,659
MU 15	Murcia 576	Totana	La Bastida	A	2,08602	0,84162	18,636
FA 13	Almería FA 1432/5	Cuevas	Fuente Álamo	Sardinia-Calabona	2,08390	0,84389	18,604
MU 49	Murcia	Mazarrón	Cabezo Negro	B	2,09784	0,85004	18,480
MU 48	Murcia CN-72-I-II-29	Mazarrón	Cabezo Negro	B	2,09809	0,85061	18,485
BA 2	Almería 52505	Níjar	El Barranquete	B	2,09579	0,85105	18,381
AM 15	Almería 13763	Antas	El Argar	B	2,09631	0,85156	18,456
MU 21	Murcia No Inv. No.	Lorca	Rincón de Almendricos	B	2,09596	0,85208	18,314
MU 32	Murcia No Inv. No.	Murcia	Monteagudo	B	2,09793	0,85216	18,383
FA 15	Almería FA 1477/1	Cuevas	Fuente Álamo	B	2,09697	0,85346	18,318
MU 50	Murcia 29-X-77	Mazarrón	Cabezo Negro	Huelva	2,10307	0,85589	18,260
AM 20	Almería 140.25	Antas	El Argar	Huelva	2,09909	0,85600	18,301
FA 6	Almería FA 74	Cuevas	Fuente Álamo	Huelva	2,10491	0,85615	18,270
BA 4	Almería 52507	Níjar	El Barranquete	Huelva	2,10777	0,85695	18,286
BA 3	Almería 52506	Níjar	El Barranquete	Huelva	2,10790	0,85709	18,295
MU 3	Murcia No Inv. No.	Totana	La Bastida, silver ring	Huelva	2,10950	0,85715	18,202
FA 12	Almería FA 1494/6	Cuevas	Fuente Álamo	Huelva	2,10595	0,85739	18,293
MU 25	Murcia No Inv. No.	Orihuela	San Antón, silver ring	Huelva	2,10834	0,85879	18,218
MU 57	Murcia No Inv. No.	Lorca	Cantera de Murviedro	Huelva	2,10713	0,86092	18,160
FA 1	Almería FA 600/5	Cuevas	Fuente Álamo	Sardinia-Sa Duchessa	2,11806	0,86637	18,072

and a bracelet from Cantera de Murviedro (MU57). Five other artefacts contain between 0,5%-5,7% of tin. The small saw from Cantera de Murviedro (MU59) seems to have about 0,7% of Ni. All these artefacts contain on average 1-2% of arsenic.

Lead isotope compositions of these artefacts and two spiral silver rings from San Anton (MU25) and La Bastida (MU3) are listed in table 2. Each of the artefacts is characterised here by three lead isotope ratios $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$ and $^{206}Pb/^{204}Pb$. These three numbers can be represented as the parameters of points in three-dimensional (3D) space. The most convenient way of comparing these data points between themselves and with other lead isotope data, is by plotting them as mirror images of these 3D points on two 2D planes. Such two mirror diagrams for lead isotope data sets for samples of copper and lead ores from Spain and Sardinia are plotted on figure 1.

The data for Spanish ores available at present shows a certain division between the ores from SE Spain (plotting in the left hand corners on both diagrams) and SW Spain. However, Some copper ores from Huelva (Minas la Cala) and lead ores from SE (Sierra Alhamilla and Gador) show lead isotope ratios quite different to the other ores from their regions. A few lead isotope analyses of copper ores from Cerro Minada, a mine located near the road Almeria-Murcia, indicate that the lead isotope ratios of this mineralisation are different from the lead ores in the Cabo da Gata and Mazzaron. Also, the ores from certain mines in Huelva (El Trastejon, Minas de Cala, Mina de Teuler and Mina La Sultana) on the present evidence have lead isotope ratios showing a considerable range of values. Such variable lead isotope ratios are characteristic of copper minerals with very low lead concentrations and comparatively high uranium contents, often occurring together with iron sulphides. A very similar range of lead isotope ratios is known from the mines of Feinan in Jordan and Timna in Israel [15]. There is no doubt that many more lead isotope analyses of minerals from all these mines are needed, together with the LI analyses of slags from dated metallurgical sites. Only in this way it would be possible to decide if the ores from these occurrences were exploited in the Bronze Age. Many metallurgical sites are known in Huelva [9], but only a few so far in the south-east [21] and there are no lead isotope analyses available for the metallurgical debris from these sites. Therefore, the present knowledge of the patterns of lead isotope data for copper ores in southern Spain is not known well at all, but on the geological grounds it seems probable that the pattern of higher $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ values for the south-east represents the real situation.

The artefacts in table 2 are arranged not according to the sites where they were excavated, but according to the values of their lead isotope ratios. This arrangement makes it easier to compare the LI ratios of the artefacts with the ores. The column 'LI Group' indicates the position of the artefacts on the diagrams plotted on figure 1.

Two of the artefacts marked as group '0' (an awl from el Baranquette and small copper saw from Cantera de Murviedro) are not plotted on figure 1, because their lead isotope ratios are well beyond the range of these diagrams. Both of them are made from copper with 1-2% of arsenic and no other impurities in the excess of 0,2%. The lead isotope composition of the awl BA1 is quite unusual amongst any of the Mediterra-

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nean Bronze Age artefacts and indicates the lead-poor-uranium-rich type of the deposit. Amongst over 250 ingots of various types found on Nuragic Sardinian sites and analysed for their lead isotope compositions in our laboratory, there are seven ingot fragments with LI ratios very close to BA1. But there are no analyses of copper ores from Sardinia matching any of these ratios. On the other hand a few copper ores from the Libiola mine (Liguria, Italy) and some other small occurrences on the Ligurian coast show the same lead isotope characteristics. The other of these two artefacts (MU59) has also lead isotope ratios that appear amongst Sardinian ingots, but not the ores.

Two artefacts, a dagger (FA1) and an axe (FA13) from Fuente Alamo have lead isotope compositions identical respectively with ores from two Sardinian copper mines: Sa Duchessa and Calabona. There are also a number of Nuragic artefacts with the same lead isotope ratios. The axe contains 1,1% of tin, the dagger is made of arsenical copper.

Three objects from three different sites (AM9 and 18, MU15) form a group at the $^{207}Pb/^{206}Pb$ range 0,839-0,841. On figure 1 these three artefacts fall in one line just on the side of the ores from Calabona (Sardinia). Two of them are made of arsenical copper, the dagger MU15 contains also 0,9% of tin. The pattern formed by these objects and the range of their LI ratios indicates that they might originate from one ore deposit, possibly in SE Spain, but there are no ore analyses matching these three objects. At present they are described as LI Group A. A number of Iron Age artefacts from Menorca [22] and two Argaric copper objects from Gatas (Almeria) also have LI compositions consistent with the group A artefacts. The ores from Calabona (Sardinia) and two samples of ores from Cerro Minada (Almeria) seem similar in their LI ratios on the $^{206}Pb/^{204}Pb$ diagram (figure1) to artefacts in LI Group A, but on the upper diagram they are quite different isotopically. Only more lead isotope analyses of copper ores from the region of Almeria can help in finding the source of these copper artefacts.

Seven artefacts from various sites form another group falling separately from the known ore data. They are called here LI Group B. These artefacts do not coincide with any ore data available at present. Judging from the range of their lead isotope ratios and the pattern of datapoints, they might be indicating another group of ores from Huelva, but without more analyses it is impossible to decide if this is the case. Only two of the artefacts from this group contain small amounts of added tin (MU48 and 49: 1,2% and 0,5% respectively).

Finally, seven artefacts, excavated on six different sites, form a lead isotope group consistent with copper ores from Huelva. Four out of those are high tin bronzes (awl MU50, dagger AM20, bronze bar FA6, a bracelet MU57). Two silver rings from La Bastida and San Anton (MU3 and MU25) also fall into this group.

5. MIXING COPPER FROM DIFFERENT REGIONS

The question often asked by the archaeologists concerns re-using of metal: if the recycling of metals was widely spread then the isotopic compositions of artefacts will

not reflect those of the ores used for production of each primary copper metal. This statement is quite correct, but fortunately it can be tested by looking at the patterns of lead isotope data of ores and artefacts from the same region. Because each set of lead isotope data characterising a sample of ore or metal consists of three numbers, and because these numbers are linearly correlated, a 'mixture' of any two isotopically different leads originating from two pieces of metal or mineral of different lead isotope ratios, will fall on both LI plots (see figure 1) on the straight line joining the points representing the 'parent' metals or minerals. For example, the three artefacts AM18, AM9 and MU15 are on the upper diagram plotting along a line. Therefore one could suggest that the AM9 was made by melting together two pieces of metal of the same isotopic compositions as AM18 and MU15. However, on the lower diagram these three artefacts do not fall on the straight line, so the argument doesn't hold. One can make similar comparisons for larger groups of artefacts and ores. For example, the artefacts forming the LI Group B, could have been made of a mixture of metal originating from the Calabona and Huelva ores. This argument does not look very likely though, because the new metal pieces made from such a mixture are not very likely to form a coherent group; they would 'slide' up and down the lines joining the datapoints representing the ores from these two regions depending on the proportion of lead present in each set of the initial pieces. Consistent remelting and mixing of metals from different deposits would result in producing a pattern for the artefacts that looks like a random scatter of points over a range of lead isotope ratios representing the whole range characteristic of the ores used. The artefacts in Group B show a very small range of LI compositions, resembling the pattern usual for metals originating from one ore deposit.

The pattern of the lead isotope ratios representing the Bronze Age artefacts from Spain might change considerably with the increased number of analyses, but the current picture does not suggest a widely spread remelting and mixing together of copper metal originating from different regions. On the other hand, if the production of new artefacts from old relied on melting down a single piece of metal and re-casting it, or melting together pieces of metal from the same deposit (or predominantly so), then the lead isotope compositions of new artefacts will be identical with the initial ones. Therefore, for example the typology of an artefact made from a Sardinian copper ingot (or a broken Sardinian axe) in south-east Spain will look exactly the same as any other artefacts made in south-east Spain, but its LI composition will remain Sardinian.

The type of artefacts in each of the isotopic groups here does not indicate very clearly such situation. The two artefacts in LI Group 0 show very low $^{208}Pb/^{206}Pb$ lead isotope ratios that hardly can be achieved from mixing metals from different sources, they might be imports from Sardinia or north-west Italy. They are both very small artefacts (an awl and a small saw) made of arsenical copper. The three objects from LI Group A consist of two typologically Argaric artefacts (a sword and an axe) from El Argar and a small piece of copper from Terrera Ventura. Until more lead isotope analyses of copper ores from south-east Spain are available, the only possible ores to produce these lead isotope ratios seem to be the Sardinian ores. Is it likely from an archaeological point of view that these artefacts from El Argar are made of metal from Sardinia? Also, it would be useful in such case to take into the consideration the trace elemental compositions of these three objects and compare them with the elemental characteristics of nuragic copper artefacts.

6. CONCLUSIONS

The amount of lead isotope data for both: ores and Bronze Age artefacts from Spain is at present vanishingly small in comparison with the existing amount of pre-historic metal and the wealth of mineral resources on the Iberian Peninsula. But even this pilot study indicates that the pattern of exploitation and distribution of copper metal in the South Spain was quite complex. Several points can be already taken into consideration:

1. The copper metal excavated on each of the sites in SE Spain originates from several different sources.

2. Some of these sources of copper ores might be situated in the SE Spain, not far from the sites, but practically on each site also there is metal consistent with SW Spanish copper ores.

3. A small number (two out of 23) of artefacts from SE Spain is consistent with the lead isotope compositions of ores from Calabona and Sa Duchessa in Sardinia. It is quite likely that these mines were exploited in the Bronze Age, because there are also Sardinian copper artefacts with identical lead isotope ratios.

4. Another two artefacts might have come to SE Spain from the Ligurian coast, perhaps *via* Sardinia. However, this statement needs further verification in the lead isotope data for uranium-rich ores from Spain and also contemporary copper artefacts from Liguria.

7. ACKNOWLEDGEMENTS

The author gratefully acknowledges the help and cooperation of Professor R. Chapman, Dr. R. Risch and the archaeologists from the Almeria and Murcia Museums in obtaining the samples for analyses. I would like also to thank Dr. Marcos Hunt-Ortiz for his permission to use the lead isotope analyses of ores from Huelva. Professor Noel Gale was a prime mover in the first phase of this project and the analytical work took place while he was the director of the Isotrace Laboratory.

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