

# Raw material procurement and selection in Southeast Iberia's early metallurgy\*

## *Captación y selección de materias primas en la primera metalurgia del Sureste de la península ibérica*

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### ABSTRACT

The role of metallurgy in the Copper Age communities of the Iberian Southeast is a recurrent question of archaeological research in western Europe. Based on lead isotope and trace element analyses of archaeometallurgical remains, this paper addresses the territorial organisation of metallurgical production during the Copper Age (3100-2200 cal BC) in the Vera Basin (Almería, Spain), the region with the earliest metallurgical evidence in western Europe. This paper comprises the study of materials from the three main settlements with metallurgical activity in the area (Las Pilas, Santa Bárbara and Almizaraque), as well as some metal objects from these and other sites (La Encantada I, Loma de Belmonte and Las Churuletas 1).

The results support a model of small-scale regional production whereby settlements exploited the resources of their nearby surroundings (up to 30 km as the crow flies). However, metallurgical exploitation prioritised mineralisations rich in arsenic and other elements, even when other sources were

more readily accessible: for the case of Las Pilas, the exploitation of Pinar de Bédar sources instead of Sierra Cabrera, closer to the site; for the cases of Santa Bárbara and Almizaraque, the sources of Cerro Minado. The possibility that Almizaraque and Las Pilas also exploited the minerals of Herrerías, although to a lesser extent, remains open. Broader exchange networks are indicated by the data from finished objects, from which greater mobility can be inferred.

### RESUMEN

*El papel de la metalurgia en las comunidades de la Edad del Cobre del Sureste de la península ibérica es una cuestión recurrente en la investigación arqueológica en Europa occidental. A partir del análisis de isótopos de plomo y elementos traza de restos arqueometalúrgicos, este artículo aborda la organización territorial de la producción metalúrgica durante la Edad del Cobre (3100-2200 cal aC) en la cuenca de Vera (Almería, España); la región con las*

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*primeras evidencias metalúrgicas en Europa Occidental. Este artículo incluye el estudio de materiales de los tres principales asentamientos con actividad metalúrgica en la zona (Las Pilas, Santa Bárbara y Almizaraque), así como algunos objetos metálicos de estos y otros sitios (La Encantada I, Loma de Belmonte y Las Churuletas 1).*

*Los resultados sustentan un modelo de producción regional a pequeña escala mediante el cual los asentamientos explotaron varios de los recursos de su entorno cercanos (hasta 30 km en línea recta). Se priorizaron las mineralizaciones ricas en arsénico y otros elementos, incluso cuando otras fuentes eran más accesibles: para el caso de Las Pilas, la explotación de las fuentes de Pinar de Bédar en lugar de las de Sierra Cabrera, más cercanas al yacimiento; y para los casos de Santa Bárbara y Almizaraque, las fuentes de Cerro Minado. La posibilidad de que tanto Almizaraque como Las Pilas también explotaran los minerales de Herre-rías, aunque en menor medida, permanece abierta. La existencia de redes de intercambio más amplias queda reflejada por los datos de los objetos, a partir de los cuales se puede inferir una mayor movilidad.*

**Key words:** Archaeometallurgy; Chalcolithic; Southeast Iberia; Provenance Studies; Lead Isotope Analysis.

**Palabras clave:** Arqueometalurgia; Calcolítico; Sudeste de la península ibérica; Estudios de Procedencia; Análisis de Isótopos de Plomo.

## INTRODUCTION

The Vera Basin, in the Iberian Southeast, is a large tertiary basin spanning some 320 km<sup>2</sup> and traversed by three rivers: Aguas, Antas and Almanzora. It is framed by the Almagro and Almagrera mountain ranges in the North; Cabrera mountain range in the South; Bédar and Lisbona ranges in the West and the Mediterranean Sea in the East (Fig. 1).

Thanks to the extensive archaeological works that started in the late 19<sup>th</sup> century by Henry and especially Louis Siret, the extensive occupation of the Vera Basin during the Late Prehistory is well documented, with early metallurgical stages constituting a decisive moment of occupation (Camalich Massieu and Martín Socas 1999). This is the area yielding the earliest metallurgical evidence in Iberia (Ruiz Taboada y Montero Ruiz 1999) and a key region for the study of long term prehistoric social change, as it has a rich archaeological record from the Neolithic to the Bronze Age whereby progressive social stratification processes with increasing fortification of the sites, individualisation of burials and other changes in settlement and burial patterns can be observed, culminating in the El Argar society (see *e. g.* Chapman 2008; Aranda *et al.* 2015). Based on field-

work at Copper Age (c. 3100-2200 cal BC) sites such as Almizaraque (Cuevas del Almanzora), Las Pilas (Mojácar), Santa Bárbara (Huércal Overa), Zájara and Campos (Cuevas del Almanzora) as well as at Bronze Age (c. 2250-1550 cal BC) sites such as El Argar (Antas), Fuente Álamo (Cuevas del Almanzora) or Gatas (Turre), the Vera Basin has become a priority study area, with much archaeological research especially devoted to investigate the role of early metallurgy in the process of social stratification.

Some authors see metallurgy as a driving force of social change and a causal aspect of social stratification due to the socio-economic changes it could have brought about. This hypothesis is based on the premise that metalworking is a complex technology requiring full-time specialists and therefore encouraging social division of labour which, with the degree of specialisation and intensification reached during the Bronze Age, would have also entailed a territorial division of labour (*e. g.* Lull 1983, Lull *et al.* 2010; Chapman 2003; Delgado and Risch 2008; Molina González and Cámara Serrano 2009). Other authors give metallurgy a secondary social value and consider that, although it could have favoured the consolidation of elite power, under no circumstances did it entail a structural economic transformation that would have resulted in a process of social stratification. For these authors, metallurgy is considered to have been a low scale, domestic activity that required no full-time specialisation and therefore would not have generated dependence. The stratification processes of the Iberian Southeast during the Copper and Bronze Ages would have been based more on the appropriation of agricultural surpluses, and therefore both coercion and the ownership of land, or, in the case of southeastern Iberia, the control of water systems would have been decisive (*e. g.* Gilman 1987, 2001; Montero Ruiz 1994; Murillo-Barroso *et al.* 2015; Murillo-Barroso and Montero Ruiz 2017).

Archaeometallurgical studies at different sites of the Iberian Southeast have revealed a broadly similar technological tradition, which survived with relatively limited change from the Copper Age to the Bronze Age (*e. g.* Rovira 2002a, 2004). Copper metallurgy is characterised by the preferential use of oxidic ores (although sometimes naturally associated to sulphidic minerals) smelted in open vessels (crucibles or the so-called “vase-furnaces”) with air supplied by tuyeres or blowing pipes. Owing to poor control of the temperature and atmosphere, which oscillates from highly oxidising to reducing, this smelting technology generates immature slag with high viscosity and high copper losses. It therefore is a quite inefficient process. These questions have been highlighted in all the archaeometallurgical studies in the area of study (Delibes *et al.* 1989; 1991; Rovira 2002b; Müller *et al.* 2004, 2006; Murillo-Barroso *et al.* 2017). This technology, inter-

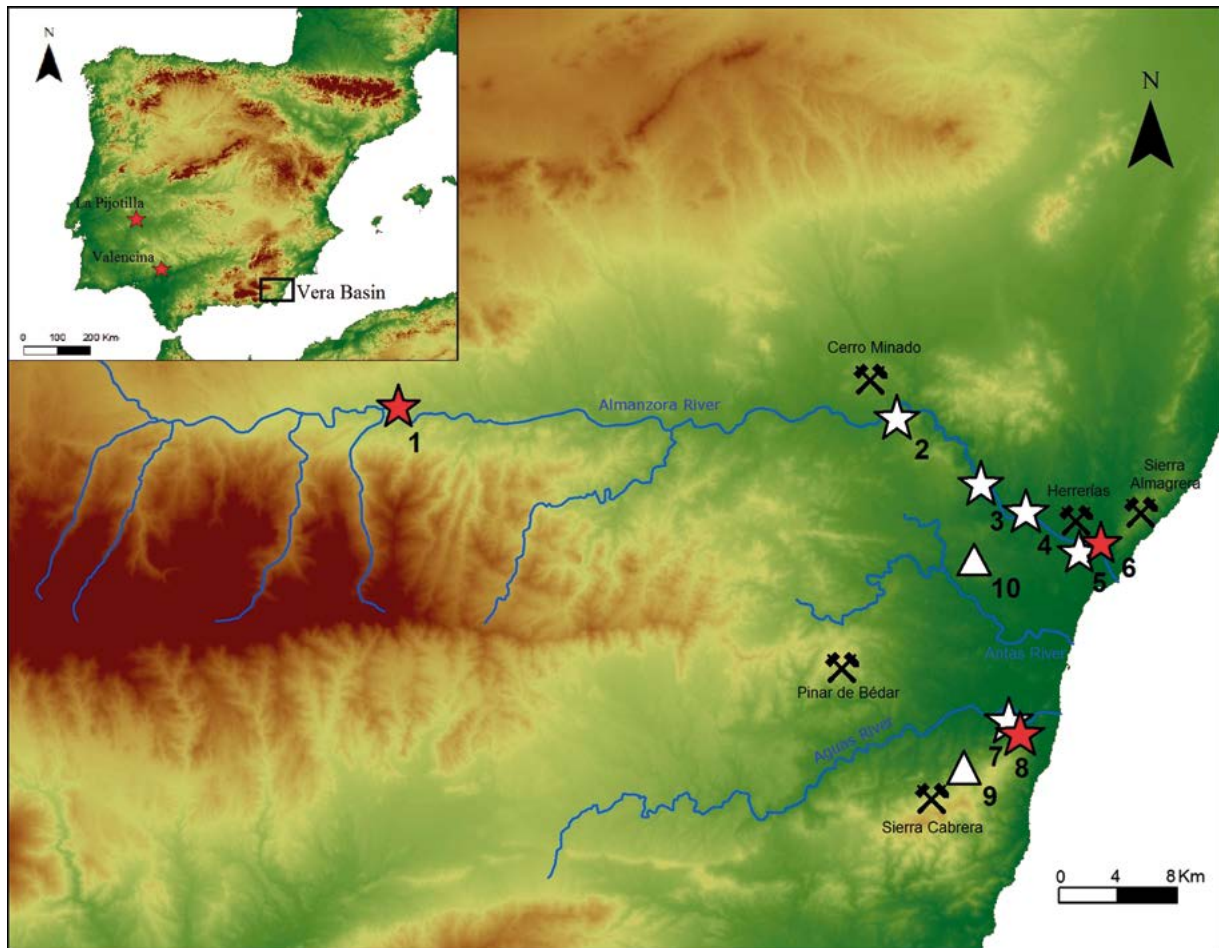


Fig. 1. Location of sites mentioned in the text. White stars: Copper Age settlements (3100-2200 cal BC); coloured stars: Copper Age tombs or necropolises (3100-2200 cal BC); Triangles: Bronze Age sites (2250-1150 cal BC). 1. Las Churuletas 2. Santa Bárbara; 3. Campos; 4. Zájara; 5. Almizaraque; 6. La Encantada; 7. Las Pilas; 8. Loma de Belmonte; 9. Gatas; 10. El Argar. Design M. Murillo-Barroso. Base map modified from CGIAR-CSI SRTM 90m 53 Digital Elevation Data (<http://srtm.csi.cgiar.org/>), with permission from SRTM (Jarvis *et al.* 2008; Reuter *et al.* 2007) (in colour in the electronic version).

preted as a low specialised and low productive one, persisted during the Argaric Bronze Age according to the archaeometallurgical analyses (Moreno *et al.* 2010; Rovira *et al.* 2015), although noting changes in object manufacturing technology (with the generalisation of annealing, a technique rarely used during the Copper Age) as well as in the spatial and regional organisation of production. This contribution aims to investigate the territorial organisation of metallurgical production in the Vera Basin during the Copper Age (3100-2200 cal BC) and to evaluate the relationship between settlements –where metallurgical activity was identified– and burial sites, where the finished objects are found. Using scientific methods (trace element and lead isotope analyses), this work focuses on the provenance identification of metallurgical finds and objects from

a broad selection of production sites and burials from the Vera Basin and surrounding areas.

## THE VERA BASIN: ARCHAEOLOGICAL AND GEOLOGICAL BACKGROUND

There are three main settlements with evidence of the full *chaîne opératoire* of metallurgical production in the Vera Basin: Las Pilas, Santa Bárbara and Almizaraque; there is also partial evidence of some stages of metallurgical works in other settlements such as Zájara and Campos. Along with these sites, there is a series of collective burial necropolises that have traditionally been associated with the populations that lived in the Vera depression and the Almanzora River basin, such

as La Encantada, Loma de Belmonte and the necropolis of the Purchena Group, among others (Fig. 1).

In this work, the metallurgical remains from the three above-mentioned settlements (Las Pilas, Santa Bárbara and Almizaraque) with production remains as well as the objects of some of the graves are investigated.

### Las Pilas (Mojácar, Almería)

The site of Las Pilas, of approximately 5 ha in extension, is located on top of a plateau 30 m above sea level, close to the estuary of the Aguas River. The site was discovered in 1989 and it was subjected to three archaeological campaigns in 1990, 1991 and 1994 (Alcaraz Hernández 1992; Camalich Massieu and Martín Socas 1999; Rovira Buendía 2007). In the last campaign, an area of 40 m<sup>2</sup> was excavated. The occupation of the site was structured in a sequence of ten phases, based on documented stages of the restructuration and re-organisation of the inhabited area. This occupation took place during the 3<sup>rd</sup> millennium BC.

In terms of constructive features, the settlement is characterised by round huts, some of them partially excavated in the ground, generally built on a stone and clay plinth, with rammed-earth walls. They usually have a central post to hold the conical roof made of vegetal lattices waterproofed with clay. Several negative structures related to grain and water storage are associated to these huts. Some large walls have been documented too, possibly related to the demarcation or defence of this sector, which clearly underwent frequent functional re-organisation of spaces where domestic and craft activities co-existed.

Ten archaeological phases have been recorded at the site, where the last excavation campaign yielded a substantial archaeometallurgical assemblage including evidence of the whole production process: stone hammers, copper ores, crucible fragments and a complete one, copper pipes, slag fragments, metal drops and a few metal objects (see Murillo-Barroso *et al.* 2017 for a technological study) (Fig. 2). Except for one copper awl and one copper mineral fragment recovered in the earliest phases (1 and 2 respectively), metal-working debris (*i.e.*, slag and a copper lump) appear from phase 5 (2905-2579 cal BC 2 $\sigma$ ) and reached the highest frequency in phase 9 (2578-2276 cal BC 2 $\sigma$ ) (Murillo-Barroso *et al.* 2017).

### Santa Bárbara (Huércal Olvera, Almería)

The site of Santa Bárbara is located on top of a plateau 221 m above sea level, next to the course of the Almanzora River. It has approximately 1.5 ha in exten-

sion, and its location offers direct access to water resources, visibility and potential control to communication routes. The site was excavated in 1990 and 1991 in a rescue operation (González Quintero *et al.* 1993; 2018). The excavations showed that the site was fenced by a wall with at least one semi-circular bastion. Other craft activities such as textile production have also been proposed in the site (González Quintero *et al.* 2018), although metallurgical production stands out due to the amount of metallurgical debris documented all over the site. The metallurgical assemblage currently under study by some of us (MMB, MDCM, DMS and MMT) includes a large amount of metallurgical debris: copper ores, complete and fragmented crucibles, slag fragments, copper droplets and few objects (Fig. 2).

### Almizaraque (Cuevas de Almanzora, Almería)

The site of Almizaraque is known since the 19<sup>th</sup> century. It was excavated by L. Siret between 1903-1906 and 1932-1933. He documented some negative storage structures (*silos*), rounded huts with some significant objects such as idols, and archaeometallurgical production remains. The site was re-excavated between 1980-1984 by Delibes and colleagues (1996) who structured its occupation in five phases and documented its maximum extension, c. 0.3 ha. Absolute dates indicate an occupation from c. 3000 to 2200 cal BC, covering the three phases in which Copper Age is usually divided (Fig. 3). Metallurgical remains are documented from the last period of the first phase up to the abandonment of the site (Delibes *et al.* 1996) and comprise evidence of the whole metallurgical process: copper ores, slag and crucible fragments, metallic copper lumps and objects (for technological studies see Delibes *et al.* 1989, 1991; Müller *et al.* 2004, 2006) (Fig. 4).

Although metallurgical activity is well documented at the three sites, it must be noted that no moulds have been found at any of them, and blowing pipe nozzles have only been documented at the site of Las Pilas. Metal objects are also scarce in these metallurgical sites (with the exception of Almizaraque, where they have been scarcely documented in modern excavations but where Siret recovered more than 70 objects). Therefore, some objects from tombs in nearby sites were included in our provenance analyses with the aim to evaluate their possible link with the production remains at these main sites.

### La Encantada necropolis (Cuevas de Almanzora, Almería)

The necropolis of La Encantada is composed of three megalithic tombs (*tholoi*) located on a mound



Fig. 2. Some archaeometallurgical remains from Las Pilas (left): a) Stone hammer, b) ore samples, c) slagged crucible and d) slag fragments, e) blowing pipe nozzles, f) a complete crucible, and Santa Bárbara (right): a) copper prills, b) ore samples, c) slagged smelting vessel fragments and d) two complete crucibles. Source M. Murillo-Barroso (in colour in the electronic version).

next to the Almizaraque settlement. They were excavated by Siret at the end of the 19<sup>th</sup> century and information is also collected by the Leisners (Leisner and Leisner 1943).

La Encantada I is a *tholos* with a circular chamber 3.5-3.6 m in diameter and a corridor 3.1 m long, divided into three sections of 0.8 m, 1.2 m and 1.1 m, respectively. About 50 individuals were documented with abundant grave goods (Leisner and Leisner 1943).

This grave has two absolute dates, one on carbon from a sample collected by Siret in 1906, unpublished, which could be associated with the oldest moments (although we are aware of the old wood problems),

and another on bone that would correspond to the Late Bronze Age and could be correlated with the later materials found in the tomb (Lorrio and Montero Ruiz 2004) (Tab. 1).

Among the metal objects preserved in the *Museo Arqueológico Nacional* (MAN, Madrid), there is a small chisel, two fragments of a thicker chisel, a punch, a fragment of an axe, a Palmela point with a fragmented tip and a second point fragmented into two pieces that the Leisners describe as a spatula (Tab. 2, Fig. 5). Typologically, the whole assemblage is consistent with 3<sup>rd</sup> millennium BC dates. Other metal objects corresponding to later reuses of these tombs such as a hoop

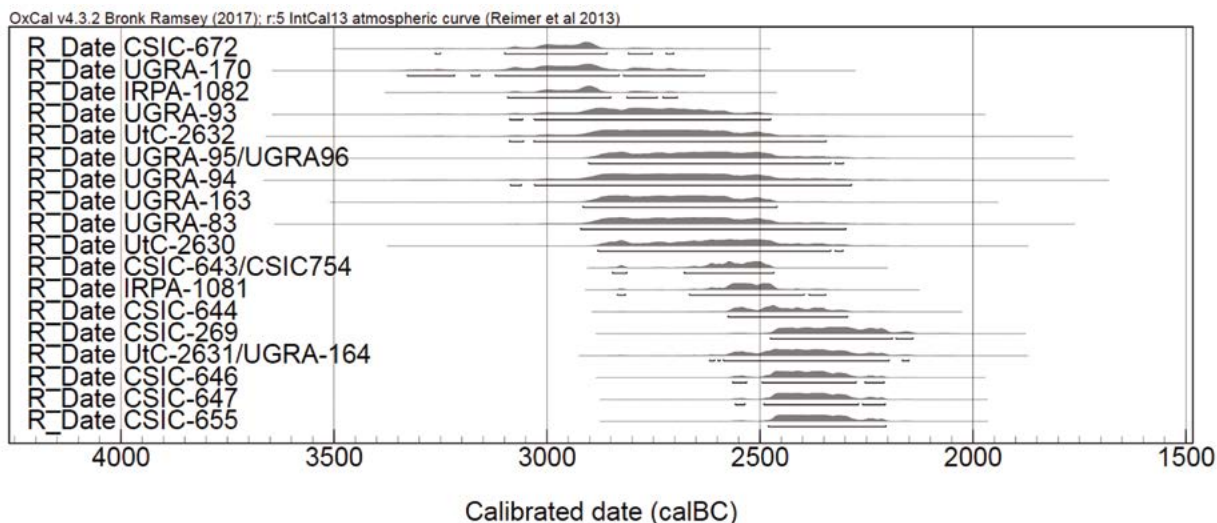


Fig. 3. Series of absolute datings from Almizaraque. Data used from *IDEArq: Infraestructura de Datos Espaciales de Investigación Arqueológica*. Madrid: CSIC.

or hook fragment, a hook, a low-lead bronze bracelet, a button and a possible fragmented nail were also recovered (Lorrio 2008). This paper presents the lead

isotope analyses of the two Palmela points from La Encantada I.

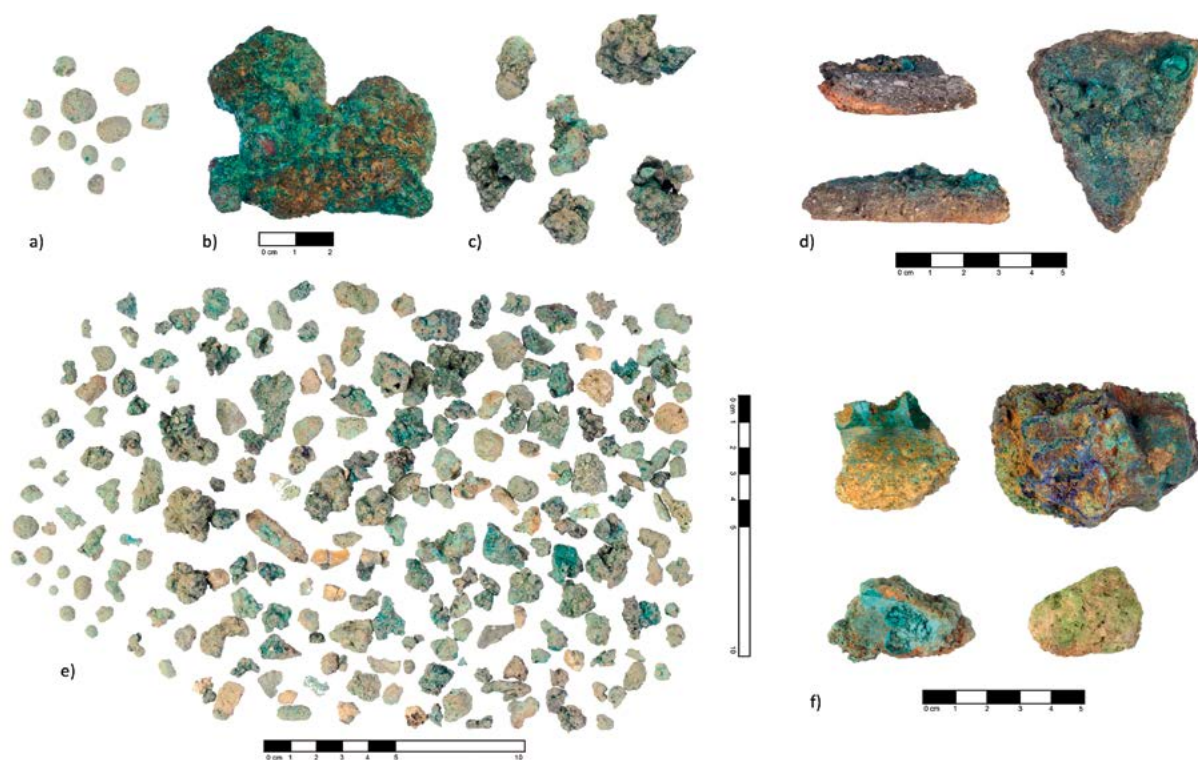


Fig. 4. Some archaeometallurgical remains from Almizaraque: a) copper prills, b) copper lump, c) slag fragments, d) slagged crucible fragments, e) slag fragments and copper prills, f) ore fragments. Source M. Murillo-Barroso (in colour in the electronic version).

ID	Date BP	Cal 2 $\sigma$ BC	Material	Reference
CSIC-655	3890 ±50	2481-2205 (95.4% prob.)	Wood/ Charcoal	Unpublished ( <i>IDEArq</i> n.d.)
CSIC-249	2830 ±60	1192-1176 (1.4% prob.) 1162-1144 (1.7% prob.) 1131-837 (92.3% prob.)	Bone	Lorrio and Montero 2004

Tab. 1. Absolute dates available for La Encantada I (*IDEArq: Infraestructura de Datos Espaciales de Investigación Arqueológica*. Madrid: CSIC. <http://www.idearqueologia.org/>). Calibrated using OxCal v4.3.2 (Bronk Ramsey 2017); r:5 IntCal13 atmospheric curve (Reimer *et al.* 2013).

### Loma de Belmonte (Mojácar, Almería)

This collective burial is close to the site of Las Pilas and was excavated by L. Siret and his foreman P. Flores (Siret 2001 [1891], 1906-07) and published by the Leisner (Leisner and Leisner 1943). It is a *tholos* with a corridor of 4 x 1.4 m, divided into two sections, which gives access to the main chamber of 5 m in diameter. In the north side of the chamber, there was a polygonal niche, approximately 1.1 x 1.2 m, formed by five slate slabs. The Leisners, according to the information gathered from L. Siret and P. Flores' field notebooks, noted that 100 individuals were documented (Leisner and Leisner 1943) with abundant grave goods, among which laminar gold beads stand out. Copper-based objects preserved in the *Museo Ar-*

Site	Object	ID	Analysis	Fe	Ni	Cu	Zn	As	Ag	Sn	Sb	Pb	Reference
La Encantada I	Axe	1984/171/1/184	AA1199	0.30	0.09	92.8	0.11	5.95	0.002	0.06	0.182	0.49	Montero Ruiz 1994
	Palmela Point	1984/171/1/183	AA1187	0.09	0.14	96.3	0.13	2.58	0.008	nd	0.059	nd	Montero Ruiz 1994
			PA26958	nd	0.13	98.1	nd	1.80	<0.15	nd	<0.15	nd	This paper
	Palmela Point	1984/171/1/182	AA1198	0.12	0.11	96.2	0.13	2.82	0.001	0.08	0.046	nd	Montero Ruiz 1994
			PA26957	nd	nd	98.3	nd	1.66	<0.15	nd	<0.15	nd	This paper
	Awl		AA1186	0.09	0.09	98.4	0.10	0.90	0.066	nd	0.061	nd	Montero Ruiz 1994
	Awl		AA1194	0.18	0.12	92.6	nd	7.02	nd	nd	0.015	nd	Montero Ruiz 1994
Awl		AA1195	0.29	0.09	92.1	nd	7.29	nd	nd	0.191	nd	Montero Ruiz 1994	
Loma de Belmonte	Sheet	86/129/LB/1/63	AA1374	0.19	0.11	95.6	0.19	3.48	0.005	nd	0.42	nd	Montero Ruiz 1994
	Palmela Point	86/129/LB/1/62	AA1377	0.05	0.11	96.2	nd	3.13	0.096	0.02	0.17	nd	Montero Ruiz 1994
			PA23904	nd	0.09	94.0	nd	5.61	<0.15	nd	<0.15	0.16	This paper
	Awl	86/129/LB/1/64	AA1375	tr	0.22	96.0	0.18	3.43	tr	nd	tr	nd	Montero Ruiz 1994
Awl	86/129/LB/1/65	AA1376	0.19	0.26	97.1	nd	1.58	tr	nd	0.02	0.48	Montero Ruiz 1994	
Las Churuletas I	Awl	1984/138/1/181	PA26953A	0.04	nd	99.8	nd	0.06	<0.15	nd	<0.15	nd	This paper

Tab. 2. Results of X-ray fluorescence analyses of copper-based objects from La Encantada I, Loma de Belmonte and Las Churuletas I hosted at the *Museo Arqueológico Nacional* (Madrid). Results expressed in wt%. Nd = Not detected. Tr. = Traces. Analyses with 'AA' conducted with a KeveX mod. 7000 spectrometer, Analyses with 'PA' with an Innov-X spectrometer (see Rovira and Montero Ruiz 2018 for further methodological detail).



Fig. 5. Metallic objects from La Encantada I, Loma de Belmonte and Las Churuletas 1. Source O. García-Vuelta (in colour in the electronic version).

queológico Nacional (Madrid) are as follows: three punches, the tip of a forth punch and a fragment of another possible mineralised punch, a fragment of a possible chisel, a folded sheet and a Palmela point (Tab. 2) (Fig. 5).

Lead isotope analyses results of metallic copper from the site of Terrera Ventura and the necropolis of El Barranquete (Stos Gale *et al.* 1999) will be also considered in this study as well as the only metallic object found in the Churuletas necropolis (Fig. 5). The awl from El Barranquete was recovered in the layer IV, i.e. the earliest one, of the chamber of tomb 9 (Almagro Gorbea 1973) which would be earlier than layer III dated between 2210-1940 cal BC  $2\sigma$  (Aranda *et al.* 2018). The awl from the Churuletas necropolis was recovered at Churuletas 1, dated between 2900-2670  $2\sigma$  cal BC (Aranda *et al.* 2017). These are the only Chalcolithic objects analysed from the Almería province and would thus be contemporary with the metallurgical sites aforementioned (Las Pilas, Santa Bárbara and Almizaraque) and potentially related to them.

### Mineralisations from the Vera Basin

The Vera Basin is renowned for its metallurgical wealth. Copper mineralisations are well-known in the area surrounding Las Pilas (Pinar de Bédar and Sierra Cabrera), Santa Bárbara (Cerro Minado) and Almizaraque (Sierra Almagrera and Herrerías) (Fig. 1). Pre-historic mining activity has only been well documented at Cerro Minado, with an absolute date of 2469-2336 cal BC  $2\sigma$  (Delgado *et al.* 2014), although L. Siret recovered a couple of mining hammers in Herrerías as

well, which could also be indicative of prehistoric mining (Montero Ruiz 2010) (Fig. 6). However, the vast intensity of Roman and especially modern mining activities, particularly in Pinar de Bédar and Herrerías, may have completely erased any prehistoric mining evidence. A geological survey was conducted in the area with the aim to better characterise these mineralisations both on their trace element patterns and their lead isotopic signatures (Murillo-Barroso *et al.* 2019). The mining districts sampled were Pinar de Bédar (Sierra de los Filabres) in the Mulhacén Complex; Lower Sierra Cabrera in the Alpujarride Complex; Herrerías

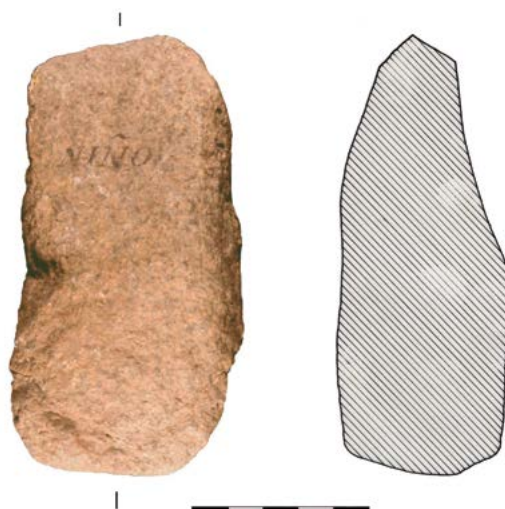


Fig. 6. Mining hammer from the Concesión Niño mine in Herrerías recovered by L. Siret. It is among materials from Herrerías at the Museo Arqueológico Nacional (Madrid). Source I. Montero Ruiz (in colour in the electronic version).

(Sierra Almagrera), also at the Alpujárride Complex but with a hydrothermal mineralisation; and Cerro Minado (Sierra de Almagro) in the Alpujárride Complex (see Murillo-Barroso *et al.* 2019 for further geological details).

The mines of Bédar are found in the Sierra de los Filabres. They were intensely worked in the 19<sup>th</sup> and 20<sup>th</sup> centuries as sources of iron and lead ores as well as for copper ores in its southeastern zone at Sierra de Alcornia from at least the 16<sup>th</sup> century (Montero Ruiz 1994; Soler Jódar 2014; Soler Jódar and Hansen 2016). No indications of prehistoric works were documented; however the later intensive mining exploitation could have eliminated such evidence. In the present, the housing development executed on this terrain between 2006 and 2008 has destroyed or covered the majority of the copper mines and altered the conditions for future investigation.

Sierra de los Filabres belongs almost entirely to the Mulhacén Complex. Ore deposits in this location are mainly associated to marbles, are stratiform, and show a mineral association of galena-chalcopyrite-malachite-azurite (Montero Ruiz 1994). In addition the presence of cerussite, zinc-olivenite, olivenite, barite and smithsonite has been documented in smaller quantities. X-ray fluorescence (XRF) (Montero Ruiz 1994) as well inductively coupled plasma mass spectrometry (ICP-MS) (Murillo-Barroso *et al.* 2019) ore analysis revealed the high levels of zinc, arsenic, lead and uranium in these copper mineralisations.

The chain of Sierra Cabrera is the continuation of the Sierra Alhamilla towards the east, and separates the Vera Basin of the Aguas River to the north from that of the Carboneras River to the south. The Alpujárride Complex emerges in a band at the foot of the northern slopes of the range, and is followed by the Mulhacén Complex towards the south. From the Triassic, there is a carbonate formation composed of black and greyish-brown rocks with very dense stratification and with a few interspersions of blue phyllite layers. It is in these phyllites layers where small outcrops of copper ores are found. Copper minerals from Sierra Cabrera display lower levels of impurities (arsenic, zinc or lead) when compared to other mineralisations at the Vera Basin (Montero Ruiz 1994; Murillo-Barroso *et al.* 2019).

Sierra Almagrera, adjacent to Herrerías, is geologically uniform, mainly composed by materials of the Paleozoic bedrocks of the Alpujárride Complex. Mineralisations from Sierra Almagrera are rich in galena, silver and copper sulphides with a host rock of graphite-rich phyllite and quartzite. Chalcopyrite has been documented in the mine Ramo de Flores and native copper, alongside with azurite and malachite in the Jaroso Ravine. Arsenic is not detected in copper ores

analysed from Sierra Almagrera although lead is usually associated to copper minerals (*e. g.* La Estrella Mine<sup>1</sup>, Montero Ruiz 1994).

Herrerías, adjacent to the seams of sulphites and sulphosalts of Pb–Sb–Ag in the Sierra Almagrera, constitutes a very well defined mineral zone inside the Vera-Garrucha basin (Martínez Frías 1991; López Gutiérrez *et al.* 1993) where copper ores have also been identified. In Herrerías a deposit of oxides and hydroxides of Fe–Mn, sulphides of base metals, and native silver can be found. Both Herrerías and Sierra Almagrera, dike type deposits, are considered epigenetic, with a similar temperature of ore deposition (200–300 °C) and very similar <sup>34</sup>S isotope values in the barites which accompany the mineralisations in both deposits. Due to this, a similar hydrothermal genesis for both, probably in a submarine environment, has been proposed (Martínez Frías 1992). This similitude in the genesis of the two deposits is also reflected in similarity and proximity in their lead isotopic fields (Murillo-Barroso *et al.* 2019). The intensive mining of Herrerías makes the identification of copper samples difficult and would have erased any evidence of prehistoric mining works.

The last copper mineralisation in the Vera Basin is Cerro Minado in the Sierra de Almagro, mostly occupied by the Alpujárride Complex. Two major tectonic units have been distinguished, a lower and an upper. The most attractive copper and cobalt ores are found in the upper unit, composed basically of schists and quartzites, phyllites and dolomites (IGME 1980). The primary deposits of copper, cobalt, silver and mercury are found filling a dense network of fissures which is a consequence of, amongst other factors, the brecciation of the dolomites, what has allowed the primary paragenesis to suffer an intense oxidizing and carbonating alteration. More than 100 types of minerals have been identified at Cerro Minado (Favreau *et al.* 2013) including native copper, malachite, cuprite, olivenite, etc. Elemental composition analysis revealed the high levels of arsenic, cobalt and nickel, due to the coexistence of erythrite and annabergite with copper minerals and arsenates (Murillo-Barroso *et al.* 2019).

Recent studies have documented stone peaks and hammers from Cerro Minado, most of them on the surface and two associated to a gallery (Delgado *et al.* 2014). Absolute dates confirm its exploitation during the Copper Age, contemporaneous with the occupation of several sites in the area such as Almizaraque, Las

<sup>1</sup> A. Obón Zúñiga. *Los inicios de la metalurgia del cobre en el Suroeste europeo. Aproximación experimental a la metalurgia de Almizaraque (Almería)*. Unpublished Ph. D. Thesis, Universidad de Zaragoza, 2017. Thesis supervised by Ignacio Montero Ruiz and José María Rodanés Vicente.

Pilas or Santa Bárbara. Its exploitation has been also proposed in the neighbouring region of the Guadalentin Valley, Murcia (Escanilla 2016) but based only on elemental analyses and lacking isotopic confirmation.

The lead isotopic fields of these mining districts were defined and three different isotopic fields were identified: Cerro Minado, Herrerías/Sierra Almagrera and Pinar de Bédar/Sierra Cabrera. Probably due to a similar genesis, Herrerías and Sierra Almagrera cannot be isotopically discriminated, and the same applies to Pinar de Bédar and Sierra Cabrera. However, discrimination is possible on the basis of their different trace element patterns (for detailed discussion see Murillo-Barroso *et al.* 2019).

## MATERIALS AND METHODS

More than 1000 archaeometallurgical finds were recovered during the archaeological excavations of the three metallurgical sites (Las Pilas, Santa Bárbara and Almizaraque). The collections include ore fragments (maximum fragment weight of 260 g), slag fragments, slagged crucible sherds and complete crucibles, stone hammers and grinding stones, copper droplets or lumps, and only in the case of Las Pilas, fragments of ceramic blowpipe nozzles (MNI five) as well as few metallic objects (Figs. 2, 3 and 5).

Selected materials were sampled for further analyses at the UCL Institute of Archaeology's Wolfson Archaeological Science Laboratories. We employed a stratigraphic sampling frame that considered archaeological and typological information, as well as the results of 591 screening analyses by portable x-ray fluorescence (pXRF) using an Olympus Innov-X Systems Delta Premium. Samples were selected from across the compositional groups qualitatively identified by pXRF. The sample set amounts to a total of 48 samples, including 21 samples of copper ores, 17 slag fragments and 10 copper objects or lumps from the three metallurgical sites, which were selected for lead isotope analyses, seeking to cover the whole production process in each site.

The sample is completed with four copper objects from the nearby necropolis of La Encantada I, Loma de Belmonte and Las Churuletas 1. Data from the literature from Almizaraque, Terrera Ventura and El Barranquete are also used for comparative purposes. In addition, trace element analysis was conducted for 18 archaeological ore samples from Almizaraque, Las Pilas and Santa Bárbara in order to compare them with the trace element patterns of the geological samples in the area (Tab. 3).

Trace element analyses of ore samples were conducted by inductively coupled plasma mass spectrom-

etry (ICP-MS) at the Sgiker-Geochronology and Isotope Geochemistry Facility of the University of the Basque Country UPV/EHU (Spain).

For each sample, 80 mg were digested in concentrated HNO<sub>3</sub> from Merck Pro-Analysis in closed Savillex PFA vessels set on a hot plate at 120°C during 24 hours. Digestion was conducted two consecutive times due to the low dissolution level in some samples. This is probably due to the high silicate portions of the mineral gangue, although our interest is on the soluble portions of metalliferous minerals. Subsequently, the solution obtained was brought to dryness and taken up in diluted HNO<sub>3</sub>. The solution obtained was centrifuged and small amounts of undissolved residues were found in most samples. Only the fraction soluble in nitric acid, i.e., the dissolved fraction, was analysed. Dilution after dissolution was done gravimetrically using an electronic balance with precision of 0.1 mg in order to prevent errors induced by volumetric dilutions. All reagents used in sample preparation were distilled in the laboratory by sub-boiling distillation using an Acidest quartz distiller in order to avoid contamination. Deionised water was obtained using a Millipore Elix device and polished to obtain a resistivity  $\geq 18$  M Ohm-cm with a Barnstead EasyPure system.

Quantitative determination of analytes of interest was done by means of a Thermo XSeries 2 inductively coupled plasma mass spectrometer (ICP-MS) equipped with collision cell (CCT), an interphase specific for elevated total dissolved solids (Xt cones) and shielded torch. A concentric nebuliser and quartz expansion Peltier-cooled chamber were employed. Rh solution, used as internal standard, and multielemental solutions for the initial tuning and calibration of the mass spectrometer, and for quality control (QC) of the results were prepared from 1000 ppm high purity Merck multi-element standard solutions for ICP, stabilised in diluted HNO<sub>3</sub> 2 to 6%.

The internal standard was added by means of an automatic online addition kit in order to prevent random errors (further details on the instrumental method are given in García de Madinabeitia *et al.* 2008). Error estimation for each element is established using the error propagation equation of Miller and Miller 2004. Uncertainty of the results corresponds to a 95% confidence level (see analytical errors 2SE in Supplementary File, SF.1).

Most of the lead isotope analyses were conducted via multi-collector inductively-coupled plasma - mass spectrometry (MC-ICP-MS) at the Geochronology and Geochemistry Service (SGIker) at the University of the Basque Country (Spain).

Aliquots of 0.05 g of the samples were prepared, which were dissolved with 0.5 mL of 7N HNO<sub>3</sub> overnight in a 70°C heating plate. After digestion the liquid has evaporated at 70°C. The residues were redis-

ID	Site	Chronology	Object	TE	LIA	pXRF	Reference
LP 6491	LP	Phase 9	Copper ore	X	X		This paper
LP 6740	LP	Phase 9	Copper ore	X	X		This paper
LP 7706	LP	Phase 9	Copper ore	X	X		This paper
LP 6972	LP	Phase 9	Copper ore	X	X		This paper
LP 6816	LP	Phase 9	Slag fragment		X		This paper
LP 6918	LP	Phase 9	Slag fragment		X		This paper
LP 6974	LP	Phase 9	Slag layer		X		This paper
LP 6735	LP	Phase 9	Slag layer		X		This paper
LP 6608	LP	Phase 9	Slag layer		X		This paper
LP 2997	LP	Phase 9	CuAs awl		X		This paper
LP 6726	LP	Phase 8	Copper ore	X	X		This paper
LP 8479	LP	Phase 7	Copper ore	X	X		This paper
LP 8815	LP	Phase 5	Slag fragment		X		This paper
LP 8862	LP	Phase 5	CuAs lump		X		This paper
LP 11454	LP	Phase 1	CuAs awl		X		This paper
HOSB_21/288	HOSB	Phase 1	Copper ore	X	X		This paper
HOSB_11/48	HOSB	Phase 1	Copper ore	X	X		This paper
HOSB_4/39	HOSB	Phase 1	Copper ore	X	X		This paper
HOSB_8/30	HOSB	Phase 1	Copper ore	X	X		This paper
HOSB_1/197	HOSB	Phase 1	Copper ore	X	X		This paper
HOSB_4/64	HOSB	Phase 1	Copper ore	X	X		This paper
HOSB_21/78	HOSB	Phase 1	Slag fragment		X		This paper
HOSB_21/283	HOSB	Phase 1	Slag fragment		X		This paper
HOSB_1/441	HOSB	Phase 1	Slag fragment		X		This paper
HOSB_1/168	HOSB	Phase 1	Slag fragment		X		This paper
HOSB_17/173	HOSB	Phase 1	Slag fragment		X		This paper
HOSB_16/112	HOSB	Phase 1	Slag fragment		X		This paper
HOSB_23/32	HOSB	Phase 1	Copper lump		X		This paper
HOSB_25/4	HOSB	Phase 1	Copper lump		X		This paper
HOSB_25/5	HOSB	Phase 1	Copper lump		X		This paper
AA 1099B	ALMZ	Phase 5	Copper ore		X		This paper
AA 1100C	ALMZ	Phase 5	Copper ore		X		This paper
AA1106D	ALMZ	Phase 3	Slag fragment		X		Montero Ruiz and Murillo-Barroso 2010
ALMZ_1443D	ALMZ	Phase 2	Copper ore	X	X		This paper
ALMZ_1096	ALMZ	Phase 2	Copper ore	X	X		This paper
ALMZ_1443 I	ALMZ	Phase 2	Copper ore	X	X		This paper
ALMZ_PA1443F	ALMZ	Phase 2	Slag fragment		X		This paper

Tab. 3. [1] Materials analysed and used in this paper. TE = trace elements, LIA = Lead isotope analyses, pXRF = portable X-ray fluorescence, LP = Las Pilas, HOSB = Santa Bárbara, ALMZ = Almizaraque, CHUR1 = Las Churuletas 1, LE = La Encantada I, LB = Loma de Belmonte, TV = Terrera Ventura, EB9 = El Barranquete 9.

ID	Site	Chronology	Object	TE	LIA	pXRF	Reference
AA1080B	ALMZ	Phase 2	Slag fragment		X		Montero Ruiz and Murillo-Barroso 2010
PA2619	ALMZ	Phase 2	Copper lump		X		Montero Ruiz and Murillo-Barroso 2010
PA3060	ALMZ	Phase 2	Chisel		X		This paper
ALMZ_AA1104E	ALMZ	Phase 1	Slag fragment		X		This paper
ALMZ_AA1094	ALMZ	Phase 1	Slag fragment		X		This paper
ALMZ_1076A	ALMZ	Pre-Beaker dump	Copper ore	X	X		This paper
ALMZ_AA1084	ALMZ	Pre-Beaker dump	Copper lump		X		This paper
ALMZ_M6	ALMZ	Uncertain Phase	Copper ore	X	X		This paper
ALMZ_M16	ALMZ	Uncertain Phase	Copper ore	X	X		This paper
ALMZ_M32	ALMZ	Uncertain Phase	Copper lump		X		This paper
PA0320	ALMZ	Siret Collection	Copper ore		X		Montero Ruiz and Murillo-Barroso 2010
1984-138-1-81	CHUR1	2900-2670 $2\sigma$ cal BC	Copper awl		X	X	This paper
1984-171-1-183	LE	Copper Age	Palmela point		X	X	This paper
1984-171-1-182	LE	Copper Age	Palmela point		X	X	This paper
1986-129-LB-I-62	LB	Copper Age	Palmela point		X	X	This paper
AM-09	TV	Copper Age	Copper lump		X		Stos Gale <i>et al.</i> 1999
A-52557 (BA1)	EB9	Chamber, level IV	Copper awl		X		Oxalid

Tab. 3. [2] Materials analysed and used in this paper. TE = trace elements, LIA = Lead isotope analyses, pXRF = portable X-ray fluorescence, LP = Las Pilas, HOSB = Santa Bárbara, ALMZ = Almirazaque, CHUR1 = Las Churuletas 1, LE = La Encantada I, LB = Loma de Belmonte, TV = Terrera Ventura, EB9 = El Barranquete 9.

solved in 0.5 N HBr and processed by conventional liquid chromatography with AG1-X8 ion exchange resin (Manhès *et al.* 1984), obtaining final solutions of 2 mL of 6N HCl with the purified Pb. These solutions were taken to dryness.

The lead samples were dissolved in 1.5 mL of 0.32N HNO<sub>3</sub> and if necessary they were diluted to a final concentration of 150-200 ng Pb/g solution. The samples were introduced as wet aerosols in a Neptune (Thermo Fisher Scientific) MC-ICP-MS using a PFA micronebuliser with nominal suction of 100  $\mu$ L/min (Elemental Scientific) and double pass dual cyclone-Scott expansion chamber. Actual baselines (electronics + chemistry) have been subtracted from the measurements of a chemical blank for 60 s prior to each sample. The spectrometric measurements were carried out in 105 cycles with an integration time of 8 s per cycle.

A Thallium reference material NBS997 with a normalised ratio of  $^{205}\text{Tl}/^{203}\text{Tl} = 2.3889$  was used for the internal mass correction (see Chernyshev *et al.* 2007 and references therein for further methodological detail). The reliability and reproducibility of the method were

verified by regular measurements of the certified reference material NBS981 interspersed between the measurements of the samples, and in the same conditions. The average uncertainties (2SE) for the NBS981 during this study are 0.0004 for the  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio, 0.0005 for the  $^{207}\text{Pb}/^{204}\text{Pb}$ , 0.0014 for the  $^{208}\text{Pb}/^{204}\text{Pb}$ , 0.0001 for the  $^{208}\text{Pb}/^{206}\text{Pb}$  and 0.00003 for the  $^{207}\text{Pb}/^{206}\text{Pb}$  (NBS981 values used are those proposed by Baker *et al.* 2004). Uncertainties of measurements are smaller than symbols used in all graphs.

The lead isotope analyses of the three Palmela points from the necropolis of La Encantada and Loma de Belmonte was conducted at the Department of Earth Sciences (University of Geneva, Switzerland) in collaboration with Massimo Chiaradia. After dissolution in a mixture of 1 mL 7M HCl and 1 mL 14M HNO<sub>3</sub>, samples were dried, conditioned in a few drops of 4M HBr and Pb was separated from the matrix using micro-columns loaded with AG-MP1-M resin in a hydrobromic medium. The purified Pb was redissolved in a 2% HNO<sub>3</sub> solution spiked with a Tl standard. Isotope ratios were measured using a Thermo Neptune Plus MC-ICP-MS

in static mode. Instrumental fractionation was corrected in-run by using  $^{203}\text{Tl}/^{205}\text{Tl} = 0.418922$  and the  $^{204}\text{Hg}$  interference on  $^{204}\text{Pb}$  was corrected by monitoring  $^{202}\text{Hg}$ . The NBS981 standard was used to check the long-term external reproducibility of measurements, which is 0.0048% for  $^{206}\text{Pb}/^{204}\text{Pb}$ , 0.0049% for  $^{207}\text{Pb}/^{204}\text{Pb}$  and 0.0062% for  $^{208}\text{Pb}/^{204}\text{Pb}$ . The in-run corrected Pb isotope ratios were further corrected for external fractionation (due to a systematic difference between measured and accepted ratios of NBS981) by a value of +0.5‰ amu using the NBS981 values proposed by Baker *et al.* 2004. Procedural blanks are <200 pg and are insignificant compared to the amount of sample Pb.

## RESULTS

### Trace element composition

The ore samples recovered in archaeological contexts display similar compositional characteristics. In all cases the samples are of polymetallic copper ores, most of them with elevated levels of arsenic (Tab. 4). Nonetheless some significant differences are observed. In order to better visualise these differences, the concentration of the most significant trace elements in the copper ores was normalised to the recommended composition of the upper continental crust in (Rudnick and Gao 2014). This normalisation aims to highlight the deviations in the chemical composition of copper ores from mean continental crust values. Normalised element patterns of ores from Las Pilas, Almizaraque and Santa Bárbara are plotted in Fig. 7, and compared with element patterns of copper mineralisations of the Vera Basin.

At the three sites, the ore samples show elevated levels of cobalt and nickel, especially in the samples from Santa Bárbara (with median values of 1136 ppm Co and 1391 ppm Ni, as opposed to 63 ppm Co and 299 ppm Ni at Las Pilas and 537 ppm Co and 170 ppm Ni at Almizaraque), which is consistent with the mineralisations of the region, especially for Cerro Minado, with high Co and Ni values and where erythrite and annabergite have been identified in copper ore samples (Murillo-Barroso *et al.* 2019).

The levels of zinc and lead are relatively high in the ores from Las Pilas (median values of 2.1% Zn and 0.7% Pb), as opposed to lower levels in the samples from Almizaraque (0.3% Zn and 110 ppm Pb) and especially from Santa Bárbara (0.1% Zn and 51 ppm Pb). Those high levels of Zn and Pb are consistent with Pinar de Bédar copper mineralisations. In contrast, the levels of arsenic and silver are very similar in the samples from the three sites: median values of 4.2% As and 40 ppm Ag at Las Pilas, 3.5% As and

105 ppm Ag at Almizaraque and 3.4% As and 119 ppm Ag at Santa Bárbara. One should also highlight the elevated levels of uranium in the samples from Las Pilas (median 95 ppm U), significantly higher than those from Santa Bárbara (5 ppm U) or Almizaraque (2 ppm U), although this high U content has been also documented in the copper ores of Pinar de Bédar (Murillo-Barroso *et al.* 2019).

A linear discriminant analysis (LDA) was conducted over the geological sample set to optimise separation between known groups, with a 96.5% of the variance explained (Fig. 8). Comparison of the trace element patterns of the archaeological ore to the geological samples shows a resemblance between the archaeological ores from Las Pilas and the geological deposits at Pinar de Bédar (particularly the levels of zinc, lead and uranium), which is consistent with the closer proximity between the Las Pilas site and these mines (Figs. 7 and 8).

The cluster formed by the majority of the samples from Almizaraque and Santa Bárbara most closely resembles the trace element pattern of Cerro Minado, although it is not possible, with the data available, to conclusively distinguish the deposits of Herrerías and Cerro Minado on the basis of their trace element compositions. The most useful element to discriminate between these two deposits appears to be lead: while the average lead level is 34 ppm at Cerro Minado, the content of this element is significantly higher at Herrerías, with an average of 530 ppm (see Murillo-Barroso *et al.* 2019). Previous analyses would seem to corroborate this trend: specifically, those conducted by Siret on ores from Herrerías (*cf.* Montero Ruiz 1994) show an average of 15% Pb, while in those from Cerro Minado analysed using XRF by I. Montero no lead was detected (with a detection limit of 0.1%, Montero Ruiz 1994). As such, it could be considered that lead is a differentiating element between the deposits of Herrerías and Cerro Minado.

In this regard, one may highlight that, in our set of samples from Almizaraque and Santa Bárbara, the samples from Santa Bárbara show low median lead values (50.9 ppm) while the samples from Almizaraque show higher median values (110 ppm).

The deposits of Sierra Cabrera are well differentiated on the basis of their pattern of trace elements, with none of the archaeological samples falling in this cluster.

These differences in composition and in trace element patterns appear to suggest that metallurgists from Las Pilas were using primarily ores from Pinar de Bédar, even though copper deposits are available much closer, at Sierra Cabrera. At Almizaraque the possibility remains open, given the differences in lead levels observed, that exploitation of the resources at both

Site	ID	Na	Mg	Al	P	K	Ca	Fe	Mn	Co	Ni	Cu	Zn	As	Sr	Ag	Sb	Ba	Pb	Bi	Th	U
		ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	%	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
Las Pilas	LP6726	60.6	7644	123	355	344	21950	4708	20.2	32.7	182	5.82	56070	41220	25890	5.45	27.8	542	29100	25.7	<0.057	86.1
	LP6491	65.3	4681	494	807	375	2634	13070	17.9	62.2	391	25.5	60520	70610	319	46.6	151	58.7	38290	90.4	0.190	104
	LP6740	65.8	3640	5001	333	2257	1443	7157	18.8	19.2	15.7	22.9	449	386	99.1	0.290	1.80	14.8	342	0.850	0.940	20.2
	LP7706	54.4	46780	570	568	377	11860	2965	44.1	63.3	207	13.1	37820	43830	463	33.9	21.2	1426	15040	699	0.370	437
	LP6972	153	1984	1319	3461	669	5541	9409	183	4546	9731	23.8	4599	68290	498	310	60.2	117	539	783	0.390	127
	LP8479	39.4	1182	593	636	248	3506	77310	109	408	760	28.3	913	4998	107	273	441	7.50	210	121	0.150	10.0
	<i>Average</i>	<b>73.2</b>	<b>10985</b>	<b>1350</b>	<b>103</b>	<b>712</b>	<b>7822</b>	<b>19103</b>	<b>65.7</b>	<b>855.3</b>	<b>1881</b>	<b>19.9</b>	<b>26728</b>	<b>38222</b>	<b>4562</b>	<b>111</b>	<b>117</b>	<b>361</b>	<b>13920</b>	<b>286</b>	<b>0.340</b>	<b>131</b>
	<i>StD</i>	<b>40.6</b>	<b>17681</b>	<b>1830</b>	<b>1206</b>	<b>770</b>	<b>7840</b>	<b>28735</b>	<b>67.6</b>	<b>1813.9</b>	<b>3854</b>	<b>8.61</b>	<b>28187</b>	<b>30096</b>	<b>10449</b>	<b>141</b>	<b>167</b>	<b>559</b>	<b>16594</b>	<b>355</b>	<b>0.328</b>	<b>157</b>
	<i>Median</i>	<b>62.9</b>	<b>4160</b>	<b>581</b>	<b>601</b>	<b>375</b>	<b>4523</b>	<b>8283</b>	<b>32.1</b>	<b>62.7</b>	<b>299</b>	<b>23.4</b>	<b>21209</b>	<b>42525</b>	<b>391</b>	<b>40.3</b>	<b>44</b>	<b>87.9</b>	<b>7789</b>	<b>106</b>	<b>0.280</b>	<b>95.0</b>
Almizaraque	ALMZ_1443D	249	253	110	60.3	238	65.1	5926	18.4	99.4	128	61.9	3598	4157	10.7	23.3	192	20.1	13.8	147	<0.057	1.50
	ALMZ_M_6	267	1075	147	593	183	281	4606	17.9	287	212	67.3	2653	34850	27.7	494	176	12.4	115	126	<0.057	4.26
	ALMZ_1076A	775	7596	377	496	1035	1288	29030	385	786	643	62.0	16150	44210	54.5	31.2	659	10.2	73.1	60.6	0.456	3.91
	ALMZ_1096	402	76.8	16.4	51.5	1016	291	4001	0.958	26.6	47.3	6.663	381	9421	30.5	17.7	121	1.20	6.74	46.1	<0.057	0.103
	ALMZ_1443I	535	758	208	181	518	75.2	22860	107	933	435	46.1	7975	82060	19.5	292	869	14.7	109	215	0.507	2.74
	ALMZ_M_16	1631	1144	96.1	378	2670	520	16130	26.5	5.62	3.01	10.3	122	35410	200	179	498	51.5	111	244	<0.057	2.0
	<i>Average</i>	<b>643</b>	<b>1817</b>	<b>159</b>	<b>293</b>	<b>943</b>	<b>420</b>	<b>13758</b>	<b>92.7</b>	<b>356</b>	<b>245</b>	<b>42.4</b>	<b>5146</b>	<b>35018</b>	<b>57.2</b>	<b>173</b>	<b>419</b>	<b>18.3</b>	<b>71.6</b>	<b>140</b>	<b>0.161</b>	<b>2.4</b>
	<i>StD</i>	<b>521</b>	<b>2863</b>	<b>124</b>	<b>230</b>	<b>922</b>	<b>456</b>	<b>10601</b>	<b>148</b>	<b>404</b>	<b>247</b>	<b>27.2</b>	<b>6093</b>	<b>27947</b>	<b>71.7</b>	<b>192</b>	<b>305</b>	<b>17.4</b>	<b>49.9</b>	<b>79.8</b>	<b>0.249</b>	<b>1.5</b>
	<i>Median</i>	<b>468</b>	<b>917</b>	<b>129</b>	<b>280</b>	<b>767</b>	<b>286</b>	<b>11028</b>	<b>22.5</b>	<b>537</b>	<b>170</b>	<b>61.9</b>	<b>3125</b>	<b>35130</b>	<b>29.1</b>	<b>105</b>	<b>345</b>	<b>13.5</b>	<b>110</b>	<b>136</b>	<b>-</b>	<b>2.35</b>
Santa Bárbara	HOSB_21/288	46.6	991	293	197	76.7	166	18130	97.5	286	390	35.1	2648	58610	28.0	0.363	645	22.3	17.6	102	0.161	2.29
	HOSB_11/48	211	1612	294	430	64.5	307	75170	133	1849	1911	27.5	308	3954	46.0	45.2	70.6	49.3	93.9	6.10	0.119	7.08
	HOSB_4/39	106	890	4042	332	1815	191	6097	8.069	76.9	870	22.8	429	800	27.4	0.734	7.90	26.5	5.02	13.5	3.84	5.81
	HOSB_8/30	472	11490	677	210	169	8847	4112	492	2254	2913	42.2	2313	14710	79.9	588	20.2	33.5	55.9	1027	0.594	3.52
	HOSB_1/197	147	31700	283	92.7	1315	5097	4261	631	3328	2003	45.5	14900	59130	60.3	308	112	38.5	26.6	204	0.228	4.03
	HOSB_4/64	240	866	263	135	116	499	33560	28.6	424	569	17.4	1025	54000	70.1	193	1497	235	45.9	1320	<0.057	10.8
	<i>Average</i>	<b>204</b>	<b>7924</b>	<b>975</b>	<b>233</b>	<b>593</b>	<b>2518</b>	<b>23555</b>	<b>232</b>	<b>1369</b>	<b>1443</b>	<b>31.7</b>	<b>3603</b>	<b>31867</b>	<b>51.9</b>	<b>189</b>	<b>392</b>	<b>67.6</b>	<b>40.8</b>	<b>445</b>	<b>0.824</b>	<b>5.60</b>
	<i>StD</i>	<b>149</b>	<b>12371</b>	<b>1510</b>	<b>126</b>	<b>770</b>	<b>3650</b>	<b>27744</b>	<b>263</b>	<b>1310</b>	<b>989</b>	<b>11.1</b>	<b>5617</b>	<b>28238</b>	<b>21.9</b>	<b>230</b>	<b>592</b>	<b>82.7</b>	<b>31.9</b>	<b>576</b>	<b>1.49</b>	<b>3.10</b>
	<i>Median</i>	<b>179</b>	<b>1301</b>	<b>294</b>	<b>204</b>	<b>143</b>	<b>403</b>	<b>12113</b>	<b>313</b>	<b>1136</b>	<b>1391</b>	<b>31.3</b>	<b>1669</b>	<b>34355</b>	<b>53.2</b>	<b>119</b>	<b>91.4</b>	<b>36.0</b>	<b>50.9</b>	<b>615.3</b>	<b>0.194</b>	<b>4.90</b>

Tab. 4. Elemental compositions of ore samples from archaeological contexts by inductively-coupled plasma - mass spectrometry (ICP-MS). See Supplementary File SF.1 for the complete table of results. Elements below detection limit were taken as 0 for average and median calculations.

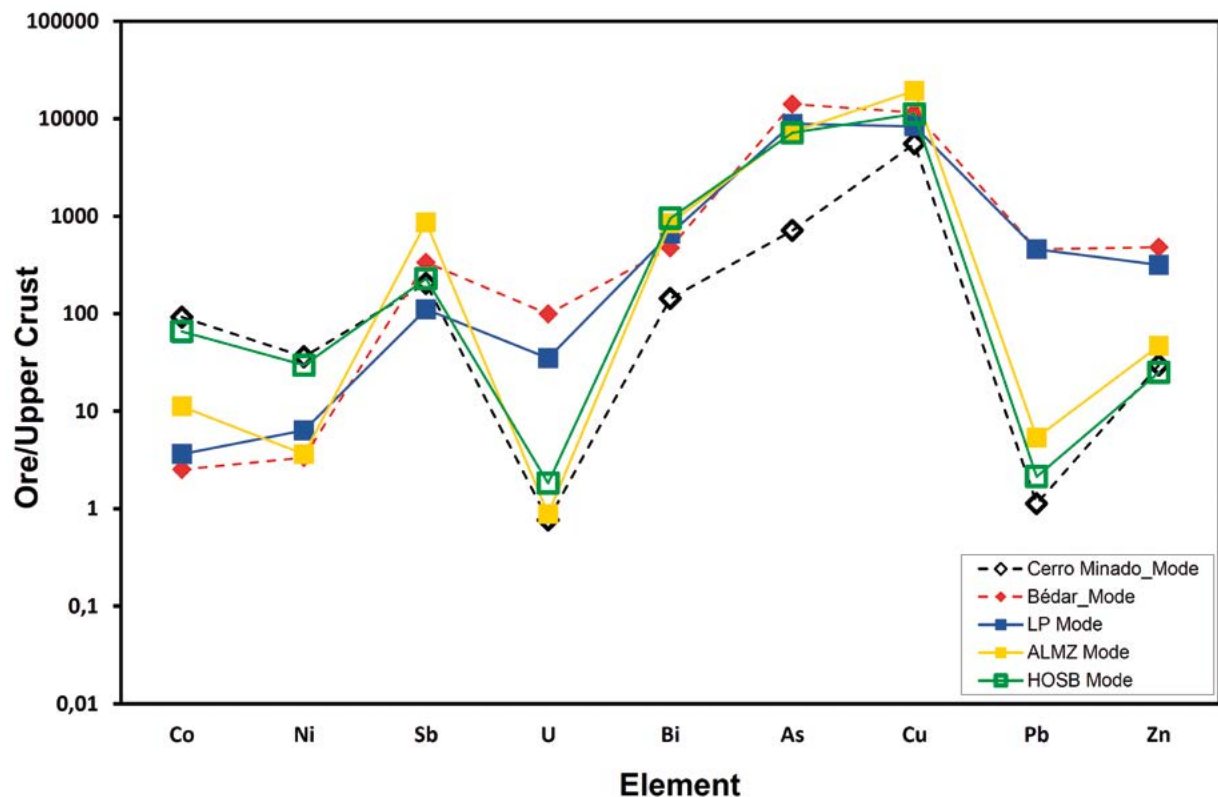


Fig. 7. Normalised multi-element diagram showing patterns in ores from Las Pílas (LP), Almizaraque (ALMZ) and Santa Bárbara (HOSB) in comparison to the patterns shown by the copper mineralisations of the Vera Basin (Cerro Minado and Pinar de Bédar). Element concentrations normalised to the recommended composition of the upper continental crust in Rudnick and Gao 2014. Geological data from Murillo-Barroso *et al.* 2019 (in colour in the electronic version).

Herrerías and Cerro Minado was taking place. Finally, at Santa Bárbara, it appears that use is being made exclusively of resources from Cerro Minado, which lies barely 1 km from the site.

Beyond these initial observations, lead isotope analyses allow more robust provenance inferences.

### Lead isotope analysis

Generally, lead isotope analyses of archaeometallurgical remains of these archaeological sites do not show very high radiogenic values, with  $^{206}\text{Pb}/^{204}\text{Pb}$  ranging from 18.44 to 18.95 (Tab. 5). However, relevant observations can be presented. Regarding the production debris (ore and slag fragments), a tight cluster can be identified at the site of Las Pílas, where most samples plot c. 18.8 on the  $^{206}\text{Pb}/^{204}\text{Pb}$  axis. All these samples are from the same phase (Phase 9 dated from 2578 to 2276 cal BC  $2\sigma$ , Murillo-Barroso *et al.* 2017) and thus could correspond to a single event. The situation is different for the other sites: production debris from Almiza-

raque plot mostly between 18.55-18.75 on the  $^{206}\text{Pb}/^{204}\text{Pb}$  axis, and those from Santa Bárbara mostly between 18.6-18.8 in  $^{206}\text{Pb}/^{204}\text{Pb}$  (Fig. 9).

Conversely, the three metal objects from Las Pílas do not match the tight cluster of ores and slag fragments, plotting at 18.56, 18.66 and 18.70 on the  $^{206}\text{Pb}/^{204}\text{Pb}$  axis, respectively. This could indicate that these three objects were produced during a different metallurgical operation or maybe elsewhere, although two of them plot close to one slag fragment (18.55) and one ore sample (18.65) from the same site. Interestingly none of the metal objects analysed matches the production debris cluster from Las Pílas; notwithstanding the small sample size, this could indicate that the metal related to this production event was not consumed at any of these three sites of the area. Conversely, two of the four objects from Almizaraque and the three objects from Santa Bárbara can be related to the production debris of each of these sites (although two of the objects from Santa Bárbara being in the limit) indicating an on-site consumption of the metal produced (Fig. 9).

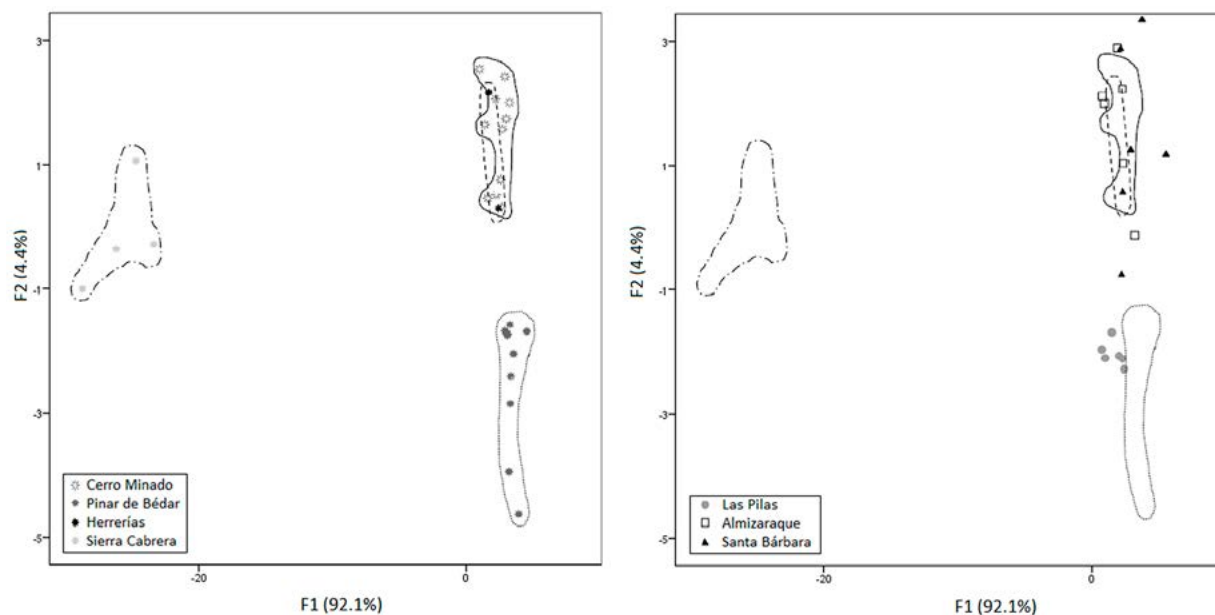


Fig. 8. Linear Discriminant Analysis. Left: LDA of ore samples from Cerro Minado, Herrerías, Sierra Cabrera and Pinar de Bédar. Data from Murillo-Barroso *et al.* 2019. Right: LDA of ore samples from the archaeological sites of Las Pilas, Santa Bárbara and Almizaraque projected on top of ore samples from Cerro Minado, Herrerías, Sierra Cabrera and Pinar de Bédar.

Isotopic fields for the copper mineralisations of the internal zone of the Betic Belt close to the sites have been defined elsewhere (Murillo-Barroso *et al.* 2019). Using thermal ionization mass spectrometry (TIMS) data from the literature and MC-ICP-MS results from our own sample set, three isotopic fields were distinguished: Cerro Minado, Pinar de Bédar/Sierra Cabrera and Herrerías/Almagrera. When plotting archaeological samples against these isotopic fields, several observations can be made: the majority of the metal-production waste from Las Pilas, whose values group into a cluster around 18.8 ( $^{206}\text{Pb}/^{204}\text{Pb}$ ), coincide in all bivariate plots with the isotopic field of Pinar de Bédar/Sierra Cabrera (Fig. 10). Although isotopically it is not possible to differentiate between the copper deposits of Pinar de Bédar and Sierra Cabrera given their similar genesis, it is possible, as already mentioned above, to distinguish them by their trace element patterns (Figs. 8 and 9): the copper ores at Pinar de Bédar have much higher mean levels of arsenic (7.6%), zinc (3.4%) and lead (3.4%) than those from Sierra Cabrera (455 ppm As, 276 ppm Zn and 31 ppm Pb on average) (Murillo-Barroso *et al.* 2019). These compositional differences allow us to posit that the origin of the archaeological metalworking remains from Las Pilas was Pinar de Bédar given that the average content of arsenic, zinc and lead from the ores and fragments of slag recovered at this site also show relatively high

average values for these three elements (3.8% As, 2.7% Zn and 1.4% Pb on average for the ores [Tab. 4], and 9.7%  $\text{As}_2\text{O}_3$ , 19.7% ZnO and 6.5% PbO on average in the fragments of slag [Murillo-Barroso *et al.* 2017]). Nevertheless, although the majority of the metalworking remains from Las Pilas can be associated with the deposits of Pinar de Bédar, we also note the minor presence of ores from other mining areas. This applies to sample LP8479 which can be associated with the isotopic field of Cerro Minado (with lower levels of lead and zinc and relatively elevated levels of Co and Ni), and the sample of copper LP6972, which falls in the overlap zone between Cerro Minado and Herrerías/Almagrera. The sample appears to correspond rather more with Herrerías/Almagrera in the lead isotope plots (Fig. 10), although its composition resembles more closely the ores from Cerro Minado, with high levels of Co and Ni. However, the samples from Las Pilas show a trend along a lead growth curve like the ores from Cerro Minado while the ores from the Bédar range seem to form a secondary isochron (Fig. 10). As such, it is plausible that the isotopic field of Cerro Minado may expand in this direction if further samples are analysed, although the composition of both ores and slags from Las Pilas are more consistent with the mineralisations of Pinar de Bédar than to the ones at Cerro Minado regarding their high levels of Zn, As, Pb and U.

ID	Site	Sample	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	Reference
LP 6726	Las Pilas	Copper ore	18.8153	15.6950	39.0799	2.07703	0.83416	This paper
LP 6491	Las Pilas	Copper ore	18.8128	15.6944	39.0780	2.07720	0.83424	This paper
LP 6740	Las Pilas	Copper ore	18.9508	15.7000	39.0462	2.06039	0.82846	This paper
LP 7706	Las Pilas	Copper ore	18.8182	15.6956	39.0803	2.07673	0.83406	This paper
LP 6972	Las Pilas	Copper ore	18.7670	15.6900	39.0099	2.07864	0.83604	This paper
LP 8479	Las Pilas	Copper ore	18.6588	15.6919	38.7930	2.07907	0.84099	This paper
LP 11454	Las Pilas	Copper awl	18.6663	15.6854	38.8236	2.07987	0.84031	This paper
LP 2997	Las Pilas	Copper awl	18.7082	15.6994	38.9978	2.08452	0.83917	This paper
LP 8862	Las Pilas	Copper lump	18.5679	15.6894	38.6972	2.08409	0.84498	This paper
LP 8815	Las Pilas	Slag fragment	18.5523	15.6861	38.6741	2.08460	0.84551	This paper
LP 6816	Las Pilas	Slag fragment	18.8127	15.6957	39.0822	2.07744	0.83432	This paper
LP 6918	Las Pilas	Slag fragment	18.8073	15.6798	39.0283	2.07516	0.83371	This paper
LP 6974	Las Pilas	Slag layer	18.8167	15.6959	39.0844	2.07711	0.83415	This paper
LP 6735	Las Pilas	Slag layer	18.8135	15.6960	39.0831	2.07740	0.83430	This paper
LP 6608	Las Pilas	Slag layer	18.8181	15.6957	39.0825	2.07686	0.83407	This paper
HOSB_21/288	Santa Bárbara	Copper ore	18.6577	15.6977	38.7829	2.07865	0.84135	This paper
HOSB_11/48	Santa Bárbara	Copper ore	18.5107	15.6793	38.6479	2.08787	0.84704	This paper
HOSB_4/39	Santa Bárbara	Copper ore	18.7877	15.7001	38.9981	2.07573	0.83566	This paper
HOSB_8/30	Santa Bárbara	Copper ore	18.8603	15.7061	38.9966	2.06765	0.83276	This paper
HOSB_1/197	Santa Bárbara	Copper ore	18.6868	15.6903	38.8121	2.07698	0.83964	This paper
HOSB_4/64	Santa Bárbara	Copper ore	18.6944	15.6931	38.8696	2.07921	0.83945	This paper
HOSB_21/78	Santa Bárbara	Slag fragment	18.6111	15.6920	38.7541	2.08231	0.84315	This paper
HOSB_21/283	Santa Bárbara	Slag fragment	18.6168	15.6973	38.7636	2.08218	0.84318	This paper
HOSB_1/441	Santa Bárbara	Slag fragment	18.7158	15.6997	38.8963	2.07826	0.83885	This paper
HOSB_1/168	Santa Bárbara	Slag fragment	18.6616	15.6953	38.8058	2.07944	0.84105	This paper
HOSB_17/173	Santa Bárbara	Slag fragment	18.5859	15.6975	38.7315	2.08391	0.84459	This paper
HOSB_16/112	Santa Bárbara	Slag fragment	18.6221	15.6936	38.7591	2.08135	0.84274	This paper
HOSB_23/32	Santa Bárbara	Copper lump	18.6409	15.6957	38.7956	2.08121	0.84201	This paper
HOSB_25/4	Santa Bárbara	Copper lump	18.5354	15.6706	38.6778	2.08669	0.84544	This paper
HOSB_25/5	Santa Bárbara	Copper lump	18.7335	15.6912	38.8121	2.07180	0.83760	This paper
ALMZ_1443D	Almizaraque	Copper ore	18.7042	15.6927	38.8605	2.07763	0.83899	This paper
ALMZ_M6	Almizaraque	Copper ore	18.7019	15.6935	38.8648	2.07812	0.83914	This paper
ALMZ_1076A	Almizaraque	Copper ore	18.6547	15.6929	38.7879	2.07926	0.84123	This paper
ALMZ_1096	Almizaraque	Copper ore	18.7509	15.6935	38.9688	2.07823	0.83695	This paper
ALMZ_1443 I	Almizaraque	Copper ore	18.6369	15.6909	38.7498	2.07920	0.84193	This paper

Tab. 5. [1] Multi-collector inductively-coupled plasma - mass spectrometry (MC-ICP-MS) lead isotope analysis results.

ID	Site	Sample	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	Reference
ALMZ_M16	Almizaraque	Copper ore	18.5600	15.6943	38.7384	2.08720	0.84560	This paper
ALMZ_AA1100C	Almizaraque	Copper ore	18.7448	15.6938	38.9627	2.07859	0.83723	This paper
ALMZ_AA1099B	Almizaraque	Copper ore	18.8407	15.6703	38.9601	2.06787	0.83173	This paper
ALMZ_PA0320	Almizaraque	Copper ore	18.578	15.6828	38.687	2.0824	0.84416	Montero Ruiz and Murillo-Barroso 2010
ALMZ_AA1104E	Almizaraque	Slag fragment	18.7135	15.6878	38.9105	2.07928	0.83831	This paper
ALMZ_AA1094	Almizaraque	Slag fragment	18.4408	15.6824	38.6296	2.09479	0.85042	This paper
ALMZ_PA1443F	Almizaraque	Slag fragment	18.6809	15.6886	38.8990	2.08229	0.83982	This paper
ALMZ_AA1106D	Almizaraque	Slag fragment	18.6827	15.668	38.8371	2.07868	0.83863	Montero Ruiz and Murillo-Barroso 2010
ALMZ_AA1080B	Almizaraque	Slag fragment	18.6497	15.6745	38.7731	2.07893	0.84047	Montero Ruiz and Murillo-Barroso 2010
ALMZ_PA2619	Almizaraque	Copper lump	18.6668	15.6649	38.7906	2.07796	0.83918	Montero Ruiz and Murillo-Barroso 2010
ALMZ_M32	Almizaraque	Copper lump	18.6994	15.6852	38.8986	2.08021	0.83881	This paper
ALMZ_AA1084	Almizaraque	Copper lump	18.5524	15.6785	38.6671	2.08421	0.84509	This paper
ALMZ_PA3060	Almizaraque	Copper chisel	18.7479	15.7103	38.9885	2.07962	0.83798	This paper
1984/138/1/181	Churuletas 1	Copper awl	18.0961	15.6230	38.1075	2.10584	0.86333	This paper
1984/71/1/183	La Encantada I	Palmela point	18.4548	15.6616	38.5720	2.09009	0.84865	This paper
1984/71/1/182	La Encantada I	Palmela point	18.4072	15.6869	38.6173	2.09792	0.85221	This paper
86/129/LB/I/62	Loma de Belmonte	Palmela point	18.7167	15.7004	38.8472	2.07588	0.83886	This paper
	Terrera Ventura	Copper lump	18.659	15.6896	38.8723	2.0833	0.84086	Stos Gale <i>et al.</i> 1999
A-52557(BA1)	El Barranquete	Copper Awl	21.285	15.8294	38.7442	1.82026	0.74369	Stos Gale <i>et al.</i> 1999

Tab. 5. [2] Multi-collector inductively-coupled plasma - mass spectrometry (MC-ICP-MS) lead isotope analysis results.

## DISCUSSION

The site of Las Pilas is located in the foothills of the Sierra Cabrera at barely 1 km from areas where copper deposits have been identified, while the mines of Pinar de Bédar are found at a distance of 10 km as the crow flies. There are several reasons why the metalworkers from Las Pilas would choose to make use of the ores from Pinar de Bédar despite having available copper resources in their immediate surroundings. It is possible that this was a voluntary and intentional

choice, given the greater richness in arsenic of the Pinar de Bédar ores as compared to those of Sierra Cabrera, allowing the direct smelting of arsenical copper. Nonetheless, it is difficult to evaluate whether the visibility of these resources in prehistory was significantly greater than those of Sierra Cabrera. Nowadays the potential of the copper deposits at Pinar de Bédar is greater than that of the veins of copper which appear spread amongst the phyllites of the Sierra Cabrera, but as has been mentioned, the intense modern exploitation

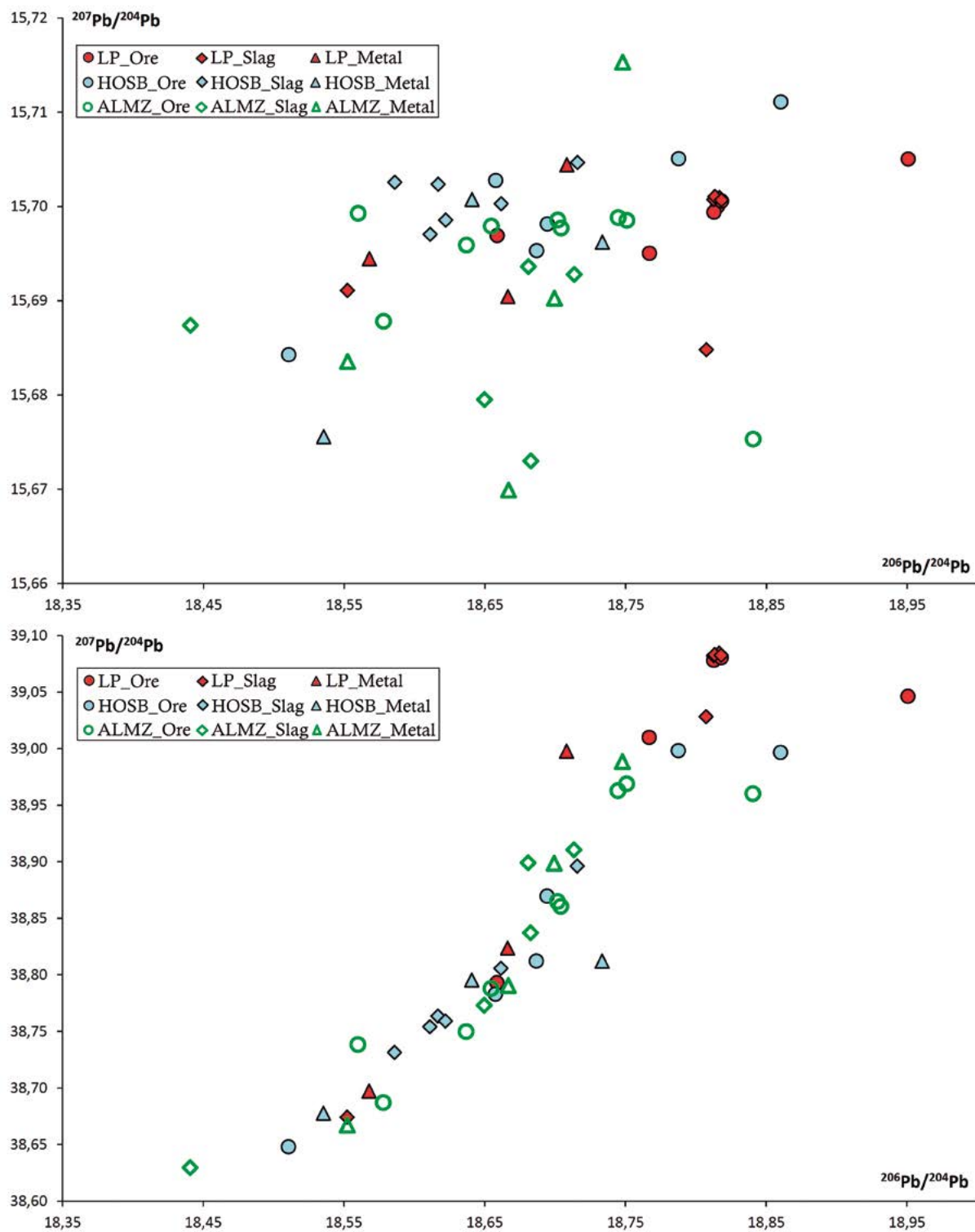


Fig. 9. Lead isotope ratios of archaeometallurgical remains (ore, slag fragments and metals) from Las Pilas (LP), Santa Bárbara (HOSB) and Almizaraque (ALMZ). Analytical errors are smaller than symbols (in colour in the electronic version).

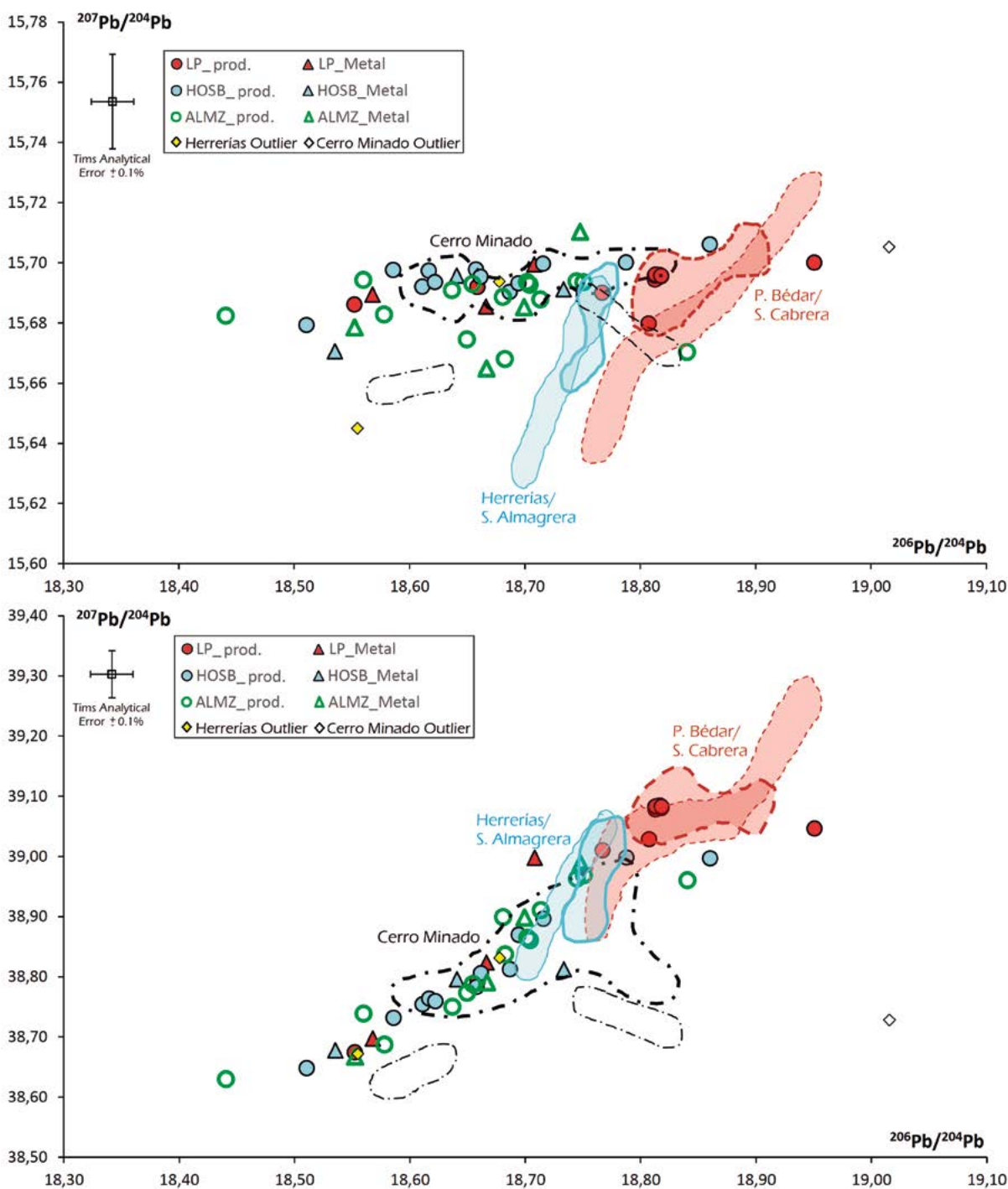


Fig. 10. Lead isotope ratios of archaeometallurgical remains (production debris and metals) from Las Pilas (LP), Santa Bárbara (HOSB) and Almizaraque (ALMZ) plotted against the isotopic fields of Cerro Minado, Pinar de Bédar/Sierra Cabrera and Herrerías/Almagrera. Isotopic fields based on multi-collector inductively-coupled plasma - mass spectrometry (MC-ICP-MS) have thicker border lines than isotopic fields defined by thermal ionization mass spectrometry (TIMS) analyses. Isotopic fields defined based on data from Murillo-Barroso *et al.* 2019 and Oxalid. MC-ICP-MS analytical errors are smaller than symbols (in colour in the electronic version).

of the zone makes it impossible to evaluate the state of the deposits in prehistoric times.

Returning to the lead isotope analysis results, the bulk of the samples of metal production remains from Santa Bárbara and Almizaraque resemble the isotopic range of Cerro Minado (Fig. 10). In the case of Santa Bárbara, all of the slag samples and four out of the six ore samples analysed can be associated with Cerro Minado. Of the other ore samples, HOSB8/30 falls in a peripheral zone of Pinar de Bédar/Sierra Cabrera isotopic field, although its trace element pattern does not match either of these deposits. The second ore sample, HOSB11/48, would fall in a zone which cannot currently be associated with any known isotopic field in Iberia.

In the case of Almizaraque, six out of the nine ore samples analysed can be associated with Cerro Minado although two of them (AA1100C, with 1.4% As [Delibes *et al.* 1989] and ALMZ\_1096, with 0.9% As [Tab. 4]) fall in the limit of Herrerías/Almagrera and Cerro Minado. Of the three other samples, AA1099B, with relatively elevated As, Pb and (to an extent) Zn contents (Delibes *et al.* 1989), resembles the isotopic field of Pinar de Bédar/Sierra Cabrera although falling in a peripheral area in some plots. Nonetheless, the samples ALMZ\_M16 and PA0320 fall close to Cerro Minado, without fitting with any known isotopic field, which opens the possibility that ores from other deposits were also being exploited, although to a lesser extent. In the case of the slag samples from Almizaraque, four of them can be associated with the isotopic field of Cerro Minado: PA1443F and AA1104E match with Cerro Minado, while AA1106D and AA1080B fall partially in the space between the geological samples analysed using MC-ICP-MS and TIMS (Fig. 10). However, taking into account the greater analytical error in the older TIMS analyses, a link to Cerro Minado cannot be completely ruled out. Finally, the last slag sample (ALMZ\_AA1094) falls in a zone which cannot be associated with any known isotopic field in the region.

In a study of Egyptian samples it has been proposed that the contribution of lead from ceramics could distort the isotopic signature of the slag samples, especially when the mineral resources used have a low lead content (*e. g.* Rademakers *et al.* 2017), something which is true in the cases of the ores from Cerro Minado and from Sierra Cabrera (c. 30 ppm on average). However, in our case, a similar spread is observed for both ore and slag samples from Almizaraque and Santa Bárbara, thus indicating that the explanation derives rather from the use of diverse mineral resources.

Overall, it appears that Cerro Minado is the main source of ores employed at Almizaraque and Santa Bárbara, although ores from other deposits could be

arriving sporadically. Santa Bárbara is located barely 1 km, and Almizaraque around 20 km as the crow flies, from Cerro Minado. In the case of Santa Bárbara it is clear that they were exploiting the nearest copper mineral resources to the site. However, in the case of Almizaraque the situation is similar to that of Las Pilas, since the Herrerías mines and the deposits of Sierra Almagrera are barely 500 m from Almizaraque. In spite of this, it appears that the majority of the metal production remains from this site match better the isotopic field of Cerro Minado. The preferential choice of some ore sources in this case does not appear justified by the arsenic levels in Cerro Minado (2.5% on average (Murillo-Barroso *et al.* 2019). Although the As levels of Sierra Almagrera ores are indeed typically low (<0.1% As on average in the XRF analyses by Montero Ruiz 1994 and <0.01% As in the recent XRF analyses from the mine La Estrella<sup>2</sup>), the deposits at Herrerías, adjacent to Almizaraque, also show moderate levels of arsenic (1.5% on average, Murillo-Barroso *et al.* 2019). Again the importance of variables such as the visibility, accessibility and potential of the deposits in prehistoric times must be highlighted, although these variables are difficult to evaluate in present times.

We have to bear in mind as well the difficulty in defining the isotopic field of Herrerías, which cannot be differentiated from Sierra Almagrera, and from which two of the analysed samples are displaced significantly towards the zone which covers the isotopic field of Cerro Minado. In figure 10 the two 'outliers' from Herrerías are also represented, which could be indicators of the direction in which the isotopic field might expand should further analyses be conducted. Were that so, the overlap with the isotopic field of Cerro Minado could turn out to be considerable; based on the limited dataset available, the differences in the trace element patterns are limited too (with the apparent exception of the mentioned lead levels). It would be necessary to expand the ore sampling from Herrerías to achieve greater definition, although given the extensive modern exploitation of the area it will be difficult to obtain new samples for analysis.

Overall, at the three sites studied, copper sources with naturally elevated levels of other elements, especially arsenic, zinc or lead, were prioritised ahead of other sources with lower levels (especially of arsenic), even when the latter were at a closer distance. This model differs from that proposed for the Valle del Guadalentín (Murcia): here, Escanilla (2016) has proposed the deliberate mixing of arsenic-rich ores from the distant sources of Cerro Minado (beyond around 50

<sup>2</sup> Vide n.º 1.

km) with local, arsenic-poor copper carbonates, yielding metal with lower arsenic levels than if Cerro Minado ores were smelted directly. Nonetheless, this constitutes a model based exclusively on elemental analyses, which should be verified isotopically.

Turning to the lumps of metallic copper and finished objects from the three sites, it is significant that none of the metallic remains coincide with the isotopic fields of Pinar de Bédar/Sierra Cabrera or Herrerías/Almagrera, especially given that the use of these resources has indeed been verified, at least in the case of the site of Las Pilas. Instead, metallic samples from all three sites can be linked to Cerro Minado. At least one sample from each site can be associated with this isotopic field in all bivariate plots, with a second from each site falling in peripheral zones in some diagrams. Further, it is significant that three objects (one from each site) have very similar isotopic values, clustering around 18.55 ( $^{206}\text{Pb}/^{204}\text{Pb}$ ), a zone around which metal production remains from the three sites also group, and which does not coincide with any known isotopic field in the close area, unless new analyses from Cerro

Minado or Herrerías expand their isotopic fields in this direction. This could indicate the existence of other deposits exploited, at least occasionally, by the three sites and which are either completely exhausted or yet to be characterised.

With regards to the isotopic signature of the remaining objects analysed, it is worth highlighting that the punch recovered in the grave at Churuletas 1 does not resemble any of the pieces from the Southeast nor the metal production remains from the Vera Basin, having an isotopic signature more similar to the objects from southwestern Iberia which could be associated with deposits of an older geological age. Figure 11 shows how objects from the southwest, in this case represented by samples from the provinces of Seville and Badajoz identified with circles (Hunt 2003; Hunt *et al.* 2009; Hunt *et al.* 2012), tend to cluster mostly in the range 18.1-18.5 ( $^{206}\text{Pb}/^{204}\text{Pb}$ ) and 38.1-38.7 ( $^{208}\text{Pb}/^{204}\text{Pb}$ ), while the majority of the objects from the Southeast (identified by triangles) do so at 18.5-18.8 ( $^{206}\text{Pb}/^{204}\text{Pb}$ ) and 38.5-39.0 ( $^{208}\text{Pb}/^{204}\text{Pb}$ ). The punch from Churuletas 1, with values of 18.0961 ( $^{206}\text{Pb}/^{204}\text{Pb}$ )

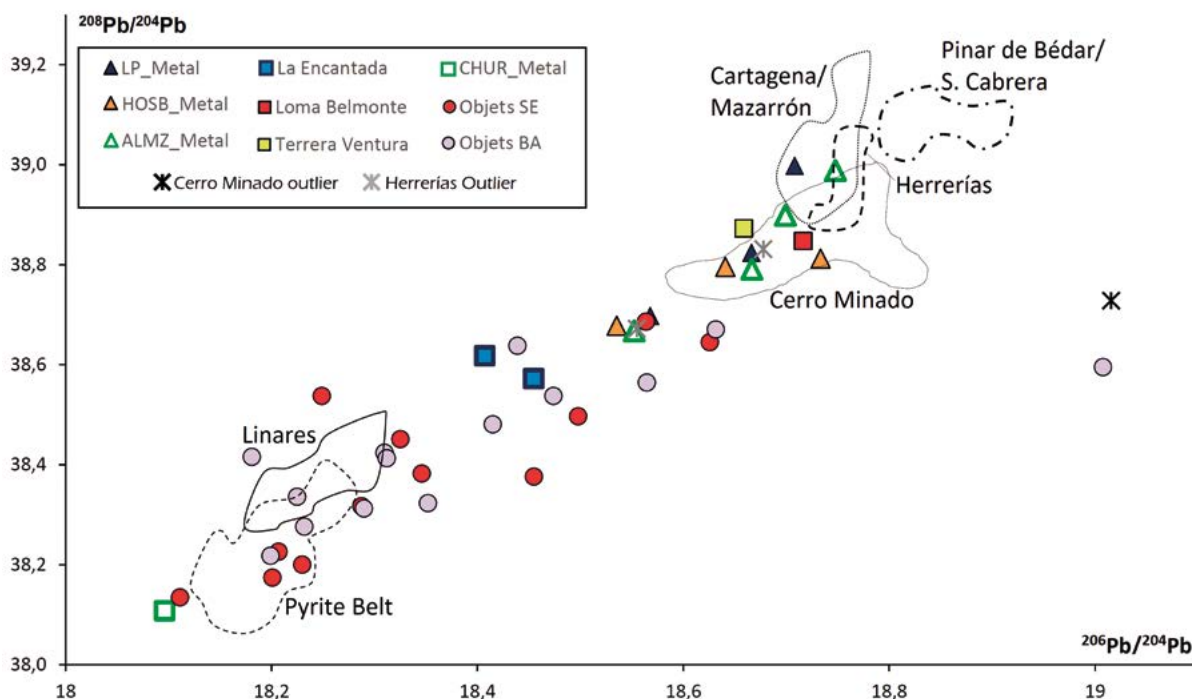


Fig. 11. Lead isotope ratios of metals from the southeastern (squares and triangles) and southwestern (circles) Iberia plotted against the isotopic fields of Cartagena/Mazarrón, Cerro Minado, Pinar de Bédar/Sierra Cabrera, Herrerías/Almagrera, the Iberian Pyrite Belt and Linares. Isotopic fields based on data from Stos Gale *et al.* 1995; Hunt Ortiz 2003; Santos Zaldegui *et al.* 2004; Murillo-Barroso *et al.* 2019 and Oxalid. Southwestern objects are from provinces of Badajoz and Seville (data from Hunt Ortiz 2003; Hunt Ortiz *et al.* 2009; Hunt Ortiz *et al.* 2012). BA = Badajoz province; SE = Seville province; LP = Las Pilas; HOSB = Santa Bárbara; ALMZ = Almizaraque; CHUR = Churuletas 1 (in colour in the electronic version).

and 38.1075 ( $^{208}\text{Pb}/^{204}\text{Pb}$ ) is displaced significantly from the majority of the objects from the Southeast, with one of the spearheads from the dolmen at La Pastora (Valencina de la Concepción, Seville) being the only object showing a relatively similar isotopic composition.

For the remaining artefacts from the area, only the Palmela-type arrowhead from Loma de Belmonte and the sample from Terrera Ventura can be related to the deposits in the Vera Basin, which in both cases coincide with the isotopic field of Cerro Minado. In the case of La Encantada I, the two arrowheads resemble three objects recovered in La Pijotilla (Badajoz), one punch, one saw, and a possible ingot, for which Ossa Morena has been proposed as their ore source (Hunt 2003).

## CONCLUSIONS

The aim of this paper was to evaluate the provision and management of copper ore resources at three metallurgical sites in the region where the earliest metallurgy in Iberia is documented. Having archaeological remains related with the whole productive process as well as a few final metal objects, it has been possible to both identify the copper sources exploited and to evaluate if the objects found at each site had been produced on-site or obtained by other means.

Based on the combination of lead isotope analyses and trace element compositions we can establish the following:

1. As suggested by prehistoric mining evidence at Cerro Minado, its exploitation during the Copper Age by the sites of Santa Bárbara and Almizaraque is well confirmed.

2. The exploitation of the copper resources of Pinar de Bédar by the site of Las Pilas is also confirmed despite the absence of prehistoric mining evidence in this district. Intense modern mining works starting in the 16<sup>th</sup> century and developing in the area especially during the 19<sup>th</sup> and the 20<sup>th</sup> centuries might have erased any prehistoric mining evidence in the field, which adds value to lead isotope analysis in combination with trace elements composition as an indirect proof of prehistoric copper mining.

3. There is no isotopic concordance between metallurgical remains and finished objects at every site. While most of the production debris from Las Pilas clusters quite tightly together matching the isotopic field of Pinar de Bédar, none of the objects can be associated to this cluster or isotopic field. That would suggest a metallurgical production focused mainly on the exploitation of specific resources and a movement of objects within a network probably including Santa

Bárbara and Almizaraque. These two sites shared the resources of Cerro Minado, as confirmed in objects and production debris, therefore suggesting self-sufficient production.

4. Copper resources with higher levels of zinc, lead and/or (especially) arsenic appear to have been prioritised over other copper resources with lower levels, despite being geographically at further distance from the sites. This is the case for the exploitation of copper ores of Pinar de Bédar instead of Sierra Cabrera at Las Pilas, or Cerro Minado instead of Herrerías or Almagrera at Almizaraque.

5. Regional 'provinces' of metal exploitation and circulation can be inferred when comparing southeastern and southwestern metal objects, although some sporadic long-distance exchange would have happened, as exemplified by the awl from Churuletas I and the Palmela points from La Encantada I.

All in all, the analyses of Copper Age production remains and artefacts from the Southeast of the Iberian Peninsula allow us to propose a model of local production using mainly, but not only, copper resources from the nearby surroundings. Despite the prioritisation of sources rich in arsenic and other elements, even though found at a greater distance from the settlements, the distances are no more than 30 km as the crow flies. In the case of Las Pilas, this preference for resources rich in arsenic, zinc and lead from Pinar de Bédar over those from Sierra Cabrera, might also be due to the greater metal-producing potential of these copper resources as compared to the low-grade copper ores from Sierra Cabrera, which would have a lower yield.

In the case of Almizaraque it seems unlikely that the metal workers at this site were unaware of the existence of the copper ores at Herrerías, at barely 500 m from the settlement and with arsenic levels also notably elevated (1.5% on average), indicating that neither arsenic content nor proximity were the determining factors at the point of choosing a resource. Other issues must also have had an important role to play in the choice of these ores, such as a greater tradition of mining at Cerro Minado, social relations with other communities such as that of Santa Bárbara (perhaps involving collaboration in mine-working), or factors relating to the visibility or accessibility of the resources.

As concerns the finished objects, although some of those from Almizaraque and Santa Bárbara could be linked to metal production remains at their respective sites, the same cannot be said in the case of Las Pilas, where it seems likely that metal produced in other settlements may be arriving. This evidence points to the circulation of goods at the regional scale and may-

be even further. The intensity of this process cannot be for now determined; however, given the relative abundance of both ore resources and production sites in the same area, metal exchange is unlikely to have been driven by hierarchical relations.

From a wider perspective, this study confirms that the metal production model during the Iberian Copper Age is neither one of dependence on a single mineral source nor one of controlled access of resources given the abundance and widespread distribution of copper ore resources available. The evidence thus appears to support a model of domestic, not specialised and not monopolised, metallurgical production. Against this background, high-resolution local studies, such as the one presented here, are mandatory to progressively understand the development of metal production and consumption at the peninsular scale. In this sense, this study is only a starting point that should be expanded to address the regional and supra-regional levels.

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## ANNEX SUPPLEMENTARY FILE

In the electronic version (drop-down menu, Supplementary files), the excel table SF1 is available as complementary material:

SF1. Trace elements compositions of ore samples from archaeological contexts by inductively coupled plasma mass spectrometry (ICP-MS).