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### Siret revisited: The metals in the British Museum's Siret collection

### SIRET REVISADO: LOS METALES DE LA COLECCIÓN SIRET DEL MUSEO BRITÁNICO

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Abstract This paper presents the analytical study of the metal objects from the Siret Collection held by the British Museum. X-ray Fluorescence has been conducted to determine their elemental composition and by lead isotope analysis (by MC-ICP-MS) to determine their origin. We combine this new data with extant analyses to discuss and reinterpret the role of bronze alloys in Argaric society and the diversity of exploited mining resources.

Keywords Archaeometallurgy, El Argar Culture, Lead isotope analysis, Elemental composition, Bronze Age.

Resumen Se presenta el estudio analítico de los objetos de metal de la Colección Siret del British Museum. Las piezas se han analizado por fluorescencia de Rayos X para determinar su composición elemental y por isótopos de plomo (MC-ICP-MS) para la determinación de su procedencia. A la luz de estos nuevos datos y de los recopilados de la bibliografía se discute el papel de la aleación de bronce en la sociedad argárica, así como la diversidad de los recursos mineros explotados.

**Palabras clave** Argueometalurgia, Cultura de El Argar, isótopos de plomo, composición elemental, Edad del Bronce.

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#### **1. INTRODUCTION**

The studies undertaken by the Siret brothers, together with their foreman Pedro Flores, in the Cuenca de Vera area and largely published in their book *Les premiers âges du metal dans le sud-est de l'Espagne* (Siret and Siret, 1887; Spanish version published in 1890) remain a benchmark for current archaeological research and, in some cases, the only source of information for the study of certain archaeological sites.

Unfortunately, the collection is scattered throughout a large number of museums in Europe and the United States as a consequence of the sale of part of the finds in the last decade of the 19<sup>th</sup> century, a habitual practice at that time. In Spain, the collection is concentrated mainly in the National Archaeological Museum in Madrid (MAN), where it was taken after the death of Luis Siret in 1934, although it had previously been donated to the Spanish state in 1928. Some finds had already been donated to the Museum of History in Barcelona, following the award of the Martorell Prize for the publication of the aforementioned monograph (Siret and Siret, 1890). Those finds were incorporated into the collection of the Archaeological Museum of Barcelona (Andúgar, 2006), now the Archaeological Museum of Catalonia.

The Siret brothers gave talks and took part in congresses to publicise their finds and promote their work and finds to the most important museums and institutions of the time. For example, the purpose of their participation in the Manchester meeting of the British Association in September 1887 (*The Yorkshire Post*, 6 February 1887, p. 6) is likely to have been to raise interest in their work in United Kingdom. They participated in the anthropology session chaired by A.H. Sayce with the conference "The Early Age of Metals in the South-East of Spain" that, according to the newspaper, was translated into English by Dr. Camartelli. At these meetings they also presented finds from their excavations. This led to the Siret brothers making a complaint to the *Manchester Courier*, which had written that their finds were from the Visigothic period. On September 2, that same year the newspaper published a letter to the editor clarifying that the finds were prehistoric, so that there could be no possible confusion amont interested parties or prospective buyers.

In the United Kingdom the main lot of finds is conserved in the British Museum in London, although there are also some pieces in the University Museum of Archaeology and Ethnology in Cambridge and the Ashmolean Museum of Art and Archaeology in Oxford. In Belgium the collection is split between the *Musées Royaux d'Art et d'Histoire* in Brussels, the *Museum Vleeshuis* in Antwerp, and the *Musée Archéologique de l'Université* in Ghent. In Germany there are finds from the Siret collection in the *Staatliche Museen* and the *Völkerkunde-Museum* in Berlin and the *Vorgeschichtliches Seminar der Philipps-Universität* in Marburg. Other Siret finds are in the *Museo Pigorini* in Rome (Italy) and the Peabody Museum of Archaeology and Ethnology at Harvard University (United States).

This dispersion of the finds means that in some cases the whereabouts of certain pieces is unknown, as the sale to one museum or another did not correspond to any scientific criteria, but rather to the commercial interests of the antiquities market that prevailed in the late 19<sup>th</sup> century and even into the early 20<sup>th</sup> century. We are therefore faced with the circumstance that not only finds from the same archaeological site, but also grave goods from the same tomb were divided among different museums. This is an enormous hindrance to their comprehensive study, because it means they are subject to different heritage rules, different administrations and various degrees of 'gate-keeping' when it comes to allowing them to be studied and sampled for scientific analysis.



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ISSN: 1133-4525 ISSN-e: 2255-3924 In this paper we present the study carried out on the metallic finds in the British Museum's Siret collection. We will first offer an initial evaluation of the collection as a whole and then comment on the results obtained from the composition and lead isotope analyses. Finally, we will discuss these results in the framework of the analyses already published for the rest of the objects from the Argaric area.

#### 2. THE SIRET COLLECTION IN THE BRITISH MUSEUM

The Siret collection was acquired by the British Museum on July 4, 1889 from Henri Siret as an addition to its European Bronze Age collection. It was sold by the Siret brothers for £200 and sent to the Museum in three boxes weighing 230 kg (Raposo, 2012).

According to the documentation held at the British Museum, it was H. Siret, living at that time in Antwerp (Belgium), who first contacted Sir Henry Hoyle Howorth, one of the trustees of the museum, to express his interest in selling part of his archaeological finds to it. In a subsequent letter, the museum acknowledged the need to increase its prehistoric Spanish collection «from which there are only objects from Gibraltar and sites with Stone Age material». Initially the museum asked for a complete assemblage of Bronze Age grave goods and for representative pieces of the main object types found in the region. The museum sent a young Charles Hercules Read (1857-1929), then assistant in the Department of Antiquities (later knighted and a leading figure within the London museum world), an assistant in the Department of Antiquities at the time, to personally inspect the collection in Belgium and select the objects to be purchased. The Siret Collection in the British Museum is the result of Read's trip to Belgium, where he selected finds from various sites spanning from the Mesolithic to the Bronze Age. In some cases, the artefact type was the deciding factor, regardless of its archaeological context. A total of 331 objects arrived at the museum in June 1889; they were confirmed, drawn, reconstructed in some cases, and included in the acquisition records on July, 4 (Raposo, 2012). This date became the prefix to the collection catalogue number (1889,0704.), followed by the object (or group of objects) and the individual catalogue entry number from 1 to 252. Three replicas of the dagger from Tomb 1 at Zapata and the halberds from Tomb 575 at El Argar and Tomb 18 at Fuente Álamo, were also purchased and given inventory numbers CRM 137, CRM 138 and 1889,0704.248, respectively. Of the 39 sites published by the Sirets in 1887, the British Museum collection contains objects from 16 of them (tab. 1).

The Siret Collection is composed of ornaments, pottery and geometric flakes from the Neolithic sites of Cueva de Los Toyos, Puerto Blanco and El Garcel; pottery, ore, slag and copper objects from the Chalcolithic sites of Tres Cabezos, Campos and Parazuelos; and metal ornaments from the Late Bronze Age Tomb 1 at Qurénima. The flint object from Haverlee (Belgium) was published in 1887 by the Siret brothers (Siret and Siret, 1887) as a matter of comparison between the sites of El Garcel and La Gerundia and was acquired by the museum for the same reason. The majority of the collection is from the Argaric Bronze Age, mainly from the sites of El Argar (Antas, Almería) and El Oficio (Cuevas de Almanzora, Almería), with moulds, crucibles, some extraordinary metal objects, such as the silver diadem from Grave 51 and the sword from Grave 429 at El Argar or the halberd from Grave 42 at El Oficio.

The collection includes 57 metal objects and four items related to metallurgical activities (the two 'ingots' from Parazuelos that, on review, were identified as copper ore and a copper slag fragment, a crucible from El Argar, which has been recently published

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ISSN: 1133-4525 ISSN-e: 2255-3924 in detail [Mongiatti and Montero-Ruiz, 2020], and a stone mould from Las Anchuras), making up 17.5% of the objects in the Siret Collection (tab. 2, fig. 1). Note that some spiral fragments were identified as a single object (1889,0704.233 from El Oficio and 1889,0704.170 from El Argar) but elemental analyses revealed these being copper and bronze fragments and therefore are considered as different objects in this paper.

 Table 1. The Siret Collection at the British Museum.

| Site               | Number of objects | Metals                   |
|--------------------|-------------------|--------------------------|
| El Garcel          | 11                |                          |
| La Gerundia        | 2                 |                          |
| Cueva de los Toyos | 11                |                          |
| Parazuelos         | 2                 | Cu ore and slag fragment |
| Palaces            | 6                 |                          |
| Campos             | 30                | 1                        |
| Tres Cabezos       | 4                 |                          |
| Puerto Blanco      | 1                 |                          |
| lfre               | 1                 |                          |
| El Argar           | 125               | 33 + 1 Crucible          |
| Las Anchuras       | 3                 | Stone mould              |
| El Oficio          | 36                | 14                       |
| Fuente Álamo       | 4                 |                          |
| Lugarico Viejo     | 8                 | 2                        |
| Zapata             | 4                 | 1                        |
| Qurénima           | 5                 | 5                        |

Table 2. Metal assemblage of the Siret Collection of the British Museum and materials analysed.

| Site       | Туре                    | ID            | Context               | LIA | XRF |
|------------|-------------------------|---------------|-----------------------|-----|-----|
| Parazuelos | Cu ore                  | 1889,0704.38  | Inside the settlement |     | Cu  |
| Parazuelos | Cu slag                 | 1889,0704.39  | Inside the settlement |     | Cu  |
| Campos     | Awl                     | 1889,0704.64  | House I               |     | Cu  |
| El Argar   | Awl with bone<br>handle | 1889,0704.133 | Outside the graves    |     | Cu  |
| El Argar   | Arrowhead               | 1889,0704.134 | Outside the graves    |     | Cu  |
| El Argar   | Arrowhead               | 1889,0704.135 | Outside the graves    | Х   | Си  |
| El Argar   | Arrowhead               | 1889,0704.136 | Outside the graves    |     | Cu  |
| El Argar   | Arrowhead               | 1889,0704.137 | Outside the graves    |     | Си  |
| El Argar   | Clipping plate          | 1889,0704.138 | Outside the graves    |     | Br  |
| El Argar   | Axe                     | 1889,0704.143 | Grave 746             | Х   | Си  |
| El Argar   | Spiral                  | 1889,0704.144 | Grave 746             | Х   | Br  |
| El Argar   | Spiral Ag               | 1889,0704.145 | Grave 746             | Х   | Ag  |

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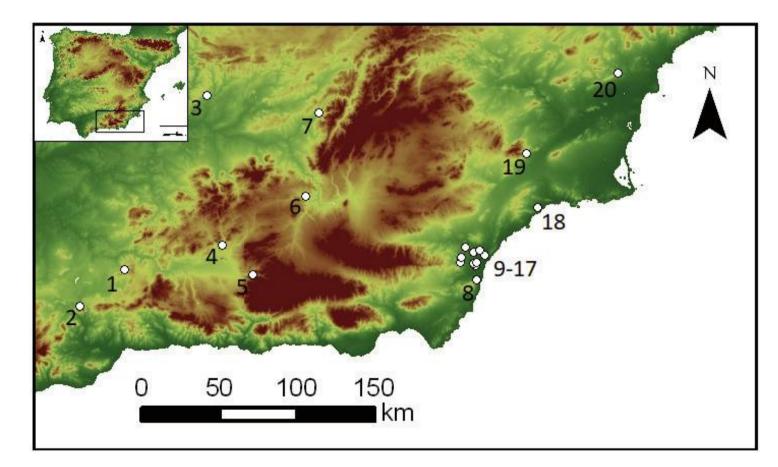
|           |                 | Table 2.       | (cont.).           |     |     |
|-----------|-----------------|----------------|--------------------|-----|-----|
| Site      | Туре            | ID             | Context            | LIA | XRF |
| El Argar  | Axe             | 1889,0704.148  | Grave 703          | Х   | Cu  |
| El Argar  | Dagger 2R       | 1889,0704.149  | Grave 703          | Х   | Cu  |
| El Argar  | Halberd 2R      | 1889,0704.153  | Grave 533          | Х   | Cu  |
| El Argar  | Sword           | 1889,0704.154  | Grave 429          | Х   | Cu  |
| El Argar  | Bracelet        | 1889,0704.155  | Grave 429          | Х   | Cu  |
| El Argar  | Ring            | 1889,0704.156  | Grave 429          |     | Br  |
| El Argar  | Ring            | 1889,0704.157  | Grave 429          |     | Cu  |
| El Argar  | Spiral Frags.   | 1889,0704.158  | Grave 429          |     | Br  |
| El Argar  | Dagger 2R       | 1889,0704.163  | Grave 711          |     | Cu  |
| El Argar  | Silver Diadem   | 1889,0704.168  | Grave 51           |     | Ag  |
| El Argar  | Spiral          | 1889,0704.170A | Grave 51           |     | Br  |
| El Argar  | Spiral          | 1889,0704.170B | Grave 51           |     | Cu  |
| El Argar  | Spiral          | 1889,0704.171  | Grave 51           |     | Cu  |
| El Argar  | Rings Frags.    | 1889,0704.171b | Grave 51           | Х   | Br  |
| El Argar  | Ring            | 1889,0704.171c | Grave 51           |     | Cu  |
| El Argar  | Dagger          | 1889,0704.177  | Grave 597          |     | Cu  |
| El Argar  | Dagger 4R       | 1889,0704.179  | Grave 694          |     | Cu  |
| El Argar  | Dagger 4R       | 1889,0704.180  | Grave 716          | Х   | Cu  |
| El Argar  | Gold ring       | 1889,0704.181  | Grave 89           |     |     |
| El Argar  | Ring            | 1889,0704.184  | Grave 471          | Х   | Ag  |
| El Argar  | Ring            | 1889,0704.185  | Grave 626          |     | Cu  |
| El Argar  | Ring            | 1889,0704.186  | Grave 649          |     | Cu  |
| El Argar  | Ring            | 1889,0704.187  | Grave 718          | Х   | Cu  |
| El Argar  | Axe             | 1889,0704.188  | Grave 958          | Х   | Cu  |
| El Oficio | Arrowhead       | 1889,0704.205  | Outside the graves |     |     |
| El Oficio | Saw             | 1889,0704.206  | Outside the graves |     | Cu  |
| El Oficio | Halberd 4R      | 1889,0704.221  | Grave 42           | Х   | Cu  |
| El Oficio | Dagger 5R       | 1889,0704.222  | Grave 42           | Х   | Cu  |
| El Oficio | Halberd         | 1889,0704.224  | Grave 62           | Х   | Cu  |
| El Oficio | Rivet Halberd   | 1889,0704.224  | Grave 62           |     | Cu  |
| El Oficio | Dagger 4R       | 1889,0704.225  | Grave 62           | Х   | Br  |
| El Oficio | Dagger 4R       | 1889,0704.226  | Grave 62           | Х   | Cu  |
| El Oficio | Dagger 3R       | 1889,0704.227  | Grave 62           | Х   | Br  |
| El Oficio | Silver bracelet | 1889,0704.228  | Grave 62           |     | Ag  |
| El Oficio | Silver ring     | 1889,0704.229  | Grave 62           |     | Ag  |
| El Oficio | Dagger 2R       | 1889,0704.230  | Grave 158          | Х   | Cu  |
| El Oficio | Awl             | 1889,0704.231  | Grave 158          |     | Br  |

Table 2. (cont.).

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| Table 2. (cont.). |                 |               |           |     |     |  |  |  |  |  |
|-------------------|-----------------|---------------|-----------|-----|-----|--|--|--|--|--|
| Site              | Туре            | ID            | Context   | LIA | XRF |  |  |  |  |  |
| El Oficio         | Spiral          | 1889,0704.232 | Grave 158 |     | Cu  |  |  |  |  |  |
| El Oficio         | Spiral 6V       | 1889,0704.233 | Grave 158 |     | Br  |  |  |  |  |  |
| El Oficio         | Spiral 6V       | 1889,0704.233 | Grave 158 |     | Cu  |  |  |  |  |  |
| El Oficio         | Spiral 5V       | 1889,0704.234 | Grave 158 |     | Cu  |  |  |  |  |  |
| Lugarico Viejo    | Dagger 2R       | 1889,0704.77  | Grave 9   |     | Cu  |  |  |  |  |  |
| Lugarico Viejo    | Axe             | 1889,0704.78  | Grave 10  | Х   | Cu  |  |  |  |  |  |
| Zapata            | Dagger 3R       | 1889,0704.87  | Grave 11  |     | Cu  |  |  |  |  |  |
| Zapata            | Rivet Dagger 3R | 1889,0704.87  | Grave 11  |     | Ag  |  |  |  |  |  |
| Zapata            | Rivet Dagger 3R | 1889,0704.87  | Grave 11  |     | Ag  |  |  |  |  |  |
| Qurénima          | Bracelet        | 1889,0704.68  | Grave 1   |     | Br  |  |  |  |  |  |
| Qurénima          | Bracelet        | 1889,0704.69  | Grave 1   |     | Br  |  |  |  |  |  |
| Qurénima          | Ring            | 1889,0704.70  | Grave 1   |     | Br  |  |  |  |  |  |
| Qurénima          | Bead            | 1889,0704.71A | Grave 1   |     | Br  |  |  |  |  |  |
| Qurénima          | Bead            | 1889,0704.71B | Grave 1   |     | Br  |  |  |  |  |  |



**Figure 1.** Location of archaeological sites mentioned in the text: 1. Necrópolis de Alcaide, 2. Cueva de Ardales, 3. Peñalosa, 4. Puerto Blanco, 5. Cerro de la Encina, 6. Cuesta del Negro, 7. Villacarrillo, 8. Gatas, 9. Lugarico Viejo, 10. Qurénima, 11. El Oficio, 12. El Argar, 13. La Gerundia, 14. El Garcel, 15. Campos, 16. Fuente Álamo, 17. Tres Cabezos, 18. Parazuelo, 19. La Bastida, 20. Laderas del Castillo.

### 3. ANALYSED FINDS AND METHODS

A total of 53 objects, the slag and the copper ore from Parazuelos and three rivets were analysed by X-ray fluorescence (XRF) to determine their composition. In addition, a sample of was extracted from 24 of the objects for lead isotope analysis (LIA) (tab. 2, fig. 2).

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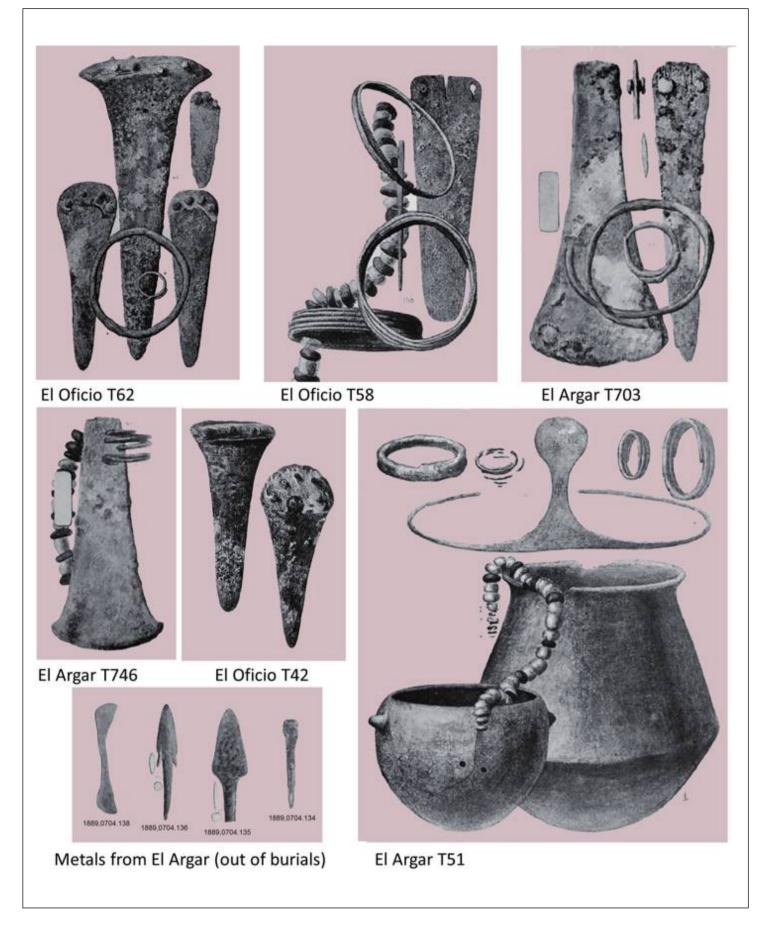


Figure 2. Grave goods of some of the tombs studied in this paper. Source: Siret and Siret (1887).

The X-ray fluorescence (XRF) analysis was carried out in the British Museum storage rooms using a portable Olympus InnovX Delta Premium DP6000 spectrometer with a rhodium anode and a silicon drift detector (SDD) from the UCL Institute of Archaeology. An analysis protocol based on the Innov-X Alloys program optimised at the UCL for the analysis of archaeological metals was applied. The program operates with an acceleration voltage of 40 kV, a current of 100 µA, a 2-mm-gauge aluminium filter and a 3-mm-diameter collimator. The acquisition time was 35 seconds per measure.

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ISSN: 1133-4525 ISSN-e: 2255-3924 The two silver objects from T62 at El Oficio and the diadem from T51 at El Argar were transferred to the Department of Scientific Research and analysed using a Bruker Artax XRF. The equipment was fitted with a molybdenum target X-ray tube and was operated at 50 kV and 0.5 mA. The objects were analysed using a collimated beam size of 1 mm and a counting time of 200 seconds. The resulting spectra were quantified using the *«fun-damental parameters with standards»* software written by Mike Cowell (formerly of the British Museum) using commercial multi-element silver alloy standards.

Some of the objects were heavily corroded and therefore the elemental analysis was carried out on the surface without removing the patina. This limits their comparison with other already published analyses, although it does allow us to obtain reference information on the metal used.

Lead isotope analysis (LIA) was conducted using Multi-Collector Inductively-Coupled Plasma Mass Spectrometry (MC-ICP-MS) at the Geochronology and Geochemistry Service (SGIker) of the University of the Basque Country (Spain) using a Neptune spectrometer (Thermo Fisher Scientific). The samples were obtained by drilling with 1-1.5-mm-diameter bits. More details about sample preparation and measurements can be found in Rodríguez *et al.* (2020). The standard deviation for all ratios was <0.005 (20), analytical errors (±0.01%), lower than symbols used in all graphs.

#### 4. RESULTS AND DISCUSSION

#### 4.1. Compositional analysis

An initial superficial compositional analysis was carried out on most of the objects with the aim of making a qualitative determination of the alloy used. In general, the state of conservation of the finds was poor and many of them were in an advanced state of corrosion, as previously reported by the Siret brothers themselves at the time they were discovered. We are aware of the limitations of superficial compositional analyses of metallic archaeological finds from which the metal has not been cleaned previously; this means the results must be taken qualitatively.

Most of the samples analysed (85%) (tab. 3) are from the Early Bronze Age period (2250-1550 BC) (El Argar, El Oficio, Lugarico Viejo, Zapata), to which we have to add three pieces from Chalcolithic contexts (3200-2250 BC) (Campos and Parazuelos) and five Late Bronze Age finds from Tomb 1 at Qurénima, as well as a fragment of metal from El Argar that we have to attribute to the mediaeval period and that we will comment on below.

The Chalcolithic items are a copper awl from Campos with 1.3% As and a sample of mineral and another of slag from Parazuelos with 8% and 2% As respectively, as well as impurities of Co, Ni and, in the case of the slag, also of Zn. The five pieces from Qurénima are bronzes with a high tin content that ranges between 8% and c. 30% wt Sn, and the two leaded bronzes beads with c. 3% and 5% wt Pb.

The El Argar-chronology pieces are the five samples of silver, 12 bronzes with Sn levels of between c. 2% and c. 11% wt Sn and 36 pieces of copper with levels of arsenic ranging from between lower values to the limit of detection at 21.7% wt As, of which 24 present values above 2% As. Of these pieces only one, Dagger 1889,0704.177 from Tomb 597 at El Argar, presents significant levels of lead, with a 2.4% wt Pb.

The silver pieces do not present significant levels of copper, except for the two rivets of the dagger from T11 at Zapata, in which the proportions could be overvalued due

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ISSN: 1133-4525 ISSN-e: 2255-3924 to the influence of the copper of the blade. The high levels of bromine detected in the spirals correspond to the surface corrosion on the pieces. Gold is another element of interest in the composition of the silver. In general, Argaric metals do not have significant proportions of gold, as the mineral of origin does not have them either. The native silver from Herrerías contains less than 0.15% Au based on the EDXRF analyses carried out in Mannheim (Murillo-Barroso *et al.*, 2014, p. 263, tab. 1) and the Argaric silver objects analysed in the same laboratory detected with a maximum content of 0.37% and an average value of 0.14% Au. The ring from La Cuesta del Negro published by Arribas *et al.* (1989) is within that high range (0.36% Au). The bracelet from El Oficio analysed with the Artax XRF, however, contains proportions of 0.7-0.8% Au. There are a few other pieces from the Bronze Age with high gold values: two of the rivets from a dagger from Villacarillo with 3-4% Au, the same as the rivets from the dagger from La Cueva de Ardales (Málaga) with 1.2% and 0.5% Au respectively (Montero-Ruiz *et al.*, 1995; Murillo-Barroso, 2013).

A consequence of the alteration of the original composition in the patina are the exceptionally high levels of arsenic of some pieces, reaching as much as 21.7%, which appears to correspond to a superficial arsenic enrichment process. Fortunately, we have three pieces previously analysed by Otto and Witter (1952) by OES. In table 4 we can observe the over-evaluation of the arsenic in the XRF analysis, with a certain correlation in which the highest values in the patina also correspond to higher arsenic values in the metal. This superficial enrichment process should also be considered for the high levels of tin detected in Late Bronze Age metals from Qurénima. Of these pieces, two also have high levels of lead, as is habitual in Late Bronze Age metallurgy.

Two recent studies have calculated the deviation between the average value of the tin in the patina and in the apparently healthy metal following the polishing of the patina in the pXRF analysis: in the case of the 41 samples analysed by Orfanou and Rehren (2015), the deviation was of the order of 55% more tin on the surface, reaching 74% in the case of the 14 samples compared by the *Iberian Peninsula Archaeometallurgy Project* (Rovira and Montero-Ruiz, 2018, p. 234).

In terms of the reliability of the surface analyses and the tendency for the enrichment of certain elements such as tin or arsenic, we must remain cautious if it is not possible to carry out any type of comparison with objects from the same archaeological site with a minimum amount of cleaning. But in certain cases, the tendency of tin or arsenic enrichment can be completely contrary, as we detect in the sword with five rivets from Tomb 429 at El Argar (Siret and Siret, 1890, tab. 68). Initially was analysed by the laboratory of M. Paul Claes, showing proportions of 7.58% Sn. Our pXRF analyses of different points of the surface revealed very pure copper without arsenic and without tin, except for a shot in the middle of the blade where 0.2% Sn was quantified. Faced with this contradiction, we decided to use the metal shaving extracted for the lead isotope analysis to confirm whether there was any tin in the metal. In the knowledge that they were not the optimum analytical conditions for a correct quantification, the shaving included in the small plastic tube was initially analysed with the Artax of the British Museum, giving a result of 5.4% Sn and 0.5% As. Subsequently, the analysis of those shavings under the Institute of History's scanning electron microscope (SEM-EDX) gave an average value of 7.9  $\pm$  0.3% Sn with remains of arsenic that cannot be quantified with precision but are less than 0.8%. This tin content is like that published by the Siret brothers and, at the same time, confirms that the bronze contains small quantities of arsenic, as the British Museum's analysis indicated. In this patina we detect the loss of tin and arsenic in the surface corrosion.

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### Table 3. Results of elemental composition analysis by p-XRF. Results in %wt. Nd = Not detected.

|                    |                |                 |       | Table 5. Result  |     | •   |      |     |      |      |      |     |          |     |     |
|--------------------|----------------|-----------------|-------|--|-----|-----|------|-----|------|------|------|-----|----------|-----|-----|
| Site               | ID             | Object          | Metal | Fe   | Со  | Ni  | Cu   | Zn  | As   | Ag   | Sn   | Sb  | Hg or Br | Pb  | Bi  |
| Campos             | 1889,0704.64   | Awl             | Cu    | 0.5  | nd  | nd  | 98.0 | nd  | 1.3  | nd   | nd   | nd  | 0.1      | nd  | 0.1 |
| El Argar           | 1889,0704.138  | Sheet           | Br    | 1.6  | nd  | nd  | 86.6 | 5.9 | nd   | nd   | 3.8  | 0.2 | nd       | 1.6 | nd  |
| El Argar           | 1889,0704.134  | Arrohead        | Cu    | 2.5  | nd  | nd  | 96.2 | nd  | 0.5  | nd   | nd   | nd  | 0.2      | 0.2 | nd  |
| El Argar           | 1889,0704.135  | Arrohead        | Cu    | 4.1  | nd  | nd  | 92.6 | 0.3 | 2.2  | nd   | nd   | nd  | 0.2      | 0.1 | nd  |
| El Argar           | 1889,0704.137  | Arrowhead       | Cu    | 1.3  | nd  | nd  | 95.9 | nd  | 2.5  | nd   | nd   | nd  | 0.2      | nd  | nd  |
| El Argar           | 1889,0704.136  | Arrohead        | Cu    | 1.7  | nd  | nd  | 95.0 | nd  | 2.9  | nd   | nd   | nd  | 0.2      | 0.1 | nd  |
| 3                  | •              | Anonead         |       | 1.7  |     |     | 93.0 |     | 6.1  |      |      |     |          |     |     |
| El Argar           | 1889,0704.133  |                 | Cu    |  | nd  | nd  |      | nd  |      | nd   | nd   | nd  | nd       | nd  | nd  |
| El Argar T51       | 1889,0704.170A | Spiral          | Br    | 0.1  | nd  | nd  | 90.4 | nd  | 0.4  | nd   | 8.8  | 0.1 | nd       | nd  | nd  |
| El Argar T51       | 1889,0704.171B | Frags. Spiral   | Br    | 0.3  | nd  | nd  | 87.6 | nd  | nd   | nd   | 10.7 | nd  | nd       | nd  | nd  |
| El Argar T51       | 1889,0704.170B | Spiral          | Cu    | 0.7  | nd  | nd  | 98.9 | nd  | nd   | nd   | 0.2  | nd  | nd       | nd  | nd  |
| El Argar T51       | 1889,0704.171  | Ring            | Cu    | 0.6  | nd  | nd  | 93.1 | nd  | 6.0  | nd   | nd   | nd  | 0.3      | nd  | nd  |
| El Argar T51       | 1889,0704.171  | Spiral          | Cu    | 0.4  | nd  | nd  | 96.9 | nd  | 2.6  | nd   | nd   | nd  | nd       | 0.1 | nd  |
| El Argar T429      | 1889,0704.156  | Ring            | Br    | 0.5  | nd  | nd  | 97.2 | nd  | nd   | nd   | 2.0  | nd  | nd       | 0.3 | nd  |
| El Argar T429      | 1889,0704.158  | Frags. Spiral   | Br    | 0.6  | nd  | nd  | 95.2 | nd  | nd   | nd   | 3.1  | nd  | nd       | nd  | nd  |
| El Argar T429      | 1889,0704.158B | Bracelet        | Br    | 0.3  | nd  | nd  | 96.2 | nd  | nd   | nd   | 3.6  | nd  | nd       | nd  | nd  |
| El Argar T429      | 1889,0704.158D | Bracelet        | Br    | 0.2  | nd  | nd  | 94.4 | nd  | nd   | nd   | 4.5  | nd  | nd       | nd  | nd  |
| El Argar T429      | 1889,0704.158C | Bracelet        | Br    | 0.5  | nd  | nd  | 91.7 | nd  | nd   | nd   | 6.6  | nd  | nd       | nd  | nd  |
| _                  | 1889,0704.154  |                 | Cu    | 0.4  |     |     | 99.2 |     |      |      |      | nd  |          | 0.1 |     |
| El Argar T429      |                | Sword           |       |  | nd  | nd  |      | nd  | nd   | nd   | nd   |     | nd       |     | nd  |
| El Argar T429      | 1889,0704.155  | Bracelet        | Cu    | 0.3  | nd  | nd  | 99.1 | nd  | nd   | nd   | 0.5  | nd  | nd       | nd  | nd  |
| El Argar T429      | 1889,0704.157  | Ring            | Cu    | 0.2  | nd  | nd  | 89.8 | nd  | 1.2  | nd   | nd   | nd  | nd       | nd  | nd  |
| El Argar T471      | 1889,0704.184  | Ring            | Ag    | 2.0  | nd  | nd  | 0.1  | nd  | nd   | 62.1 | nd   | nd  | 34.9     | 0.1 | 0.4 |
| El Argar T533      | 1889,0704.153  | Halberd 2R      | Cu    | 0.2  | nd  | nd  | 76.5 | nd  | 18.9 | nd   | nd   | nd  | nd       | nd  | nd  |
| El Argar T597      | 1889,0704.177  | Dagger          | Cu    | 1.0  | nd  | nd  | 88.8 | 0.7 | 5.2  | nd   | nd   | nd  | nd       | 2.4 | nd  |
| El Argar T626      | 1889,0704.185  | Ring            | Cu    | 1.4  | nd  | nd  | 97.8 | nd  | nd   | nd   | nd   | nd  | nd       | nd  | nd  |
| El Argar T649      | 1889,0704.186  | Ring            | Cu    | 0.6  | nd  | nd  | 94.2 | nd  | 5.1  | nd   | nd   | nd  | nd       | nd  | nd  |
| El Argar T694      | 1889,0704.179  | Dagger 4R       | Cu    | 4.5  | nd  | nd  | 92.7 | 0.3 | 1.8  | nd   | nd   | nd  | 0.3      | nd  | nd  |
| El Argar T703      | 1889,0704.148  | Axe             | Cu    | nd   | nd  | nd  | 99.1 | nd  | 0.8  | nd   | 0.1  | nd  | nd       | nd  | nd  |
| El Argar T703      | 1889,0704.149  | Dagger 2R       | Cu    | nd   | nd  | nd  | 98.3 | nd  | 1.1  | 0.6  | nd   | nd  | nd       | nd  | nd  |
| El Argar T711      | 1889,0704.163  | Dagger 2R       | Cu    | 1.8  | nd  | nd  | 95.3 | nd  | 2.7  | nd   | nd   | 0.1 | nd       | nd  | nd  |
|                    |                |                 |       | 0.3  | nd  |     | 77.9 |     | 21.7 | nd   |      | nd  |          |     |     |
| El Argar T716      | 1889,0704.180  | Dagger 4R       | Cu    |  |     | nd  |      | nd  |      |      | nd   |     | nd       | nd  | nd  |
| El Argar T718      | 1889,0704.187  | Ring            | Ag    | 2.8  | nd  | nd  | 0.1  | nd  | nd   | 62.1 | nd   | nd  | 35.4     | nd  | 0.4 |
| El Argar T746      | 1889,0704.145  | Spiral          | Ag    | 2.4  | nd  | nd  | 0.1  | nd  | nd   | 60.7 | nd   | nd  | 36.4     | nd  | 0.4 |
| El Argar T746      | 1889,0704.144  | Spiral          | Br    | 0.5  | nd  | nd  | 97.5 | nd  | nd   | nd   | 1.9  | nd  | nd       | nd  | nd  |
| El Argar T746      | 1889,0704.143  | Axe             | Cu    | nd   | nd  | nd  | 98.5 | nd  | 0.8  | 0.6  | 0.1  | nd  | nd       | nd  | nd  |
| El Argar T958      | 1889,0704.188  | Axe             | Cu    | nd   | nd  | nd  | 90.3 | nd  | 0.5  | nd   | 0.2  | nd  | nd       | 0.1 | nd  |
| El Oficio          | 1889,0704.206  | Saw             | Cu    | 2.1  | nd  | nd  | 94.5 | nd  | 2.7  | nd   | nd   | nd  | 0.3      | nd  | nd  |
| El Oficio T42      | 1889,0704.221  | Halberd 4R      | Cu    | 0.2  | nd  | nd  | 93.5 | nd  | 6.1  | nd   | nd   | nd  | nd       | 0.2 | nd  |
| El Oficio T42      | 1889,0704.222  | Dagger 5R       | Cu    | 0.5  | nd  | nd  | 82.7 | nd  | 16.7 | nd   | nd   | nd  | nd       | 0.1 | nd  |
| El Oficio T62      | 1889,0704.227  | Dagger 3R       | Br    | 0.2  | nd  | nd  | 94.1 | nd  | nd   | nd   | 1.9  | nd  | nd       | nd  | nd  |
| El Oficio T62      | 1889,0704.225  | Dagger 4R       | Br    | 0.3  | nd  | nd  | 93.3 | nd  | 0.1  | nd   | 3.4  | nd  | nd       | 0.3 | nd  |
| El Oficio T62      | 1889,0704.224  | Halberd Rivet   | Cu    | 0.1  | nd  |     | 99.0 |     | 0.6  |      |      |     | 0.3      |     |     |
|                    |                |                 |       |  |     | nd  |      | nd  |      | nd   | nd   | nd  |          | nd  | nd  |
| El Oficio T62      | 1889,0704.224  | Halberd         | Cu    | 0.3  | nd  | nd  | 89.9 | nd  | 9.7  | nd   | nd   | nd  | nd       | 0.1 | nd  |
| El Oficio T62      | 1889,0704.226  | Dagger 4R       | Cu    | 0.5  | nd  | nd  | 86.3 | nd  | 13.1 | nd   | nd   | nd  | nd       | 0.1 | nd  |
| El Oficio T158     | 1889,0704.233  | Spiral 6V       | Br    | 0.1  | nd  | nd  | 91.5 | nd  | 6.1  | nd   | 2.2  | nd  | nd       | 0.1 | nd  |
| El Oficio T158     | 1889,0704.231  | Awl             | Br    | 0.3  | nd  | 0.1 | 96.2 | nd  | 0.4  | nd   | 2.7  | 0.2 | nd       | 0.1 | nd  |
| El Oficio T158     | 1889,0704.233  | Spiral 6V       | Cu    | 1.5  | nd  | 0.1 | 94.0 | nd  | 2.6  | nd   | nd   | nd  | nd       | nd  | nd  |
| El Oficio T158     | 1889,0704.234  | Spiral 5V       | Cu    | 1.3  | nd  | nd  | 95.0 | nd  | 3.6  | nd   | nd   | nd  | nd       | 0.1 | nd  |
| El Oficio T158     | 1889,0704.230  | Dagger 2R       | Cu    | 0.3  | nd  | nd  | 95.2 | nd  | 4.5  | nd   | nd   | nd  | nd       | nd  | nd  |
| El Oficio T158     | 1889,0704.232  | Spiral          | Cu    | 0.3  | nd  | nd  | 94.2 | nd  | 4.5  | 0.4  | nd   | 0.5 | nd       | 0.1 | nd  |
| Lugarico Viejo     | 1889,0704.78   | Axe             | Cu    | 0.5  | nd  | nd  | 94.6 | nd  | 3.6  | nd   | nd   | 0.1 | 0.1      | 0.2 | 0.1 |
| Lugarico Viejo     | 1889,0704.77   | Dagger 2R       | Cu    | 1.1  | nd  | nd  | 92.2 | nd  | 6.6  | nd   | nd   | nd  | nd       | nd  | nd  |
| Zapata T11         | 1889,0704.87   | Dagger 3R       | Cu    | 0.7  | nd  | nd  | 96.2 | nd  | 3.0  | nd   | nd   | nd  | nd       | nd  | nd  |
|                    |                |                 |       |  |     |     |      |     |      |      |      |     |          |     |     |
| Zapata T11         | 1889,0704.87   | Dagger 3R Rivet | Ag    | 3.5  | nd  | nd  | 54.1 | 0.1 | 0.9  | 40.2 | 0.7  | nd  | 0.2      | nd  | nd  |
| Zapata T11         | 1889,0704.87   | Dagger 3R Rivet | Ag    | 0.4  | nd  | nd  | 42.5 | nd  | 0.8  | 56.3 | nd   | nd  | nd       | nd  | nd  |
| Site               | ID             | Object          | Metal | Fe   | Со  | Ni  | Cu   | Zn  | As   | Ag   | Sn   | Sb  | Hg or Br | Pb  | Bi  |
| Qurénima           | 1889,0704.69   | Bracelet        | Br    | 1.6  | nd  | nd  | 89.4 | nd  | nd   | nd   | 8.0  | 0.2 | nd       | 0.8 | nd  |
| Qurénima           | 1889,0704.68   | Bracelet        | Br    | 1.0  | nd  | nd  | 83.6 | nd  | nd   | nd   | 13.8 | nd  | nd       | nd  | nd  |
| Qurénima           | 1889,0704.71A  | Bead            | Br    | 0.4  | nd  | nd  | 80.2 | nd  | nd   | nd   | 15.0 | nd  | nd       | 2.9 | nd  |
|                    |                |                 |       |  |     |     |      |     |      |      |      |     |          |     |     |
| Qurénima           | 1889,0704.71B  | Bead            | Br    | 3.9  | nd  | 0.1 | 58.6 | 0.1 | nd   | nd   | 29.3 | 0.1 | nd       | 5.2 | nd  |
| Qurénima           | 1889,0704.70   | Ring            | Br    | 2.9  | nd  | nd  | 63.9 | 0.2 | nd   | nd   | 29.7 | nd  | nd       | 0.1 | nd  |
| <b>c'</b> .        | ID             | Object          | Metal | Fe   | Со  | Ni  | Cu   | Zn  | As   | Ag   | Sn   | Sb  | Hg or Br | Pb  | Bi  |
| Site               |                |                 |       | and the second | 1   |     | 1    |     |      | 1    | 1    |     |          |     |     |
| Site<br>Parazuelos | 1889,0704.39   | Slag            | Cu    | 57.2   | 0.9 | 1.0 | 36.8 | 1.6 | 2.0  | nd   | nd   | nd  | nd       | nd  | nd  |

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ISSN: 1133-4525 ISSN-e: 2255-3924 Table 4. Comparison of surface analyses on patina by pXRF (this paper) and metal analyses by atomic emissionspectrometry (AES) by Otto and Witter (1952). Bld = Below limit of detection; tr = Traces.

| Site           | ID            | Туре    | Ni    | Cu   | As   | Ag   | Sn  | Sb   | Bi   | Reference           |
|----------------|---------------|---------|-------|------|------|------|-----|------|------|---------------------|
| Lugarico Viejo | 1889,0704.78  | Axe     | 0.034 | 94.6 | 3.6  | bld  | bld | 0.08 | 0.08 | This paper          |
|                |               |         | tr    | 98.4 | 1.2  | 0.14 |     | tr   | 0.05 | Otto & Witter, 1952 |
| El Argar T533  | 1889,0704.153 | Halberd | bld   | 76.5 | 18.9 | bld  | bld | bld  | bld  | This paper          |
|                |               | 2R      |       | 96.5 | 3.3  | tr   |     |      | 0.01 | Otto & Witter, 1952 |
| El Oficio T62  | 1889,0704.224 | Halberd | bld   | 91.2 | 8.1  | bld  | bld | bld  | bld  | This paper          |
|                |               |         |       | 96.8 | 3    | tr   |     |      | tr   | Otto & Witter, 1952 |

A similar case, although less extreme, was described in the study of the sword from Santa Ana (Herrerías), also part of the Siret collection in the MAN. The exterior greenish patina offered an Sn content of between 0.42 and 0.99% Sn, while the metallic nucleus ranged between 7.52 and 8.51% Sn and the Siret analysis quantified as 7.87% Sn (Montero-Ruiz *et al.,* 2016, p. 75, tab. 1).

Taking into account these circumstances, we have to be cautious when assessing the percentages of some of the objects in the Siret collection, especially in the combination of tin and arsenic in the same piece.

Copper and tin bronze alloys with high quantities of arsenic are not exceptional in this metallurgy (Montero-Ruiz, 1994, pp. 261-262) and they have also been detected in Spiral 1889,0704.233 from Tomb 158 at El Oficio, a bronze with 2.9% Sn and 6.1% As. However, in five of the argaric bronzes analysed a copper-tin alloy with no significant presence of arsenic is observed. The presence/absence of arsenic may indicate a change in the copper resources mined for the manufacture of these bronzes as, during the Chalco-lithic and the Bronze Age, pure copper objects were an exception in this geographic area (Murillo-Barroso *et al.*, 2020; Perucchetti *et al.*, 2020). In this respect, of particular significance are the high levels of arsenic and cobalt in the mineral sample from Parazuelos, with nearly 9% and 1.4% respectively, and the presence of nickel, zinc and arsenic in the piece of slag from the same archaeological site. These impurities are characteristic of the mineralisations in the area, with cobalt, nickel and arsenic being common impurities in the ore from Cerro Minado, and zinc is recurrent in the ore from Pinar de Bédar (Murillo-Barroso *et al.*, 2019).

It is worth briefly mentioning the axe from T958 at El Argar (fig. 3). This urn tomb, which contained two burials, is not included in the Siret brothers' 1887 publication, as it was excavated in July of that year. Unlike the rest of pieces, the axe appears with the metal visible, without patina, showing a surface porous structure. We do not have the Sirets' detailed sketch or any of their comments, as we do for their other published finds; we only have the descriptive sketch of the find made by their foreman Pedro Flores (field diary Ref. 1944/45/FD00590). This surface could be consequence of a chemical cleaning before or after its entry in the museum. The XRF analysis contains 0.5% As and 0.2% Sn.

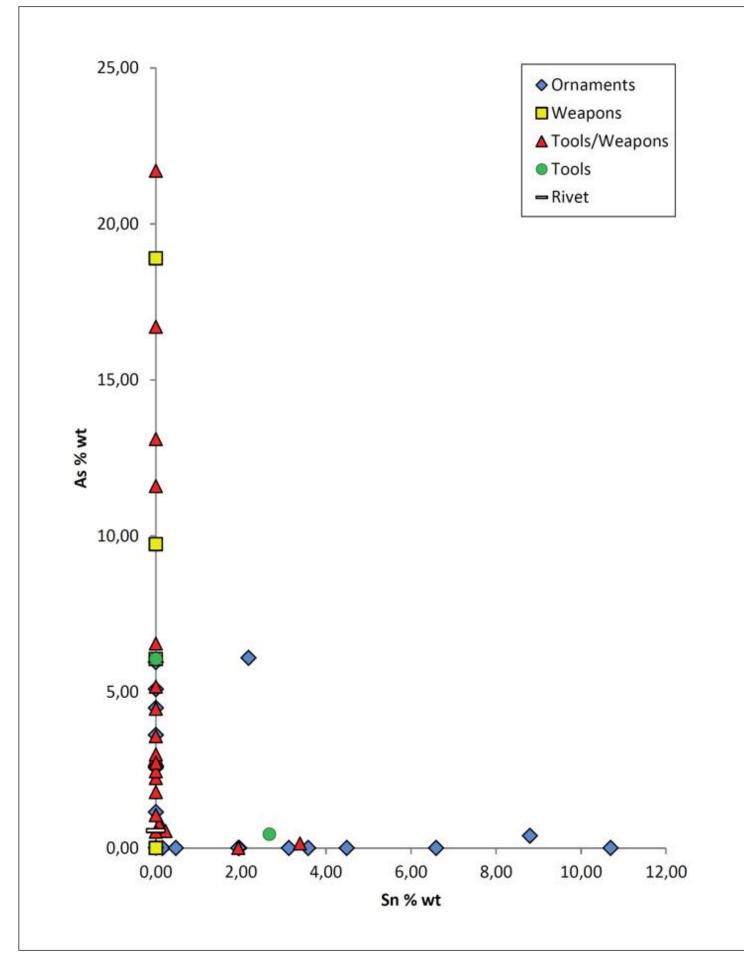
The piece of scrap metal sheet 1889,0704.138 from El Argar, containing c. 6% Zn, c. 4% Sn and c. 2% Pb has a composition that is not compatible with a prehistoric date. Excavations in 1991 (Schubart *et al.*, 2014) documented an Islamic settlement that was dated to the between the 8<sup>th</sup> and the 10<sup>th</sup> centuries and that may account for the presence of intrusive metalwork at the site (Menasanch, 2000). The Siret brothers (1890, p. 159) also provided information on the presence of objects from later periods in the description of the finds from El Argar and they noted that some needles recovered from the site were made of brass.

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ISSN: 1133-4525 ISSN-e: 2255-3924 It is of note that the majority of the 18 bronze objects analysed are adornments and only three are functional objects (two daggers and an awl). If we limit ourselves to the objects from the Argaric period, only 27% of the copper objects are adornments, compared to 73% of the bronze pieces (fig. 4). This series of analyses confirms something that has been proposed in other studies (Montero-Ruiz, 1994; Lull *et al.*, 2010; Aranda *et al.*, 2012; Montero-Ruiz *et al.*, 2019): the bronze alloy preference for the manufacture of adornments. The potential functional improvement of bronze over copper or arsenicated copper in terms of the greater durability and efficiency of the bronze objects does not appear to have been one of the incentives for the development of that alloy, which has a preferentially ornamental use.



Figure 3. Axe from T958 at El Argar and detail of its surface. Source: MMB.



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**Figure 4.** Comparison of metals typology and their % in As or Sn. Note how most of the ornaments present high levels of Sn while most of the tools and/or weapons present high levels of As.

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#### 4.2. Lead isotopes and metal provenance

With respect to the lead isotope analysis (tab. 5), only samples of Argaric chronology were selected: 5 silver and 19 copper-based objects. The bulk of the samples are distributed in the range 18.10 to 18.50 in the Pb<sup>206/</sup>Pb<sup>204</sup> ratio and 15.62 to 15.68 in the ratio in the Pb<sup>207/</sup>Pb<sup>204</sup>, the most extreme values are the axe from Lugarico Viejo (17.82 in the Pb<sup>206/</sup>Pb<sup>204</sup>) and some tin bronzes from El Argar and El Oficio with highest values in the Pb<sup>206/</sup>Pb<sup>204</sup> (> 15.70) (fig. 5). We should emphasise the greater similarity of the silver items, which form a cluster between 18.21 and 18.29 in the Pb<sup>206/</sup>Pb<sup>204</sup> ratio (fig. 5), and three of the four bronzes analysed, with values between 18.41 and 18.55 in the same ratio. The sole exception is the bronze spiral 1889,0407.144 from Tomb 746 at El Argar, with a value of 18.31, that places close to the five pieces of silver. A comprehensive study was made of Argaric silver in Murillo-Barroso (2013) and Bartelheim *et al.* (2012), and a new paper has been published summarising Bronze Age silver on the Iberian Peninsula that incorporates these new data (Montero-Ruiz *et al.*, 2019); therefore our comments here will focus on the copper-based metals with the aim of clarifying some of the questions regarding Argaric metallurgical production that have been debated for a long time.

| ID             | Site              | Туре        | Context | Alloy | Pb <sup>208/</sup> Pb <sup>206</sup> | Pb <sup>207/</sup> Pb <sup>206</sup> | Pb <sup>206/</sup> Pb <sup>204</sup> | Pb <sup>207/</sup> Pb <sup>204</sup> | Pb <sup>208/</sup> Pb <sup>204</sup> |
|----------------|-------------------|-------------|---------|-------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|
| 1889,0704.135  | El Argar          | Arrowhead   |         | Cu    | 2.09953                              | 0.85347                              | 18.3615                              | 15.6710                              | 38.5504                              |
| 1889,0704.171B | El Argar          | Ring frags. | T51     | Br    | 2.09467                              | 0.85135                              | 18.4369                              | 15.6964                              | 38.6193                              |
| 1889,0704.154  | El Argar          | Sword       | T429    | Cu    | 2.09210                              | 0.84986                              | 18.4443                              | 15.6750                              | 38.5873                              |
| 1889,0704.155  | El Argar          | Bracelet    | T429    | Cu    | 2.09579                              | 0.85065                              | 18.4171                              | 15.6666                              | 38.5984                              |
| 1889,0704.184  | El Argar          | Ring        | T471    | Ag    | 2.10476                              | 0.85646                              | 18.2459                              | 15.6270                              | 38.4034                              |
| 1889,0704.153  | El Argar          | Halberd 2R  | T533    | CuAs  | 2.10264                              | 0.85836                              | 18.2299                              | 15.6478                              | 38.3311                              |
| 1889,0704.149  | El Argar          | Dagger      | T703    | Cu    | 2.10747                              | 0.86293                              | 18.1047                              | 15.6231                              | 38.1552                              |
| 1889,0704.148  | El Argar          | Axe         | T703    | Cu    | 2.09754                              | 0.85269                              | 18.3645                              | 15.6592                              | 38.5203                              |
| 1889,0704.180  | El Argar          | Dagger 4R   | T716    | Cu    | 2.09288                              | 0.85163                              | 18.3890                              | 15.6606                              | 38.4860                              |
| 1889,0704.187  | El Argar          | Ring        | T718    | Ag    | 2.10780                              | 0.85652                              | 18.2537                              | 15.6347                              | 38.4753                              |
| 1889,0704.143  | El Argar          | Axe         | T746    | Cu    | 2.10347                              | 0.85541                              | 18.2868                              | 15.6427                              | 38.4658                              |
| 1889,0704.144  | El Argar          | Spiral      | T746    | Br    | 2.09710                              | 0.85451                              | 18.3124                              | 15.6482                              | 38.4031                              |
| 1889,0704.145  | El Argar          | Spiral      | T746    | Ag    | 2.09565                              | 0.85466                              | 18.2951                              | 15.6362                              | 38.3403                              |
| 1889,0704.188  | El Argar          | Axe         | T958    | Cu    | 2.10269                              | 0.85439                              | 18.3442                              | 15.6730                              | 38.5721                              |
| 1889,0704.221  | El Oficio         | Halberd 4R  | T42     | CuAs  | 2.09892                              | 0.85117                              | 18.3798                              | 15.6444                              | 38.5778                              |
| 1889,0704.222  | El Oficio         | Dagger 5R   | T42     | Cu    | 2.08418                              | 0.84225                              | 18.6651                              | 15.7206                              | 38.9014                              |
| 1889,0704.224  | El Oficio         | Halberd     | T62     | CuAs  | 2.10721                              | 0.86267                              | 18.1347                              | 15.6443                              | 38.2137                              |
| 1889,0704.225  | El Oficio         | Dagger 4R   | T62     | Br    | 2.08674                              | 0.84642                              | 18.5554                              | 15.7057                              | 38.7203                              |
| 1889,0704.226  | El Oficio         | Dagger 4R   | T62     | CuAs  | 2.10002                              | 0.85615                              | 18.2788                              | 15.6494                              | 38.3858                              |
| 1889,0704.227  | El Oficio         | Dagger 3R   | T62     | Br    | 2.09724                              | 0.85338                              | 18.4135                              | 15.7138                              | 38.6175                              |
| 1889,0704.228  | El Oficio         | Bracelet    | T62     | Ag    | 2.10685                              | 0.85831                              | 18.2188                              | 15.6374                              | 38.3842                              |
| 1889,0704.229  | El Oficio         | Ring        | T62     | Ag    | 2.10542                              | 0.85723                              | 18.2488                              | 15.6434                              | 38.4213                              |
| 1889,0704.230  | El Oficio         | Dagger 2R   | T158    | CuAs  | 2.09523                              | 0.85006                              | 18.3756                              | 15.6203                              | 38.5011                              |
| 1889,0704.78   | Lugarico<br>Viejo | Axe         | T10     | CuAs  | 2.12455                              | 0.87566                              | 17.8175                              | 15.6021                              | 37.8541                              |

 Table 5. Results of Lead Isotope Analysis by MC-ICP-MS.

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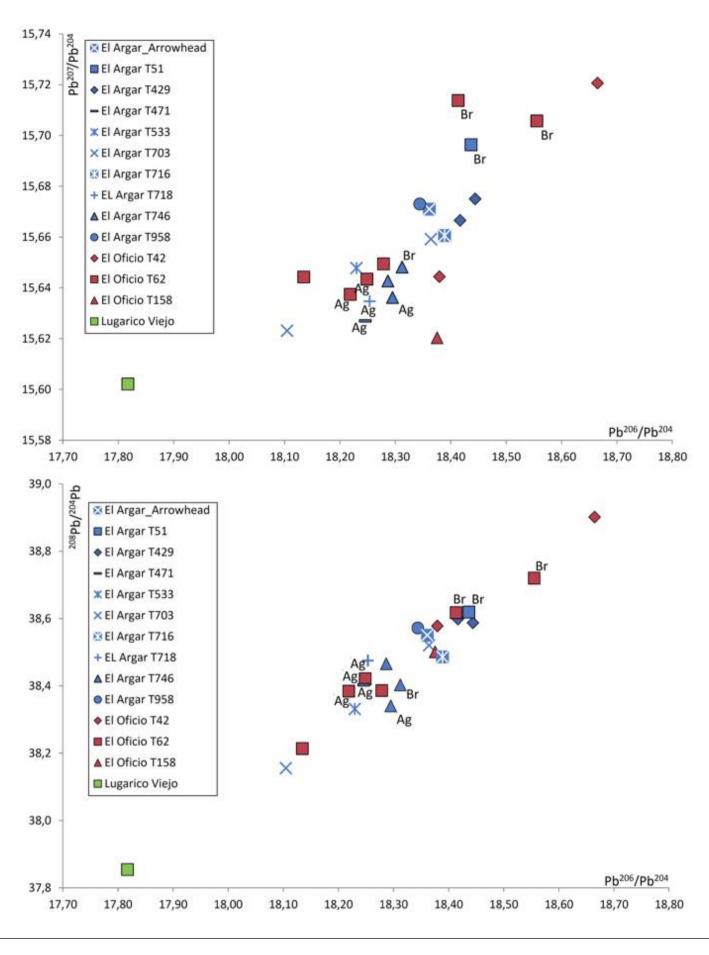


Figure 5. Lead Isotope ratios of objects analysed.

### 4.2.1. Centralised model of supply from the Alto Guadalquivir

The concept of a centralised supply stemming from the Alto Guadalquivir was put forward by Lull and colleagues (for example Lull and Risch 1996 on the Argaric State, or Lull *et al.*, 2010) as soon as the first analyses of lead isotopes were available in the Oxford laboratory in the framework of the *Gatas Project* (Stos-Gale *et al.*, 1999). The interpretation suggested the Linares area as the main supply zone, without ruling out other more distant possible areas such as the southwest of the Iberian Peninsula at this first moment

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ISSN: 1133-4525 ISSN-e: 2255-3924 (Stos-Gale *et al.*, 1999). The idea was reinforced by the Peñalosa excavations (Contreras 2000; Moreno Onorato *et al.*, 2010) that showed a settlement with intensive metallurgical activity and with an exceptional record, due to its quantity (Moreno Onorato *et al.*, 2017). This hypothesis relies on the assumption of a supraregional organisation of metallurgical production (Lull *et al.*, 2010; Lull *et al.*, 2011) with a high technical and social division. Although it provides for the possibility that some local mineral from Almería and Murcia may have been used, it emphasises the weight of Linares as the main mining region during the Early Bronze Age (Lull *et al.*, 2010, p. 333) and especially the control of metal production by a social class.

The supraregional model was questioned from the outset (Montero-Ruiz, 1999), because it was not supported by the published lead isotope analyses. On the contrary, it was deduced that the metal could come from different provenances and that those from Linares were not in the majority (Montero-Ruiz and Murillo-Barroso, 2010), although it was detected in settlements far away, such as Fuente Álamo. An alternative model highlighting regional production, domestic and low efficiency technology was proposed (Montero-Ruiz, 1999; Murillo-Barroso *et al.*, 2015; Murillo-Barroso *et al.*, 2020).

This debate was based on a small number of analyses of metal objects (mainly from the provinces of Murcia and Almería) and limited geological reference information (Stos-Gale *et al.,* 1995). New research about metals from settlements in Granada province allowed us to confirm the diversity of origins and the complexity of their study due to the lack of a geological reference database (Murillo-Barroso *et al.,* 2015).

The geological reference information has increased substantially, especially from the copper mineralisations of the Betic Internal Zones that runs from the province of Málaga to Murcia (Murillo-Barroso *et al.,* 2019; Rodríguez Vinceiro *et al.,* 2018; Renzi *et al.,* 2016; Brandherm *et al.,* 2022; Murillo-Barroso *et al.,* in press), and from the NE area of the iberian peninsula (Canals and Cardellach, 1997; Montero-Ruiz, 2017; Montero-Ruiz, 2018).

With the study of the British Museum's Siret collection we are significantly adding data to the available information and we can make somewhat more detailed comparisons to determine whether the supply of metal in this coastal zone of the Cuenca de Vera depended on the metal obtained in the Alto Guadalquivir. In this study we also include the five samples from the archaeological site of Fuente Álamo and three from El Argar published by Stos-Gale *et al.* (1999), although we must take into account the larger analytical error of these samples analysed by TIMS (e.g. Murillo-Barroso *et al.*, 2019). More recently 3 new analyses from El Argar in the Siret Collection at the Pigorini Museum have been published (Müller-Kissing, 2022).

Table 5 compiled the metals from the archaeological sites of El Argar, Fuente Álamo and El Oficio differentiated by their alloy (silver, copper or bronze). To avoid confusion, we have not included the analyses from the archaeological site of Gatas that, although available in OXALID, they are not classified by the chronological phases of their excavators. To contrast the provenance from the Alto Guadalquivir copper resources we use the LIA from the mines in the Linares district (Santos Zaldegui *et al.*, 2004; García de Madinabeitia *et al.*, 2021) and the published copper ores and metals from the settlement of Peñalosa located in the same district (Hunt *et al.*, 2011).

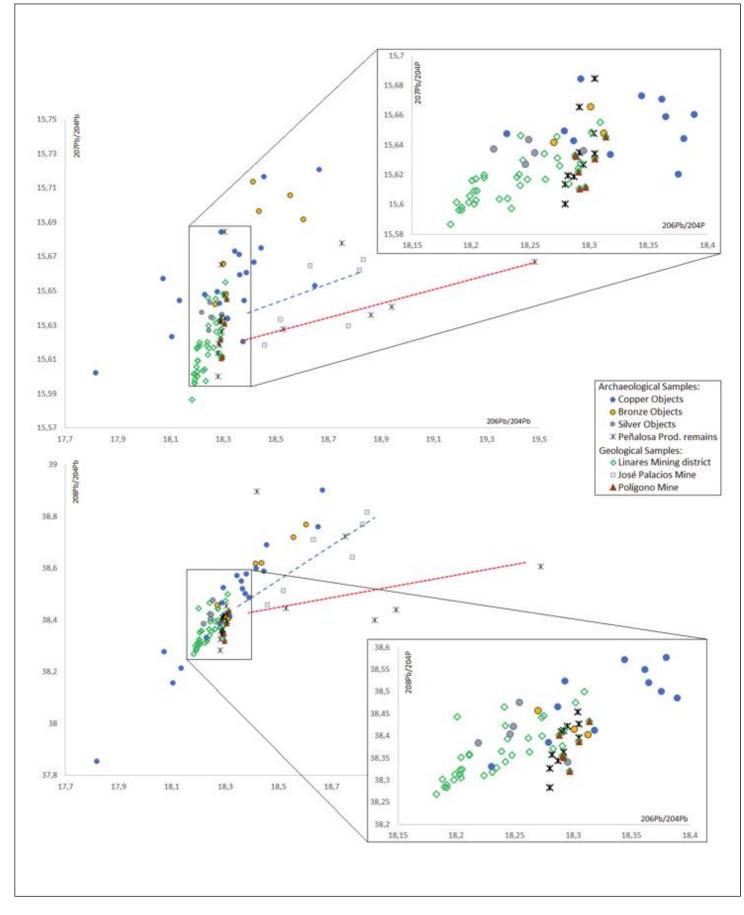
Peñalosa shows two clear tendencies (fig. 6). On the one hand, a group of samples align vertically in fig. 6a with similar values on the Pb<sup>206/</sup>Pb<sup>204</sup> ratio but changing in the Pb<sup>207/</sup>Pb<sup>204</sup>. The samples from Peñalosa are distributed only in a limited zone of the whole isotopic field of the district of Linares, and match with the Polígono mine samples. The second trend covers the distribution towards the right of the graph with radiogenic values

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ISSN: 1133-4525 ISSN-e: 2255-3924 that exceed of 19 in the <sup>206</sup>Pb/<sup>204</sup>Pb ratio and reach as high as 22.8 (off the graph). These results are not clearly linked with the copper ores from the José Martín Palacios mine, for whose exploitation we have archaeological proofs and radiocarbon dates (2205-2030 cal BC) (Arboledas *et al.*, 2015), with an earlier chronology than the main Bronze Age phases of Peñalosa. This second group of ores found in the settlement show at least a second copper resource but with a completely different isotopic signature than the Linares and other close mining districts in the Central Iberian Zone, such as Alcudia Valley or Pedroches. The data from José Martín Palacios and Polígono mines, and from the unknown resource could demarcate a mixing area if metal from both types of minerals were mixed. However, no object is clearly located in this mixing region.

Of the 30 objects compared, six of them clearly match to the minerals processed in Peñalosa from the Polígono mine: the copper axes from T479 and T673 at El Argar, a bronze spiral from T746 and a bronze dagger from El Argar (AM20), a copper dagger from Fuente Álamo T58 (FA-15), finally a four rivets copper dagger from T62 at El Oficio (BM226) could be also linked to the Polígono mine. The copper halberd/dagger (FA-12) and a bronze awl (FA 74) from Fuente Álamo could be related with other mines in the Linares district, but a copper axe (nº 48 Pigorini Museum) and a copper halberd from T533 at El Argar would not have a clear provenance from this mining district, but they could come from mines in the Central Iberian zone.

We could quantify this metal supply from Peñalosa to the Cuenca de Vera in the 20% and up to 26% if we consider the whole Linares mining district, with objects of both copper and bronze. Similar figures have also been detected in other regions (Brandherm *et al.*, 2022).



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**Figure 6.** Lead isotope values of copper, bronze and silver objects analysed in comparison to Peñalosa metallurgical remains, the Linares mining district and José Martín Palacios mine.

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#### 4.2.2. Diversity of origins in the same set of grave goods and the same archaeological site

Having corroborated that the Alto Guadalquivir is only one of the main areas of metal provenance, it is important to consider whether this is a distorting effect caused by the available sample. An indirect way of assessing this is to compare the data from a single set of grave goods. Pieces with similar isotopic signatures would indicate homogeneity in the supply, while the opposite situation would reinforce the idea of a supply from different origins and therefore outside any regulated control.

The available sample is small but can begin to provide some information. From T429 at El Argar we only have two pieces (a sword and a bracelet) with LIA, although the grave goods are rounded off with other small adornments such as rings or spirals that have not been analysed. The only inventoried items from T703 at El Argar in the British Museum are an axe and a dagger, but not the two adornments that appear in the sketch by Siret and Siret (1887); all the grave goods from T746 at El Argar have been analysed: a bronze dagger and spiral and another silver spiral. From double tomb T42 at El Oficio both the halberd and the dagger, which belonged to different individuals according to Pedro Flores' plan (fig. 7), have been analysed. Finally, although it is a double tomb, at T62 in El Oficio, thanks to Pedro Flores' plans, we know the layout of the four copper-based pieces that link, on the one hand, the halberd and the bronze dagger with four rivets (BM 224 and BM 225), and on the other, the copper dagger with four rivets and the bronze dagger with three rivets (BM226 and BM227).

In figure 8 we can see that the items from the same tomb are separated by some distance from each other. Regardless of the specific geographic identification of the origin, we could assume that they were made with metal from different copper ores. Only in T746 and T429 at El Argar is there certain proximity between the values. The axe and the spiral from T746 can be linked to Linares and the Polígono mine and clearly have a close origin. The sword and the bracelet from T429 could also have the same origin, although we need a detailed comparison, as they are in a zone in which various mineralisations overlap.

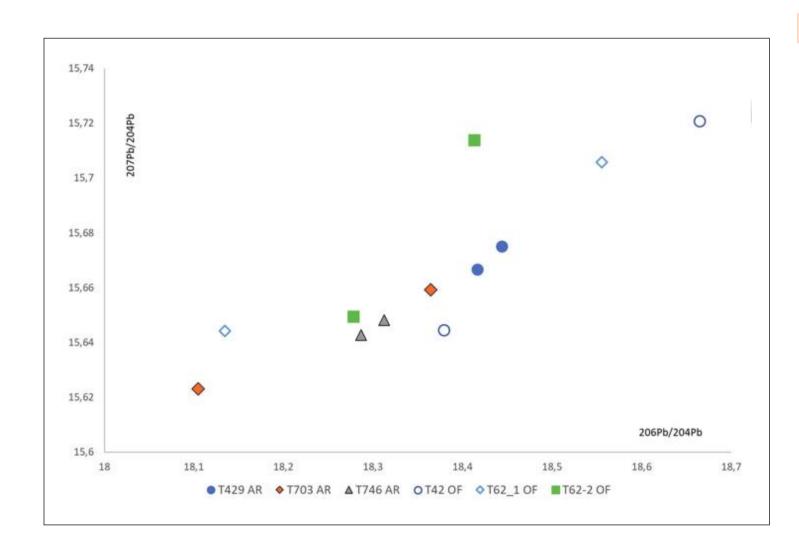
The objects from T42 at El Oficio belong to different individuals and the metal has a different origin. Assuming some degree of family or kindship relationship between the occupants of the tomb, the result indicates that the same metal sources were not maintained from generation to generation. If to this picture we add the case of T21 and T18 at El Cerro de la Encina, in which the metal also presented a diversity of origins (Murillo-Barroso et al., 2015), we detect a tendency towards the lack of a predominant or exclusive area supplying any one individual, or even the same family, and even less so in an archaeological site. We were already aware of this situation in Fuente Álamo (Montero-Ruiz and Murillo-Barroso, 2010, 48) which has at least four different origins. El Argar and El Oficio also demonstrate that diversity; all three sites share metal from Linares and in all three we find metal linked to the coastal areas of Almería and Murcia (which includes Cerro Minado) that are represented in values of >18.6 <sup>207</sup>Pb/<sup>204</sup>Pb. In figure 9, however, we detect a different distribution for each of the archaeological sites, despite their geographic proximity. Particularly striking is the distribution at El Oficio into two clearly separated groups, while the (more numerous) finds from El Argar present a more continuous distribution, although there are also pieces with extreme values.

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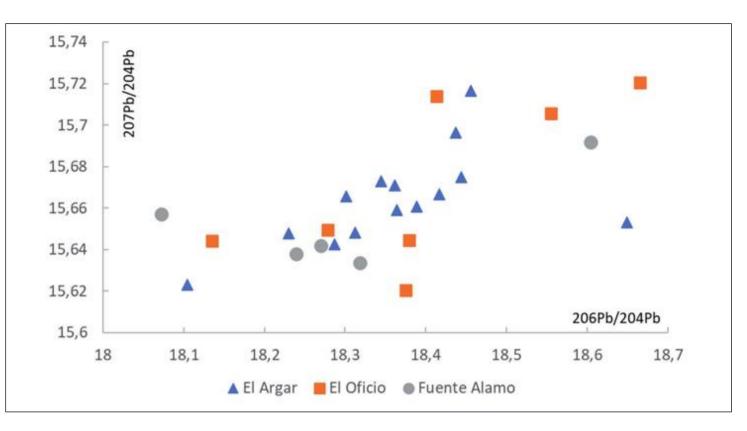
Figure 7. Sketch drawing of double tomb T42 at El Oficio according to Pedro Flores.



**Figure 8.** Lead isotope values of metal objects from tombs 429, 703 and 746 at El Argar, and tombs 42, and 62 at El Oficio. Note how objects from the same grave good differ from each other.



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**Figure 9.** Lead isotope ratios of objects at El Argar, El Oficio and Fuente Álamo. Note the different tendencies in the three of them.

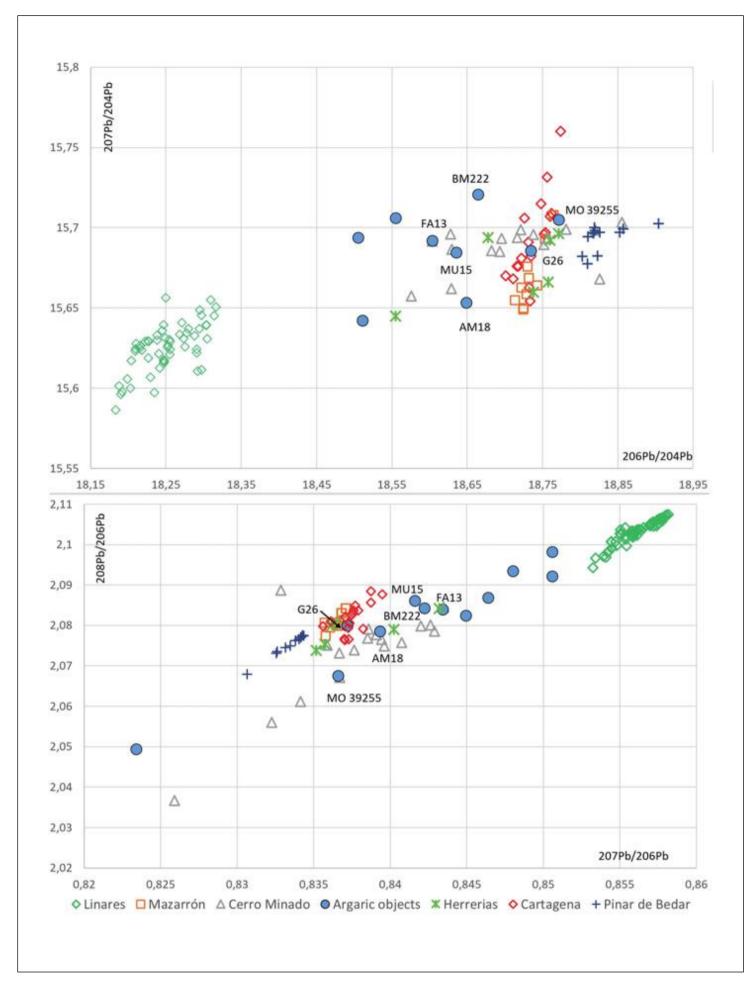
#### 4.2.3. The copper minerals in the Cuenca de Vera

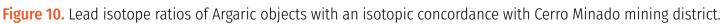
The supply of the copper minerals available in the Cuenca de Vera is another aspect that should be discussed in a little more detail within a panorama of diversity. C14 dates (Delgado-Raak et al., 2014) and lead isotope analysis (Murillo-Barroso et al., 2020) confirm the exploitation of Cerro Minado during the Copper Age, although Lull et al. (2010) suggested that the mine was not worked during that stage of the Bronze Age. LIA only identify three pieces with that origin (fig. 10): an axe from T68 at Fuente Álamo (FA-13), the sword with five rivets from El Argar (AM18) and a dagger with five rivets from T42 at El Oficio (BM-222). Figure 10 shows the best discrimination between Cerro Minado and the minerals from nearby zones, such as Sierra Almagrera, Mazarrón and Cartagena, and includes for comparison the awl from Gatas (M2) that was identified from the outset as originating in the Mazarrón-Cartagena area (Stos-Gale et al., 1999). This proportion is quite low (11%) but should be taken into account when considering the factors that impact on the production and circulation of metal. In this respect, we can mention another two objects found in distant places that were probably manufactured with metal from Cerro Minado: the bracelet from T21 at El Cerro de la Encina (MO 39255) in Granada province (Murillo-Barroso et al., 2015) and a dagger with three rivets from La Bastida (MU15).

There are two factors that could be obscuring production with local metal. The first could be that metal as a prestige element circulated outside its production area and we would expect to find more items with this signature at archaeological sites outside the Cuenca de Vera; the second would be that this circulation of prestige goods that were deposited in the tombs due to their identifying value did not affect other metals with a more instrumental use, such as saws, arrowheads or chisels that were not included with grave goods and that would more reliably reflect the local metal. For the time being the studies have focused on grave goods and therefore we do not know whether this hypothesis is credible. We only have an arrowhead from the settlement of El Argar with lead isotope studies (BM-135) and they do not coincide with the minerals from the Cuenca de Vera or the mines of Murcia but could match other betic resources in Almería.

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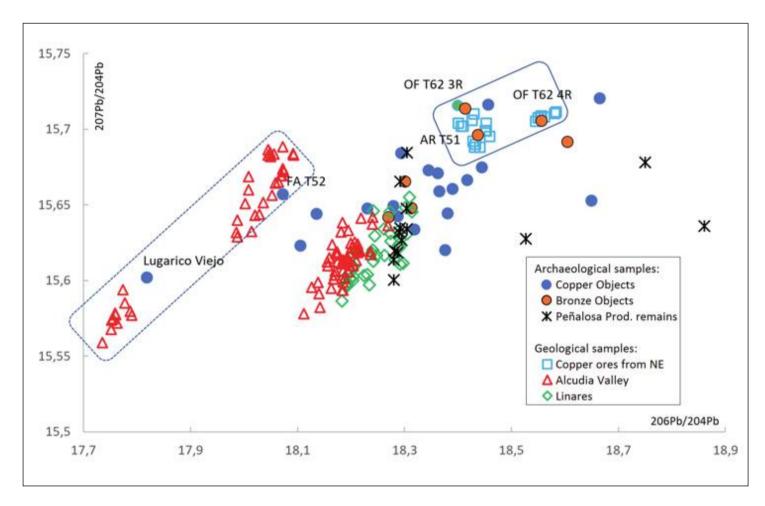
### 4.2.4. Other resources

Finally, we would like to comment on the items that show values lower than 18.15 (<sup>206</sup>Pb/<sup>204</sup>Pb). Those metals are associated with older mineralisations (>400 ma), including the Alcudia Valley, an area that could justify the origin of both the axe from Lugarico Viejo and the dagger in T52 at Fuente Álamo (fig. 11).

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ISSN: 1133-4525 ISSN-e: 2255-3924 Moreover, some of the bronzes without arsenic, such as those from T62 at El Oficio or the spiral from T51 at El Argar, are placed in the distribution of minerals from the northeastern Iberian Peninsula (the Catalan coastal mountain ranges and the Pyrenees) (fig. 11). A detailed study of these early tin bronzes was developed in Montero-Ruiz y Murillo-Barroso (2022). For the time being, we can neither link to data from the Iberian Peninsula the values of the halberd from T62 at El Oficio, nor those of the dagger from T703 at El Argar.

These items attest the complexity of the study of origins and the change of focus needed to understand the metallurgical production in El Argar.



**Figure 11.** Lead isotope ratios of Argaric objects with an isotopic concordance with other mineralisations outside the Argaric territory.

#### 5. CONCLUSIONS

With the results published in this study, not only have we substantially expanded the analytical base of the Argaric finds in the province of Almería, but we have also made possible a new concept of Argaric metallurgy. This study therefore highlights the importance of the Siret collection and the need to study it in greater depth, given that the quality of its record and the volume of finds it encompasses still today offers the potential to open up new lines of research and to confirm or refute hypotheses already put forward.

The data contributed reflect the multiplicity of resources in use and the complexity of the exchange networks that would have been involved in the supply of metal during the Bronze Age. The ores from Linares and its surrounding area were not in the majority in the Argaric area and therefore it seems we cannot infer dependency between the archaeological sites on the coasts of Almería and Murcia and those of the interior and the Linares

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ISSN: 1133-4525 ISSN-e: 2255-3924 area (e.g. Peñalosa) for the supply of metal. Nevertheless, the contribution of ore from the coastal zones of Almería and Murcia, where we attest a mining-metallurgical tradition from the Chalcolithic on, is also limited. Pending a continuing expansion of the geological information database that will allow more effective comparisons, especially for the pieces that still cannot be associated with any known mineralisation on the Iberian Peninsula, this study brings to light the existence of a significant number of metallic objects probably manufactured with mineral resources from outside the Argaric area, despite there being abundant mineral resources. The incidence of exogenous metal in quantities considerably larger than traditionally thought, requires us to re-evaluate the character of Argaric society and its relationship with the dynamics of metallurgical supply and production.

In the case of the bronzes, it appears evident that the added value of a scarce resource such as tin is an aspect to be explored in the social valorisation of this alloy and its majority use in items of ostentation and personal adornment. In an area with abundant copper resources, a long mining and metallurgical tradition and the technical and technological knowledge needed to manufacture metal objects, it is possible that the value of this significant number of foreign objects may have lain more in the social relations established with the groups with whom the metal was traded than in the in the intrinsic value of the metal objects per se.

It will be necessary to better define the possible origins of those pieces that we currently define as exogenous based on geological criteria and investigate whether there are anomalies in the copper mineralisation of southeastern Spain that could lead to a change in that evaluation. In any case, whether they are exogenous or local, what can be confirmed is the diversity of mineral resources that were in circulation and that could allow us to evaluate the possible exchange routes between the different Argaric territories. An additional avenue that requires further research is the presence and incidence of metal of Argaric origin in other areas of the Iberian Peninsula. This would enable an assessment of the role these exogenous interchanges played in the development of the Argaric society, as well as in the value placed on metal by those societies.

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### Authors' contributions

- Conception and design: IMR, MMB.
- Data analysis and interpretation: IMR, AL, NW, DH, MMB.
- Paper writing: IMR, MMB.
- Paper critical review: IMR, AL, NW, DH, MMB.
- Paper final approval: IMR, AL, NW, DH, MMB.
- Administrative, technical, and logistic support: NW, DH.
- Graphic design: IMR, MMB.
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