



## Non-ferrous metallurgy from the Phoenician site of La Fonteta (Alicante, Spain): a study of provenance

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### ABSTRACT

The lead isotope composition of 22 samples from the excavation of the Phoenician site of La Fonteta (Guardamar del Segura, Alicante, Spain) has been analysed in a preliminary study of their provenance. These materials span chronologically from the first half of the 8th century BC to the middle of the 6th century BC.

The samples have been selected to include materials used in lead and silver production, as well as in copper-based metallurgy. Therefore, lead droplets, galena nodules and fragments of litharge have been analysed, together with a fragment of a copper ingot, an object, a melting waste composed of a Cu–Pb alloy, and two fragments of a material that we have provisionally labelled ‘Pb–Cu cupellation debris’.

All these materials have been initially analysed by X-Ray Fluorescence-Spectrometry to identify their bulk compositions. Some of them have been also analysed by SEM–EDX and ICP–OES. Subsequently, lead isotope analyses (LIA) have been performed using Thermal Ionization Mass Spectrometry (TIMS). The new LIA data obtained has been compared with published lead isotope data from the Mediterranean area. The identification of different groups suggests a significant complexity of the internal and external trade routes during the Orientalising period. For this reason, the provenance study of these materials is especially important in shedding light on the commercial dynamics that the Phoenicians established with native people to control the raw materials and to commercialise finished products.

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### 1. Introduction

The Phoenician settlement of La Fonteta is situated in the Natural Park of the sand dunes of Guardamar, on the right riverside of the Segura estuary, 28 km south of Alicante (Fig. 1). This site – built on the coast, next to a river – had an unquestionable geo-strategic position that permitted control over trade and access to mining resources through connections established with native communities. Proper commercial axes between the hinterland and the peninsular coastal area were established, taking advantage of the settlement’s position, by exploiting inland wealth and establishing wider commercial links by sea.

The habitat is surrounded by an impressive defence system whose excavation permitted a basic chronological distinction of the habitation phases of the site. Its chronology spans approximately from the first half of the 8th century to the middle of the 6th

century BC. The period prior to the erection of the walls – called Archaic Fonteta (AF), from its beginning to the third quarter of the 7th century BC, phases I–III – has been clearly distinguished from the later one, which corresponds to Recent Fonteta (RF) and spans from the last quarter of the 7th century to the middle of the 6th century BC, phases IV–VII. The last period of occupation of the site corresponds to the destruction and collapse of the walls and to its abandonment, coinciding with a major sand dune invasion that affected the settlement and its surroundings (González Prats, 1999).

During the archaeological campaigns at the site, and mainly due to the discovery of some metallurgical waste deposits, abundant quantities of materials related to metal production have been collected. The housing structures have also provided significant evidence regarding these activities. For example, the use of some rooms could be associated with metallurgical operations, based on the presence of firing structures, as demonstrated by the finding of abundant archaeometallurgical debris *in situ* (González Prats, 1999–2000).

At present, the data available on metallurgy at La Fonteta permits us to assert that, since its first occupation and during all its occupation phases, production and trade of metal were important

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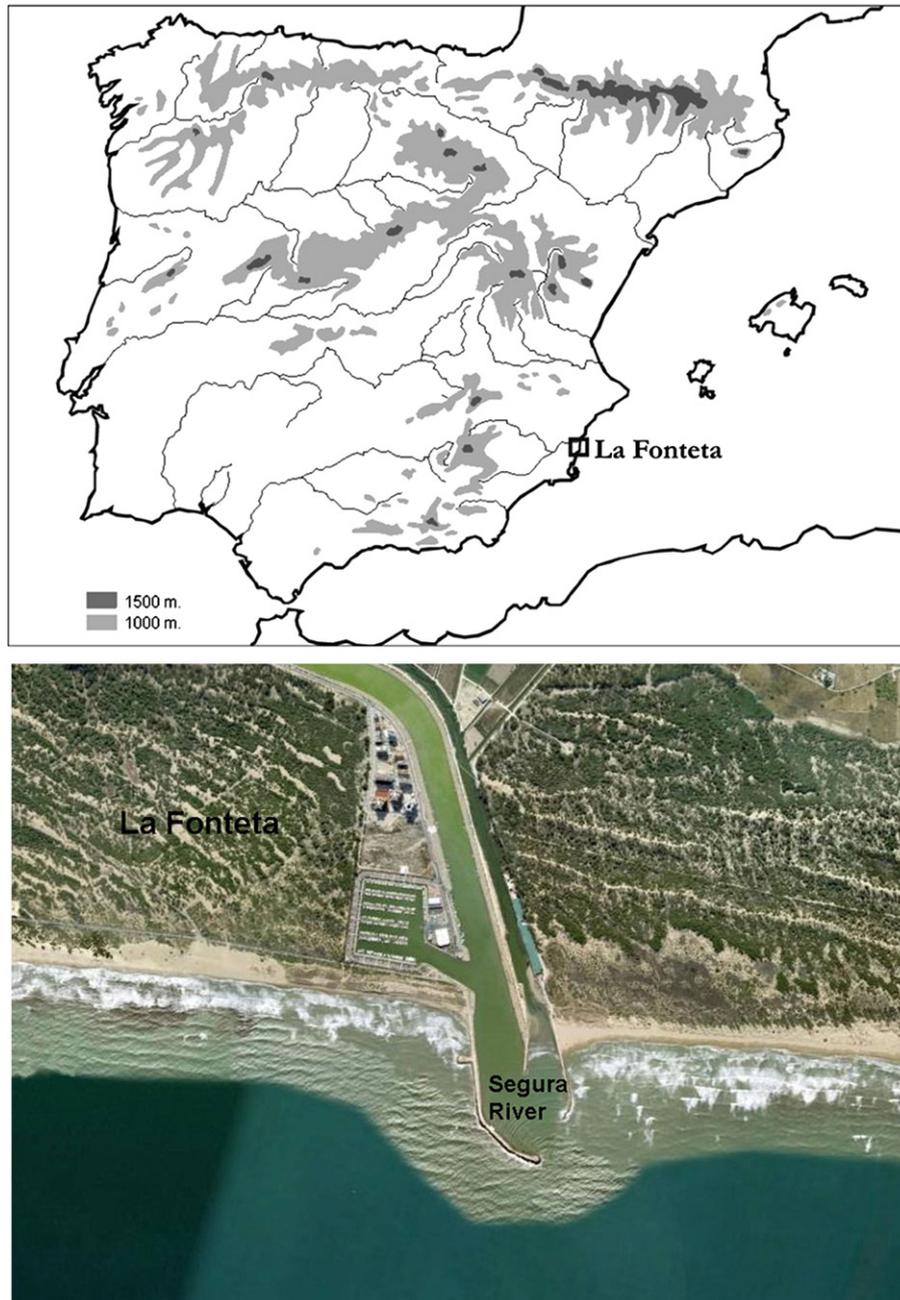


Fig. 1. Location of the Phoenician settlement of La Fonteta.

aspects of the economy of the habitat. The archaeometallurgical finds can be mainly related to the processing of iron, copper, copper–lead alloys, and of binary and ternary bronzes. Lead and silver production also seem to have taken place, as suggested by the discovery of 12 galena nodules, some litharge fragments, cupellation remains, and of a large number of metallic lead droplets (González Prats and Ruiz Segura, 1999; Fig. 2).

Amongst these, the specimens employed in pyrometallurgical processes are particularly worthy of attention because of their quantity and variety: approximately 60 fragments of sandstone and clay moulds, about 240 fragments of crucibles with different sizes and shapes, and more than 400 fragments of tuyeres (Renzi, 2007).

In addition, approximately 100 kg of slags have been collected. Some of them can be generally connected to copper-based

metallurgy, others to iron smithing activities. To date, due to identification problems from both a morphological and analytical point of view, no slags have been found that can be clearly classified as the result of iron reduction operations (Renzi and Rovira Llorens, 2009). Metallic items and melting wastes are also present (Fig. 3): raw materials – such as metal masses of Cu–Pb or binary and ternary bronzes – and some finished metal artefacts (copper, bronze, iron and lead objects) have been collected.

The scientific study of this group of materials provides valuable information about the technological level reached by the Phoenicians in metallurgical production between the 8th and the 6th century BC. Nevertheless, the preliminary state of the archaeological research at the site does not allow us to draw definitive technological conclusions.



Fig. 2. Archaeometallurgical findings from La Fonteta: fragments of litharge and cupels; lead droplets; galena nodules.

The area excavated until now is limited, and the characteristics of La Fonteta outlined here cannot be considered as representative of the whole settlement. In fact, a French team of archaeologists, who also worked at La Fonteta, draws attention in a monographic study about the site to the scarcity of archaeometallurgical remains collected during the excavation (Rouillard et al., 2007).

## 2. Selection of the samples

The archaeometallurgical samples selected for the provenance study by isotopic characterisation of lead include materials related to lead and silver production and copper alloys. Among the specimens available, samples ascribed to different chronological phases of the settlement's occupation sequence have been selected, distinguishing between the two main occupation periods of the site: Archaic Fonteta and Recent Fonteta.

As regards lead–silver metallurgy, the selection of samples was carried out in order to help clarify the relation between the findings within the *chaîne opératoire* for the production of these metals: galena as raw material, lead as the result of smelting, and the litharge fragments as secondary products of a metallurgical process

for obtaining silver – the cupellation – in which lead has a fundamental role as a collector of the noble metal.

The selection of the galena nodules is especially significant in this research. The mineral had been moved to a site without mineral resources in its vicinity, and points to production and material redistribution being carried out at the site. Furthermore, the suggested provenance of raw material is more reliable than the one of other metallic wastes or debris of metallurgical processes, in which various materials of different provenance could be mixed.

The analyses of metallic lead have been performed in order to verify if its isotopic composition was matching the galena, and thus to find out if the raw mineral found at the site was potentially used for smelting. However, it has to be noticed that until now no lead slags have been identified at La Fonteta to support this hypothesis.

Elemental analyses showed a low–silver content in the galena, the average being 170 ppm Ag. This content would not have been considered valuable enough for silver extraction (Rehren and Prange, 1998), therefore some fragments of litharge have been analysed in order to clarify whether a direct relation existed between this lead oxide, the galena nodules and the lead droplets. In other words, in view of the lack of argentiferous galena, we wanted to test whether the metallic lead could have been used to



Fig. 3. Archaeometallurgical findings from La Fonteta: moulds; tuyeres; crucibles; iron slags; copper-based slags; melting wastes.

obtain silver through cupellation processes from other resources, such as cupro-argentiferous minerals. The abundant presence of metallic and slaggy materials composed of Cu–Pb or Cu–Pb–Sn alloys, often with a very low tin content, seems to point in this direction. This group of selected samples includes two fragments of a material that – due to its morphological, structural and chemical characteristics – we identified as possible remains of cupellation vessels or hearth linings which had absorbed base metal oxides. This material has been provisionally labelled ‘Pb–Cu cupellation debris’ because of its composition, consisting mainly of lead and copper oxides (Table 1). Apart from containing relevant quantities of both elements, its crystal phases frequently appear in the cupellation hearth material used to oxidise argentiferous lead (Renzi et al., 2007). For this reason, the hypothesis that this debris was a sub-product of processes for extracting silver from cupro-argentiferous minerals has been considered.

Table 1

SEM–EDX results of the bulk analyses (wt%) of the Pb–Cu cupellation debris from La Fonteta.

Sample	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	K <sub>2</sub> O	CaO	FeO	CuO	PbO
F41778	1.9	4.5	14.5	n.d.	8.5	n.d.	23.6	47.0
F-41716-1	1.2	4.0	17.1	0.6	7.5	1.2	22.1	46.3

Finally, a fragment of a copper ingot, a melting waste and a metallic item composed of a Cu–Pb alloy have also been sampled. Their chemical compositions are shown in Table 2.

### 3. Methodology

Lead isotope ratios of a first group of samples from La Fonteta have been measured at the *Institut für Mineralogie* of the *Westfälische Wilhelms-Universität* in Münster by M. Bode, using a Multi-collector VG Sector 54 by solid-source Thermal Ionization Mass Spectrometry (TIMS), equipped with 9 Faraday collectors.

This group includes 18 samples: 6 galena nodules, 3 droplets of metallic lead, 4 fragments of litharge, a fragment of a copper ingot, a metal item and a melting waste composed of a Cu–Pb alloy, and a fragment of Pb–Cu cupellation debris.

Litharge and galena nodules were prepared by crushing and pulverizing small portions of material in an agate ball mill, to a particle size of c. 62 µm. Metallic samples were taken by drilling with stainless steel bits of 1.5 mm in diameter; a different bit was used for each specimen in order to avoid contamination.

Lead isotope measurements of galena, litharge and metallic lead were carried out without chemical separation. Prior to analysis, approximately 3 mg of these samples were dissolved in 10 ml of 6 N

**Table 2**  
Chemical composition of the copper-based materials from La Fonteta (wt%). ICP–OES analyses.

Sample	Fe	Ni	Cu	Zn	Pb	As	Sb	Sn	Ag	Bi	P	S	Total
F31123	0.002	0.014	60.7	<0.001	40.1	0.026	<0.001	0.092	0.029	<0.001	0.005	0.02	101.0
F11063	0.028	0.014	56.9	0.008	30.0	0.015	0.008	0.037	0.029	0.008	0.028	0.51	87.6
F1408	0.002	0.013	92.9	0.033	1.03	0.002	<0.001	<0.001	0.065	<0.001	0.004	0.84	94.9

HCl and diluted with H<sub>2</sub>O–Milli-Q up to 30 ppm of lead. Column chromatography has been used to separate the lead from the other elements present in the copper-based materials. The chemical procedure for lead separation has been carried out in Bochum by M. Behmenburg, at the Material Science Laboratory of the *Deutsches Bergbau-Museum* Department of Archaeometallurgy. A detailed description of a similar procedure for lead separation can be found in Prange (2001) and in Pernicka et al. (1984).

Finally, lead obtained by column chromatography was dissolved in 4–6 ml of 6 N HCl. 1 µl of these dissolutions was loaded on single Re filaments with the silica gel/H<sub>3</sub>PO<sub>4</sub>-method (2 µl of silica gel and 1 µl of 0.25 N H<sub>3</sub>PO<sub>4</sub>) and dried out in several steps (Cameron et al., 1969). During the measurements, the run temperature of the filament was kept between 1200 °C and 1300 °C. Acceleration voltage was about 8 kV.

The samples were measured in static collection mode using four Faraday cups, one for each analysed lead isotope (approximately 2–4 V in <sup>208</sup>Pb). External mass fractionation correction was calculated using the NBS 982 standard (Todt et al., 1996), resulting in a correction factor of 1.0011/amu. The internal standard error (2SE precision) was always lower than 0.01% after 6 blocks of 15 measurements. Reproducibility is shown in Table 3.

On the basis of the first LIA results obtained in Münster, we decided to widen the samples in order to clarify various questions about the provenance of some materials relevant to our research. Consequently, a second series of four lead isotope analyses was carried out at the Geochronology and Isotope Geochemistry Research Facility of the University of the Basque Country (Bilbao, Spain). This second series includes 2 samples of metallic lead, a fragment of litharge and a fragment of Pb–Cu cupellation debris. The measured ratios were obtained using a Finnigan MAT 262 Thermal Ionization Mass Spectrometer (TIMS), equipped with 8 Faraday cups and a Secondary Electron Multiplier.

As in the previous case, the procedures for sample preparation vary according to the type and composition of the analysed samples. In the case of metallic lead, a clean area was selected to extract the samples by drilling the metal with stainless steel bits of

1 mm in diameter. The litharge was ground in an agate ball mill; subsequently, the samples were dissolved in 3 ml of 7 N HNO<sub>3</sub> to be measured in the spectrometer.

The sample preparation process for the Pb–Cu cupellation debris has been more complex, since it was necessary to carry out a chemical separation of the lead that involves different stages of preparation. A more detailed description of this procedure can be found in Montero Ruiz et al. (2007) and in Santos Zalduegui et al. (2004b).

The diluted samples were separately loaded onto out-gassed Re single filaments with 3 µl of silica gel, and heated up to approximately 2.3 mA before introducing them in the TIMS.

As with the previous examinations, for this second series of analyses the samples were also measured in static collection mode using four Faraday cups. The data obtained were corrected for mass fractionation by comparison with replicate analyses of the Pb NBS 981 standard and its theoretical value proposed by Todt et al. (1996). In the case of the samples that constituted the object of our study, the values obtained for the NBS 981 standard, analysed in the same period of time for a total of 6 measurements, are shown in Table 3.

Preceding the lead isotopic study, chemical analyses have been performed by Dr. Ignacio Montero Ruiz and Prof. Dr. Salvador Rovira Llorens using the X-ray fluorescence spectrometer Metorex XMet 920MP of the National Archaeological Museum of Madrid in order to determine the main elemental composition of the selected samples.

The samples have also been analysed by inductively coupled plasma optical emission spectrometry (ICP–OES), using an IRIS/AP–HR (Thermo Jarrell Ash Corporation, Franklin, MA, USA) with a high-resolution Echelle Monochromator. These analyses have been performed by W. Steger and Dr. M. Prange at the Material Science Laboratory of the *Deutsches Bergbau-Museum* Department of Archaeometallurgy.

Moreover, in order to study their structural phases and composition, the litharge and the fragments of Pb–Cu cupellation debris have been examined by scanning electron microscopy. The FEI Quanta 200 environmental microscope of the National Museum of Natural Sciences of Madrid has been used to this purpose; it is equipped with an Oxford Instruments Analytical–Inca system and has been operated by microscopists M. Furió and L. Tormo.

**Table 3**  
Reproducibility (2SD) and mean values of standard measurements (NBS 982, NBS 981) at the Münster and Bilbao laboratories compared to standard values measured by Todt et al. (1996).

	<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>206</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb
NBS 982 (Todt et al., 1996)	1.000160	0.467006	36.7492	17.1621	36.7555
Mean values of NBS 982 after 185 measurements (Münster laboratory)	0.99868	0.46676	36.623	17.094	36.574
Standard deviation (2SD) (Münster laboratory)	0.0011	0.003	0.0439	0.0238	0.0682
RSD (%) (Münster laboratory)	0.1072	0.0716	0.1200	0.1390	0.1864
NBS 981 (Todt et al., 1996)	2.16701	0.914585	16.9356	15.4891	36.7006
Mean values of NBS 981 after 6 measurements (Bilbao laboratory)	2.16351	0.91396	16.911	15.456	36.587
Standard deviation (2SD) (Bilbao laboratory)	0.002	0.0008	0.016	0.022	0.070
RSD (%) (Bilbao laboratory)	0.0924	0.0875	0.0946	0.1423	0.1913

#### 4. Geological framework

The geology of the Guardamar del Segura area (Alicante), where the Phoenician settlement of La Fonteta is situated, is characterised by Tertiary basins, with rocks mainly formed by limestone, marls and sandstones.

As the metallogenic maps of the Alicante province show (IGME, 1972–1973b), there are no important metalliferous sources in the immediate surroundings of the site. Within a radius of 30 km from Guardamar, northward in the Crevillente area (Alicante), some small massive deposits of iron assigned to the Jurassic period have been identified; westward, close to the Orihuela mountain range (Alicante), some iron mineralisations formed during the Triassic period. Moreover, in Santomera, also in the Orihuela area and included in the Alpujarride complex, stratigraphic deposits of Cu(Au) have been identified. However, no lead ores have been identified so far in the vicinity of La Fonteta (IGME, 1972–1973a; Fig. 4).

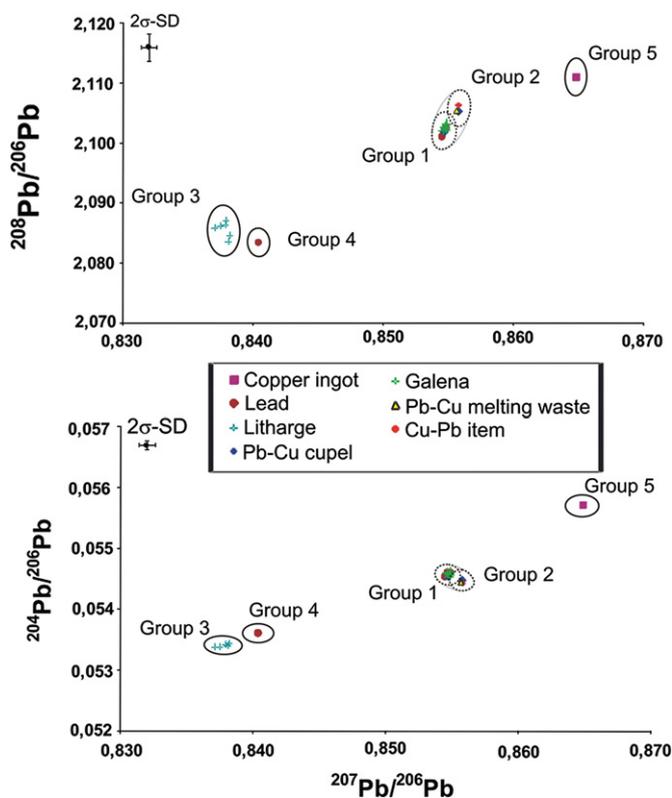




**Table 4**

Lead isotope data of the materials from La Fonteta.

Sample	Object	Sector	Chronology	Phase	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{204}\text{Pb}/^{206}\text{Pb}$
F-25090	Galena	5B habitat	RF	IV/VI	2.1027	0.8548	18.3254	15.6640	38.5321	0.0546
F-62073	Galena	1N waste deposit	AF	II	2.1030	0.8549	18.3250	15.6660	38.5383	0.0546
F-12752	Galena	14N/E habitat	AF	II	2.1020	0.8546	18.3170	15.6545	38.5031	0.0546
F-41567	Galena	54S/W waste deposit	AF	I	2.1032	0.8548	18.3348	15.6733	38.5613	0.0545
F-35287	Galena	54N waste deposit	AF	II	2.1025	0.8548	18.3197	15.6601	38.5169	0.0546
F-62026	Galena	1W waste deposit	AF	II	2.1021	0.8549	18.3123	15.6548	38.4944	0.0546
F-41536	Lead	54S/W waste deposit	AF	I	2.1021	0.8547	18.3193	15.6575	38.5094	0.0546
F-41722	Lead	54S/W waste deposit	AF	I	2.1019	0.8547	18.3163	15.6551	38.4983	0.0546
F-50306	Lead	1W waste deposit	AF	II	2.1024	0.8549	18.3152	15.6572	38.5066	0.0546
F-11000	Lead	8B/C habitat	RF	VI	2.1010	0.8545	18.3354	15.6677	38.5222	0.0545
F-11113	Lead	8B/C habitat	RF	VI	2.0834	0.8404	18.6547	15.6776	38.8659	0.0536
F-25081	Litharge	8B/C habitat	AF	III	2.0863	0.8379	18.7107	15.6779	39.0359	0.0534
F-1401	Litharge	7E habitat	RF	VI	2.0870	0.8382	18.7342	15.7030	39.0989	0.0534
F-62053	Litharge	65 ext. walls	RF	VI	2.0845	0.8371	18.7282	15.6773	39.0392	0.0534
F-25089	Litharge	5B habitat	RF	V/VI	2.0858	0.8381	18.7347	15.7022	39.0760	0.0534
F-41540	Litharge	54S/W waste deposit	AF	I	2.0835	0.8375	18.7229	15.6806	39.0094	0.0534
F-25097	Litharge	5A habitat	AF	III/IV	2.0861	0.8379	18.7211	15.6866	39.0538	0.0534
F-41778	Pb–Cu cupellation debris	54S/W waste deposit	AF	II	2.1053	0.8558	18.3569	15.7098	38.6462	0.0545
F-41716-1	Pb–Cu cupellation debris	54S/W waste deposit	AF	I	2.1019	0.8547	18.3374	15.6734	38.5430	0.0545
F-11063	Cu–Pb item	14N/W habitat	AF	III	2.1063	0.8558	18.3737	15.7244	38.7010	0.0544
F-31123	Cu–Pb melting waste	5A/S habitat	AF	III/IV	2.1053	0.8557	18.3672	15.7165	38.6693	0.0544
F-1408	Copper ingot	A3 survey	RF	VI	2.1109	0.8649	17.9473	15.5221	37.8857	0.0557

**Fig. 6.** Lead isotope ratios of the archaeometallurgical findings from La Fonteta. Groups of materials.

The lead isotope study of the first group of samples (Fig. 6) shows that its members come from the same area, allowing us to directly relate the raw material (galena) to the finished product (lead metal). Furthermore, Group 1 also includes a fragment of Pb–Cu cupellation debris. This group can be assigned to the mines in the Almería province, in particular to Sierra de Gádor or Sierra Alhamilla (Fig. 7). In both mining areas, mixed deposits with lead and copper mineralisations are present. Sierra de Gádor mainly consists of dolomites and limestone, in which stratiform mineralisation of copper carbonates (such as azurite and malachite) and lead and zinc sulphides (as galena and sphalerite) are embedded (Martín-Rojas et al., 2007). Dolomitic limestone, mica schist and phyllites with iron mineralisations, argentiferous galena, sphalerite and chalcopyrite, often associated with barite, make up the geological composition of the Sierra Alhamilla area (Gallegos Díaz, 1986).

The ratios of the Pb–Cu cupellation debris fragment included in Group 1 coincide with geological data from Sierra de Gádor, whereas the copper-based items of Group 2 form a tight cluster in the diagram but do not coincide with any specific mining area (Fig. 7). According to the principles described by Stos-Gale (2001), no mineralisations are known for Group 2 to suggest a provenance hypothesis of ‘mixing line’ with lead likely originating from the Gádor area. This mixing would be an interesting option if we consider that the other Pb–Cu cupellation debris can be clearly included in Group 1. However, at present, it seems we are dealing with ore deposits whose origin is still unknown, although its proximity to lead isotope ratios of Group 1 could suggest that it is a mining area close to the south-east or, at least, an area of similar geological age.

A global comparative analysis has also been performed for Group 3, formed exclusively by the six fragments of litharge. As with Group 2, no exact match was found, but these samples from La

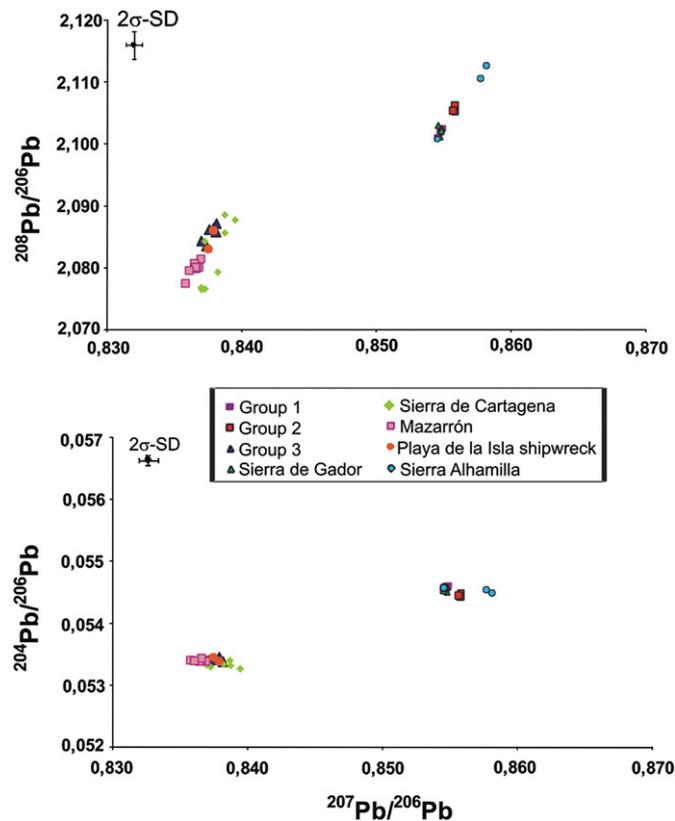


Fig. 7. Lead isotope ratios of groups 1 and 2 compared with ores from Sierra de Gádor and Sierra Alhamilla (Almería); lead isotope ratios of Group 3 and Playa de la Isla shipwreck compared with galena ores from Sierra de Cartagena and Mazarrón (Murcia).

Fonteta appear similar in their lead isotope ratios to mineralisations from the south-east area of the Iberian Peninsula. Their isotopic values seem to be generally linked to the mining deposits of Cartagena-Mazarrón (Murcia), an area that is geographically close to La Fonteta (Fig. 7).

The data obtained matches results of the analyses of fragments of two litharge cakes that were part of the cargo of the Phoenician shipwreck found in the Puerto de Mazarrón area (Cartagena). The shipwreck samples have been delivered for their analysis to Salvador Rovira, whom we thank for offering us the possibility of publishing the results. The 'Phoenician ship 2', dated to the 7th century BC, sank in the waters of Playa de la Isla with a cargo of some pottery and more than 1000 kg of lead ingots and litharge cakes (Negueruela et al., 2004). Our data suggest that the litharge fragments found in the shipwreck from Playa de la Isla and those at La Fonteta come from the same mining area (Fig. 7). However, as the diagram shows, none of these samples is fully consistent with the comparative data for mineralisations available for the Murcia province. Both minerals from the Sierra de Cartagena area and Mazarrón are close to the litharge samples but do not completely coincide with this third group.

Furthermore, some materials from Ampurias could originate from the same geological area (Montero Ruiz et al., 2008: 315). Lead objects, litharge and a silver coin have isotopic values that are similar to the samples forming Group 3. Moreover, some of the lead ingots recovered from the Mahdia shipwreck also show an isotopic signature similar to the Fonteta litharge (Begemann and Schmitt-Strecker, 1994), but they do not provide any further information for determining the provenance of the raw material. It is worth noticing that the litharge fragments from La Fonteta are not an isolated case; there

are numerous materials probably originating from areas of this south-eastern mining region that have not been isotopically characterised yet. This area could have been an active producer of lead and silver, at least from the 8th to the 1st century BC.

The complexity and varied mineralogy of the deposits in the Murcia mining district make it difficult to carry out an exhaustive characterisation of the area. Moreover, there is no data available that can cover the width and variety of local mining resources. For example, the Cabezo de Aborica (Bolnuevo) mines, the Lomo de Bas-Águilas (Sierra del Cantar) or Sierra Espuña mining areas have not been characterised yet; the characterisation of the mining deposits of both Murcia and Almería provinces carried out to date is insufficient for our purposes.

The isotopic results of Group 4, consisting of a single lead droplet, are not consistent either with minerals from the Gádor-Alhamilla area or from Cartagena-Mazarrón. They also do not coincide with other comparative geological data available for the Iberian Peninsula, or with any Iberian or extra-peninsular geological deposits known. Even if more mineralisations should probably be analysed to define its provenance, it can be noticed in the diagram that the sample presents an alignment with some geological samples. This linear distribution of points suggests this metal could be the result of a lead mixing, melting together metals from different areas (Fig. 8). According to the 'mixing line' explained by Stos-Gale (2001), among the possible provenances we believe this lead droplet could be the result of mixing lead coming from the Sierra de Gádor – identified as the possible source for Group 1 – with lead from Sierra Almagrera (Almería).

The fifth and last group also consists of a single sample, a copper ingot fragment. Its lead isotope ratios do not seem to coincide with any mining region of the Iberian Peninsula characterised until now, being quite distinct from the peninsular geological deposits, with the exception of some mines of the Ossa Morena zone (Tornos and Chiaradia, 2004; Klein et al., 2009). Nevertheless, the sample does not match the isotopic ratios of the geological data characterising this region (Fig. 9). Hence, Ossa Morena has to be excluded as its possible provenance area. The position of this sample in the diagrams does not suggest either, as in the last case, the hypothesis of a metal mixing from different geographical provenances.

Plotting all the Iberian pre-Roman metal objects in our database in order to test whether there was any coincidence with some of them showed that the ingot fragment is not consistent with any of these objects, but falls in an empty area of the diagram (Fig. 9). Therefore, an extra-peninsular origin has to be assumed for this sample.

To date, only two areas present isotopic values close to the ratios of the copper ingot fragment: the Wadi Arabah (Gale et al., 1990; Hauptmann et al., 1992; Hauptmann, 2007) and some mines from Sardinia (Valera et al., 2005; Begemann et al., 2001). Archaeological and geological samples from two ancient copper mining areas in the Wadi Arabah have been analysed: Timna in Israel and Faynan in Jordan, situated west and east of the Wadi, respectively. Both areas of the Wadi once formed a single geological deposit and they consequently present a similar although very wide range of isotopic values (Hauptmann et al., 1992). In some cases, this similarity can make an exact distinction between the possible provenance of a sample from the Faynan or the Timna area more difficult (Artzy, 2006).

The case of Sardinia is also complex (Valera et al., 2005), as there are various mines presenting lead isotope ratios compatible with the analysed copper ingot. These mines belong mainly to two large mining regions, the Sulcis-Iglesiente in the Cagliari province (Reigraxius, Perda, Arenas, Sa Duchessa, Montevecchio and San Giorgio mines) and the Nuoro province, in the northern part of the island (Funtana Raminosa and Canale Barisone-Torpè mines).

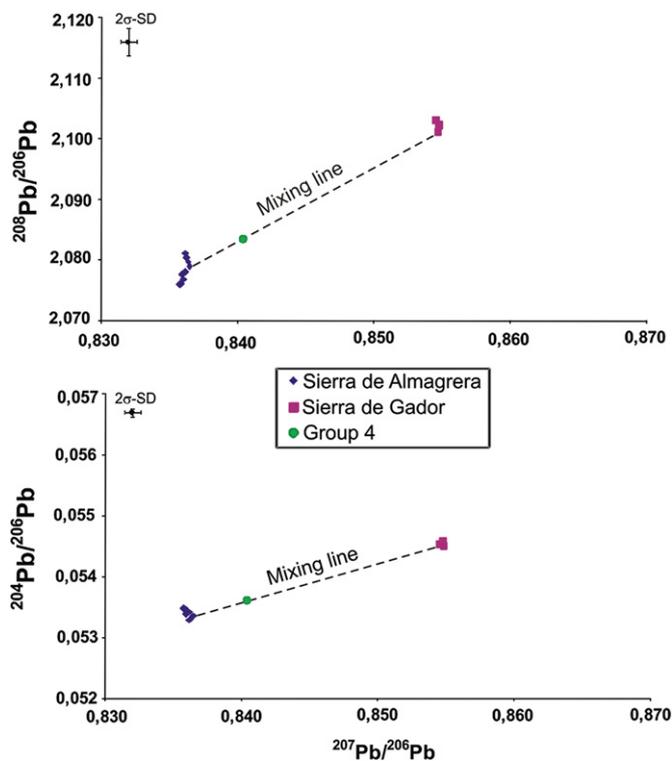


Fig. 8. Lead isotope ratios of ores from Sierra Almagrera, Sierra de Gádor and the lead droplet from La Fonteta (Group 4) – the mixing principle.

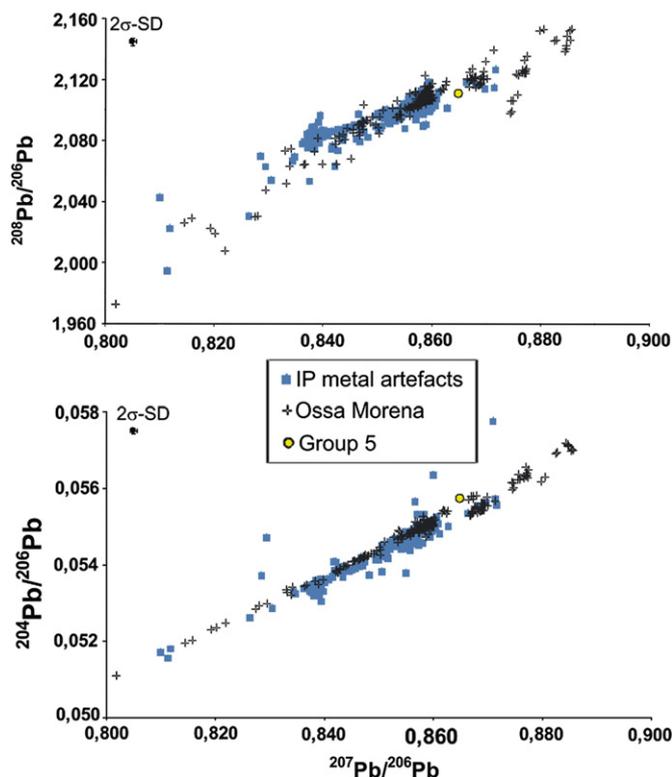


Fig. 9. Lead isotope ratios of ores from Ossa Morena and Group 5; lead isotope ratios of pre-Roman metal artefacts from the Iberian Peninsula compared with Group 5.

According to the lead isotope diagram of geological data from these two mining areas (Fig. 10), some Sardinian mineralisations display a partial overlap with the isotopic fields of the Wadi Arabah, although this overlap is minor with Faynan. Moreover, the actual isotopic values of the ingot fragment fall into an intermediate area between the mineralisations of the two regions, but do not coincide with any of them. It is also worth noticing that the sample from La Fonteta seems to show a slightly more consistent relation to the Timna area. In any case, the problem of assigning the provenance of a sample to Sardinian or Wadi Arabah mineralisations is not an isolated scenario. A similar situation occurred when studying some copper-based objects coming from Tel Jatt in Israel (Artzy, 2006).

The chemical composition of the ingot shows proportions of elements compatible with data provided by Hauptmann (2007: 79) for Timna and Faynan ores, including the lead. Nevertheless, the high lead content (>1%) can also appear in some copper mineralisations from Sardinia (Begemann et al., 2001: 75, table 5). Yet, at present, there are no chemical reference analyses available for trace elements of Sardinian copper ores. Therefore, for the time being, the study of these elements does not provide any valid evidence to determine the sample's provenance.

### 6. Technological interpretation

In order to clarify the groups and nature of the materials analysed, the metallurgical processes they are related to must be briefly mentioned. According to the results of XRF analyses carried out on the galena samples, the nodules collected at La Fonteta are not argentiferous. Therefore, this mineral is not related to the production of silver; it was imported only to produce lead, as the lead isotope ratios of the metallic lead droplets seem also to confirm, showing good coincidence with the isotopic values of the galena (Fig. 6). It is also worth noticing that the silver content of these lead droplets (lower than 120 ppm in all the samples analysed) is coherent with the composition of the galena.

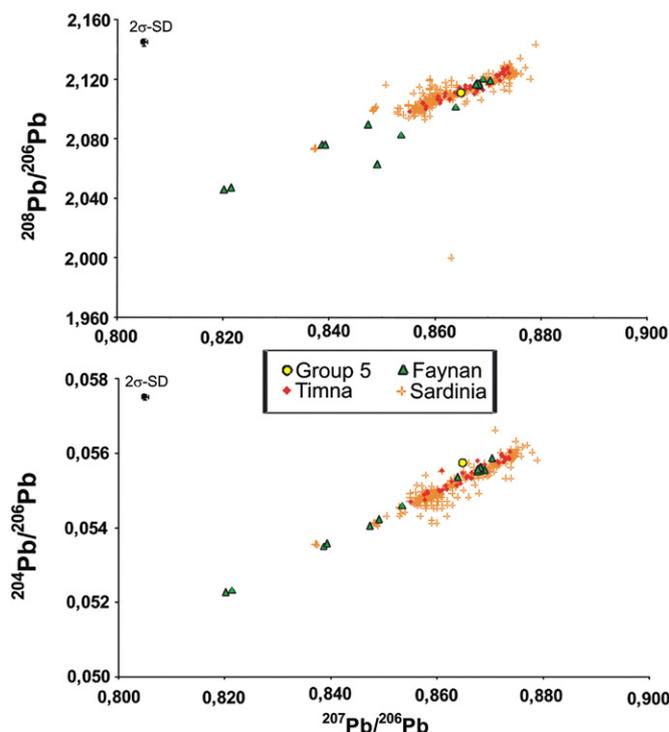


Fig. 10. Lead isotope ratios of Timna, Faynan and Sardinia ores compared with Group 5.

As regards the 'Pb–Cu cupellation debris', it is still unclear what kind of metallurgical process it can be related to. We considered the option that these materials could be the result of refining debased silver alloyed with copper (Bayley and Eckstein, 1997, 2006). However, this hypothesis seems improbable as, during the chronological span that is the object of our research, the debasement of silver by adding copper was not common in the Iberian Peninsula. XRF analyses of 67 silver objects in the *Archaeometallurgy of the Iberian Peninsula* project, whose chronology spans from the 8th to the 6th century BC, showed in most cases almost pure silver (>97%Ag); only few items (4.5%) have a copper content higher than 5% Cu, and just in one case the amount of copper exceeds 10%.

An alternative possible hypothesis could be that this debris is the secondary product of operations of silver extraction from cupriferos ores. The cupro-argentiferous minerals would be smelted to a copper–silver metallic mass. In the following stage, lead would be added to the raw metal to act as a collector for the noble metal. If we accept this option, cupellation debris formed mainly of copper–lead oxides and a silver regulus would be obtained.

Following this, a further process could also be suggested for reducing again the Pb–Cu oxides to metal or employing these sub-products to produce ternary bronzes by adding some tin-rich material. The presence at La Fonteta of various Cu–Pb metallic masses, such as the abovementioned copper–lead alloy F31223, of ternary bronzes and Cu–Pb and Cu–Pb–Sn slags seems to point to this hypothesis (Renzi et al., 2007). Consequently, the analysed copper-based alloys could be indirectly related to these processes for obtaining silver by exploiting cupro-argentiferous ores.

Due to the focus of this paper on provenance studies, no further details about this hypothetical process for obtaining silver can be given. This is a separate scientific problem, and we hope to be able to provide more information as our research will progress. However, according to the archaeometallurgical evidence of sites such as La Loba (Fuenteovejuna, Córdoba; Blázquez et al., 2002) and Monte Romero (Huelva; Kassianidou, 2003), exploitation of complex copper minerals with an appreciable content of silver is already known in the period related to our study. Moreover, at La Fonteta copper slags have been recovered and, in one case, silver grains have been detected embedded in copper sulphide inclusions trapped into the slaggy matrix of the sample; these are all elements that could support the proposed hypothesis (Renzi et al., 2007).

Then, if we accept that these materials can be considered the secondary product of a process of silver extraction from cupro-argentiferous ores, the fragment of Pb–Cu cupellation debris included in Group 1 could corroborate that the galena was exploited to obtain lead and that this lead could have been used in the extraction of silver from complex Cu–Ag ores. However, there is no clear evidence to establish a possible technological relation between the materials from Group 1 and Group 2. At this point, Group 2 material has to be assumed to originate from a different mining area whose provenance cannot be defined yet.

The litharge fragments forming Group 3 are certainly related to metallurgical activities for obtaining silver. As we have seen for the previous group of materials, the desilvering of argentiferous ores (generally galena) produces a metallic bullion composed of lead and silver, and lead oxide or litharge and cupellation hearth material as secondary products. The isotopic study of the litharge samples shows that their provenance is different from that of the materials in Group 1. This is consistent with the suggestion that the galena nodules found at La Fonteta were not used as a source of silver, as they do not contain a sufficient amount of the precious metal.

Yet, the presence of litharge at La Fonteta does not necessarily imply they were producing silver *in situ*, as this material could have been imported for different uses. Long distance transportation of litharge is documented by the discovery of the

Phoenician shipwreck from Puerto de Mazarrón, Cartagena, whose cargo included numerous lead ingots and litharge cakes (Negueruela et al., 2004). The isotopic ratios of the litharge from La Fonteta are similar to the ones from the shipwreck, even if the exact reasons for transporting this material have not been clarified yet. There are various possibilities to explain this. Litharge could have been re-smelted to obtain metallic lead and used, for example, in the production of leaded bronzes and Cu–Pb alloys. During this chronological period, the use of industrial lead is limited; few lead items have been collected at La Fonteta – mainly lead sinkers for fishing nets and weights – and no architectural use of this metal is documented. From the classical written sources, we know that in later periods litharge was also employed for medical uses, as a balm or as a pigment. However, it is unlikely that all the litharge could be re-used for these daily uses (Rehren et al., 1999: 302). Another possible explanation for the re-use of the litharge could be its addition for direct smelting of sulphidic lead ores, such as galena. Lead oxide reacts with lead sulphide to form metallic lead and sulphur dioxide gas, producing a similar effect to the roasting undertaken to reduce the sulphur content of the ore. Furthermore, the addition of the litharge to the smelting charge would permit the recovery of the silver eventually trapped in the lead oxide, making the whole process economically more profitable (Rehren et al., 1999: 307).

The presence at La Fonteta of a copper ingot can be easily explained. During this chronological period, the production of binary and ternary bronzes was widespread across the settlement, and the manufacture of copper objects is also documented.

## 7. Conclusions

The study of metallurgical finds from La Fonteta by lead isotope analyses allows us to explore some chronological aspects of raw material supply for lead and silver production. In particular, this study assigns the mineral exploitation mainly to two different mining regions, excluding the copper ingot.

According to the currently available data, it can be suggested that during the archaic phase (AF) the Phoenicians were exploiting galena from the Almería mining area to obtain lead, most likely from the Sierra de Gádor or Alhamilla. In this first period, the inhabitants of La Fonteta were probably also exploiting cupro-argentiferous ores coming from this area; unfortunately, the origin of these minerals cannot be specified further. Work is in progress to understand the possible technological relationship between these two groups.

In this regard, it is worth mentioning that the Phoenician settlement of Abdera (Adra) is situated at the estuary of the Adra River, the natural route to and from the mining resources of the Sierra de Gádor. This site has also provided various archaeometallurgical materials (Suárez Márquez et al., 1989), and may have served as a link or trading point to La Fonteta.

From phases III–IV – corresponding to the transition between the two main occupation periods of the site, Archaic Fonteta and Recent Fonteta – a major restructuring of the site occurred, as shown by the construction of the walls. This coincided with a major change in raw material supply and technological activity. To date, the recent phase has yielded only a limited number of archaeometallurgical materials. No galena or metallic lead have been found that could be clearly ascribed to RF. Lead fragments coming from later stratigraphic levels cannot be ascribed to a reliable chronology due to their disturbed recovery contexts; some of these layers are earth deposits that have been turned over, filling levels or waste deposits.

Of the litharge finds, two fragments belong to RF, three to the transition phase and just one fragment to AF. The isotopic study of

these samples shows that during the entire occupation sequence of the site no changes have been documented for the provenance area of the raw material employed for their production. For all the cases studied until now, the Murcia province appears to be the most probable mining region for the origin of these materials.

The current collection of materials and isotopic data suggests that in the transition phase (III–IV) the use of galena and lead from the Almería province gradually diminishes, until its complete disappearance in the subsequent phases. According to the provenance of the litharge fragments analysed, these resources would be substituted in a more consistent way by the minerals of the Murcia region. This change in the mineral supplying area and the associated technological changes are also indicated by the absence of Pb–Cu cupellation debris in the recent period of Fonteta.

According to the results obtained so far, the most probable hypothesis seems to be that from the transition period (phases III–IV) between AF and RF onward, the processing of cupro-argentiferous ores and lead and galena from the Almería province at La Fonteta ceased. Instead, lead could have been imported as litharge cakes, and copper would have been obtained in ingot form from other sources that still have to be identified.

The copper ingot fragment was found in one of the first surveys performed at La Fonteta, in a deposit that was turned over due to its use as a waste deposit. Therefore, its date cannot be precisely established, even if it was initially ascribed to phase VI (RF – first half of 6th century BC). The possibility that this fragment belongs to an earlier period cannot be excluded, as a similar find was collected in another sector of the site and was dated to phase III (AF – mid-7th century BC).

The provenance of this ingot sample has not been identified with certainty. At the present stage of research on isotopic characterisation of Iberian copper ore deposits, the data available does not suggest a peninsular origin for this sample. Instead, it seems to point toward two possible areas of origin: Sardinia or the Wadi Arabah, in particular the Timna region. Both possibilities are quite coherent with the historical context of the Phoenician site of La Fonteta. In any case, for the time being it can be safely affirmed that the copper ores used for producing the ingot are not related to the geological area exploited for the other groups of materials analysed. Thus, the suggested exploitation of argentiferous copper minerals during AF would have been aimed mostly at silver production, with copper being only a by-product, supplementing imported ingot copper from elsewhere.

In conclusion, the provenance study of 22 samples of different typology (litharge, lead, galena and copper-based materials) collected at La Fonteta indicates the use of at least five clearly differentiated mineral sources.

1. The first can be related to mineralisations of the Almería province, but it has not been possible to specify whether they were exploiting Sierra de Gádor or Sierra Alhamilla ores, or resources from both areas. This source was mostly used during the archaic phase of La Fonteta to provide low-silver galena and lead metal.
2. Another source, supplying the copper-based material in Group 2, cannot be geographically located, even if it corresponds to a mining area with isotopic values close to the Sierra de Gádor-Sierra Alhamilla ones, presumably in the same Almería mining region as Group 1. Its exploitation is predominant during the Archaic Fonteta period, and is potentially linked to the use of lead and galena of Group 1.
3. The third source, our Group 3, matches the Cartagena-Mazarón mining district, and is represented at La Fonteta by litharge fragments dated mostly to the transitional period and Recent Fonteta layers. Its function within La Fonteta is at present

unknown, but matches an established trade in litharge in the western Mediterranean.

4. The fourth source matches minerals from Sierra Almagrera (Almería). At La Fonteta, a single lead droplet can be linked to this source, if we consider the hypothesis of mixing lead from this area with lead from the Sierra de Gádor.
5. Finally, the copper ingot seems to come from an extra-peninsular source; its isotopic ratios are close to the values of some mineralisations of Sardinia and Timna.

Although the number of analyses available is still limited and the sampling is not sufficient to characterise the entire collection of materials from La Fonteta, this provenance study reveals a complex and dynamic framework of Phoenician metallurgy and commercial economy between the 8th and the 6th century BC.

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