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From Galena to Lead: Divergent Paths in Early Metallurgy in the Western Mediterranean

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Keywords

Palaeolithic, Neolithic, early metallurgy, technology, provenance, galena, Catalonia, Iberian Peninsula, lead isotope analysis

Abstract

This paper examines the use of galena, beginning in the Middle Paleolithic, before the onset of metallurgy in the Iberian Peninsula, and shows that it was a well-known raw material during the Neolithic, both in funeral and in household contexts. The identification of probable provenance suggests a non-long distance movement, with the Molar-Bellmunt-Falset mining district in Tarragona as the main source area for extracting this ore since the Palaeolithic. Although southern France and other European regions share a similar use of galena in Neolithic times, each took different approaches to the use of the raw material. In some areas (southern France, Sardinia) lead was used for metal production, while in others (Iberia, continental Italy) it was not. These differences can be explained by different social choices in each region, reinforcing the idea that innovation is not simply a question of technology.

Introduction

Galena, lead sulphide (PbS), is one of the main lead ores on Earth. It has two properties that attract attention: its weight and its bright metallic lustre. However, it is soft, and under pressure it breaks along three perfect cleavages at 90-degree angles (Haldar, 2020) (Figure 1). This makes it difficult to shape using lithic technology and an unsuitable material for everyday use. From a metallurgical point of view, galena can be easily melted under conditions similar to those needed for copper ores (Craddock, 1995, pp.205-206; Atzeni, 2005, p.26), while metallic lead has a much lower melting point (327.5 °C) than copper. This low melting temperature helps in working the metal over a simple domestic fire without the need for complex structures.

In this paper, we explore the use and possible provenance of galena before the beginning of metallurgy in the Iberian Peninsula, and especially in north-eastern Iberia. The special focus is on Palaeolithic finds as well as the newly discovered finds from the Neolithic period. In this context, we also raise the question of the spatial relationship with potential mining areas. The use of galena, hitherto not considered or valued as raw material in Western Europe prehistoric societies, prompts us to consider its relationship with the first documented lead metallurgy contemporaneous with copper metallurgy in neighbouring France and to discuss the differences between both areas in the adoption of metallurgical technology.

General overview about the prehistoric use of galena

Galena is found at archaeological sites from as early as the Middle Palaeolithic and there is evidence of its continued use until the Chalcolithic (~3000 BC) across wide geographically and culturally different areas, including North America (Walthall, 1981), Europe with special frequency in southern France (Baudais, Kramar and Gallay, 1990; Roscian, Claustre and Dietrich, 1992; Wyss, 1999, p.236; Convertini and Georjon, 2018), North Africa and the Near and Middle East as far as Pakistan (Jarrigue, 2008; Molist, et al., 2009; Bains, et al., 2013; Radivojević,

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Figure 1. Fracture structure of a galena sample from Can Sadurní (13CS-H10-IIJ-11a4-92) under the scanning electron microscope (SEM). Photo: O. García-Vuelta, Instituto de Historia-CSIC, Madrid.

et al., 2017; Çevik, et al., 2020; Charpentier, 2020). This confirms that it was a well-known mineral before its metallurgical use. Sometimes this galena is found together with other materials used as pigments (mainly ochre), although it is also documented in funeral and domestic contexts as unprocessed blocks in the investigation area of this study, especially during the Neolithic. Despite the difficulties of working it using lithic technology, attempts were made to manufacture ornamental elements (necklace beads or balls), although these were not common compared to the use of other more suitable raw materials, as in the cases of Ulucak Huyuk (Çevik, et al., 2020) or Çatalhöyük (Bains, et al., 2013). This ornamental use coincides in the Near East with the use of native copper, sometimes in the same contexts (Molist, et al., 2009; Radivojević, et al., 2017).

Lead was part of the experimentation that led to the development of metallurgy, perhaps before copper if we accept the bracelet from Level 12 at Yarim Tepe I in Iran dated to c. 5700 BC (Merpert and Munchaev, 1987; Radivojević, et al., 2021, p.52), although its application and adoption were very irregular depending on the period and area (Yahalom-Mack, et al., 2015). In this context, an early use of and familiarity with galena could have encouraged experimentation with heat to obtain metallic lead, but the new metal needed a functional use to be applied. In the Balkans experiments with galena to obtain pigments may have been the main objective (Hansen, et al., 2019; Kramberger, Berthold and Spiteri, 2021) although the metal could also had been processed in Belovode (Radivojević, and Roberts, 2021). In Sardinia lead was geared to a functional use such as clamps to repair pottery vessels (Valera, Valera and Rivoldini, 2005; Pearce, 2017). In southern France personal ornamentation was the main category of lead objects during the Chalcolithic and was combined with other copper and lithic ornaments, mainly in the Upper Hérault and the Gard Valleys (Roscian, Claustre and Dietrich, 1992). In continental Italy neither galena nor lead have been employed, although some antimony and silver ornaments were in use (Dolfini, 2020). In northeastern Iberia and other Iberian regions, galena was hardly known in Neolithic contexts prior to this paper, and metallic lead was not adopted either for ornamental or for any other use.

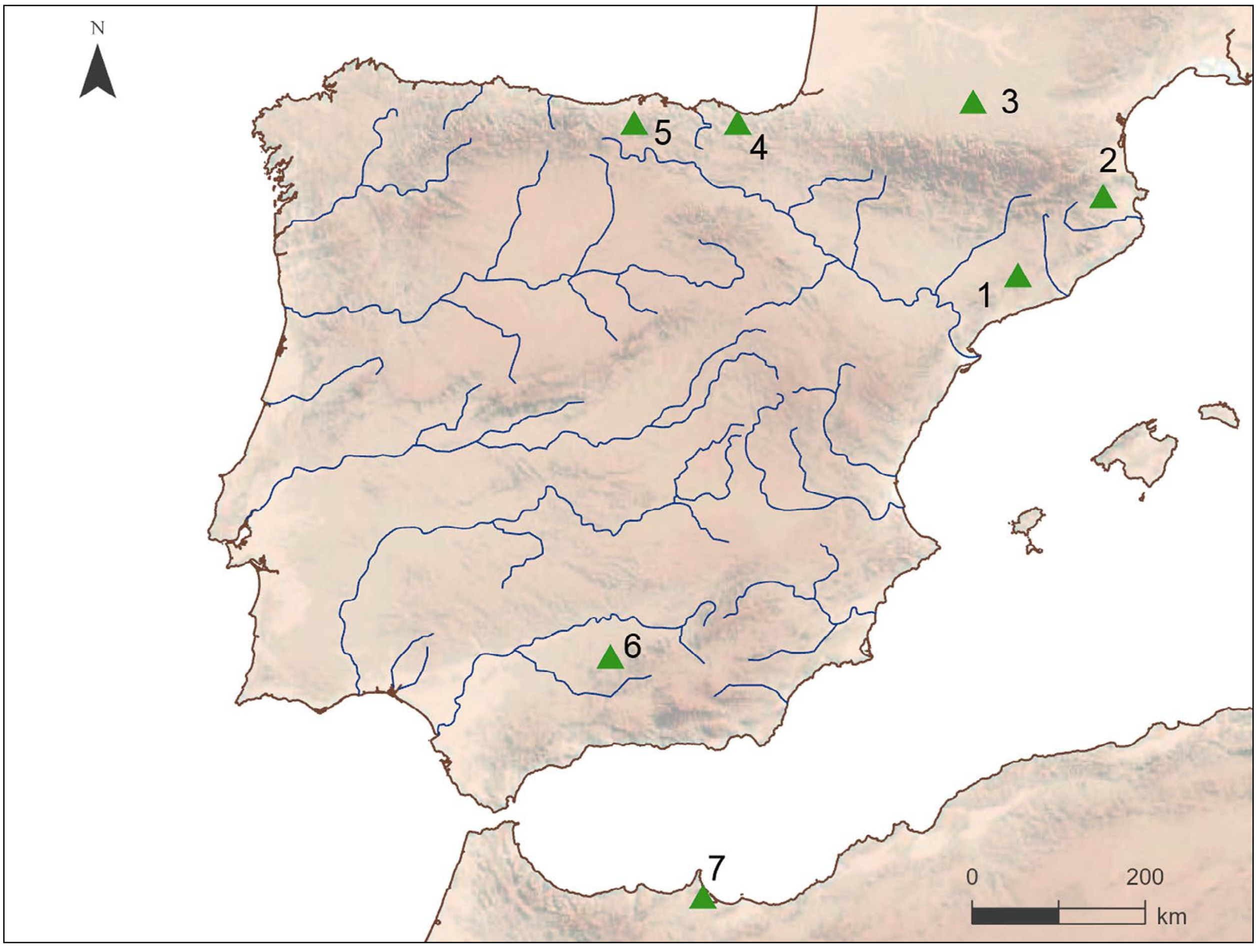

Figure 2. Location of the Palaeolithic sites where galena was found in Western Mediterranean: 1-Abric Romani, 2-Bora Gran dén Carreras and Coma d´Infern, 3-Grotte de Rigeaux, 4-Lezetxiki, 5-El Castillo, 6-Pirulejo, 7-Ifri n'Ammar.

Galena artefacts from the Iberian Peninsula

Galena from Middle and Late Palaeolithic contexts

As noted, galena is not a suitable material for making ornaments with the lithic technology used for other types of stone. However, there is evidence of its use for other purposes since at least as early as the Middle Palaeolithic, although this use is not always well determined. Several blocks have been identified in Level J of Abric Romani (Capellades, Barcelona) (Figure 2: 1; Table 1). Three fragments found in sublevel Ja have been published (Arteaga, et al., 2001, p.20; Vaquero, et al., 2012, p.211, Fig.7.11). They were identified as galena by micro X-ray diffraction analysis (µXRD) and by scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX), indicating that the outer surface was formed by cerussite. The largest piece measures 38.2 x 32 x 26 mm and weighs 130.4 g (Figure 3); the smallest 20.3 x 12.3 x 11.4 mm and 10.3 g; while the third one measures 38 x 21.5 x 10.7mm and weighs 31.5 g. The chronology of this sublevel Ja has been established between 53,000-50,000 BP. A fourth fragment found in the 2004 excavation campaign on Level 0, Grid T42 (Figure 3) is mentioned by Chacón, et al. (2013, Tab.2). This fragment would have been older, from around 56,000 BP according to the Bayesian chronology model proposed for the site (Gómez de Soler, et al., 2020). At the Abric Romani finds (Arteaga, et al., 2001), it has been suggested that the galena was used to produce fire (like pyrite); however, its fragility when struck makes this unlikely.

But earlier utilisation of galena is suggested by the block found in Level XXVII (lower OI) of the Middle Palaeolithic deposits of Ifri n'Ammar Cave (Morocco) (Figure 2: 7) which according to TL dating could be >150 Ka (Richter, et al., 2010).

Other Galena finds in a Palaeolithic context have been documented in Level 3a of the Lezetxiki Cave (Mondragón, Gipuzkoa) (Figure 2: 4), for which the affiliation between Mousterian or Aurignacian is under discussion (Esparza, 1993, p.41). The galena finds from Level 18 of Cueva del Castillo (Puente Viesgo, Santander) (Figure 2: 5) belong to this transitional period (Cabrera, 1984).

On the Iberian Peninsula, there is a context that indicates a possible functional use. In the Magdalenian strata of El Pirulejo Cave (Priego, Córdoba) (Figure 2: 6), lead contamination was detected in the sediment, probably due to the handling of galena (Monge, et al., 2015, p.6). A further functional indication is given by the joint presence of ochre and galena at the Magdalenian-period rock shelter of Bora Gran d'en Carreras de Serinya (Alsius i Torrent, 1906, p.60) and at Coma d'Infern a Les Encies (Soler i Masferrer, 1980, p.54), both in the province of Girona (Figure 2: 2).

Some studies suggest that galena was transported some distance from the geological deposits where it was extracted and collected, as is the case of the 2.5-kilo fragment found in the Magdalenian levels of Grotte de Rigeaux (Lespugue, Haute-Garonne) (Figure 2: 3), which is thought may have come from the Pyrenees, some 50 km away (Saint-Périer, 1912, p.153).

Galena was commonly found in the Upper Palaeolithic of Western Europe. As evidence of its use and geographic distribution, one must consider both the 42 galena fragments found in the Aurignacian phase of Walou Cave (Trooz, Belgium) and the Chatelperron phase finds from La Roche-au-Loup Cave (Merry-sur-Yonne, France) (Poplin, 1988; Goemaere and Jadin, 2014). In an adult burial in Tomb V at Arene Candide (Italy), ochre, galena and graphite were found together (Palma di Cesnola, 2001, p.278).

The widespread finds of galena fragments from various contexts of the European Palaeolithic reveal some possible uses in burial contexts, although in most cases we have no concrete evidence of their real function. Galena may have been used as a body dye in this context, as is well documented among Native North Americans, but where it is always mixed with some kind of binder (Koerper and Strudwick, 2006). In the Palaeolithic finds, however, it would not have been necessary to prepare a dye because when galena is crushed it generates fine shiny cleavage flakes that can be spread and by themselves generate a visual effect on surface.

Figure 3. Pieces of galena found in Abric Romani (Middle Palaeolithic). Photo: I Montero-Ruiz.

In addition, some of the galena blocks we have listed were found together with other elements without any apparent functionality, such as fossils or quartz crystals. In these cases, it is likely that they were collected and kept as curious or exotic objects (Moncel, et al., 2012) and served as amulets or talismans, a function also attributed to the galena used by North American natives (Koerper and Strudwick, 2006, pp.9-10). Like ochre (Hodgskiss, 2020), galena may have had different cosmetic, hygienic, ritualistic, etc. uses that are difficult to confirm archaeologically.

Galena from Neolithic contexts

The presence of galena at Neolithic sites in Catalonia has gone almost unnoticed in previous research, although Gallart (1983-84) recorded fragments at the site of La Planeta (Artesa de Lleida) (Figure 4: 3), an open-air settlement where a possible dwelling was excavated. Among the finds made at that site, there are three blocks of galena (Gallart, 1983-84, p.43, Fig.6) of variable sizes and weights (maximum 375 g); two of them are cubes and the third has an irregular shape (Table 1). The same publication cites another fragment of galena identified among the surface finds at the Neolithic site of Pleta del Paco (Les Borges Blanques) in the same area (Figure 4: 7).

Another galena fragment comes from Tomb X of the Neolithic necropolis of Masdenvergenc (Amposta, Tarragona)(Figure 4: 1) (Molist and Clop, 2010, p.316). The necropolis consists of pit graves and is dated between the 5th and the first half of the 4th millennium BC. Esteve Gálvez (2000, pp.100-129) excavated the remains of 33 tombs there, most of them were in very poor condition, in total, human remains were found in only ten of them. A piece of galena was documented in Tomb X, which was an oval pit measuring 1.62 x 2 m and bounded by 15 not very large stones and divided into two symmetrical spaces by five smaller stones (Figure 5). The eastern part of the pit did not contain any remains; in the western part, there was a grave with a somewhat shrunken corpse on its right side. Around the neck of the corpse there was a 5-row necklace, 5.40 m long, made of 1360 disc-shaped pieces of cardium and pectuncle shells (Esteve Gálvez, 2000, p.109, Figs. 9 and 10) (Figure 5). At chest height, the corpse was holding a small pottery vessel with both hands. Next to it, there was a short, poorly made flint blade and a small 15 x 15 x 11 mm galena cube with a weight of 20 g (Table 1).

During our studies, since 2015, we have been following the trail of galena finds in Neolithic contexts and have identified several new sites; some are in the excava-

Table 1. Description of the galena from the northeastern Iberian Peninsula and analytical techniques used. The site numbering (Nº) coincides with the map in Figure 4. Province (B= Barcelona; GI= Girona; LE= Lleida; TA= Tarragona).

Nº	Site	Province	Length (mm)	Width (mm)	Thickness (mm)	Weight (g)	Technique	LIA
1	Masdenvergenc PA25507	TA	15	15	11	20.1	XRF	x
2	El Molló	TA	1.3	0.9	0.5	3.1		
2	El Molló PA25772	TA	1.5	0.9	0.6	3.38	XRF	x
3	La Planeta PA25775	LE	60	40	34	375.8	XRF	x
3	La Planeta PA25776	LE	28	22	20	69.2	XRF	x
3	La Planeta PA25777	LE	35	40	20	218.7	XRF	
4	Tossal de L´Embrosi PA25780	LE	42.6	39.5	23	84.8	XRF	x
5	Serra de Puigverd PA25779	LE	35	19	10	42.0	XRF	
6	Serra de Castelldans PA25778	LE	37	33	31	203.7	XRF	x
7	Pleta del Paco	LE						
8	Vall Major I PA26455	LE	31.2	30.1	22.9	96.4	XRF/ICP-MS	x
8	Vall Major I PA26456	LE	31.9	22.3	19.2	68.5	XRF/ICP-MS	x
9	Vall Major III PA25781	LE	49.6	36.5	22.7	104.2	XRF	x
10	Comella Ruf PA25783	LE	64.8	39.6	20	180.7	XRF	
11	Serra dels Tinells PA25782	LE	29.6	24.6	18.2	59.1	XRF	x
12	Abric Romani NJ V50 540	B	38.2	32	26	130.4	SEM/ICP-MS	x
12	Abric Romani Ja 70 041	B	38	21.5	10.7	31.5	SEM/ICP-MS	x
12	Abric Romani Ja K61 36	B	20.3	12.3	11.4	10.3	SEM	x
12	Abric Romani O 42 nº1 7/5 30-8	B					SEM	x
13	Can Sadurni 11a4-92 PA26631	B	15	16	14	17.1	XRF/ICP-MS	x
13	Can Sadurni 11a1-17 PA26632	B	24	17	18	33.1	XRF/ICP-MS	x
13	Can Sadurni MM26 PA27677	B				108.3	SEM	
13	Can Figueres	B	19	16	17	29.3		x
14	La Draga PA25784	GI	66	42	40	341.5	XRF	x

tion phase and therefore some of the finds have yet to be published.

Chronologically, the earliest find within the Neolithic is the galena block found at the Neolithic site of La Draga (Figure 4: 14), a large settlement with an estimated area of 15,000 m^2 on the shore of Lake Banyoles (Girona) (Bosch, Chinchilla and Tarrús, 2006). The cube-shaped

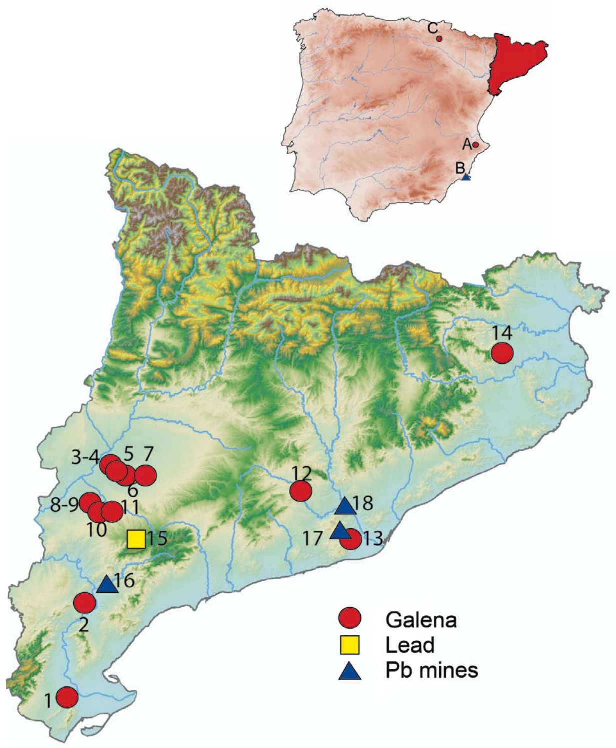

Figure 4. Location of the archaeological sites where galena fragments have been found and the mines related to their possible provenance. A. Coveta de l'Or (Beniarrés, Alacant), B. Cartagena , C. Los Cascajos (Los Arcos, Navarra); 1. Masdenvergenc (Amposta), 2. El Molló (Mora la Nova), 3. La Planeta (Artesa de Lleida), 4. Tossal de l'Ambrosi (Artesa de Lleida), 5. Serra de Puigvert (Puigverd de Lleida), 6. Serra de Castelldans (Castelldans), 7. Pleta del Paco (Les Borges Balnques), 8. Vall Major I (Sarroca de Lleida), 9. Vall Major 3 (Sarroca de Lleida), 10. Comella Ruf (Torrebesses), 11. Serra dels Tinells (Granyena de les Garrigues), 12. Abric Romaní (Capellades), 13. Can Sadurní (Begues), 14. La Draga (Banyoles), 15. Coveta de l'Heura (Ulldemolins), 16. Molar-Bellmunt-Falset (MBF) mining district, 17. Garraf Mines, 18. La Martorellense (Martorell).

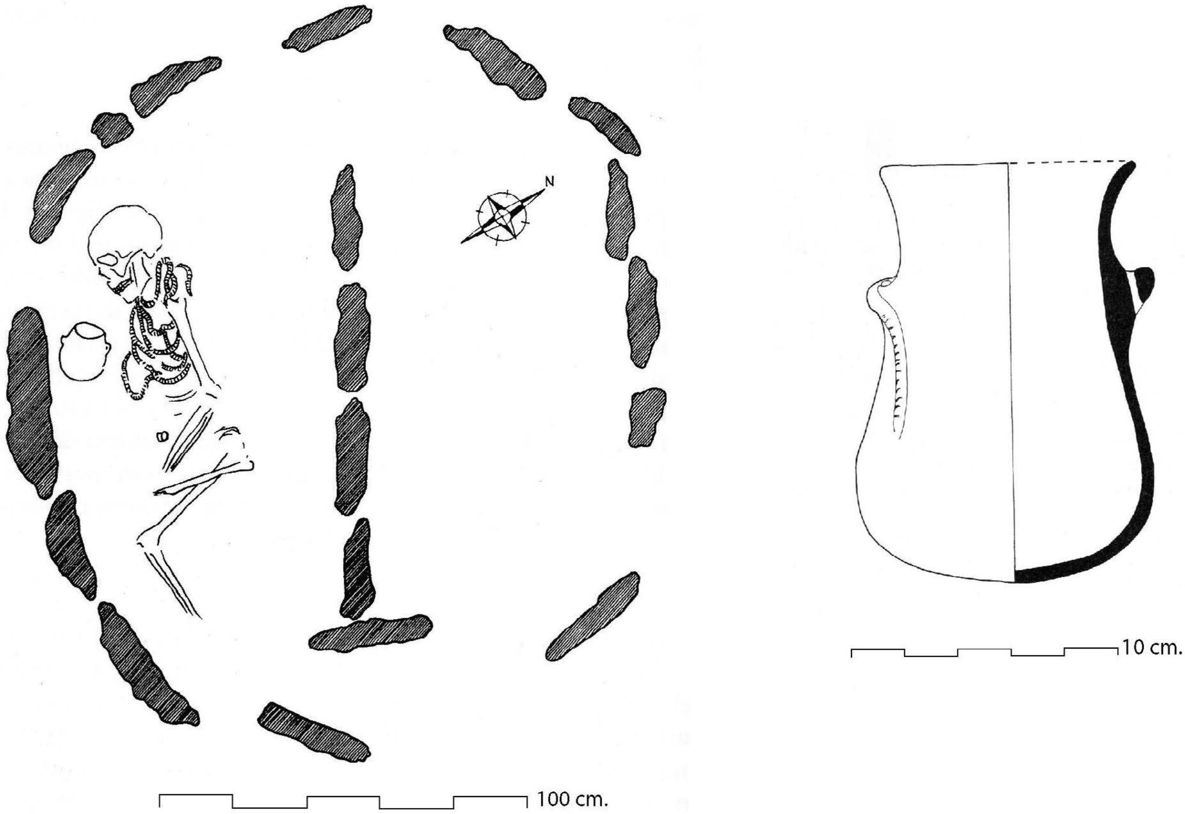

Figure 5. Mas Masdenvergenc (Amposta, Tarragona) Burial X (after Esteve Gálvez, 2000, Figs.9 and 10).

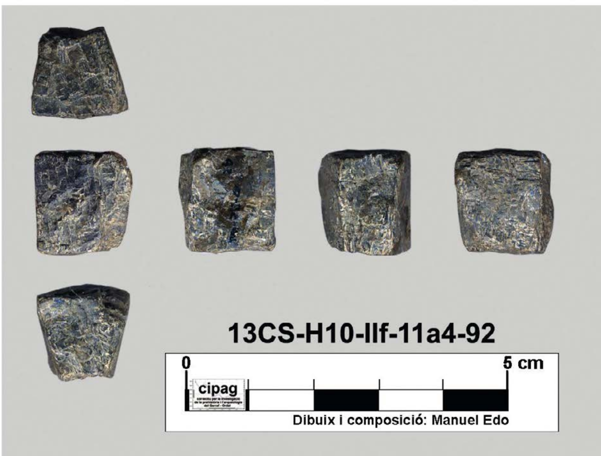

Figure 6. Galena samples from Neolithic levels at Can Sadurni (Begues, Barcelona). Photo: M. Edo.

galena fragment weighing 341.5 g was found in Sector A (grid JC44, UE2001) in an area with structures associated with a midden. Two different building traditions overlap in this settlement. In the earliest, huts were built on wooden platforms and in the more recent, they were built over the previous structures on stone-paved surfaces. These phases overlap from a chronological point of view, covering a period from 5210 to 4800 cal BC and both can be attributed to the Early Neolithic-Cardial facies (Andreaki, et al., 2020 and 2022; Terradas, Palomo and Piqué, 2020).

From the Cave of Can Sadurní (Begues, Barcelona) (Figure 4: 13) (Edo, et al., 2019) come two small, roughly cubic-shaped fragments (13CS-H10-IIJ-11a4-92 and

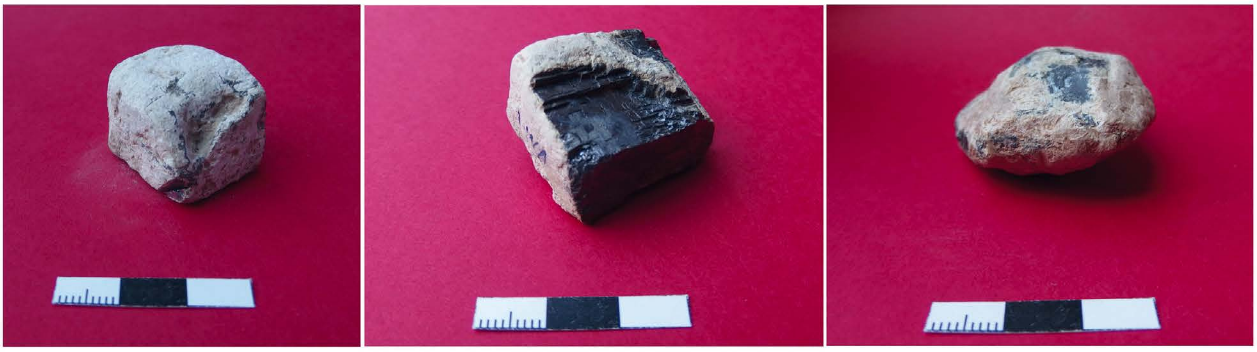

Figure 7. Galena samples from La Planeta (PA257762), Vall Major I (PA26456) and Vall Major 3 (PA25781) (from left to right). Photo: J. Gallart.

13CS-I11-IIF-11a1-17) weighing 17 and 33 g respectively (Figure 6). They both were found in the layers of the post-Cardial Phase I (Layers 11 a4 and 11 a1). The first one is connected with the evidence of stabling cattle, but the second, found in Layer 11 a1, is related to a funerary context. This galena fragment cannot be associated with a particular burial, as most human bones are fragmented, largely unconnected, and associated with very mixed grave goods. It is therefore likely that this fragment was part of a set of grave goods. This would provide evidence of galena in both domestic and funerary contexts. The dates obtained from these layers (Edo, et al., 2019) place the occupation period between 4300-4000 cal BC for Phase 11 a1, and between 4550-4350 cal BC for Phase 11 a4. There is a third fragment of galena (CS-MVB_SE-773) that was not found during the recent works but is part of the collection of finds made before the systematic excavation. Its chronological attribution is questionable, given the extensive sequence of the archaeological site.

Another unpublished fragment of galena (29.3 g) was found in the Can Figueres Cave (Begues, Barcelona) (Blasco, Edo and Villalba, 2011), near Can Sadurní (Figure 4). It came from Layer 4 where there were various burials dating between the Late Neolithic and the Chalcolithic.

The last fragment of galena found during a systematic excavation are from El Molló (Mora la Nova, Tarragona) (Figure 4: 2), an open-air settlement attributed to an Epicardial Neolithic (Piera, et al., 2017; Molist, et al., 2020). The fragments of galena were found in a domestic structure, in Sector I. Although it has not yet been possible to obtain a ^{14}C dating, based on the contextualisation with the items of the archaeological record (mainly pottery and stone), the occupation is dated to the Late Epicardial Ancient Neolithic (first half of the 5th millennium BC).

The archaeological surveys carried out over the years on the western Catalan plain (Lleida province) (Figure 4: 4-11) (Bosch, et al., 2022) have identified several Neolithic sites with galena blocks. In total, nine galena pieces have been found in eight different sites in this area, in addition to those already mentioned in La Planeta (Table 1) (Figure 7). In all cases, they are surface archaeological sites of variable sizes, located on fluvial terraces or gentle slopes near rivers and torrents that flow into the River Segre. They are also in places that are highly suitable for agriculture. The typochronological classification of the associated finds (pottery, knapped stone, polished objects, hand mills, shell and lithic ornaments, etc.) allows a chronological classification of the galena finds into the early Epicardial and post-Cardial Neolithic (late 6th-first half of the 5th millennium BC) (Bosch, et al., 2022). There is a significant variation in the size and weight of the galena fragments recovered, ranging from the 203 g of a piece from Serra de Castelldans to the 42 g of one from Serra de Puigverd.

Galena was not only found in the Catalan area, although the compiled information is scarce and not as exhaustive as that presented for the northeast of the Iberian Peninsula. For example, one can highlight the most interesting finds from Cova de L'Or (Beniarrés, Alicante) (Figure 4: A), discovered in the 1950s and 60s. They are one cube and three spherical pieces (Figure 6) associated with Early Cardial Neolithic strata dating from 5400-5200 BC (Martí Oliver, 1977). The finds are deposited in the Alcoi Museum (Inv. No. 17482) and presents the following characteristics: a highly magnetic iron oxide ball weighing 130 g; a second, smaller ball with reddish tones and a polished surface that, when scratched, reveals silvery tones and has been identified by SEM-EDX as galena (weight = 79.4 g, size 31 x 28.1 x 25.8 mm); and third a 103.5 g cubic fragment of iron oxide (Figure 8).

Another ball from the Cova de l'Or is deposited in the Museu de Prehistòria – Servei d'Investigacions Prehistòriques (SIP) in Valencia. It was found in the Crevice F (No. 105,418) and has also been identified as galena by SEM-EDX. This piece has a partially fractured surface and is more irregular in its manufacture than the previous one. We were able to take a sample for lead isotope

Figure 8. Items from the Cova de L´Or (Beniarrés, Alicante) in the Alcoi Museum. Left to right: iron ball, galena ball and iron cube. Photo: T. Orozco.

testing from this last piece. The most remarkable thing is, apart from its age, that both galena pieces were worked and polished to give them a spherical shape.

Another galena fragment was found in Los Cascajos (Los Arcos, Navarre) (Figure 4: C), a Neolithic open-air settlement where a total of 36 individuals were recovered from graves grouped in a cemetery (García-Gazólaz and Sesma-Sesma, 2007). Grave goods were generally scarce but in structure 182 a young woman was buried with some bone beads, two fragments of pottery and a rounded fragment of galena[1]. There is no ^{14}C date for this burial, but the context suggests a date in the Early Neolithic, probably ca. 4650–4400 cal BC (subphase Iii) (Fernández-Crespo, et al., 2019).

Galena fragments have also been recorded in the Basque Country, although here it is difficult to make a clear chronological distinction between the Neolithic and the Early Bronze Age. A fragment of galena and a quartz crystal were found during the excavation of the Loa dolmen in 1922 (Altuna, et al., 1982). Another find came from the open-air settlement of Haltzerreka, where a block of galena and another of lignite were recovered. That site is dated to the Early Bronze Age (Mujika, et al., 2009).

Analytical investigations methods

It was possible to sample most of the fragments compiled in Table 1 (Figure 4) for lead isotope analysis. Only a few samples were large enough for parallel analysis to identify trace element composition via ICP-MS (Table 2) with greater precision than preliminary XRF or SEM-EDX analyses. The pXRF analyses were performed with an INNVOV-X portable equipment from the Museo Arqueológico Nacional in Madrid (Rovira Llorens and Montero-Ruiz, 2018). The SEM EDX used is a Hitachi S-3400N-Type II with a EDX spectrometer Bruker Quantax 4010 (SSD) from the MICROLAB (IH-CSIC). The mass spectrometry analyses were carried out in the laboratories of the General Geochronology Service at the University of the Basque Country (SGIker), with a MC-ICP-MS Neptune for lead isotope analyses (Rodríguez, et al., 2020) and a Q-ICP -MS iCAP Qc for elemental analyses (García de Madinabeitia, Sánchez Lorda and Gil Ibarguchi, 2008).

The provenance of galenas found in archaeological contexts

The presence of galena at Palaeolithic and Neolithic sites distributed over a large area of Catalonia led us to question the use of local resources, since there are numerous lead mines in the region (Mata Perelló, 1990) mainly exploited during the 19th and 20th centuries, but some of them with finds dated to the Roman period (Rafel and Armada, 2010). Indirect evidence based on lead isotopes analysis moves the origin of lead mining back to the 8th-6th century cal BC (Montero-Ruiz, et al., 2008; Murillo-Barroso, et al., 2016; Rafel, et al., 2019).

Chemical analysis

The elemental characterisation of the archaeological and geological galena by Q-ICP-MS (Table 2) confirms that they all are lead minerals without appreciable admixture of zinc (ruling out the presence of sphalerite ZnS). They all have a low silver content (<500 ppm) and show a range in their various quantified elements, including the rare earths.

Among the results, the high copper content detected in samples AR 70-041 from Abric Romaní and PA22823 from the Tivissa stands out, while it is absent in the Vall Major I sample. The uranium content is high in the same sample from Abric Romaní and in that of La Draga (>10 ppm), while bismuth stands out in sample PA26455 from Vall Major I (>100 ppm). The Linda Mariquita 3 and Tivissa samples present the highest proportions in most of the elements linked to the rare earth elements (REE).

Lead isotope analysis

The isotopic characterisation of geological samples from lead mines in the northeastern Iberian Peninsula had already been undertaken in research projects led by some of the authors (Nuria Rafel and Ignacio Montero).

Table 2: Elemental characterisation of some geological and archaeological galena by Q-ICP-MS (SGIKer - UPV/EHU). Values in ppm, MDL: method detection limit.

Sample	Na	Mg	Al	Ca	Ti	V	Cr	Co	Ni	Cu	Zn	Ga	As	Se
Linda Mariquita- 1	122.0	99.0	710.6	196.1	6.15	2.28	1.60	2.79	25.15	699.8	619.9	1.06	10.19	3.79
Linda Mariquita- 2	104.6	61.6	93.2	282.7	2.42	0.20	0.54	0.15	21.75	182.7	17.82	0.48	0.52	4.68
Linda Mariquita- 3	119.8	58.3	163.6	299.9	5.04	1.76	1.38	0.63	5.32	141.6	93.61	0.59	8.33	1.69
Mina Martollerense	19.5	73.4	303.2	5425.3	0.71	0.52	0.57	12.2	11.77	208.7	51.45	0.23	15.30	<MDL
Abric Romani AR 70-041	48.7	12.1	6.26	483.1	<MDL	43.7	9.78	262.5	65.70	1568.8	3.28	0.09	11.94	<MDL
Abric Romani AR V50-540	48.6	9.5	4.54	261.0	<MDL	31.0	10.0	<MDL	0.60	147.2	1.53	0.04	1.29	<MDL
Vall Major I PA26455	41.3	140.0	<MDL	1608.0	59.1	2.81	2.08	1.26	4.36	<MDL	3.70	2.28	8.46	61.4
Vall Major I PA26456	47.2	127.8	<MDL	1428.1	54.0	2.51	2.12	1.19	3.46	<MDL	2.83	2.16	8.58	43.4
Tivissa PA22823	323.9	537.1	4805.7	1363.0	23.9	30.6	7.23	0.40	3.25	2913.6	689.61	0.54	<MDL	2.54
La Draga PA25784	272.5	101.8	177.3	597.4	7.35	2.70	3.87	0.12	21.81	198.8	16.65	0.45	1.97	2.13
Can Sadurni CS-17	57.2	137.4	8.49	1555.3	51.0	3.00	2.40	1.48	4.14	246.1	9.92	2.02	8.63	37.9
Can Sadurni CS-92	120.5	179.2	80.9	3319.6	59.9	3.90	3.59	1.35	3.84	<MDL	34.47	2.23	9.68	43.9

Sample	Rb	Sr	Y	Zr	Ag	Cd	In	Cs	Ba	La	Ce	Pr	Nd	Sm	Eu
Linda Mariquita- 1	3.10	180.9	6.17	1.41	49.11	5.57	0.33	1.94	171.9	1.64	5.10	0.72	3.40	1.16	0.46
Linda Mariquita- 2	3.34	201.8	0.65	0.08	51.26	6.05	0.47	1.47	30.54	0.86	0.94	0.20	0.82	0.25	0.14
Linda Mariquita- 3	3.54	416.0	2.51	0.52	38.16	4.86	0.31	1.07	19.11	10.7	3.06	3.07	11.27	2.25	0.55
Mina Martollerense	1.17	10.1	3.24	0.31	301.6	2.23	0.59	0.43	70.11	0.69	2.10	0.34	1.65	0.62	0.22
Abric Romani AR 70-041	<MDL	331.1	0.22	0.03	97.51	1.21	0.03	0.03	18.41	0.08	0.16	0.03	0.15	0.09	0.02
Abric Romani AR V50-540	<MDL	142.0	0.01	0.01	108.6	1.42	0.02	0.02	10.89	0.03	0.05	0.01	0.02	0.01	0.00
Vall Major I PA26455	1.23	3.6	0.44	0.86	396.4	11.1	0.81	0.24	0.42	0.15	0.12	0.08	0.41	0.46	0.10
Vall Major I PA26456	1.19	1.6	0.36	0.80	222.8	8.71	0.23	0.21	0.27	0.13	0.11	0.08	0.38	0.42	0.10
Tivissa PA22823	3.01	181.6	3.39	<MDL	27.67	0.00	0.00	0.00	27.13	5.75	4.25	1.19	6.60	1.35	<MDL
La Draga PA25784	3.48	99.2	0.20	0.19	19.06	4.53	0.02	0.99	18.47	0.21	0.50	0.05	0.24	0.05	0.01
Can Sadurni CS-17	1.05	2.2	0.43	1.01	231.4	9.40	0.50	0.20	7.03	0.20	0.16	0.08	0.41	0.37	0.09
Can Sadurni CS-92	1.48	7.1	0.55	2.04	177.4	4.91	0.58	0.25	19.12	0.30	0.27	0.11	0.54	0.47	0.10

Sample	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Hf	Tl	Bi	Th	U
Linda Mariquita- 1	1.34	0.23	1.23	0.22	0.56	0.07	0.41	0.07	0.13	0.48	10.3	0.11	1.85
Linda Mariquita- 2	0.26	0.04	0.19	0.03	0.06	0.01	0.03	<MDL	0.12	0.37	9.4	0.00	0.34
Linda Mariquita- 3	1.26	0.16	0.70	0.10	0.27	0.04	0.22	0.03	0.11	0.33	10.6	0.02	0.34
Mina Martollerense	0.71	0.11	0.57	0.09	0.23	0.03	0.18	0.03	0.00	1.16	22.8	0.11	0.05
Abric Romani AR 70-041	0.11	0.02	0.10	0.01	0.03	0.00	0.01	0.00	0.00	0.12	14.2	0.00	16.67
Abric Romani AR V50-540	0.01	0.00	0.00	0.00	0.00	<MDL	0.00	0.00	0.00	0.11	25.9	0.00	0.59
Vall Major I PA26455	0.28	0.04	0.16	0.04	0.11	0.03	0.14	0.03	0.18	<MDL	531.5	0.25	0.18
Vall Major I PA26456	0.26	0.04	0.15	0.04	0.10	0.03	0.12	0.03	0.11	0.37	70.5	0.23	0.16
Tivissa PA22823	1.12	<MDL	0.55	<MDL	<MDL	<MDL	<MDL	<MDL	0.00	0.00	84.3	<MDL	4.12
La Draga PA25784	0.06	0.01	0.04	0.01	<MDL	0.01	0.01	<MDL	0.23	1.04	10.4	0.02	12.41
Can Sadurni CS-17	0.27	0.03	0.14	0.03	0.10	0.03	0.12	0.03	0.72	0.37	59.3	0.21	0.15
Can Sadurni CS-92	0.31	0.04	0.16	0.04	0.11	0.03	0.14	0.03	0.35	0.34	15.6	0.25	0.19

Together with the data collected from the references (Table 3), this allows us to make a relatively comprehensive comparison between the main mining areas in the different units of the Catalan coastal mountain ranges in the provinces of Girona, Barcelona and Tarragona (Canals and Cardellach, 1997; Montero-Ruiz, et al., 2009a; 2009b; Montero-Ruiz, 2017) and the Pyrenees on the Spanish and French sides (Marcoux, Joubert and Lescuyer, 1991; Cardellach, Canals and Pujals, 1996; Munoz, et al., 2016). In order to test the hypothesis of a local provenance, it was necessary to incorporate new samples from mines in the province of Barcelona

Table 3. Lead isotopes of geological samples from the NE of Iberia: MBF= Molar-Bellmunt-Falset; CCR= Catalan Coastal Ranges; Provinces: An= Andorra; B=Barcelona; GI= Girona; L= Lleida; TA= Tarragona.

Mine	Zone	Region	Mineral	Province	Id.	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{206}Pb$	Reference	Technique
Berta	Central	CCR	Galena	B		18.5168	15.7154	38.9213	0.84871	2.10195	This paper	MC-ICP-MS
Berta	Central	CCR	Galena	B		18.51	15.725	38.951	0.8495	2.1043	Canals and Cardellach, 1997	TIMS
Berta	Central	CCR	Galena	B		18.511	15.727	38.942	0.8496	2.1037	Canals and Cardellach, 1997	TIMS
Berta	Central	CCR	Galena	B		18.509	15.714	38.888	0.8490	2.101	Canals and Cardellach, 1997	TIMS
Berta	Central	CCR	Galena	B		18.495	15.703	38.861	0.8490	2.1012	Canals and Cardellach, 1997	TIMS
Berta	Central	CCR	Galena	B		18.45	15.702	38.863	0.8511	2.1064	Canals and Cardellach, 1997	TIMS
Can Franquesa	Central	CCR	Galena	B	PA12757	18.4592	15.6952	38.7398	0.85025	2.09861	This paper	MC-ICP-MS
Gava	Garraf	CCR	Galena	B	PA 27669	18.3538	15.6821	38.5309	0.85443	2.09934	This paper	MC-ICP-MS
Martorell	Central	CCR	Galena	B		18.3174	15.6993	38.5846	0.85707	2.10645	This paper	MC-ICP-MS
Martorell	Central	CCR	Galena	B		18.3241	15.6994	38.5913	0.85676	2.10604	This paper	MC-ICP-MS
Martorell	Central	CCR	Galena	B		18.318	15.678	38.534	0.8559	2.1036	Canals and Cardellach, 1997	TIMS
Martorell	Central	CCR	Galena	B		18.366	15.708	38.629	0.8553	2.1033	Canals and Cardellach, 1997	TIMS
Martorell	Central	CCR	Galena	B		18.335	15.704	38.622	0.8565	2.1065	Canals and Cardellach, 1997	TIMS
Martorell	Central	CCR	Galena	B		18.366	15.707	38.609	0.8552	2.1022	Canals and Cardellach, 1997	TIMS
Martorell	Central	CCR	Galena	B		18.325	15.698	38.588	0.8566	2.1058	Canals and Cardellach, 1997	TIMS
Mina Torrent Bosc	Central	CCR	Galena	B	PA12758	18.46	15.696	38.747	0.85025	2.09891	This paper	MC-ICP-MS
Mina Torrent des Bruix	Central	CCR	Galena	B	PA13227	18.4532	15.6987	38.7512	0.85072	2.09992	This paper	MC-ICP-MS
Rigros	North	CCR	Galena	B		18.678	15.723	39.1	0.8418	2.0934	Canals and Cardellach, 1997	TIMS
Rigros	North	CCR	Galena	B		18.628	15.768	39.19	0.8465	2.1038	Canals and Cardellach, 1997	TIMS
Vallirana	Garraf	CCR	Galena	B	PA 27666	18.3523	15.6812	38.5277	0.85445	2.09934	This paper	MC-ICP-MS
Vallirana	Garraf	CCR	Galena	B	PA 27671	18.2429	15.6313	38.4014	0.85684	2.10500	This paper	MC-ICP-MS
Mina Castell	North	CCR	Galena	GI	PA12410	18.5155	15.6865	38.851	0.8472	2.0983	Montero, et al., 2009b	TIMS
Mina Castell	North	CCR	Galena	GI	PA12411	18.5208	15.6853	38.8296	0.8469	2.0965	Montero, et al., 2009b	TIMS
Mina Leonor	North	CCR	Galena	GI	PA12384	18.5402	15.6854	38.8917	0.8460	2.0977	Montero, et al., 2009b	TIMS
Mina Pepito	North	CCR	Galena	GI	PA12387	18.4879	15.6797	38.7388	0.84810	2.09530	This paper	MC-ICP-MS
Mina Pepito	North	CCR	Galena	GI	PA12386	18.4819	15.6922	38.7648	0.8491	2.0974	Montero, et al., 2009b	TIMS
Mina Pepito	North	CCR	Galena	GI	PA12388	18.478	15.6824	38.7354	0.8487	2.0962	Montero, et al., 2009b	TIMS
Mina W	North	CCR	Galena	GI	PA12762	18.483	15.6923	38.7693	0.84901	2.09751	This paper	MC-ICP-MS
Mina Leonor	North	CCR	Galena	GI	PA12377	18.5388	15.6854	38.8948	0.8461	2.098	Montero, et al., 2009b	TIMS
Pideval Este	North	CCR	Galena	GI	PA12761	18.5361	15.6783	38.8729	0.84582	2.09709	This paper	MC-ICP-MS
Puig Perals	North	CCR	Galena	GI	PA12389	18.5188	15.6863	38.8432	0.84705	2.09745	This paper	MC-ICP-MS

Mine	Zone	Region	Mineral	Province	Id.	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{206}Pb$	Reference	Technique
Puig Perals	North	CCR	Galena	GI	PA12390	18.5167	15.6822	38.8336	0.8503	2.0972	Montero, et al., 2009b	TIMS
Balcoll	MBF	CCR	Native Silver	TA	PA11658	18.3584	15.6789	38.5289	0.85404	2.09860	Montero-Ruiz, Murillo Barroso and Bartelheim, 2014	MC-ICP-MS
Balcoll	MBF	CCR	Native Silver	TA	PA11657	18.3582	15.6696	38.4994	0.85356	2.09708	Montero-Ruiz, Murillo Barroso and Bartelheim, 2014	MC-ICP-MS
Barranco Hondo	MBF	CCR	Galena	TA	PA12274	18.31	15.681	38.494	0.85643	2.10236	Montero-Ruiz, 2017	MC-ICP-MS
Barranco Hondo	MBF	CCR	Galena	TA	PA11997	18.301	15.662	38.44	0.85578	2.1004	Montero-Ruiz, et al., 2009a	TIMS
Barranco Hondo	MBF	CCR	Galena	TA	PA11998	18.313	15.677	38.487	0.85607	2.1017	Montero-Ruiz, et al., 2009a	TIMS
Cueva del Paraguas	MBF	CCR	Galena	TA	PA11968	18.299	15.667	38.448	0.85618	2.1011	Montero-Ruiz, 2017	TIMS
Jalapa	MBF	CCR	Galena	TA	PA12288	18.311	15.68	38.493	0.85632	2.10213	Montero-Ruiz, 2017	MC-ICP-MS
Jalapa	MBF	CCR	Galena	TA	PA12289	18.336	15.672	38.499	0.85471	2.09964	Montero-Ruiz, 2017	MC-ICP-MS
Linda Mariquita	MBF	CCR	Galena	TA	PA12272	18.316	15.664	38.451	0.85523	2.0993	Montero-Ruiz, 2017	TIMS
Linda Mariquita	MBF	CCR	Galena	TA	PA12273	18.308	15.661	38.446	0.85539	2.0999	Montero-Ruiz, 2017	TIMS
Linda Mariquita	MBF	CCR	Galena	TA	PA11956	18.3571	15.6859	38.5557	0.85449	2.1003	Montero-Ruiz, 2017	MC-ICP-MS
Linda Mariquita	MBF	CCR	Galena	TA	PA11956	18.3534	15.6953	38.5896	0.85516	2.10267	Montero-Ruiz, 2017	MC-ICP-MS
Linda Mariquita	MBF	CCR	Galena	TA	PA10482	18.338	15.675	38.502	0.85481	2.0996	Montero-Ruiz, 2017	TIMS
Linda Mariquita	MBF	CCR	Galena	TA	LM-1	18.3212	15.6822	38.5017	0.85596	2.10148	This paper	MC-ICP-MS
Linda Mariquita	MBF	CCR	Galena	TA	LM-2	18.3319	15.683	38.5058	0.85550	2.10047	This paper	MC-ICP-MS
Linda Mariquita	MBF	CCR	Galena	TA	LM-3	18.3238	15.6783	38.4956	0.85562	2.10085	This paper	MC-ICP-MS
Linda Mariquita	MBF	CCR	Galena	TA	PA11954	18.323	15.68	38.503	0.85572	2.1013	Montero-Ruiz, et al., 2009a	TIMS
Linda Mariquita	MBF	CCR	Galena	TA	PA11954	18.322	15.677	38.495	0.85567	2.1011	Montero-Ruiz, et al., 2009a	TIMS
Linda Mariquita	MBF	CCR	Cobre	TA	PA11996	18.368	15.658	38.506	0.85244	2.0963	Montero-Ruiz, et al., 2009a	TIMS
Linda Mariquita	MBF	CCR	Galena	TA	PA12007	18.431	15.688	38.655	0.85116	2.0973	Montero-Ruiz, et al., 2009a	TIMS
Linda Mariquita	MBF	CCR	Galena	TA	PA12008	18.322	15.688	38.524	0.85626	2.1026	Montero-Ruiz, et al., 2009a	TIMS
Linda Mariquita	MBF	CCR	Galena	TA	PA12008	18.316	15.68	38.498	0.85611	2.1019	Montero-Ruiz, et al., 2009a	TIMS
Mina Regia	MBF	CCR	Galena	TA	PA11992	18.301	15.671	38.46	0.85631	2.1015	Montero-Ruiz, et al., 2009a	TIMS
Mina Regia	MBF	CCR	Galena	TA	PA11993	18.314	15.686	38.511	0.85651	2.1028	Montero-Ruiz, et al., 2009a	TIMS
Mineralogia	MBF	CCR	Galena	TA	PA11985	18.32	15.692	38.533	0.85658	2.1033	Montero-Ruiz, et al., 2009a	TIMS
Mineralogia	MBF	CCR	Galena	TA	PA11985	18.317	15.689	38.521	0.85652	2.1030	Montero-Ruiz, et al., 2009a	TIMS
Mineralogia	MBF	CCR	Galena	TA	PA11987	18.309	15.676	38.483	0.85623	2.1019	Montero-Ruiz, et al., 2009a	TIMS
Mineralogia	MBF	CCR	Galena	TA	PA11987	18.318	15.688	38.52	0.85642	2.1028	Montero-Ruiz, et al., 2009a	TIMS

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Mineralogia	MBF	CCR	Galena	TA	PA11988	18.301	15.667	38.453	0.8561	2.1011	Montero-Ruiz, et al., 2009a	TIMS
Raimunda	MBF	CCR	Galena	TA	PA11994	18.333	15.691	38.545	0.8559	2.1024	Montero-Ruiz, et al., 2009a	TIMS
Raimunda	MBF	CCR	Galena	TA	PA11994	18.333	15.692	38.545	0.8559	2.1024	Montero-Ruiz, et al., 2009a	TIMS
Raimunda	MBF	CCR	Galena	TA	PA11995	18.317	15.672	38.483	0.8556	2.1009	Montero-Ruiz, et al., 2009a	TIMS
Mina Besso	Montsant	CCR	Cobre	TA	PA20277	18.341	15.676	38.47	0.85471	2.0975	Montero-Ruiz, 2017	MC-ICP-MS
Mina Besso	Montsant	CCR	Galena	TA	PA20281	18.352	15.675	38.484	0.85412	2.0970	Montero-Ruiz, 2017	MC-ICP-MS
Minas des Closos	Montsant	CCR	Galena	TA	PA20606	18.4816	15.6894	38.7559	0.84892	2.09701	This paper	MC-ICP-MS
Minas des Closos	Montsant	CCR	Galena	TA	PA20607	18.4774	15.6867	38.7443	0.84897	2.09685	This paper	MC-ICP-MS
Vail de Ribes	Andorra	Pyrinees	Galena	An	377-g	18.406	15.702	38.595	0.8531	2.0969	Romer and Soler, 1995	TIMS
Vail de Ribes	Andorra	Pyrinees	Galena/ sphalerite	An	377-sp	18.399	15.716	38.589	0.8542	2.0973	Romer and Soler, 1995	TIMS
Vail de Ribes	Andorra	Pyrinees	Galena	An	377-g2	18.401	15.704	38.574	0.8534	2.0963	Romer and Soler, 1995	TIMS
Vail de Ribes	Andorra	Pyrinees	Galena	An	478-a	18.426	15.706	38.59	0.8524	2.09432	Romer and Soler, 1995	TIMS
Vail de Ribes	Andorra	Pyrinees	Sphalerite	An	478-b	18.416	15.683	38.51	0.8516	2.09112	Romer and Soler, 1995	TIMS
Les Ferreres	Eastern	Pyrinees	Cu	GI	PA21878	18.5839	15.7114	38.8241	0.84543	2.08913	Montes Landa, et al., 2021	MC-ICP-MS
Les Ferreres	Eastern	Pyrinees	Cu	GI	PA21879	18.5807	15.7103	38.8173	0.84552	2.08912	Montes Landa, et al., 2021	MC-ICP-MS
Les Ferreres	Eastern	Pyrinees	Cu	GI	ROC-1	18.55473	15.70804	38.78196	0.84658	2.09014	Montes Landa, et al., 2021	MC-ICP-MS
Les Ferreres	Eastern	Pyrinees	Cu	GI	ROC-3	18.55585	15.70868	38.782	0.84656	2.09001	Montes Landa, et al., 2021	MC-ICP-MS
Bielsa-Parzán	Central	Pyrinees	Galena	HU		18.488	15.753	38.563	0.85207	2.0858	Subias, et al., 2015	MC-ICP-MS
Bielsa-Parzán	Central	Pyrinees	Galena	HU		18.518	15.678	38.912	0.84664	2.1013	Subias, et al., 2015	MC-ICP-MS
Bizielle	Central	Pyrinees	Galena	HU		18.472	15.691	38.722	0.84945	2.0962	Subias, et al., 2015	MC-ICP-MS
Bizielle	Central	Pyrinees	Galena	HU		18.48	15.747	38.751	0.85211	2.0969	Subias, et al., 2015	MC-ICP-MS
Panticosa	Central	Pyrinees	Galena	HU		18.437	15.768	38.792	0.85524	2.1040	Subias, et al., 2015	MC-ICP-MS
Portalet	Central	Pyrinees	Galena	HU		18.411	15.685	38.585	0.85194	2.0958	Subias, et al., 2015	MC-ICP-MS
South Mine Peak	Central	Pyrinees	PbZn	HU	LC5022-3	18.087	15.69	38.299	0.86747	2.11750	Garcia San Segundo, et al., 2014	MC-ICP-MS
South Mine Peak	Central	Pyrinees	PbZn	HU	LC5022-2	18.094	15.692	38.321	0.86725	2.1179	Garcia San Segundo, et al., 2014	MC-ICP-MS
Tebarray	Central	Pyrinees	Galena	HU		18.079	15.662	38.251	0.86631	2.1158	Subias, et al., 2015	MC-ICP-MS
Tebarray	Central	Pyrinees	Galena	HU		18.094	15.684	38.326	0.86681	2.1182	Subias, et al., 2015	MC-ICP-MS
Tebarray	Central	Pyrinees	Galena	HU		18.117	15.71	38.384	0.86714	2.1187	Subias, et al., 2015	MC-ICP-MS
Tebarray	Central	Pyrinees	Galena	HU		18.138	15.737	38.505	0.86763	2.1229	Subias, et al., 2015	MC-ICP-MS
Yenefrito	Central	Pyrinees	Galena	HU		18.379	15.718	38.449	0.85522	2.0920	Subias, et al., 2015	MC-ICP-MS

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Yenefrito	Central	Pyrinees	Galena	HU		18.421	15.695	38.616	0.85202	2.0963	Subias, et al., 2015	MC-ICP-MS
Yenefrito	Central	Pyrinees	Galena	HU		18.424	15.696	38.612	0.85193	2.0957	Subias, et al., 2015	MC-ICP-MS
Yenefrito	Central	Pyrinees	Galena	HU		18.429	15.709	38.659	0.85241	2.0977	Subias, et al., 2015	MC-ICP-MS
Yenefrito	Central	Pyrinees	Galena	HU		18.44	15.723	38.708	0.85266	2.0991	Subias, et al., 2015	MC-ICP-MS
Font dels Lladres	Central	Pyrinees	Galena	L	FLL-2	18.064	15.696	38.288	0.86891	2.1196	Cardellach, Canals and Pujals, 1996	TIMS
Liat	Central	Pyrinees	Galena	L	LT-16	18.042	15.671	38.213	0.86858	2.118	Cardellach, Canals and Pujals, 1996	TIMS
Liat	Central	Pyrinees	Galena	L	LT-2	18.054	15.671	38.213	0.86801	2.1166	Cardellach, Canals and Pujals, 1996	TIMS
Margalida	Central	Pyrinees	Galena	L	MG-13	18.021	15.656	38.157	0.86876	2.1174	Cardellach, Canals and Pujals, 1996	TIMS
Mauricio	Central	Pyrinees	Galena	L	M-11	18.045	15.67	38.198	0.86838	2.1168	Cardellach, Canals and Pujals, 1996	TIMS
Victoria	Central	Pyrinees	Galena	L	V-24	18.08	15.676	38.226	0.86704	2.1143	Cardellach, Canals and Pujals, 1996	TIMS

(Table 3). Some of these new geological samples were provided by the Barcelona Seminary Geology Museum (https://www.mgsb.es/), as it was often not possible to collect samples on site due to the closure or loss of access to most sites. From a comparative and interpretative perspective, it should be mentioned that the analyses performed using Thermal Ionisation Mass Spectrometry (TIMS) are less precise than those performed with MC-ICP-MS. This concerns in particular geological reference samples when ratios with the ^{204}Pb isotope as denominator are used, as shown by the comparison of TIMS analyses of OXALID with those of MC-ICP-MS analyses of samples from the mines of the Vera Basin (Almería) presented by Murillo-Barroso, et al. (2019). However, the new galena samples analysed in this project, when compared to previous TIMS analyses from the same mines, show no significant differences. This is reflected in the results published by Canals and Cardellach (1997) of the analyses of samples from the Martorell or Berta mines and the new LIA from the Linda Mariquita mine.

The analyses of archaeological samples (Table 4) show a concentration between 18.31 and 18.33 for the $^{206}Pb/^{204}Pb$ ratio and 15.68 and 15.69 for the $^{207}Pb/^{204}Pb$ ratio (Figure 8). Only 4 of the 18 Palaeolithic and Neolithic galena samples from northeastern Iberia diverge from this general trend. In three of them – La Draga (Girona), Can Sadurni-92 (Barcelona) and Vall Major 3 (Lleida) – the differences are very slight and only the fragment without context from Can Sadurni can be considered as truly different. This major concentration of the finds coincides fully with the main hub of the Molar-Bellmunt-Falset (MBF) mines in Tarragona, whose value distribution approach to close to 18.43 in the $^{206}Pb/^{204}Pb$ ratio (Figure 9). Considering the global isotopic field of the MBF area, the three samples mentioned above with somewhat different values could be integrated and the entire collection of Neolithic and Palaeolithic chronology could be explained within this single provenance. Yet, it cannot completely rule out a different origin for any of these three samples, perhaps related to mines in the Barcelona province, as we will discuss below. The rest of the lead mines, both in the Pyrenees and Girona and other areas of Tarragona and Barcelona can be ruled out as possible sources (Figure 9).

Of the sample material from the archaeological sites located in the same province as Barcelona, the four Middle Paleolithic samples from Abric Romani, one Neolithic galena from Can Sadurni, and another from Can Figueres are completely compatible with the MBF district and do not overlap with any of the mines in Barcelona province, including the Martorell mines mentioned earlier. Although the number of samples that characterise this isotopic field is low (7), compared to the MBF district (32), the trends shown in Figure 10 for the MBF and the Martorell mine are different and the distribution of most of the Neolithic galena is clearly outside the range of the Martorell mines. LIA of other mines in Barcelona province, located in the northern Garraf Massif (Vallirana, Gava and Cervelló) close to the Can Sadurni and Can Figueres archaeological sites, could be aligned

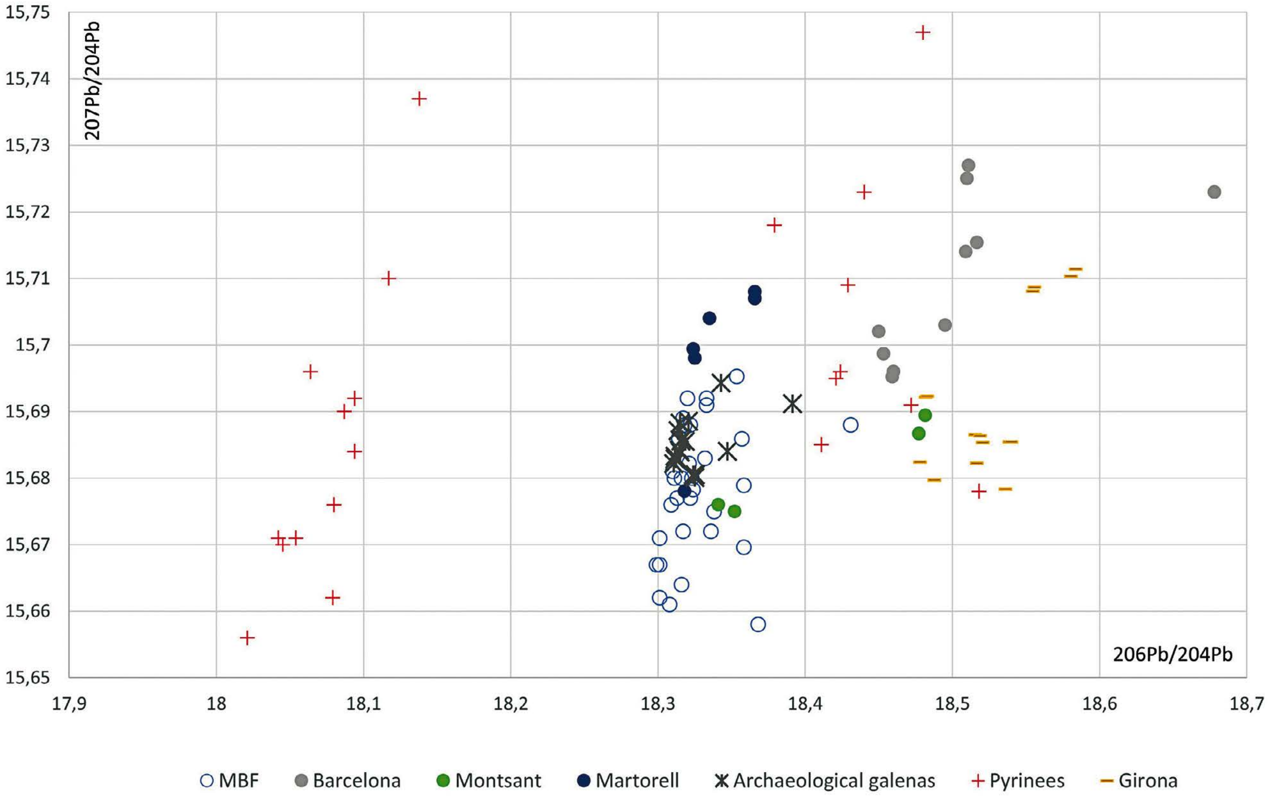

Figure 9. Results of the lead isotope analyses of galena found in archaeological sites from the Northeast in relation to the lead mines in the same area.

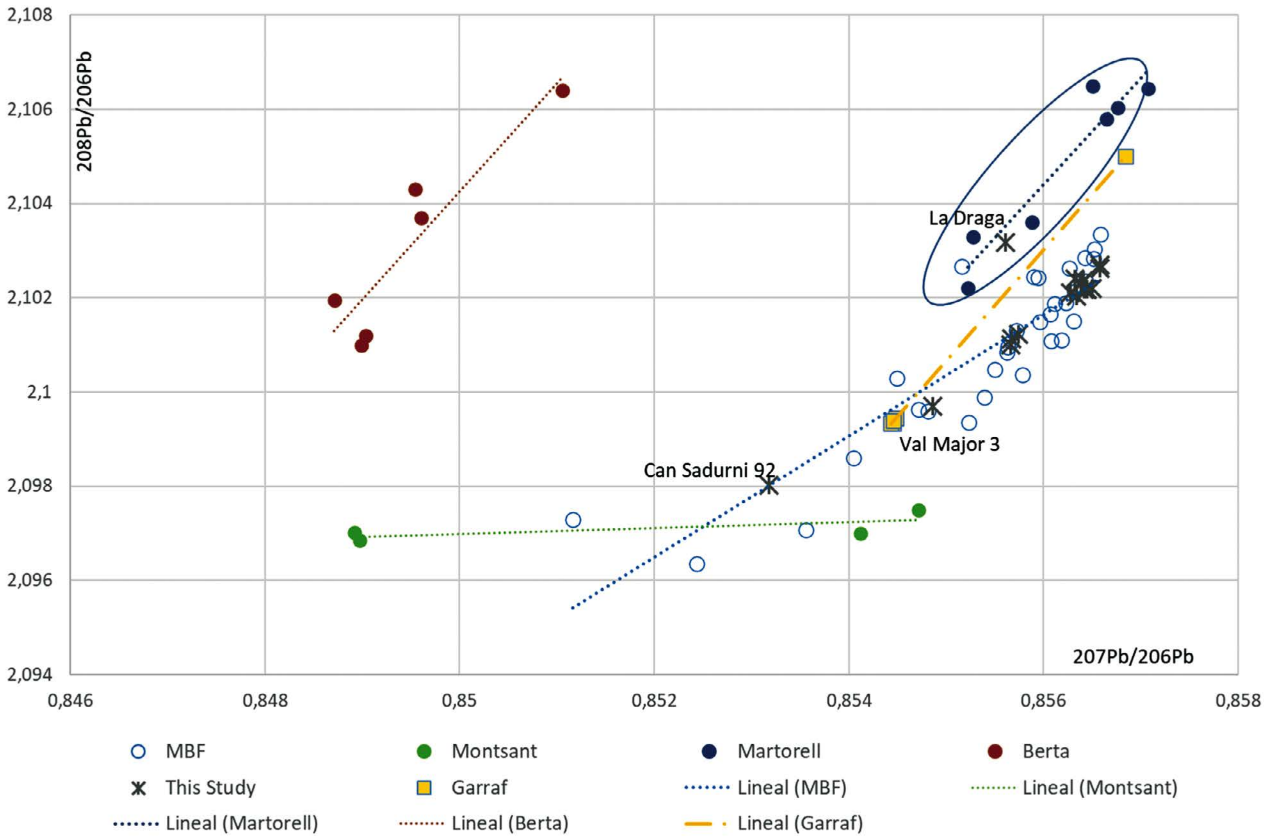

Figure 10. The graph shows the differences between the isotopic fields of the El Molar (MBF), the Martorell mines and the Berta mine and the position of the galena found in archaeological sites from the Northeast.

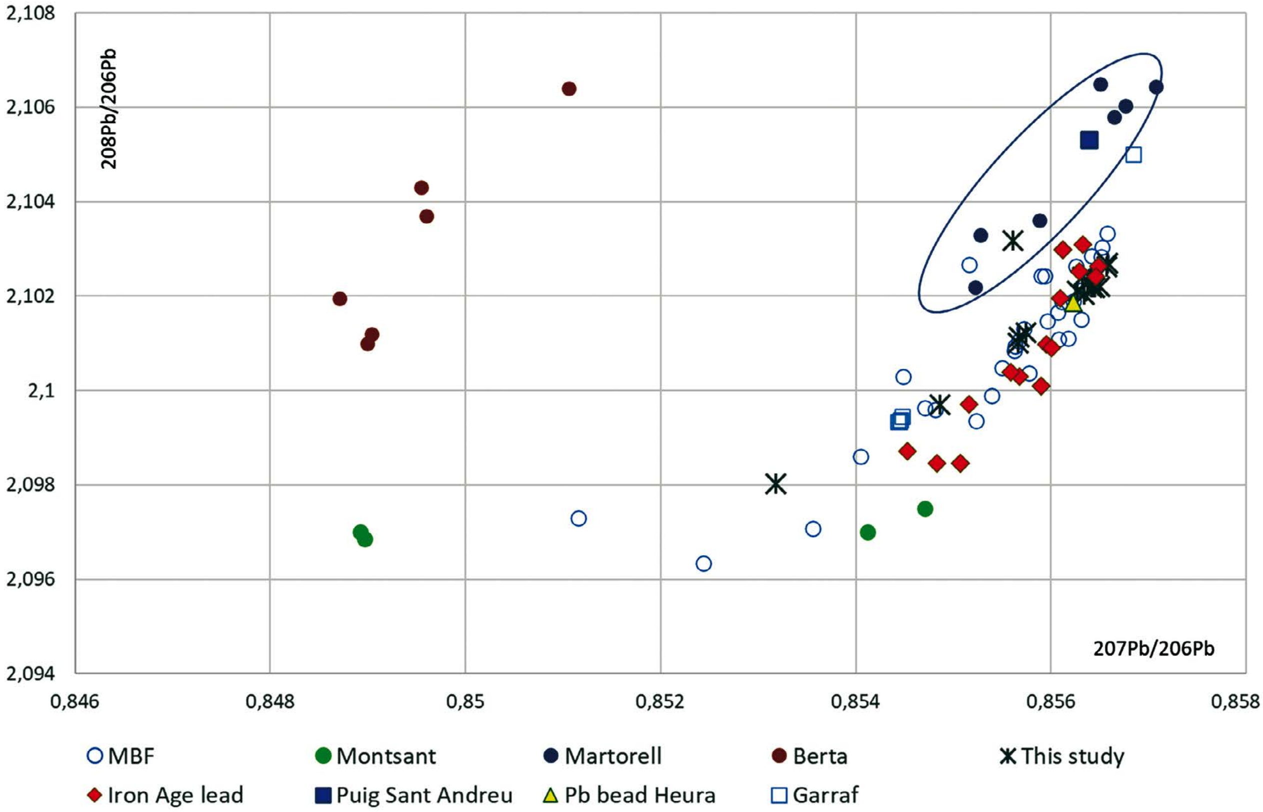

Figure 11. The graph shows the comparison of the distribution of the Palaeolithic and Neolithic galenas found in archaeological sites from the Northeast and the Iron Age finds.

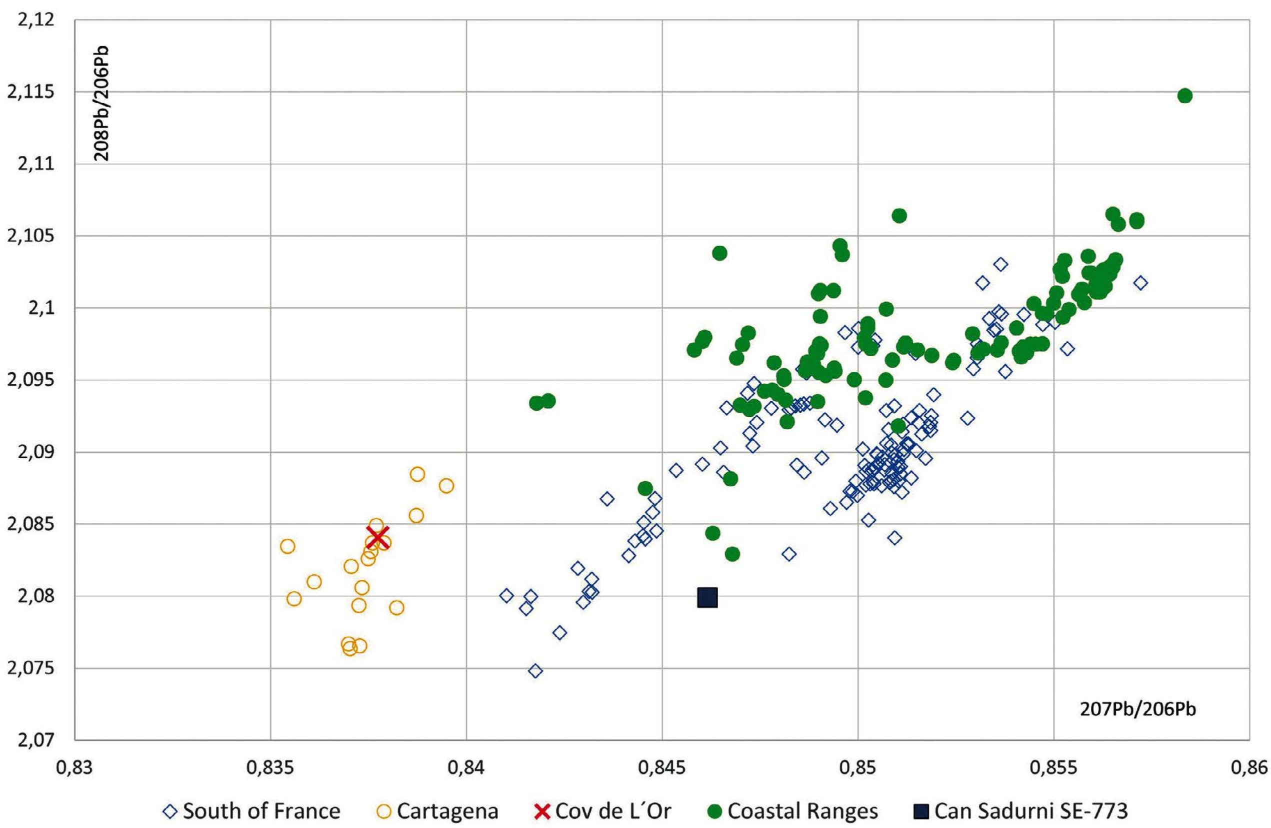

Figure 12. Graphical comparison of the lead isotope ratios of the galena from Cova de L'Or and the galena without context from Can Sadurni.

with the same trend as Martorell and partially overlap with MBF samples in an area that only affects the galena from Can Sadurni-92 and Vall Major 3 (Figure 10).

Conclusion

Spatial relationship between archaeological sites and mining sites

In summary, although the Martorell, Berta, Vallirana, Gava and Cervelló mines are geographically the closest to the Can Sadurni, Can Figueres and Abric Romani archaeological sites, most of the galena from those three sites fits the Molar-Bellmunt-Falset (MBF) isotopic field, whether they are from the Palaeolithic or the Neolithic.

If we compare the galena of this study with other archaeological samples from later periods, a fully coincidence between them can be appreciated (Figure 11). This is relevant due to some lead isotope analysis of galena, lead and Pb slag from the Iron Age site of El Calvari de El Molar (Tarragona), located just 500 metres from the Linda Mariquita mine at one end of the MBF district, fully overlap. Some other samples spread over the MBF isotopic field, including those from Castellet de Banyoles (Tivissa, Tarragona), the site that controlled the exploitation of these mines in the 3rd century cal BC, just before the Romans (Montero-Ruiz, et al., 2008), makes it more likely that the provenance of Vall Major 3 (Figure 11) could be this mining district with a long historical exploitation. In addition, the lead isotope ratio from the bead from La Coveta de L'Heura (Tarragona) (Figure 11), the only known metallic lead object from the Chalcolithic, matches those of the lead ores from the MBF mines (Montero-Ruiz, 2017) and is grouped with the majority of the Neolithic and Palaeolithic galena of this study.

The only evidence of an ancient exploitation of the Martorell mines comes from a piece of galena found at the archaeological site of Ullastret (Puig de Saint Andreu, Girona) dated to the 3rd century BC (unpublished analysis, Table 4, Figure 11). It could have a provenance similar to that of the La Draga galena, although not the same like the sample of Can Sadurni 92. We do not have data to identify the use of other mines in the Garraf Massif during prehistory, and only the geographical factor of proximity leaves open the possibility that galena was extracted in the Neolithic. Regarding the galena from La Draga, an indirect reasoning that may support its origin from Martorell mines is the absence of flint remains from the MBF zone where there are abundant sources of siliceous rocks, sources providing a specific flint, whereas some remains of siliceous rocks from the lower Llobregat Basin have been recovered on the site. Specifically, there are some knapped flakes of Triassic jasper that outcrop on the Montjuïc hills, in Barcelona (Terradas, et al., 2012).

The exception to this model is the sample without context CS-MVB_SE-773 from Can Sadurni (Figure 11), whose provenance is complex because it presents unusual values for the Iberian Peninsula, with very low values in the ^{208}Pb isotope (thorogenic). It should be remembered that there is no reliable chronological context for the find and that it comes from an undocumented find context, which makes its assignment to an archaeological find complex questionable. Some samples from Mont Lozere and the Cevennes in the French Massif Central could have a certain similarity, but they do not offer a clear fit, seen especially in Figure 12 using ratios without the ^{204}Pb isotope. Some of the copper mineral samples from Mina Turquesa in the Montsant area or from the Albiol mines in Tarragona also show singular values in the ratios with the ^{208}Pb isotope and could be an option for the existence of mineralisation with that isotopic range. Although our reference samples are copper, in Albiol galena is mentioned in the Mas d'en Galofre mine (Mata Perelló, 1990). In addition, the distance between Can Sadurni and Albiol is just 70 km.

In conclusion, regardless of whether all the galena has a single origin (MBF) or a number of them could have another in the Martorell, Garraf or Albiol mines, there are indications that the mobility of the raw material can be assumed over medium distances.

La Draga lies 120 km in a straight line from the Martorell mines and 250 km from the MBF mines. For the Abric Romani (Palaeolithic), the distance to MBF would be closer, 90 km in a straight line, and even less for the rest of the Neolithic sites in Lleida province, the closest being El Molló, less than 10 km in a straight line from the El Molar mines.

The galena from Cova de L'Or (Beniarrés, Alicante) was found in a very different geographical area, which also lacks lead deposits in its immediate surroundings. Lead isotope analyses (Table 4) link the possible origin of this galena to the Cartagena (Murcia) mining district (Figure 12), 150 km in a straight line to the south. Here, too, we can assume medium mobility in the movement of the raw material.

About the early metallurgy in Western Mediterranean

The MBF mining district was the probable origin of most of the Neolithic galena identified in Catalonia, with other minor options around Martorell in the Barcelona prov-

Table 4.- Archaeological galenas from Iberia. Lead isotopes analysis by MC-ICP-MS. Province: B= Barcelona; GI= Girona; L=Lleida; TA= Tarragona; V=Valencia. Chronology: PAL= Paleolithic; NEO= Neolithic; H2= Iron Age II.

Sample	Prov.	Chrono.	Reference	$^{208}Pb/^{206}Pb$	Error (2SE)	$^{207}Pb/^{206}Pb$	Error (2SE)	$^{206}Pb/^{204}Pb$	Error (2SE)	$^{207}Pb/^{204}Pb$	Error (2SE)	$^{208}Pb/^{204}Pb$	Error (2SE)
Abric Romani	B	PAL	AR Ja K61 36	2.10219	0.00010	0.85645	0.00003	18.3107	0.0011	15.6822	0.0012	38.4926	0.0036
Abric Romani	B	PAL	O 42 nº1 7/5 30-8	2.10220	0.00007	0.85650	0.00002	18.3114	0.0010	15.6833	0.0010	38.4950	0.0032
Abric Romani	B	PAL	AR V50-540	2.10218	0.00008	0.85644	0.00002	18.3116	0.0009	15.6829	0.0010	38.4944	0.0027
Abric Romani	B	PAL	AR Ja 70-041	2.10218	0.00012	0.85642	0.00003	18.3124	0.0011	15.6831	0.0013	38.4958	0.0041
Can Figueres	B	NEO	SCF-8	2.10210	0.00004	0.85627	0.00001	18.3184	0.0009	15.6855	0.0008	38.5072	0.0021
Can Sadurni	B	NEO	13CS-I11-IIF-11a1-17	2.10240	0.00003	0.85633	0.00001	18.3208	0.0005	15.6886	0.0005	38.5177	0.0013
Can Sadurni	B	NEO	13CS-H10-IIJ-11a4-92	2.09802	0.00003	0.85318	0.00001	18.3914	0.0006	15.6912	0.0006	38.5854	0.0016
Can Sadurni		?	CS-MVB_SE-773	2.08006	0.00005	0.84623	0.00001	18.538	0.0008	15.6875	0.0008	38.5602	0.0022
La Draga	GI	NEO	A-6932	2.10318	0.00005	0.85561	0.00001	18.3429	0.0010	15.6943	0.0010	38.5783	0.0026
Planeta 2	L	NEO	PA25776	2.10203	0.00004	0.85634	0.00001	18.3150	0.0011	15.6839	0.0009	38.4987	0.0025
Planeta 1	L	NEO	PA25775	2.10237	0.00004	0.85641	0.00001	18.3156	0.0008	15.6856	0.0007	38.5061	0.0018
Tossal de L´Embrosi	L	NEO	PA25780	2.10123	0.00005	0.85574	0.00001	18.3239	0.0009	15.6805	0.0009	38.5026	0.0024
Serra Tinells	L	NEO	PA25782	2.10100	0.00005	0.85566	0.00002	18.3251	0.0009	15.6801	0.0008	38.5011	0.0022
Serra de Castelldans	L	NEO	PA25778	2.10113	0.00005	0.85567	0.00002	18.3253	0.0009	15.6805	0.0009	38.5040	0.0024
Vall Major-3	L	NEO	PA25781	2.09969	0.00005	0.85486	0.00001	18.3470	0.0010	15.6840	0.0009	38.5229	0.0024
Vall Major-I	L	NEO	PA26455	2.10263	0.00004	0.85658	0.00001	18.3138	0.0006	15.6871	0.0006	38.5071	0.0016
Vall Major-I	L	NEO	PA26456	2.10269	0.00004	0.85658	0.00001	18.3150	0.0005	15.6883	0.0006	38.5108	0.0016
El Molló	TA	NEO	PA25772	2.10229	0.00004	0.85638	0.00001	18.3145	0.0008	15.6842	0.0008	38.5023	0.0021
Masdenvergenc	TA	NEO	PA25507	2.10230	0.00004	0.85638	0.00001	18.3164	0.0009	15.6859	0.0009	38.5067	0.0023
Cova de L´Or	V	NEO	CL-SIP	2.08408	0.00005	0.83774	0.00001	18.7288	0.0011	15.6899	0.0010	39.0322	0.0027
Puig de Sant Andreu	GI	H2	PSA-UB	2.1053		0.8564		18.342		15.708		38.616	

ince. Copper is also present in the MBF district and lead isotope studies suggest the working of the copper mines at least as early as the Late Chalcolithic and Bell Beaker periods (for example, Palmela point from del Tossal de les Venes and dagger from the Cova de l'Arbonès) (Montero-Ruiz, 2017, Figs. 48, 50, 53; Soriano, et al., 2022).

Southern France and northeastern Iberia are adjacent regions with cultural links. Both areas are rich in galena ores that were used in Neolithic contexts during the 5th and 4th millennia Cal BC. The former had galena and malachite beads (Roscian, Claustre and Dietrich, 1992) and probably the metallurgical technology[2] prepared the way to transform them in lead and copper ornaments rather frequent in the Fontbouisse culture (3rd millennium BC), while in the latter they were absent (Murillo-Barroso and Montero-Ruiz, 2012; Montero-Ruiz, Murillo-Barroso and Ruiz-Taboada, 2021). Blocks of galena were found in Iberia, but not manufactured beads, as well as hundreds of variscite beads instead of malachite during the Neolithic (Borrell, et al., 2019). The only case of a lead bead (Coveta de l´Heura) made with local raw materials (from MBF) (Montero-Ruiz, 2017), and the only three copper beads from Cau d'en Serra (Martín Cólliga, et al., 1999, p.157; Murillo-Barroso and Montero-Ruiz, 2012, p.58) pending a provenance study to confirm a local or foreign origin, suggest that a similar process to the French one could have occurred, but in the end it did not. The different choice suggests the communities living in each area did not share the same social values for metal.

Ornaments are important components in the active and contingent constitution of identities and their social values are generally interpreted in relation to individual or group identities (Mattson, 2021). During the 5th and 4th millennia BC in northeastern Iberia people's mobility was limited, although extensive raw material trading networks were established with other European regions (e.g. Alpine jade axe heads, Provencal Barremian-Bedoulian flint and Sardinian obsidian). Within Catalonia, a medium-scale distribution of flint from Priorat (Tarragona) or variscite from Gava (Barcelona) is well known (Terradas, et al., 2016; Borrell, et al., 2019; Díaz-Zorita Bonilla, et al., 2021). Lead isotope analyses suggest that the galena from the southern Priorat (MBF) would have been part of the same raw material distribution network during the Neolithic.

These divergent processes in the use and production of copper and lead ornaments reinforce the relevance of social factors to explain the adoption of technical innovations. The availability and knowledge of the raw materials are a precondition to metallurgical adoption or innovation, although the technological knowledge of metallurgy was not necessarily applied to them. The success of innovation always depended on social adoption but was not necessarily based on economic criteria. The links or interregional interaction between southern France and northeastern Iberia were supported by other lithic raw materials and were not symmetrical in the circulation of goods. Galena, or other related lead ores, and the subsequent metallurgical technology to obtain lead were not part of this common system, although, together with other features, they define the cultural diversity identified in the archaeological record.

Finally, the diverse use of galena and lead detected in several parts of Europe and the Mediterranean during the Neolithic reveals a multilinear evolution of technology without a diffusionist paradigm. This perspective suggests that the narrative linking metallurgy to social progress or social complexity in Western Europe must be redefined, as Radivojević and Roberts (2021, p.258) do in the case of the Balkans.

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Notes

1 We thank Manuel Rojo and Jesus Sesma for the unpublished information about this fragment of galena.

2 Recent archaeological works on the Le Planet (Aveyron, France) site confirms the smelting of lead and copper in a chronological frame of the first half of the 3rd millennium cal BC (Costa, et al., 2021).

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