Mineral provenance of Roman lead objects from the Cinca River basin (Huesca, Spain)

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Abstract

A set of 50 lead artifacts, out of which 29 were *glandes plumbeae*, found at archaeological sites located in the Cinca River basin (Huesca, Spain) were analyzed by MC-ICP-MS to determine lead isotope ratios. A comparison with lead ore deposits exploited by the Romans in the Iberian Peninsula allowed to differentiate two main groups of samples: those manufactured with Pb from nearby Pyrenean or Iberian Range ores (30%), and those from mining areas of Cartagena-Mazarrón in southeastern Spain or S'Argentera in the island of Ibiza (70%). This finding supports the existence of lead-ore extraction in the Central Pyrenees in the Roman period and enhances our understanding of metal supply networks in the region and army provisioning during the Sertorian war.

Keywords: *Cerro de la Alegría*; *glandes plumbeae*; lead isotope ratios; MC-ICP-MS; Sertorian War; sling bullets; Pyrenean mines

1. Introduction

The identification of raw materials sources of metal objects through archaeometric methods is leading to important advances in the field of archaeology (Rehren and Pernicka, 2008). In particular, lead isotope analysis is nowadays commonly used for provenance studies of archaeological artifacts and is a well-established procedure in archaeometallurgy (Albarede, et al., 2012, Baker, et al., 2006, Orfanou, et al., 2020, Stos-Gale, et al., 1995, Stos-Gale and Gale, 2009). In the last decades, some archaeological research projects have included the comparison of lead isotope ratios of artifacts with those of ore deposits, showing that this may allow the identification of the most likely origin of the raw metals (see summary by Klein (2007)), always taking into consideration the archaeological context.

Roman mining has been the subject of many studies (Erdkamp, et al., 2015, Healy, 1978, Hirt, 2010). In Roman Spain, a number of mining areas have been identified as suppliers of lead (silver) (Domergue, 1990, Domergue, et al., 2013), mainly *Cartago Nova* (Southeast Volcanic Province, Cartagena-Mazarrón deposits) and the Central Iberian Zone (Linares-La Carolina, Los Pedroches, Alcudia-Almadén) (Sinner, et al., 2020). Other deposits which were exploited in Roman times include, for instance, those of the Moncayo area in the Iberian Range (e.g., Valdeplata ravine mines) (Sanz Pérez, 2003) or the one of S'Argentera in Ibiza (Balearic Islands) (Hermanns, 2014).

Regarding the Central Pyrenees, several lead mines have been exploited in this area, whose mineralogy has been studied by Johnson, et al. (1996), Subías *et al*. (Fanlo, et al., 1998, Subías, et al., 2015, Subías, et al., 2010, Subías, et al., 1997) and other authors (Cardellach, et al., 1996, García-Sansegundo, et al., 2014, Marcoux, et al., 1991, Munoz, et al., 2016). Nonetheless, according to –for instance– Sablayrolles (2001) and Domergue, et al. (2013), there would be no definite evidence for galena exploitation in the Roman period, despite the presence of two lead ingots with the seal OSCA (Veny Melià, 1979), Latin name for the earlier Iberian settlement of *Bolskan* (modern Huesca, 55 km NW from Cinca River). However, during the Iberian and Roman period, the city of *Bolskan/Osca* coined silver coins (Konrad and Plutarch, 1994), and Cato mentions silver and iron-mines north of the Ebro in the early second century BC. Moreover, Livy in three passages referring to the same period also comments on silver from *Osca*, whose origin should be traced to argentiferous galenas from the Pyrenees (Davies, 1935).

The aim of the present study has been to use archaeometrical techniques to support the possibility of the exploitation of lead ores in the Central Pyrenees in Roman times. A set of 50 lead objects (including 29 *glandes plumbeae*) deposited in the Centro de Estudios de Monzón y Cinca Medio (CEHIMO) collection in Monzón (Huesca, Spain) have been analyzed. The rationale behind these analyses is not only to fill a literature gap[1], but also to contribute to the understanding of how supply networks between the peoples of the Cinca River basin and the rest of *Hispania* developed. Further, given that the historical context for some of the lead artifacts is the Sertorian war (between 80 and 71 BC), a conflict in which Balearic slingers were involved, the analysis of the provenance of these sling bullets can promote our knowledge of this Roman period in the Iberian Peninsula and of the provisioning of the armies involved.

2. Materials and Methods

2.1. *Geographical and archaeological context*

The studied artifacts were found at *Cerro de la Alegría* (CdlA), *Lo Pingato* (Enate), Santalecina, *Ripoll-Saso* and Estiche archaeological sites, all located in the Cinca River basin (Figure 1).

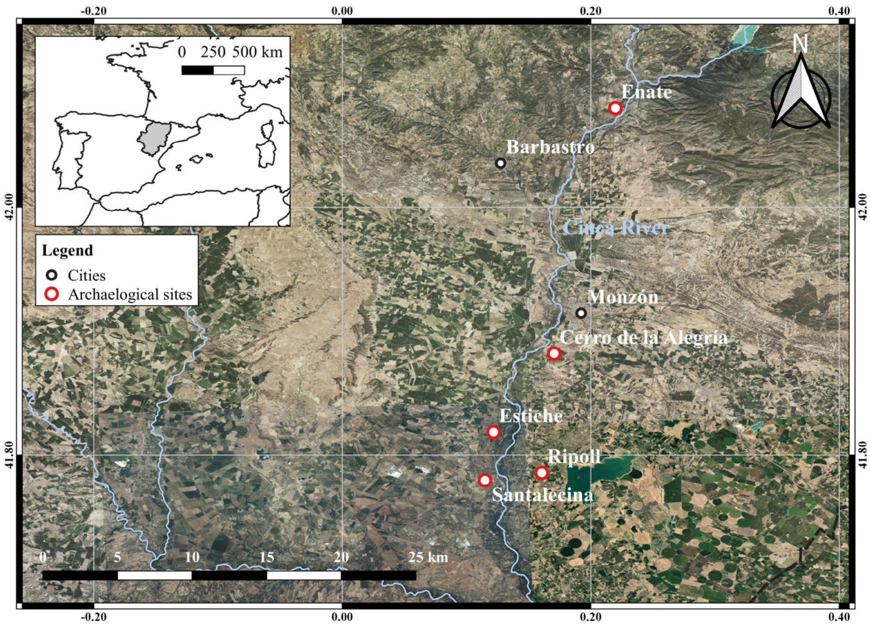


Figure 1. Location of the archaeological sites in which the lead artifacts included in the study were recovered, together with some modern cities. *Inset*: Location of Aragón in Spain.

Cinca River (*Cinga*) is a tributary of Segre River (*Sicoris*), which flows southwards from the Central Pyrenees to the Ebro River. The Ebro River Basin was conquered by the Romans after the Second Punic War and was part of the theater of the Roman Civil War between Sertorius and Pompeius. Years later (42 BC), the junction between *Cinga* and *Sicoris* rivers, near the old city of *Ilerda*, territory of the Illergetes, was the battleground between Julius Caesar and the Pompeian army (Perrin, 1882). During the Roman Empire, in *Hispania Citerior* Province, an important number of villas (*villae*) appeared in this territory, such as those named Del Rey in Enate

[1] According to [Montero Ruiz, I., 2018. La procedencia del metal: consolidación de los estudios con isótopos de plomo en la Península Ibérica, Revista d'arqueologia de Ponent, 311-328., Fig. 2], based on up-to-date data from a national archaeometallurgy project, no objects from Aragón have been analyzed.

(inhabited up to the 4th century), Tosal de los Moros in Santalecina (*Saltus Licinius*) and Estiche, all located on the right bank of the Cinca.

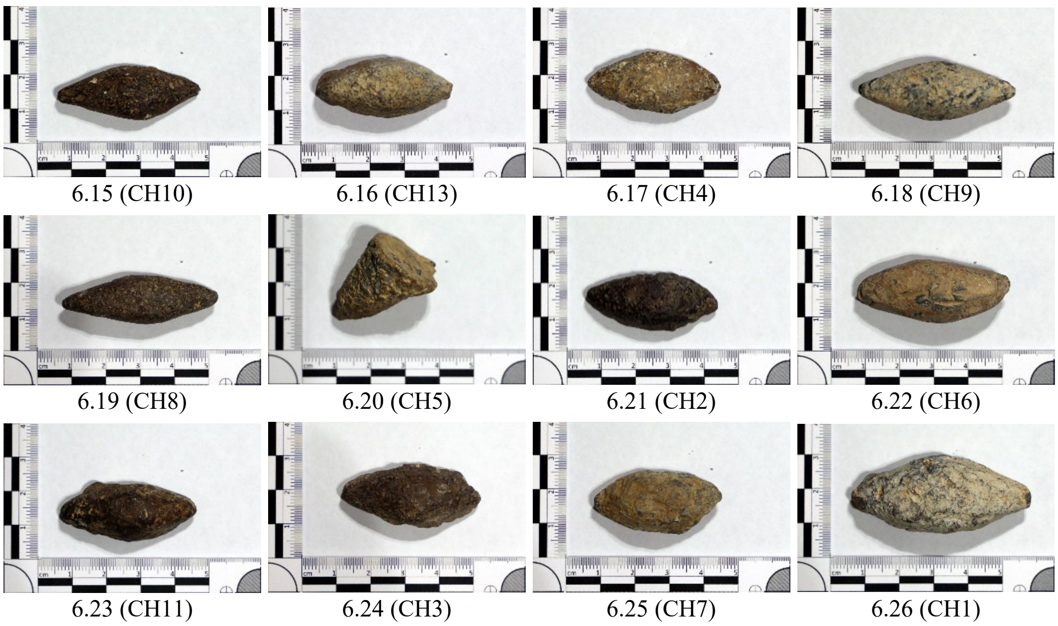
In the Roman period, the important causeway from *Ilerda* to *Osca* crossed the Cinca River near the modern city of Monzón. On the left bank of the Cinca, south of Monzón, are located the archaeological sites of *Ripoll-Saso* and *Cerro de la Alegría*. This later site (UTM coordinates 31N 265342.0720, 4640673.317) corresponds to the *Tolous* mansion, an Iberian *oppidum,* related to aforementioned Roman road. The site is a presumable Roman Republican military camp connected to the battles between Quinto Sertorio and the *optimates* Quinto Cecilio Metelo Pio and Cneo Pompey in 80-71 BC, as a consequence of Sulla Civil War (Contreras, et al., 2006a, Contreras, et al., 2006b).

The objective of the proconsul Quinto Cecilio Metelo Pio was to establish a series of enclaves, the *propugnacula imperi*, to control the most strategic points of the territory and, possibly, *Tolous* would have fulfilled that function. The finds may correspond to a supply of slingers involved in the contest, in line with the information provided by classical sources. Q. Claudio Cuadrigario, in one of the fragments of book XIX, alludes to a passage from the Sertorian Wars of the year 79 BC in which the troops of Metelo Pio besieged those of Quinto Sertorio (Contreras, et al., 2006b, Vilar, 2013). Cuadrigario also indicated that the slings were more effective if they were thrown firing from the bottom up pointing to the wall from the outside (Quesada Sanz, 1997). Finally, the joint work of Metelo Pio in *Hispania Ulterior* and Pompeyo in *Hispania Citerior* was able to put an end to the war with the assassination of Sertorio by Perperna in the year 73 BC at Osca.

2.2. *Studied samples*

The samples object of this study, from the CEHIMO collection (Monzón, Spain), included 29 *glandes plumbeae* (out of which 18 were found at *Cerro de la Alegría*, 6 in Enate/*Lo Pingato* and 4 in *Ripoll-Saso*) (Figure 2 and Figures S1-S3), plus one belonging to a private collection. A morphological study of some of the *glandes plumbeae* from the same CEHIMO collection was carried out by Contreras, et al. (2006a), who assigned them mostly to Völling 1c type, but with examples of Völling 1a, 2a and 2b types too.

Dimensions and weights of some of the *glandes plumbeae* from *Cerro de la Alegría* archaeological site are summarized in Table S1. Since a Pyrenean origin has been advocated for larger pieces (Vilar, 2013), the isotopic analyses for samples 6.26 (CH01) and 6.27 (CH12) may shed light into this hypothesis.



6.15 (CH10) 6.16 (CH13) 6.17 (CH4) 6.18 (CH9)

6.19 (CH8) 6.20 (CH5) 6.21 (CH2) 6.22 (CH6)

6.23 (CH11) 6.24 (CH3) 6.25 (CH7) 6.26 (CH1)

Figure 2. *Glandes plumbeae* from *Cerro de la Alegría* archaeological site.

Another 21 lead artifacts with origin in the three aforementioned locations and in Santalecina and Estiche (Figure S4) were also analyzed. Most of them were loom weights, although a few staples used to repair pottery or marble were also included in the study.

It should be clarified that the artefacts studied were randomly collected at the surface.

2.3. *Lead isotopic analyses*

The archaeological pieces were sampled by drilling to obtain fresh and chemically representative metal. Drilling chips from the first few micrometers were discarded to avoid surface material potentially affected by, for instance, corrosion processes or conservation treatment. A Thermo Scientific Neptune multicollector ICP–MS instrument (Thermo Electron Corporation, Bremen, Germany), operated in low-resolution mode ($m/\Delta m = 400$), was employed for the measurement of the lead isotope ratios.

The digestion of the samples was carried out in 15 mL Savillex flat-bottom Teflon (PFA) screw-cap vials, where about 10 to 150 mg sample were placed together with 8 mL of 3:1 HCl/HNO_3. The closed vials were heated at 110 °C for 24 h and, after evaporation to dryness, 2 mL of 1M HNO_3 were added. Then they were heated again at 110 °C for 30 min, leaving the samples prepared for the chemical separation of lead from the sample matrix. This was carried out using the Pb specTM selective resin. Both resin conditioning and lead-sample matrix separation used protocols are described in detailed elsewhere (Huelga-Suarez, et al., 2014).

For the MC-ICP-MS lead isotope ratios measurements, the pure lead fractions obtained were diluted with 0.42M HNO_3, adjusting the Pb concentration to 200 $\mu g\ L^{-1}$. All samples were run in a sample-standard bracketing (SSB) sequence with a 200 $\mu g \cdot L^{-1}$ Pb isotopic standard solution of NIST SRM 981 (common lead isotopic standard). In addition, the NIST SRM 997 (thallium isotopic standard) reference material was also added (adjusting its concentration to 100 $\mu g \cdot L^{-1}$) to correct the effect of mass discrimination through external normalization. After the measurement of each solution, the sample introduction system was washed with a 0.42M HNO_3 solution until the intensities of the different monitored *m/z* ratios reached white values again.

2.4. *Statistical analysis*

The statistical evaluation of potential ore sources (using mean values for the lead isotopic ratios) was performed via cluster analysis in SPSS software (IBM, Armonk, NY, USA). The starting point was a hierarchical cluster analysis (HCA) with randomly selected data in order to find the best method for clustering (cluster method: furthest neighbor; distance type: Euclidean). K-means analysis was then performed on the entire original dataset (considering all four lead isotopes, as recommended by Albarede, et al. (2020)) so as to assign probable provenances to the archaeological pieces.

3. Results and Discussion

The Pb isotopic ratios for the CEHIMO collection pieces are presented in Table 1. These results have been compared with those reported in an earlier isotopic study conducted by Müller, et al. (2014) on six different pieces from the CEHIMO collection (not included in the present study) (Table 2).

Table 1. Lead isotopic ratios for the sling bullets and loom weights collection deposited in the CEHIMO museum, from the experimental results of this study, and suggested origin of the mineral.

Reference	Type of artifact	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	Cluster*	Suggested provenance
CH01/6.26 CdlA	Sling bullet	18.4904	15.6939	38.7432	1	PYR/IBR/NE
CH02/6.21 CdlA	Sling bullet	18.7171	15.6935	39.0220	5	SE
CH03/6.24 CdlA	Sling bullet	18.7122	15.6930	39.0115	5	SE
CH04/6.17 CdlA	Sling bullet	18.5802	15.6806	38.8460	2	PYR/IBR
CH05/6.20 CdlA	Sling bullet	18.7540	15.7071	39.0701	5	SE
CH06/6.22 CdlA	Sling bullet	18.5062	15.6785	38.8620	2	PYR/IBR
CH07/6.25 CdlA	Sling bullet	18.7183	15.6942	39.0239	5	SE
CH08/6.19 CdlA	Sling bullet	18.7053	15.6862	38.9685	6	SE/Ibiza
CH09/6.18 CdlA	Sling bullet	18.6742	15.6815	38.9681	6	SE/Ibiza
CH10/6.15 CdlA	Sling bullet	18.6711	15.6901	38.9677	6	SE/Ibiza
CH11/6.23 CdlA	Sling bullet	18.6952	15.6824	38.9986	5	SE
CH12/6.27 CdlA	Sling bullet	18.5380	15.6815	38.7966	1	PYR/IBR/NE
CH13/6.16 CdlA	Sling bullet	18.7302	15.6941	39.0216	5	SE
CH14 CdlA	Loom weight	18.4010	15.6582	38.5827	4	PYR/IBR
CH15 Enate	Loom weight	18.7383	15.6980	39.0521	5	SE
CH16 CdlA	Loom weight	18.7083	15.6968	38.9354	6	SE/Ibiza
CH17 Estiche	Loom weight	18.4383	15.6695	38.6474	4	PYR/IBR
CH18 R-Saso	Loom weight	18.4212	15.6770	38.6262	4	PYR/IBR
CH19 CdlA	Loom weight	18.5926	15.6776	38.8712	2	PYR/IBR
CH20 SantaL	Loom weight	18.5051	15.6834	38.8404	2	PYR/IBR
CH21 SantaL	Loom weight	18.7184	15.6920	39.0353	5	SE
CH22 SantaL	Loom weight	18.6739	15.6910	38.8401	2	PYR/IBR
CH23 SantaL	Loom weight	18.6160	15.6840	38.8221	2	PYR/IBR
CH24 CdlA	Loom weight	18.7301	15.6915	39.0473	5	SE
CH25 CdlA	Loom weight	18.7179	15.6856	39.0326	5	SE
CH26 CdlA	Loom weight	18.7236	15.6811	39.0136	5	SE
CH27 CdlA	Staple	18.7934	15.6871	39.0276	5	SE
CH28 R-Saso	Loom weight	18.4843	15.6965	38.7491	1	PYR/IBR/NE
CH29 R-Saso	Staple	18.4380	15.6695	38.7323	1	PYR/IBR/NE
CH30 Enate	Sling bullet	18.7426	15.6873	39.0626	5	SE
CH31 Enate	Sling bullet	18.7173	15.6973	39.0765	5	SE
CH32 Enate	Sling bullet	18.7280	15.6845	39.0367	5	SE
CH33 Enate	Sling bullet	18.3987	15.6831	38.7489	1	PYR/IBR/NE
CH34 Enate	Sling bullet	18.7382	15.6830	39.0435	5	SE
CH35 Enate	Sling bullet	18.7605	15.6982	39.0499	5	SE
CH36 CdlA	Staple	18.7610	15.6979	39.0409	5	SE
CH37 CdlA	Sling bullet	18.7083	15.6842	39.0094	5	SE
CH38 CdlA	Sling bullet	18.6912	15.6946	39.0007	5	SE
CH39 CdlA	Sling bullet	18.7179	15.6952	39.0223	5	SE
CH40 CdlA	Staple	18.7088	15.6947	39.0354	5	SE
CH41 R-Saso	Loom weight	18.5331	15.6948	39.0122	2	PYR/IBR
CH42 CdlA	Loom weight	18.7094	15.6974	39.0532	5	SE
CH43 R-Saso	Sling bullet	18.7580	15.6950	39.0754	5	SE
CH44 R-Saso	Sling bullet	18.6750	15.6943	38.9651	6	SE/Ibiza
CH45 R-Saso	Sling bullet	18.7390	15.6840	39.0369	5	SE
CH46 R-Saso	Sling bullet	18.7084	15.6830	39.0108	5	SE
CH47 CdlA	Sling bullet	18.7402	15.6998	39.0351	5	SE
CH48 CdlA	Staple	18.7174	15.6940	39.0211	5	SE
TIR CdlA	Sling bullet	18.7308	15.6949	39.0300	5	SE
G1 CdlA	Sling bullet	18.7243	15.6840	39.0234	5	SE

CdlA, R-Saso and SantaL stand for *Cerro de la Alegría*, *Ripoll-Saso* and Santalecina archaeological sites. PYR, IBR, NE and SE stand for Pyrenees, Iberian Range, northeastern Spain (Catalonian Coastal Ranges) and southeastern Spain (Cartagena-Mazarrón) ores.

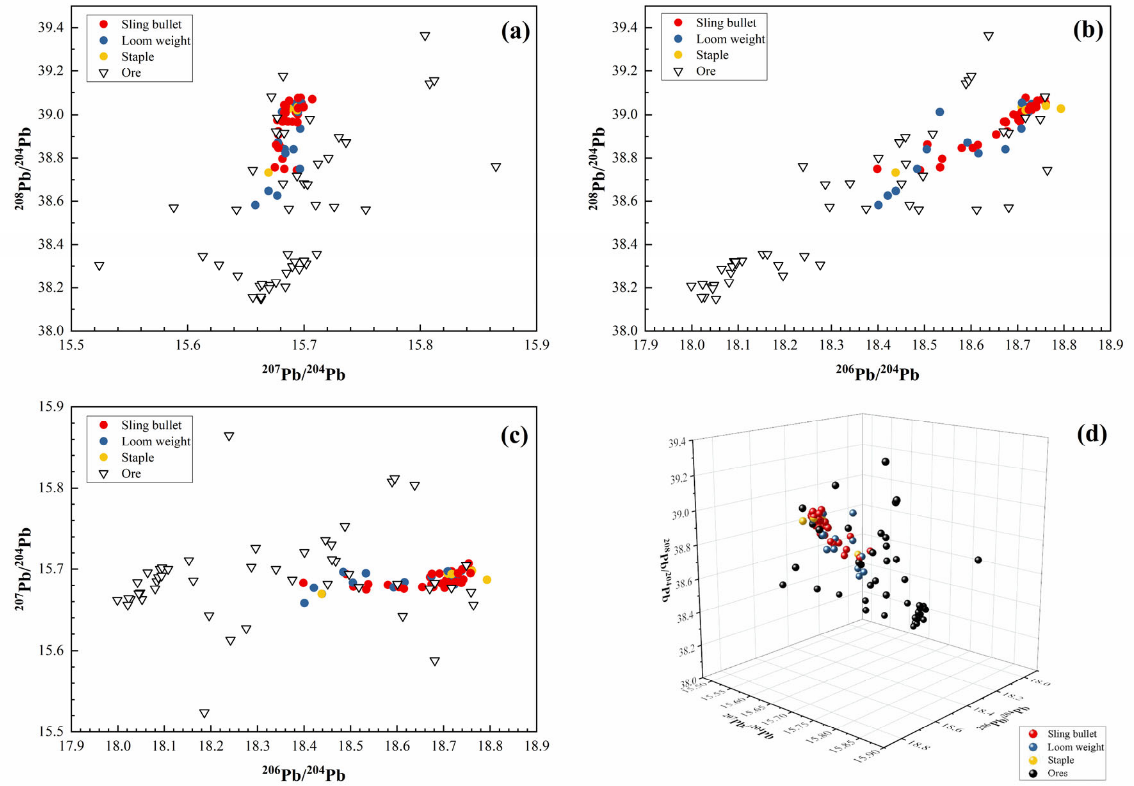
* Cluster refers to clusters in Figures S5 and S6 and Table S2. Clusters 3, 7, 8, 9 and 10 are not listed because they only consist of mineral deposits.

Table 2. Isotopic ratios of some sling bullets from CEHIMO Museum (different from those referred in Table 1); source: (Müller, et al., 2014)) and suggested provenance of the mineral.

Artifact	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	Cluster	Suggested provenance
MO-01 CdlA	18.604	15.678	38.846	2	PYR/IBR
MO-04 CdlA	18.678	15.678	38.924	6	SE/Ibiza
MO-08 R-Saso	18.702	15.677	38.974	6	SE/Ibiza
MO-09 R-Saso	18.534	15.675	38.757	1	PYR/IBR/NE
MO-14 CdlA	18.654	15.678	38.909	6	SE/Ibiza
MO-15 CdlA	18.615	15.676	38.860	2	PYR/IBR

CdlA and R-Saso stand for *Cerro de la Alegría* and *Ripoll-Saso* archaeological sites. PYR, IBR, NE and SE stand for Pyrenees, Iberian Range, northeastern Spain (Catalonian Coastal Ranges) and southeastern Spain (Cartagena-Mazarrón) ores.

In order to identify the possible origin for above lead artifacts, their Pb isotopic ratios were compared with those of mineralizations from the Iberian Peninsula and the French Central Pyrenees reported in the literature (summarized in Table 3[2]). Binary and ternary plots with the Pb isotope compositions of the samples under study, the sling bullets reported by Müller, et al. (2014) and representative mineral deposits are shown in Figure 3.



[2] Mean values have been used for some mineral deposits in order to smooth regional variability, and to some extent minimize analytical issues [Albarede, F., Blichert-Toft, J., Gentelli, L., Milot, J., Vaxevanopoulos, M., Klein, S., Westner, K., Birch, T., Davis, G., de Callataÿ, F., 2020. A miner's perspective on Pb isotope provenances in the Western and Central Mediterranean, Journal of Archaeological Science 121.]

Figure 3. (**a-c**) Binary projections and (**d**) ternary plot, using ^{204}Pb normalization, of Pb isotope compositions of the archaeological pieces under study and ores reported in the literature.

Table 3. Lead isotopic data reported in the literature for Pyrenean, Iberian Range, Central Iberian Zone, Northeastern Iberian Peninsula, Southern Iberian Peninsula, and Balearic Islands deposits.

Mineralization	^{206}Pb/^{204}Pb	^{207}Pb/^{204}Pb	^{208}Pb/^{204}Pb	Reference
PYR, La Manere	18.052	15.663	38.150	
PYR, Carboire	17.999	15.662	38.209	
PYR, Pierrefitte† (*n*=4)	18.084	15.685	38.270	
PYR, Argut† (*n*=2)	18.027	15.663	38.159	
PYR, Bentaillou	18.042	15.684	38.206	
PYR, Crabioules	18.091	15.700	38.323	(Marcoux, et al., 1991)
PYR, Arrens	18.095	15.702	38.312	
PYR, Gedre	18.153	15.711	38.357	
PYR, Cheze Ecole† (*n*=2)	18.162	15.686	38.356	
PYR, Nerbiou† (*n*=2)	18.108	15.700	38.325	
PYR, Taichougnes	18.023	15.664	38.217	
PYR, Aran Valley, Margdalida	18.021	15.656	38.157	
PYR, Aran Valley, Font dels Lladres	18.064	15.696	38.288	
PYR, Aran Valley Liat† (*n*=2)	18.048	15.671	38.213	(Cardellach, et al., 1996)
PYR, Aran Valley Victoria	18.080	15.676	38.226	
PYR, Aran Valley Mauricio† (*n*=2)	18.045	15.670	38.198	
PYR, Benasque *1*	18.087	15.690	38.299	(García-Sansegundo, et al., 2014)
PYR, Benasque *2*	18.094	15.692	38.321	
PYR, Les Argentieres† (*n*=3)	18.375	15.687	38.566	(Munoz, et al., 2016)
PYR, Lacore† (*n*=6)	18.497	15.694	38.718	
PYR, Bielsa-Parzán, Ana-1	18.518	15.678	38.912	
PYR, Bielsa-Parzán, P4-7	18.488	15.753	38.563	
PYR, Bielsa-Parzán, PF5-1	18.239	15.865	38.762	
PYR, Bielsa-Parzán, P3-S	18.638	15.804	39.365	(Subías, et al., 2015)
PYR, Bielsa-Parzán, P4-S	18.589	15.808	39.142	
PYR, Bielsa-Parzán, P5-S	18.595	15.812	39.157	
PYR, Bielsa-Parzán, P5-Sd	18.601	15.682	39.177	
PYR, Bielsa-Parzán, Ana† (*n*=3)	18.451	15.682	38.682	(Martín Ramos, et al., 2019)
PYR, Benasque Eriste† (*n*=3)	18.468	15.710	38.584	
IBR, Segura de Baños† (*n*=7)	18.459	15.730	38.897	
IBR, Calcena† (*n*=4)	18.401	15.721	38.801	
IBR, Bádenas† (*n*=3)	18.446	15.736	38.872	(Subías, et al., 2010, Subias Pérez,
IBR, Cuca-Alta† (*n*=7)	18.340	15.700	38.683	et al., 1994)
IBR, Nogueras† (*n*=3)	18.287	15.703	38.678	
IBR, Santa Cruz† (*n*=3)	18.296	15.726	38.575	
NE, Catalonian Coastal Ranges† (*n*=24)	18.460	15.712	38.774	(Canals and Cardellach, 1997)
SE, Sierra de Cartagena† (*n*=6)	18.749	15.705	38.980	(Graeser and Friedrich, 1970,
SE, Mazarrón	18.681	15.588	38.572	Trincherini, et al., 2009)
SE, Cartagena-Mazarrón	18.759	15.672	39.082	(Müller, et al., 2014)
SE, Portman† (*n*=2)	18.717	15.677	38.987	
Ibiza, S'Argentera† (*n*=12)	18.681	15.683	38.916	(Hermanns, 2014)
Ibiza, S'Argentera	18.670	15.676	38.923	(Müller, et al., 2014)
CIZ, Andújar-Montoro† (*n*=5)	18.764	15.656	38.744	
CIZ, Los Pedroches† (*n*=28)	18.612	15.642	38.562	
CIZ, Alcudia-Almadén† (*n*=2)	18.186	15.524	38.305	(Klein, et al., 2009)
CIZ, Linares-La Carolina	18.242	15.613	38.347	
SW, Beja, Portugal	18.276	15.627	38.307	
SW, Riotinto† (*n*=21)	18.196	15.643	38.257	(Pomiès, et al., 1998)

PYR = Pyrenees; IBR = Iberian Range; CIZ = Central Iberian Zone; SW = Southwestern Iberian Peninsula; NE = Northeastern Iberian Peninsula; SE = Southeastern Iberian Peninsula (also referred to as SVP = Southern Volcanic Province).

† Average values.

Hierarchical cluster analysis (Figure S5, Table S2) allowed to assign probable provenances, shown in Figure S6 and indicated for each of the samples in the rightmost columns in Table 1 and

Table 2. At this point, a word of caution seems to be necessary, since it should be made clear that the Pb isotope analysis technique does not provide absolute assurance in the correlations because, in some cases, it is affected by the mixing of ores and from the melting and re-melting of pieces from diverse origins, and because the Pb isotopic composition from a given region may in some cases show considerable dispersion (Santos Zalduegui, et al., 2004). Further, considering that *Hispania* was a major Pb supplier, only mineral deposits from the Iberian Peninsula have been included in the reference ore database, but other regions of the Empire cannot be entirely dismissed as potential metal providers (for instance, some ambiguity arises because of the closeness of the southeastern Spanish field to the Tuscan isotope field). Hence, the interpretation of Pb isotope data presented herein to assess the possible provenance should be taken with some caution.

Out of the 50 archaeological pieces under study, 10 artifacts (20%) would probably have been manufactured with Pb from Pyrenean (Bielsa-Parzán, Benasque or Les Argentieres ores) or Iberian Range deposits (Cuca-Alta, Calcena, Segura de Baños, Nogueras and Bádenas ores); 5 artifacts (10%) would have used either Pyrenean, Iberian Range or Catalonian Coastal Range deposits; and 35 artifacts (70%) could be assigned either to Cartagena-Mazarrón or Ibiza deposits. The similarities in isotopic composition between these later two mineralizations has been previously reported by Ruiz De Smedt (1992).

Concerning the 6 sling bullets reported by Müller, et al. (2014), 3 would have a Pyrenean/Iberian Range/NE origin, and the other 3 would be assigned to SE/Ibiza mineralizations.

Thus, a third of the artifacts were found to have their origin in nearby Pyrenean or Iberian Range Mesozoic deposits, and two-thirds came from more remote mines, such as those of the SE (La Unión, Cartagena-Mazarrón) or Ibiza. It is worth noting that the isotope ratios of the lead pieces from the CEHIMO collection did not match the isotopic signature of Central Iberian Zone or southwestern Spain ore deposits (unlike those reported by Gomes, et al. (2017) for Conimbriga, Portugal), nor those of ores belonging to the so-called Pyrenean Paleozoic Axial Zone (Munoz, et al., 2016).

It should also be clarified that few unequivocal matches between the source ores and the objects have been found. Other Pb sources that had not yet been identified cannot be ruled out, and the possibility of some degree of mixing cannot be dismissed either (for instance, in samples CH01, CH04, CH12, CH19 or MO-09).

Regarding the *glandes plumbae* –29 from our study (Figure 2, Figures S1-S3) plus 6 from Müller, et al. (2014)–, 23% of the slingshots could be referred to Pyrenean, Iberian Range or NE mines, and the remaining 77% possibly had their origin in the Peninsular Southeast or in the Balearic Islands. While the study by Müller, et al. (2014) –not conclusive about the provenance– suggested the Portman area (Cartagena-Mazarrón) as one of the best candidates for the raw material used (as we do), a Balearic origin should not be dismissed, provided that Livy, Strabo and Diodorus Siculus mention that the Balearic mercenaries were skillful slingers (Echols, 1950). Further, the morphology of some of those sling bullets would be similar to that of sling bullets found in Sanitja (Menorca, Balearic Islands), as noted by Contreras, et al. (2006a). On the other hand, the proposed reassignment of the provenance for three of those sling bullets to Pyrenean, Iberian Range or NE mineral deposits should be referred to the omission of northern *Hispania* ores in the isotopic signatures used for comparison purposes.

Given that, as noted above, the sling bullets of the CEHIMO collection were found in fortified settlements linked to the Sertorian wars (most probably during 77/76 BC, when the Sertorian troops moved along the Ebro Valley (Müller, et al., 2014)), it may be speculated that the Sertorian side could have used nearby Pb ore deposits, while the senatorial troops (Memmius' army from *Cartago Nova*?) would have brought Pb from southeastern Spain (or from the Balearic Islands), either as finished objects and/or as raw metal (small ingots) that were casted to lead bullets at the site itself.

Apart from the *glandes plumbae*, 10 out of the 21 lead loom weights, staples and other artifacts from Cinca River basin archaeological sites possibly had their mineralization origin in Pyrenean or Iberian Range mines, which may be referred to activity of the *civitates* and *villae* in the I-IV centuries.

The fact that some of the mineralizations in the Pyrenees to which it is possible to refer the artifacts are the Bielsa-Parzán deposits, in the headwaters of the river Cinca, would suggest the presence of a Roman supply route along the river (Figure S7), used for the transportation of lead ore or ingots and other goods (for example, the transport of marble of Saint Beat from the south of *Galia* to *Caesaraugusta* has been suggested in the work by Lapuente, et al. (2015)). The supply of these materials from the mining areas of Bielsa-Parzán (*Territorium Boletanum*) to the Enate, Santalecina and Estiche *villae* and to the *Barbotum* (Coscojuela de Fantova) and *Labitolosa* (Puebla de Castro) *civitates*, until the *Tolous* road junction was reached, could be carried out using the banks of the Cinca River. Once *Tolous* was reached, the ore or ingots could be transported by a main road (*via*) to *Osca* (going up by *Caun* (Berbegal) and *Pertusa* (Pertusa)), or going down to *Ilerda* (Lérida), by *Mendiculeia* (Tamarite de Litera).

4. Conclusion

The isotopic data obtained from the CEHIMO lead artifact collection suggests that the nearest deposits, located in the Pyrenees and in the Iberian Range, probably were important Pb sources for the Cinca River basin settlements (accounting for 32% of the samples analyzed). Nonetheless, 68% of the samples would have their origin in the Cartagena-Mazarrón (or Ibiza) ores. It may be hypothesized that the distinct provenances of the sling bullets could be related to the different armies involved in a given battle during the Sertorian War. Almost half of the lead loom weights, staples and artifacts from other Cinca River basin archaeological sites had a probable mineralization origin in nearby mineral deposits, including the Bielsa-Parzán (*Territorium Boletanum*) ores, which may be referred to the pacific activity of the *villae* and *civitates* in the Cinca area in the I-IV centuries AD. The reported data would support the existence of lead-ore extraction in the Central Pyrenees in Roman times, which had been questioned in the literature. Additional analyses from other museum collections would be needed for further supporting the evidence presented in this study.

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