



AMALIA, A Matching Algorithm for Lead Isotope Analyses: Formulation and proof of concept at the Roman foundry of Fuente Spitz (Jaén, Spain)

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ABSTRACT

This article presents A Matching Algorithm for Lead Isotope Analyses (AMALIA) that yields analytical coincidences in lead isotope databases, allowing a fast selection of potential candidates for metal provenance. As a proof of concept, potential ore sources for 29 Roman lead artifacts from the archaeological site of Fuente Spitz (Jaén, Spain) are provided. Additionally, a reassessment of legacy, TIMS lead-isotope analyses is conducted by re-analysis of 26 galena samples from nearby mining districts by MC-ICP-MS. The study demonstrates the accuracy and reliability of AMALIA and stresses the need to assess the isotope ratio data obtained without lead isotopic tracers (spikes) by TIMS carefully on a case-to-case basis. At the archaeological level, our study shows that the foundries and smelting sites at Fuente Spitz and Cerro del Plomo processed galena ores from the mining districts of La Carolina and Linares to produce a variety of lead products and lead ingots that have been found at several places thorough Europe, thereby providing tangible evidence of the regional and long-distance commercial circuits that these foundries were supplying.

1. Introduction

Metals – their exploitation, trade and use – have been pivotal in the forge, development and fate of ancient states and societies (Albarède et al., 2016; Earle et al., 2015; Sherratt and Sherratt, 1993). Archaeometric research on metal objects has given invaluable information on these topics and among them, lead isotope analysis (LIA) has proven itself to be an unrivalled tool in metal provenance studies (Eshel et al., 2019; Nocete et al., 2014; Westner et al., 2020). The success of LIA is based on the unique nature of lead and its naturally occurring isotopes. There are but a few elements on Earth that show notable differences in their isotope abundances due to the presence of one or more radiogenic isotopes (Ar, Sr, Ce, Nd, Hf, Os and Pb). In this exclusive group only the siderophile element osmium and the chalcophile element lead are common in metal alloys. Whereas osmium has one natural radiogenic isotope (¹⁸⁷Os) produced after the beta decay of ¹⁸⁷Re, lead has three natural isotopes (²⁰⁶Pb, ²⁰⁷Pb and ²⁰⁸Pb) that have grown in abundance independently during the 4.5 billion-years history of Earth. Therefore, a single LIA offers three independent parameters (in practice, isotope

ratios) that characterize a mineral or a metal alloy. Since the first comprehensive dataset published by Russell and Farquhar (1960), it has been observed that lead isotope compositions allow to distinguish ore deposits and mining districts worldwide and consequently may indicate the provenance of metal objects (Brill and Wampler, 1967). In order to visualize these features in scientific publications, the 3-dimensional space of LIA is projected in two combined, bivariate diagrams that include the three independent parameters. This graphical representation is very useful for data comparison but has some drawbacks. The most obvious is the loss of information caused by the projection of spatial data in two dimensional planes. In addition, it only serves to estimate how close or how far away two analyses are from each other, but cannot resolve if they are analytically equal or comparable, a decision that is left at the discretion of the researcher. To solve this limitation, many authors employ isotopic fields to assign the LIA of a metal object to a group of ore deposits from a mining district or a larger geographical region. However, their use is discouraged (Baxter et al., 2000; Stos-Gale and Gale, 2009).

Clearly, we need to find methods that complement and improve the

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information extracted from LIA in conventional bivariate graphs. Several authors have proposed different mathematical approaches (Baxter et al., 1997; Birch et al., 2020; De Ceuster and Degryse, 2020; Delile et al., 2014; Sayre et al., 1992; Stos-Gale and Gale, 2009). Most of them employ statistic methods (e.g.: kernel density estimation, Mahalanobis distance) that still need to define isotope fields/groups and means, and preserve some of the previously exposed flaws even if data treatment is more rigorous. Others are better addressed to the comparison of raw analytical data in the 3-D space through the calculation of the Euclidean distance between two real analyses. However, this geometric approach only yields a magnitude and ignores the vectorial nature of analytical uncertainties in LIA.

We propose A Matching Algorithm for Lead Isotope Analyses (AMALIA) that overcomes the exposed drawbacks by taking into account the analytical uncertainties along each of the considered axes (isotope ratios). It also establishes interlaboratory, empirically-defined threshold values to yield a list of analytical coincidences (matches) to be further scrutinized based on archaeological grounds as potential sources for the studied metal. This last feature makes AMALIA a powerful tool when combined with increasingly larger databases.

2. The AMALIA algorithm

2.1. Formulation

The ultimate objective of LIA in provenance studies is to determine if a metal object could have been sourced from a known ore deposit. For it to be true, their lead isotopic compositions have to be analytically equal. In other words, the difference between their values should be smaller than, or equal to the analytical uncertainty (AU). For any lead isotope ratio given for two samples a and b , and expressed in logical form:

$$EQUAL \text{ IF } \left(\frac{{}^{20x}Pb}{{}^{20y}Pb} \right)_a - \left(\frac{{}^{20x}Pb}{{}^{20y}Pb} \right)_b \leq AU \left(\frac{{}^{20x}Pb}{{}^{20y}Pb} \right)_{a,b}$$

This logical statement must be true for the three, independent lead-isotope ratios. For example, for the widely-used triplet of isotope ratios containing ${}^{204}Pb$:

$$\text{IF } \left(\frac{{}^{206}Pb}{{}^{204}Pb} \right)_a - \left(\frac{{}^{206}Pb}{{}^{204}Pb} \right)_b \leq AU \left(\frac{{}^{206}Pb}{{}^{204}Pb} \right)_{a,b} \text{ THEN,}$$

$$\text{IF } \left(\frac{{}^{207}Pb}{{}^{204}Pb} \right)_a - \left(\frac{{}^{207}Pb}{{}^{204}Pb} \right)_b \leq AU \left(\frac{{}^{207}Pb}{{}^{204}Pb} \right)_{a,b} \text{ THEN,}$$

$$\text{IF } \left(\frac{{}^{208}Pb}{{}^{204}Pb} \right)_a - \left(\frac{{}^{208}Pb}{{}^{204}Pb} \right)_b \leq AU \left(\frac{{}^{208}Pb}{{}^{204}Pb} \right)_{a,b} \text{ THEN MATCH } (a = b)$$

This string of logical statements forms AMALIA. Alternatively, any other triplet of independent ratios (for example those containing ${}^{206}Pb$) can be used. AMALIA is easy to use and can be formulated in spreadsheets and programming routines for (semi)automatic identification of coincident samples in large LIA databases.

2.2. Analytical uncertainty

Given that published LIA are obtained at different laboratories and in order to compare them, the analytical uncertainty has to consider the variety of results that can be obtained at any laboratory for the same sample. This can be estimated from the published results of the common-lead interlaboratory standard, the certified reference material SRM-981, also known as NBS-981 (National Institute of Standards and Technology -formerly National Bureau of Standards-, USA). A large dataset for the SRM-981 can be downloaded from the GeoReM server at the Max Plank

Institut für Chemie in Mainz, Germany (Jochum et al., 2005). The current dataset contains more than 600 analytical results obtained by TIMS and MC-ICP-MS. They comprise uncorrected isotope ratios from those studies in which the SRM-981 was employed for off-line correction of the instrumental mass bias in the unknown samples, as well as corrected results from those studies that employed lead isotope tracers (spikes) or thallium standard for on-line, mass bias correction in both the samples and the SRM-981. We have discarded all the non-corrected values for SRM-981 that are only valid to evaluate the instrumental mass bias at each laboratory but do not represent the true isotopic composition of SRM-981. The remaining 358 LIA in our purged SRM-981 dataset are all true values for that standard, and have been obtained in 88 laboratories worldwide by TIMS or MC-ICP-MS. For each of the five commonly employed isotope ratios, data distribution is non-normal (Fig. 1). Therefore, we have considered that the best expression of the analytical uncertainty is the relative range (%) of data spread from the minimum to the maximum values. The obtained uncertainty parameters (Fig. 1) characterize the variety of results that can be produced when measuring a homogeneous sample at different laboratories and allow different specimens (e.g.: ore concentrate, archaeological object, metal prill) derived from the same original material (ore deposit) to be compared.

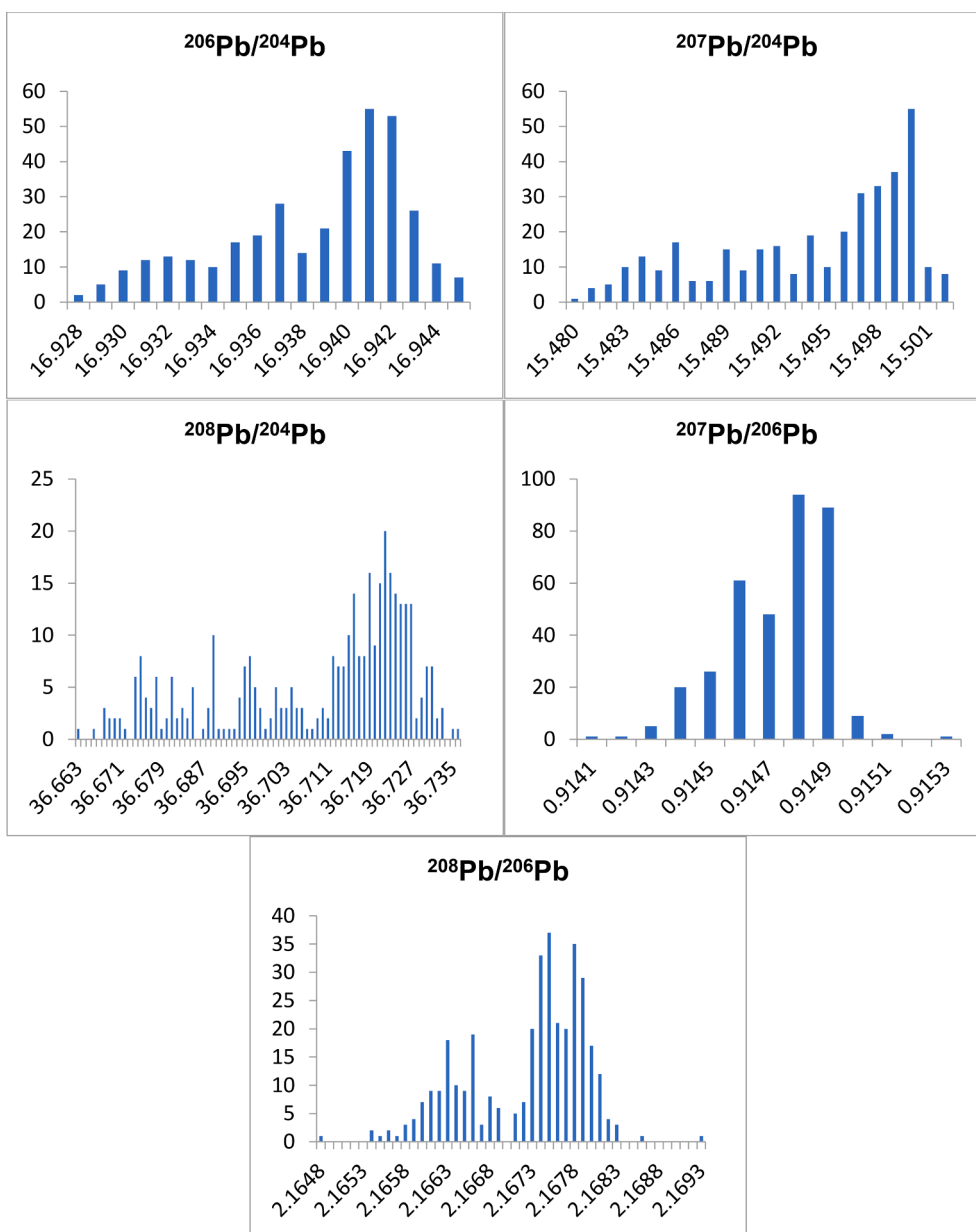
3. Proof of concept: The Fuente Spitz workshop

3.1. Archaeological background

The Roman archaeological settlement of Fuente Spitz, also known as Fuente Spí(s), Spys, Spiz and Espí(s), is located at the eastern end of La Carolina (Jaén, Spain). The site, currently buried under the Aquisgrana industrial estate, covers an unknown area that could be larger than 10 ha, based on the spread of the archaeological remains found (Fig. 2). In 1987, the sole archaeological campaign conducted to date revealed architectural structures, mostly walls and their foundations, as well as abundant Roman material culture (see below). Stratigraphically, an important phase of occupation was recorded around the middle of the 1st century CE. Currently, there is consensus among scholars in providing a chronology for Fuente Spitz between the 1st c. BCE and the late 1st c. CE, perhaps continuing into the 2nd c. CE (Gimeno Pascual and Ramos Miguel, 2021). A substantial collection of 664 objects has been recovered from Fuente Spitz over the years and is currently on display at the Museum of La Carolina. Central to our study are the many artifacts made of lead and/or related to metallurgical practices, such as foundry slags, litharge and lead remains from slag heaps, one crucible and 364 lead objects, which include seals (83), fragments of sarcophagi, water pipes, weights of different sizes (77), ingots (3), sling bullets (65), containers, tokens and clamps. They are our best and almost only evidence to study and reconstruct the activities that were once carried out at Fuente Spitz.

From a historiographical perspective, Fuente Spitz was included by Domergue (1987) in his catalogue of ancient mines and foundries in the Iberian Peninsula. Years later, the same scholar (Domergue, 2000) compared the isotopic signature of samples extracted from eleven of the 43 ingots from the Cabrera 5 wreck (Balearic Islands), with samples of ore and metal from El Centenillo, specifically from Cerro del Plomo (Domergue, 1971) and the Fuente Spitz foundry. The study revealed that the lead employed in the Cabrera 5 ingots was very probably extracted from the El Centenillo mine, or at least from the mines of the Jaén district. In later works, the Roman foundry of Fuente Spitz is cited by Arboledas Martínez (2010), connecting the site to the mining exploitations of La Carolina, where the activity of the foundries associated with the numerous mines seems to come to an end between the middle and end of the 1st century BCE. From this time onwards, the ore extracted must have been transferred to the Fuente Spitz foundry, which operated throughout the 1st century CE, to be processed.

In sum, all the historical and archaeological studies are consistent and interpret Fuente Spitz as a Roman site specializing in metallurgical



SRM-981 Interlaboratory statistics (n = 358 from 88 laboratories)					
	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb
Min	16.928	15.480	36.663	0.9141	2.1648
Max	16.945	15.502	36.736	0.9153	2.1692
Range (abs)	0.017	0.022	0.073	0.0012	0.0044
Range (%)	0.10	0.14	0.20	0.13	0.20

Fig. 1. Data distribution and statistics summary for interlaboratory analytical results of the certified reference material SRM-981.

activity, an industrial foundry responsible for the processing of the galena ores mined in La Carolina, and very similar to other nearby better-studied sites such as Cerro del Plomo (Domergue, 1971). Therefore, Fuente Spitz must be linked to the important lead and silver production of Sierra Morena, the main producing and exporting mining district in Hispania after the Cartagena-Mazarrón mines fell into decline at the end of the 1st century BCE.

3.2. Sampling strategy

We have chosen objects that, based on their typology and purpose, could have been made to be commercialized locally/regionally and/or traded to other provinces and overseas (e.g. lead sheets, remains of funerary urns, sling bullets and ingots) or for the use/functioning of the Fuente Spitz foundry and nearby sites (e.g. *instrumenta domestica*, taps, pipes, weights). The aim was to obtain as much information as possible about how a foundry and smelting site such as Fuente Spitz functioned,



Fig. 2. Fuente Spitz (today) and its 1956 orthophoto from USA Army Map Service flight.

focusing on questions of supply, ownership, production and distribution. An interesting question to explore related to supply was the possibility that the foundry would not only be supplied with the galena ores mined by the anonymous *societates* in the area (Stannard et al., 2021), but also by acquiring already manufactured lead objects to be recycled. In an attempt to address these questions, 29 objects from Fuente Spitz and 2 lead seals from the near Cerro del Plomo smelting site were selected (Table 1).

In addition, 26 galena samples obtained from the same hand specimens studied by Santos Zalduegui et al. (2004) and Velasco et al. (1996), all of them deposited at the Departamento de Geología (UPV/EHU), were later analyzed with the aim of testing the accuracy of already published TIMS data for those ore deposits.

3.3. Analytical methods

The studied metallic objects were filed down with a diamond tool while pure galena was extracted from samples by gently scratching them by hand with a pair of pliers. Given the huge amount of lead in both metallic-lead artifacts and galena samples, contamination from the materials employed in sample preparation (plastics, metallic tools) is negligible. Rinsing the metallic tools with laboratory-grade isopropanol and wiping them with lint-free cellulose paper avoids cross-contamination among samples. Each sample was transferred to a polystyrene (PS) vial and cleaned in 300 μl of 7 M HNO_3 for ten minutes. After this surficial dissolution, the supernatant was removed with a disposable Pasteur pipette and discarded. A new volume of 200 μl of 7 M HNO_3 was then added to the vial for another ten minutes and the supernatant transferred to a new PS vial and diluted in water to a final volume of 2 ml. Reagent-grade nitric acid and ultrapure water (18.2 M Ω cm) were distilled three times at sub-boiling temperature in closed perfluoroalkoxy (PFA) vessels prior to their use. The final, stable solutions were analyzed on a ThermoFisher Scientific NEPTUNE MC-ICP-MS, following the spectrometric method detailed in Rodríguez et al. (2020).

3.4. LIA database

Improvements in computational capabilities have led to the publication of several large databases containing lead isotope data that may assist in provenance studies of archaeological objects (e.g. Stos-Gale and Gale, 2009; Hsu and Sabatini, 2019; Klein et al., 2022). Starting with the seminal OXALID database (Stos-Gale and Gale, 2009), we have gathered about 11,000 lead isotope analyses from Europe and the circum-Mediterranean region, with special emphasis on the Western Mediterranean. Our databank (MedConTaCct) contains isotopic, mineralogical, archaeological and geographical information for more than 3,400 lead ores and about 2,000 metallic-lead and silver artifacts and black gloss ceramics. It is available at the webserver of the University of Victoria under data sources (<https://medcontactt.uvic.ca/>), and has been used to correlate our results for the archaeological objects found at Fuente Spitz with potential ore deposits from which lead could have been sourced.

4. Results

Almost all the analyzed archaeological artifacts from Fuente Spitz and Cerro del Plomo present similar Pb isotopic compositions (Table 1). The two outliers are the spoon Inv. 1554, which shows significantly lower $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios, but higher $^{208}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ ratios, and the sling bullet Inv. 1185, which has higher radiogenic-lead contents and plots away from the main group in the opposite direction in bivariate diagrams (Fig. 3a and 3b).

The AMALIA algorithm has been tested to compare these artifacts with already published isotope data of galena ore deposits. We are aware that some authors prefer to employ the $^{206}\text{Pb}/^{204}\text{Pb}$ - $^{207}\text{Pb}/^{204}\text{Pb}$ - $^{208}\text{Pb}/^{204}\text{Pb}$ triplet to extract all the geological information about the ores, whereas others prefer to employ those ratios with the best analytical precision, namely the $^{206}\text{Pb}/^{204}\text{Pb}$ - $^{207}\text{Pb}/^{206}\text{Pb}$ - $^{208}\text{Pb}/^{206}\text{Pb}$ triplet. Therefore, we have done a combined test with the AMALIA algorithm, employing the two aforementioned triplets. The output of the two tests shows a slight mismatch, with about 30 % of the coincidental ores being close to the threshold values of the analytical uncertainties and failing to match with the metal objects in one of the two considered 3-D spaces. In order to be as

Table 1
Pb isotopic compositions of artifacts from Fuente Spitz and Cerro del Plomo (where indicated), and reanalyzed galenas.

Code	Sample type	²⁰⁶ Pb/ ²⁰⁴ Pb	Error (2SE)	²⁰⁷ Pb/ ²⁰⁴ Pb	Error (2SE)	²⁰⁸ Pb/ ²⁰⁴ Pb	Error (2SE)	²⁰⁷ Pb/ ²⁰⁶ Pb	Error (2SE)	²⁰⁸ Pb/ ²⁰⁶ Pb	Error (2SE)
Inv. Num. 1486	Inscriptions <i>Plumbarius</i>	18,244	0,001	15,634	0,001	38,407	0,002	0,85696	0,00001	2,10520	0,00004
Inv. Num. 776	Seals Unsealed conic	18,232	0,001	15,637	0,001	38,408	0,002	0,85766	0,00001	2,10664	0,00005
Inv. Num. 779	Unsealed conic	18,232	0,001	15,635	0,001	38,402	0,003	0,85760	0,00001	2,10638	0,00005
Inv. Num. 784	XXX	18,236	0,001	15,635	0,001	38,406	0,003	0,85740	0,00001	2,10608	0,00005
Inv. Num. 798	Head/S-C seal	18,212	0,001	15,640	0,001	38,367	0,002	0,85878	0,00001	2,10668	0,00004
Inv. Num. 973	S-C seal	18,236	0,001	15,638	0,001	38,411	0,002	0,85750	0,00001	2,10629	0,00005
Inv. Num. 1145	SA-B seal	18,246	0,001	15,635	0,001	38,406	0,002	0,85689	0,00001	2,10492	0,00004
Inv. Num. 4691	SA-B seal	18,257	0,001	15,631	0,001	38,402	0,002	0,85618	0,00001	2,10346	0,00004
Inv. Num. 1926	TX/S-C seal	18,226	0,001	15,635	0,001	38,401	0,002	0,85782	0,00001	2,10690	0,00005
Inv. Num. 1919	ROA clamp-type seal	18,239	0,001	15,631	0,001	38,397	0,002	0,85696	0,00001	2,10518	0,00004
Inv. Num. 1946	S-C seal (Cerro del Plomo)	18,233	0,001	15,638	0,001	38,410	0,003	0,85763	0,00001	2,10656	0,00005
Inv. Num. 1954	Head/S-C seal (Cerro del Plomo)	18,233	0,001	15,636	0,001	38,406	0,002	0,85760	0,00001	2,10645	0,00004
Inv. Num. 1472	<i>Instrumenta domestica</i> Pot	18,231	0,001	15,635	0,001	38,403	0,002	0,85761	0,00001	2,10644	0,00004
Inv. Num. 1554	Spoon	18,048	0,001	15,620	0,001	38,157	0,002	0,86549	0,00001	2,11422	0,00004
Inv. Num. 1901	Little Jug	18,271	0,001	15,636	0,001	38,442	0,002	0,85576	0,00001	2,10394	0,00004
Inv. Num. 1904	Weight	18,248	0,001	15,636	0,001	38,411	0,002	0,85687	0,00001	2,10502	0,00004
Inv. Num. 1905	Weight	18,243	0,001	15,634	0,001	38,406	0,002	0,85697	0,00001	2,10529	0,00004
Inv. Num. 1051	Pipework/water supply Faucet	18,229	0,001	15,633	0,001	38,398	0,002	0,85759	0,00001	2,10638	0,00005
Inv. Num. 1470	Pipe	18,233	0,001	15,631	0,001	38,397	0,002	0,85730	0,00001	2,10589	0,00005
Inv. Num. 1474	Pipe	18,238	0,001	15,634	0,001	38,401	0,002	0,85718	0,00001	2,10550	0,00004
Inv. Num. 1475	Trap	18,241	0,001	15,632	0,001	38,400	0,002	0,85693	0,00001	2,10512	0,00004
Inv. Num. 1475a	Pipe 1 of trap	18,249	0,001	15,633	0,001	38,409	0,002	0,85667	0,00001	2,10478	0,00004
Inv. Num. 1163	<i>Militaria</i> Sling bullet	18,232	0,001	15,636	0,001	38,405	0,002	0,85764	0,00001	2,10653	0,00004
Inv. Num. 1180	Sling bullet	18,230	0,001	15,635	0,001	38,399	0,002	0,85762	0,00001	2,10634	0,00004
Inv. Num. 1182	Sling bullet	18,232	0,001	15,636	0,001	38,405	0,002	0,85765	0,00001	2,10654	0,00004
Inv. Num. 1184	Sling bullet	18,231	0,001	15,636	0,001	38,404	0,002	0,85767	0,00001	2,10658	0,00004
Inv. Num. 1185	Sling bullet	18,419	0,001	15,658	0,001	38,638	0,002	0,85010	0,00001	2,09776	0,00004
Inv. Num. 304/1	Remains of sarcophagi/ funeral urns Sheet	18,237	0,001	15,638	0,001	38,408	0,002	0,85748	0,00001	2,10608	0,00004
Inv. Num. 1485	Sheet	18,234	0,001	15,637	0,001	38,408	0,002	0,85757	0,00001	2,10641	0,00004
Inv. Num. 001/no id.	Sheet	18,246	0,001	15,634	0,001	38,406	0,002	0,85682	0,00001	2,10491	0,00004
	Other										

(continued on next page)

Table 1 (continued)

Code	Sample type	²⁰⁶ Pb/ ²⁰⁴ Pb	Error (2SE)	²⁰⁷ Pb/ ²⁰⁴ Pb	Error (2SE)	²⁰⁸ Pb/ ²⁰⁴ Pb	Error (2SE)	²⁰⁷ Pb/ ²⁰⁶ Pb	Error (2SE)	²⁰⁸ Pb/ ²⁰⁶ Pb	Error (2SE)
Inv. Num. 7406	Slag	18,227	0,001	15,633	0,001	38,393	0,002	0,85766	0,00001	2,10634	0,00004
CAR-1	La Carolina mining district Galena (El Sinapismo)	18,210	0,001	15,628	0,001	38,377	0,002	0,85818	0,00001	2,10744	0,00004
CAR-2	Galena (El Sinapismo)	18,247	0,001	15,639	0,001	38,414	0,002	0,85709	0,00001	2,10524	0,00005
CAR-3	Galena (Los Guindos)	18,245	0,001	15,636	0,001	38,407	0,004	0,85701	0,00002	2,10505	0,00010
CAR-4	Galena (El Centenillo)	18,224	0,001	15,628	0,001	38,383	0,003	0,85751	0,00002	2,10615	0,00009
LIN-1	Linares mining district Galena (Grupo Matacabras)	18,254	0,001	15,630	0,001	38,404	0,002	0,85625	0,00002	2,10382	0,00005
LIN-2	Galena (Grupo Matacabras)	18,295	0,001	15,637	0,001	38,432	0,002	0,85473	0,00001	2,10072	0,00004
LIN-3	Galena (Lavadero Matacabras)	18,215	0,001	15,625	0,001	38,373	0,002	0,85781	0,00001	2,10666	0,00004
LIN-4	Galena (La Esperanza)	18,436	0,001	15,652	0,001	38,572	0,002	0,84898	0,00002	2,09222	0,00005
LIN-5	Galena (Filón El Cobre)	18,271	0,001	15,641	0,001	38,423	0,002	0,85603	0,00001	2,10292	0,00004
LIN-6	Galena (Grupo Matacabras)	18,304	0,001	15,639	0,001	38,449	0,003	0,85444	0,00001	2,10060	0,00004
LIN-7A	Galena (Grupo Matacabras)	18,238	0,001	15,630	0,001	38,395	0,002	0,85700	0,00001	2,10523	0,00004
LIN-7B	Galena (Grupo Matacabras)	18,304	0,001	15,639	0,001	38,451	0,002	0,85444	0,00001	2,10075	0,00004
LIN-8	Galena (Grupo La Cruz)	18,226	0,001	15,629	0,001	38,388	0,002	0,85754	0,00001	2,10627	0,00004
LIN-9	Galena (Grupo La Cruz)	18,278	0,001	15,634	0,001	38,421	0,002	0,85539	0,00002	2,10209	0,00006
LIN-10	Galena (Grupo La Cruz)	18,256	0,001	15,629	0,001	38,395	0,003	0,85612	0,00001	2,10318	0,00005
LIN-11	Galena (Filón Arrayanes)	18,227	0,001	15,629	0,001	38,388	0,002	0,85746	0,00001	2,10612	0,00004
LIN-12	Galena (La Fernandina)	18,216	0,001	15,626	0,001	38,376	0,002	0,85785	0,00001	2,10678	0,00004
835-015	Alcudia mining district Galena (Nuestra Señora de la Paz)	18,248	0,001	15,649	0,001	38,419	0,003	0,85757	0,00002	2,10539	0,00009
835-092 (GAL19)	Galena (La Veredilla)	18,248	0,001	15,650	0,001	38,435	0,002	0,85764	0,00001	2,10624	0,00004
835-092 (GAL20)	Galena (La Veredilla)	18,208	0,001	15,627	0,001	38,368	0,002	0,85828	0,00001	2,10723	0,00004
836-051	Galena (El Encinarejo)	18,263	0,001	15,642	0,001	38,403	0,002	0,85649	0,00001	2,10274	0,00004
836-065	Galena (La Gitana)	18,237	0,001	15,648	0,001	38,423	0,002	0,85805	0,00001	2,10689	0,00005
SD-SC-2-1	Sierra de la Demanda mining district Galena (Filón San Cristóbal)	18,258	0,001	15,671	0,001	38,516	0,002	0,85832	0,00001	2,10957	0,00005
SD-SC-2-2	Galena (Filón San Cristóbal)	18,265	0,001	15,670	0,001	38,524	0,002	0,85794	0,00001	2,10915	0,00006
SD-SC-2-3	Galena (Filón San Cristóbal)	18,280	0,001	15,674	0,001	38,538	0,002	0,85743	0,00001	2,10822	0,00005
SD-SC-2-4	Galena (Filón San Cristóbal)	18,271	0,001	15,672	0,001	38,534	0,002	0,85776	0,00001	2,10902	0,00005
<i>Analytical sessions in July 2021, October 2021 and April 2023</i>		²⁰⁶ Pb/ ²⁰⁴ Pb	2 SD	²⁰⁷ Pb/ ²⁰⁴ Pb	2 SD	²⁰⁸ Pb/ ²⁰⁴ Pb	2 SD	²⁰⁷ Pb/ ²⁰⁶ Pb	2 SD	²⁰⁸ Pb/ ²⁰⁶ Pb	2 SD
SRM 981	n = 8	16,9437	0,0021	15,5006	0,0013	36,7284	0,0043	0,9148	0,0001	2,1677	0,0001

rigorous as possible, we have only admitted as potential sources those ore samples that coincide with a metal object in the two spaces under consideration (triplets). This topic is treated more in detail in the discussion section.

The AMALIA algorithm considers that the main, homogeneous group of artifacts from Fuente Spitz is analytically indistinguishable from a

large group of local mines in the La Carolina, Linares, and Alcudia districts. Also, they are analytically comparable to a smaller group of galenas from more distant mines in southwestern and northern Spain, as well as remote ores from Morocco, Wales, Germany and Chios (Greece). A summary of these initial results with matching to Spanish mines can be found in [Table S1 \(supplementary material\)](#).

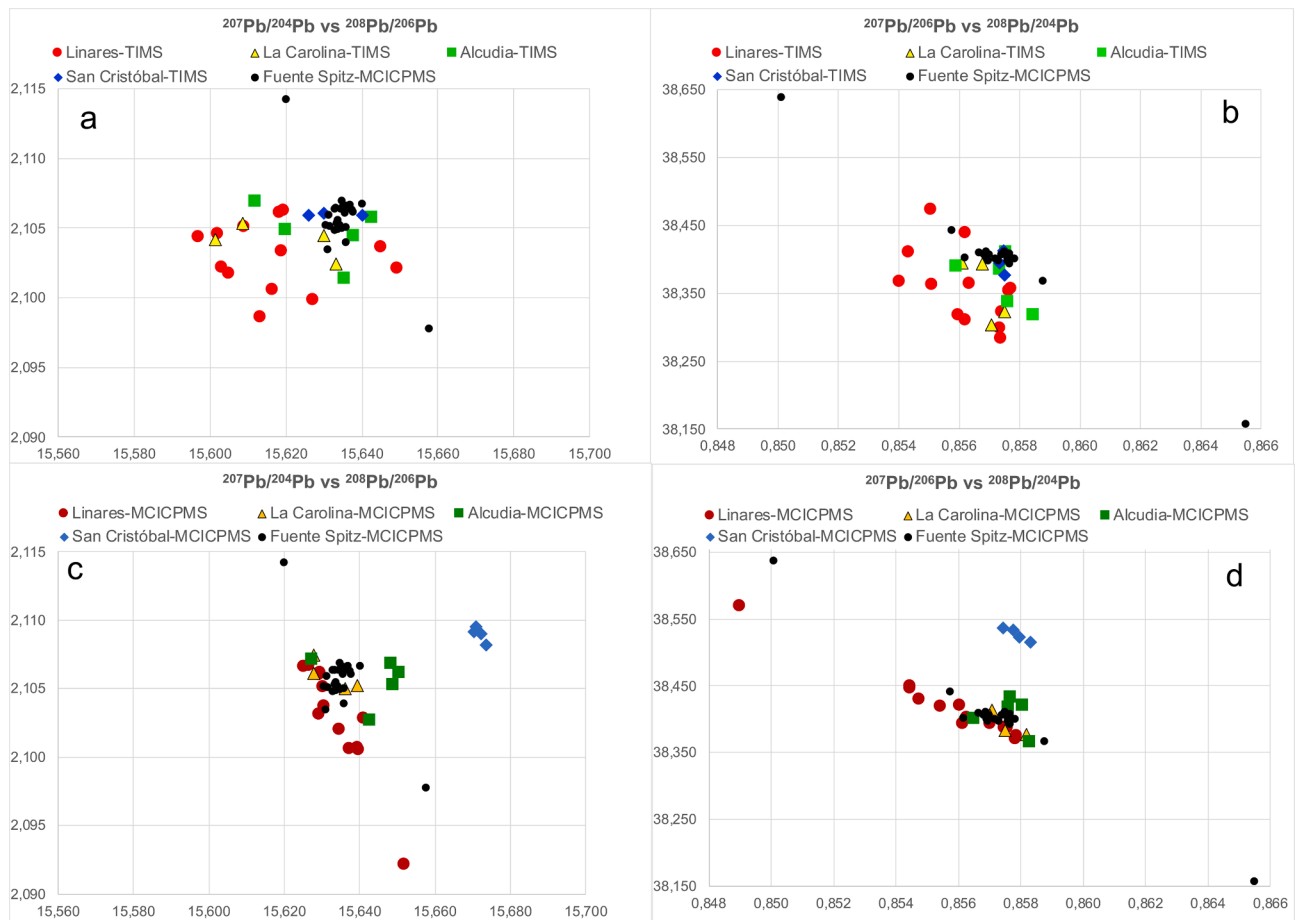


Fig. 3. Bivariate lead isotope-ratio plots showing published TIMS data for galena ores from the local Linares-La Carolina-Alcudia mining districts, and from the distant San Cristóbal mine (Fig. 3a and 3b). New MC-ICP-MS analyses of the same galena hand-samples are plotted in Fig. 3c and 3d. The analyzed objects from Fuente Spitz (MC-ICP-MS data) are displayed in the four graphs for reference.

It is important to note that despite the homogeneous isotope composition of the artifacts from Fuente Spitz, they do not match most mines from the Linares district that was extensively analyzed by García de Madinabeitia (2004; later published in Santos Zalduegui et al., 2004, but without the analytical uncertainties and with some errors in mine identification and georeferentiation). Moreover, they match the galenas from the El Centenillo mine (La Carolina district) analyzed by most authors (Dayton and Dayton, 1986; Tornos and Chiaradia, 2004; Milot et al., 2021), but they are different from the galena of El Centenillo studied by García de Madinabeitia (2004).

In an effort to check if the Linares and La Carolina districts, or even the El Centenillo mine are isotopically heterogeneous, or if there is some analytical bias in some data obtained by TIMS, new analyses were performed on a selection of the available collection of galenas studied by García de Madinabeitia (2004). The chosen samples include almost all the ores from the Linares and La Carolina mining districts (including the El Centenillo mine), and a small batch of samples from the large Alcudia district. In addition, we have analyzed four galenas from San Cristóbal mine in Sierra de la Demanda that are strikingly similar to the objects from Fuente Spitz (Fig. 3a and 3b). The new set of analyses by MC-ICP-MS is more homogeneous and shows less dispersion in bivariate diagrams. A notable shift towards higher $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios can also be seen (Table 1, Fig. 3c and 3d). The new calculations with the AMALIA algorithm offer more matches between the main group of objects from Fuente Spitz and local mines from the La Carolina and Linares districts, fewer matches with mines of the Alcudia district, and no coincidence with the small San Cristóbal mine (Table 2). In particular, 18 out of the 29 artifacts of Fuente Spitz match the new analysis of

galena sample CAR-4 of El Centenillo. In contrast, no single match was obtained previously (Table S1) with the TIMS analysis performed on CAR-4 by García de Madinabeitia (2004). The singular sling bullet Inv. 1185 has no matches with local mines but is comparable with galena from mina Miravet (Castellón) and some mines in the Basque-Cantabrian basin. Finally, there is still no single match for the spoon Inv. 1554. The complete list of analytical coincidences (about 50–70 lead ores and metals) for each artifact from Fuente Spitz is available as supplementary information to this article (Table S2).

5. Discussion

5.1. Validation of the AMALIA algorithm

The new AMALIA algorithm has demonstrated great efficacy in matching elements in a well-defined case study of a Roman smelting center located within a distance of one-day journey to active lead mines: most of the manufactured objects analyzed at the foundry of Fuente Spitz are analytically indistinguishable from galena ores from local mines in the La Carolina (El Centenillo, Los Guindos, El Sinapismo) and Linares (Matacabras, Arrayanes, La Cruz) districts. Almost all of these mines were active during the Roman period (Domergue, 1971, 1987). Moreover, the objects from Fuente Spitz are also comparable to metallic lead objects found at Cerro del Plomo, a Roman smelting site in the vicinity of the El Centenillo mine (Table 3). There are more mines that are analytically comparable to the analyzed objects from Fuente Spitz, located in Morocco, Wales, Germany and Greece. However, the long distances required to transport the heavy freight to the foundry make

Table 2

Potential provenance of archaeological objects from Fuente Spitz based on the AMALIA algorithm, after published and new data of galena from local mines. Those mines with evidence for ancient extraction are underlined. Distances estimated on current road routes. Those analyses in italics and preceded by an asterisk correspond to samples with known, poor analytical precision. See text for critical interpretation.

Analysed objects		ORIGIN (ore deposits)			
Sample	Description	Sample	Mine	Reference	Matches out of 26 objects(n)
Homogeneous group (n = 26)		La Carolina mining district (<20 km around Fuente Spitz)			
Inv. 1486	<i>Plumbarius</i>	* M31	<u>El Centenillo</u>	Milot et al., 2021	25
Inv. 1051	Faucet	JED	<u>El Centenillo</u>	Dayton & Dayton, 1986	24
Inv. 776	Seal	CAR-3	Los Guindos	This work	24
Inv. 779	Seal	CAR-2	El Sinapismo	This work	23
Inv. 973	Seal	CAR-4	<u>El Centenillo</u>	This work	18
Inv. 798	Seal	CEN-2	<u>El Centenillo</u>	Tornos & Chiaradia, 2004	11
Inv. 784	Seal	* SE-1	<u>San Gabriel</u>	García de Madinabeitia, 2004	6
Inv. 1926	Seal	CAR-1	El Sinapismo	This work	3
Inv. 1145	Seal	LIN-7A	Linares mining district (ca. 30 km S of Fuente Spitz)		
Inv. 4691	Seal	LIN-11	Grupo Matababras	This work	24
Inv. 1919	Seal	LIN-8	<u>Filon Arrayanes</u>	This work	20
Inv. 7406	Slag	LIN-12	<u>Grupo La Cruz</u>	This work	20
Inv. 1904	Weigh	LIN-3	La Fernandina	This work	12
Inv. 1905	Weigh	LIN-1	Lavadero Matababras	This work	12
Inv. 1470	Pipe	LIN-10	Grupo Matababras	This work	10
Inv. 1474	Pipe	3621	<u>Grupo La Cruz</u>	This work	10
Inv. 1475	Trap	LIN-5	La Minilla	Milot et al., 2021	3
Inv. 1475	Pipe 1 of trap	BSP2548	<u>Filón El Cobre</u>	This work	1
Inv. 304	Sheet		unknown	Hunt-Ortiz, 2003	1
Inv. 1485	Sheet	12773	Alcudia valley mining district (>80 km NW of Fuente Spitz)		
Inv. 001	Sheet	835-092a	Santa Teodora	Milot et al., 2021	22
Inv. 1472	Pot	835-015	La Veredilla	This work	21
Inv. 1163	Sling bullet	836-065	Nuestra Señora de la Paz	This work	21
Inv. 1180	Sling bullet	17125	La Gitana	This work	19
Inv. 1182	Sling bullet	* 809-027	San Teodoro	Milot et al., 2021	15
Inv. 1184	Sling bullet	11659	<i>La Cazadora/Campillo</i>	García de Madinabeitia, 2004	14
		* 809-024	La Jarosa	Milot et al., 2021	13
		836-051	<u>San Quintín (Pozo San Froilán)</u>	García de Madinabeitia, 2004	8
		835-001b	El Encinarejo	This work	5
		* 861-025	La Panadera	García de Madinabeitia, 2004	4
		11620	<i>María Aurora</i>	García de Madinabeitia, 2004	4
		27746	La Genoveva	Milot et al., 2021	3
		835-010b	El Horcajo	Milot et al., 2021	3
		835-001a	La Reina	García de Madinabeitia, 2004	2
		835-012	La Panadera	García de Madinabeitia, 2004	1
		835-030	San Luis	García de Madinabeitia, 2004	1
		835-011	<u>Romanilla (San Rafael)</u>	García de Madinabeitia, 2004	1
		* 860-031	Joffre	García de Madinabeitia, 2004	1
		11615	<i>Santa Rosa</i>	García de Madinabeitia, 2004	1
		11641	Hipólita	Milot et al., 2021	1
			San Lorenzo	Milot et al., 2021	1
			western Sierra Morena (>170 km W of Fuente Spitz)		
		GAL-41	Lomo del Perro	García de Madinabeitia, 2004	17
		BA-5	El Chaparral	Santos Zalduegui et al., 2007	3
		SOL	<u>El Soldado</u>	García de Madinabeitia, 2004	2
		MOR-2	<u>Morras de Cuzna</u>	García de Madinabeitia, 2004	1
		BRI	Santa Brigida	García de Madinabeitia, 2004	1
		MOR	Guadalupe-Morras	García de Madinabeitia, 2004	1
		GAL-39	La Herrumbrosa	García de Madinabeitia, 2004	1
			Iberian Pyrite Belt mining district (>290 km SW of Fuente Spitz)		
		* Pb-89, 401	unknown	Brill et al., 1987	3
		Pb-10	Tallesca	Marcoux and Sáez, 1994	2
		Pb-13	El Parador	Marcoux and Sáez, 1994	1
Sample	Description	Sample	Mine	Reference	
Inv. 1901	Little jug	LIN-1	Grupo Matababras	This work	
		LIN-5	<u>Filón El Cobre</u>	This work	
		LIN-9	<u>Grupo La Cruz</u>	This work	
		LIN-10	<u>Grupo La Cruz</u>	This work	
		BSP2548	unknown	Hunt-Ortiz, 2003	
		BSP2558	unknown	Hunt-Ortiz, 2003	
		836-051	El Encinarejo	This work	
		BS-11 (MP-2)	<u>Mina Polígono</u>	Jaramillo, 2005	
		Pb-13	El Parador	Marcoux and Sáez, 1994	
Sample	Description	Sample	Mine	Reference	
Inv. 1185	Sling bullet	PA20705	Miravet	Montero-Ruiz et al., 2014	
		PA20706	Miravet	Montero-Ruiz et al., 2014	
		PA22095	Miravet	Montero-Ruiz et al., 2014	
		* 1	<i>Otxamentegi</i>	Velasco et al. 1996	
		* 2	<i>Otxamentegi</i>	Velasco et al. 1996	
		* 1	<i>Sta. Bárbara</i>	Velasco et al. 1996	

(continued on next page)

Table 2 (continued)

Analysed objects		ORIGIN (ore deposits)				
Sample	Description	Sample	Mine	Reference	Matches out of 26 objects(n)	
		*	2	Sta. Bárbara	Velasco et al. 1996	
		*	Modesta	Modesta	Velasco et al. 1996	
		*	SD-MN-1R	Najerilla	Velasco et al. 1996	
		*	SD-MN5	Najerilla	Ibáñez Gómez, 1998	
		*	SD-MNm	Najerilla	Ibáñez Gómez, 1998	
Sample	Description	Sample	Mine	Reference		
Inv. 1554	Spoon	No matches				

them very unlikely as sources for raw material procurement.

There is a mismatch in data output by AMALIA depending on the independent triplet used ($^{206}\text{Pb}/^{204}\text{Pb}$ - $^{207}\text{Pb}/^{204}\text{Pb}$ - $^{208}\text{Pb}/^{204}\text{Pb}$ or $^{206}\text{Pb}/^{204}\text{Pb}$ - $^{207}\text{Pb}/^{206}\text{Pb}$ - $^{208}\text{Pb}/^{206}\text{Pb}$). The whole sets of coincidences for each triplet for the case of the Fuente Spitz artifacts is available in Table S2. The mismatch does not imply a violation of mathematics or physics in algorithm formulation. It is most likely a consequence of the way in which LIA are presented in scientific publications, that normally tabulate only one, independent triplet. In consequence, data input for the two remaining ratios that are needed to form the second triplet have to be calculated, in many cases at a loss in precision and eventually in accuracy. This is an unresolvable problem in LIA databases and also for the empiric calculation of laboratory uncertainties for SRM-981. Nevertheless, it is a minor error affecting the last significant digits in isotope ratios, far less than the analytical uncertainties. To avoid this incertitude we have successfully employed, and recommend the use of the extended algorithm to the quintet of isotope ratios $^{206}\text{Pb}/^{204}\text{Pb}$ - $^{207}\text{Pb}/^{204}\text{Pb}$ - $^{208}\text{Pb}/^{204}\text{Pb}$ - $^{207}\text{Pb}/^{206}\text{Pb}$ - $^{208}\text{Pb}/^{206}\text{Pb}$, that covers the two most commonly-used triplets and gives an internally-consistent output.

5.2. A reappraisal of legacy TIMS analyses: The importance of the uncertainty in the measurement

It is common practice in the publication of LIA in archaeological studies to provide the spectrometric measurements without their intrinsic measurement uncertainties. Many archaeologists recognize that they do not understand the meaning of the uncertainties whereas other, more experienced authors simply state that leaving the uncertainties out of the data tables render them more manageable and readable. This is bad practice, and the ISO guide 98-3 sanctioned by the International Union of Pure and Applied Chemistry (IUPAC) and the International Union of Pure and Applied Physics (IUPAP) among others, indicates that “when reporting the result of a measurement of a physical quantity, it is obligatory that some quantitative indication of the quality of the results be given so that those who use it can assess its reliability. Without such an indication, measurement results cannot be compared, either among themselves or with reference values given in a specification or standard... The concept of uncertainty as a quantifiable attribute... is a doubt about how well the result of the measurement represents the value of the quantity being measured”.

Recording the uncertainties is particularly critical in the case of TIMS data due to two factors. First, nearly if not all of the TIMS data obtained for archaeometric studies did not employ the technique of lead double- or triple-spikes, which provides the most accurate results. All those data were instead corrected externally (off-line) for the instrumental mass fractionation, by applying the empirical factors calculated for unspiked SRM 981 runs in the same analytical sessions. This method assumes that the thermal emission of ionized isotopes that is measured in the TIMS is reproducible from sample to sample and comparable to that of the SRM 981. Sample preparation of the SRM 981 involves mixing a known amount of pure lead in a fixed proportion with an ion emitter (usually a silica gel) on a rhenium filament. Ion emission from these preparations is stable. But the preparation of unknown samples for TIMS analysis is far

from reproducible: the amount of lead that is loaded on the filament is generally unknown, and it may bring impurities from the purification step, either other cations from the sample and/or organic material from the chromatographic resin. As a consequence, the mixture produced with the ion emitter on the filament may lead to unstable ion emissions, progressive emissions from different reservoirs within the filament, or weak and waning emissions. These adversities may make the ion emission and eventually the mass fractionation to diverge from that produced by the SRM 981. Second, false, underestimated $^{20x}\text{Pb}/^{204}\text{Pb}$ ratios may be obtained when measuring ^{204}Pb in weak ion emissions: at low current intensities coming into the detector, the electronic noise may contribute considerably and fatally to the total ^{204}Pb signal. Both problems (unstable sample emission and low ^{204}Pb signal) normally lead to poor counting statistics and therefore to a bad uncertainty value. Similar problems might also occur in MC-ICP-MS analysis; however, the nature of the technique allows for previous, optimal preparation of the unknown samples to reproduce the analytical conditions of the SRM 981 standard in the same session.

In sum, the fractionation factors of the SRM 981 may not accurately correct the analytical bias of every unknown sample in the TIMS and weak ion emissions may lead to underestimated $^{20x}\text{Pb}/^{204}\text{Pb}$ values. In order to identify these analytical problems, it is necessary to publish the measurement uncertainties because larger than usual uncertainties increase the doubt about the accuracy of the measurement. Consequently, an undefined number of lead isotope analyses present in databases might be inaccurate. Unfortunately, the only way to verify if imprecise analyses are accurate or not requires them to be re-analyzed one by one with any technique that allows for internal correction of mass fractionation (lead double- or triple-spikes, thallium addition). This has been tested in this work for some of the data reported by García de Madinabeitia (2004) for the galena ores of La Carolina, Linares and Alcadia districts (Spain), which showed uncertainties of up to 0.5 % in the $^{20x}\text{Pb}/^{204}\text{Pb}$ ratios. The new measurements have good uncertainties, lower than 0.1 % (Table 1) and give more confidence in the obtained ratios. Moreover, these new values present higher $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios (Fig. 2), strongly suggesting their underestimation in the original TIMS analyses and therefore their inaccuracy. The original values should be replaced by our new results in databases and in any provenance study. Similar results have been obtained for the galenas from the San Cristobal mine (Velasco et al., 1996), which were strikingly comparable to metallic objects from Fuente Spitz despite the long distance from the mine to the foundry. The authors did not report the uncertainties on the original measurements by TIMS. However, the new data also show higher $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$, suggesting again their underestimation in the original analyses. As a result, the new data for the San Cristobal mine do not match the objects from Fuente Spitz and this distant mine can be discarded as a potential source for Fuente Spitz objects.

We would not want that all this subsection is erroneously understood as a statement against using TIMS as a valid technique for LIA and metal provenancing. In fact, none of the methods is free from analytical imprecisions and/or inaccuracies (Thirlwall, 2002), and good practice in spectrometric protocols and sample preparation for TIMS may minimize the possibility of obtaining inaccurate results.

Table 3
Potential distribution of metallic lead and silver from Fuente Spitz smelting site. See text for critical interpretation.

Analysed objects		DISTRIBUTION (COMMERCE, ARMY)					
Sample	Desc.	Sample	Settlement	Country	Reference	Object	Matches (n) out of 26 objects
Homogeneous group (n = 26)		<i>Roman local lead items</i>					
Inv. 1486	<i>Plumbarius</i>	PRE1946	Cerro del Plomo - El Centenillo	Spain	This work	Lead seal	24
Inv. 1051	Faucet	PRE1954	Cerro del Plomo - El Centenillo	Spain	This work	Lead seal	24
Inv. 776	Seal	3	Cerro del Plomo - El Centenillo	Spain	Trincherini et al., 2001	Lead metal	18
Inv. 779	Seal	5	Fuente Espí (Fuente Spitz)	Spain	Trincherini et al., 2001	Galena - Archaeological	2
		<i>Roman shipped lead ingots</i>					
Inv. 973	Seal	12,981	Cabrera 5 (wreck)	Spain	Trincherini et al., 2001	Lead ingot	25
Inv. 798	Seal	12,988	Cabrera 5 (wreck)	Spain	Trincherini et al., 2001	Lead ingot	9
Inv. 784	Seal	12,813	Cabrera 5 (wreck)	Spain	Trincherini et al., 2001	Lead ingot	7
Inv. 1926	Seal	12,999	Cabrera 5 (wreck)	Spain	Trincherini et al., 2001	Lead ingot	6
Inv. 1145	Seal	12,994	Cabrera 5 (wreck)	Spain	Trincherini et al., 2001	Lead ingot	1
Inv. 4691	Seal	98–49	Cabrera 4 (wreck)	Spain	Domergue et al., 2013	Lead ingot	1
		<i>Roman military weapons</i>					
Inv. 1919	Seal	Mü09-37	Cerro de las Balas (C. de la Atalaya)	Spain	Müller et al., 2015	Sling bullet	23
Inv. 7406	Slag	Mü09-37	Cerro de las Balas (C. de la Atalaya)	Spain	Müller et al., 2015	Sling bullet	23
Inv. 1904	Weight	ARC779 PB1	Thuin Oppidum	Belgium	Paridaens et al., 2020	Sling bullet	23
Inv. 1905	Weight	MZ-14	Ategua	Spain	Müller et al., 2015	Sling bullet	22
Inv. 1470	Pipe	Mü05-19	Cerro de las Balas (C. de la Atalaya)	Spain	Müller et al., 2015	Sling bullet	10
Inv. 1474	Pipe	Mü08-46	Cerro de las Balas (C. de la Atalaya)	Spain	Müller et al., 2015	Sling bullet	1
		<i>Roman, non-local lead and lead-bearing items</i>					
Inv. 1475	Trap	Pb- 847, 1701	Rio Tinto	Spain	Brill et al., 1987	Lead metal	20
Inv. 1475	Pipe 1 of trap	CMP.1.09.R2.35.2	Castromaior	Spain	Orejas Saco del Valle et al., 2015	Bronze Aes (Calagurris mint)	15
Inv. 304	Sheet	CMP.1.09.R2.35.2	Castromaior	Spain	Orejas Saco del Valle et al., 2015	Bronze Aes (Calagurris mint)	15
Inv. 1485	Sheet	CL79/T1/77	Corta Lago	Spain	Stos-Gale, 2001	Litharge	1
Inv. 001	Sheet	DA707,8	Dangstetten	Germany	Durali-Müller, 2005	Lead object	1
		<i>Roman silver distribution</i>					
Inv. 1472	Pot	DA707,8	Dangstetten	Germany	Durali-Müller, 2005	Lead object	1
Inv. 1163	Sling bullet	TDM 57 A	Marengo	Italy	Angelini et al., 2019	Silver in decorated belt	23
Inv. 1180	Sling bullet	CMP.1.09.E36.6.16	Castromaior	Spain	Orejas Saco del Valle et al., 2015	Denarius (Lugdunum mint)	12
Inv. 1182	Sling bullet	MS042	Museum	Germany	Westner et al., 2020	Denarius (Rome mint)	11
		<i>Pre-roman silver distribution</i>					
Inv. 1184	Sling bullet	MS042	Museum	Germany	Westner et al., 2020	Denarius (Rome mint)	11
		9807	Cerro de la Virgen, grave 22b	Spain	Bartelheim et al., 2012	Silver bracelet (Bronze Age)	24
		Ring 4	Palacio III	Spain	Murillo-Barroso et al., 2015	Silver ring (Iron Age)	18
		PA20440	La Ayuela	Spain	Murillo-Barroso et al., 2016	Silver ring (Iron Age)	17
		MU-25	San Antón	Spain	OXALID	Silver ring (Bronze Age)	6
		RMG 30	Tomb 66A	Italy	OXALID	Silver chain	6
		37,919 (MA-114721)	El Oficio	Spain	Murillo-Barroso, 2013	Silver ring	2
		MO-21294 (4630–11)	Cerro de la Encina	Spain	Murillo-Barroso, 2013	Small silver nail	1
		Ring 3	Palacio III	Spain	Murillo-Barroso et al., 2015	Silver ring (Iron Age)	1
		Gr2	Palacio III	Turkey	Desaulty et al., 2011	Silver coin (Ionia Miletos mint)	1
		26	Antas de Ulla	Spain	Comendador Rey et al., 2014	Silver chain	1
		33	Antas de Ulla	Spain	Comendador Rey et al., 2009	Silver chain	1
		9824	Cerro de la Virgen, grave 22a	Spain	Bartelheim et al., 2012	Silver ring (Bronze Age)	1
		13,050	Cuesta del Negro, grave 4	Spain	Bartelheim et al., 2012	Silver ring (Bronze Age)	1
		G25	Gatas/Gatar	Spain	OXALID	Silver ring	1
		13,703	Terrera del Reloj	Spain	Bartelheim et al., 2012	Silver ring (Bronze Age)	1

(continued on next page)

Table 3 (continued)

Analysed objects		DISTRIBUTION (COMMERCE, ARMY)						
Sample	Desc.		Sample	Settlement	Country	Reference	Object	Matches (n) out of 26 objects
Sample Inv. 1901	Description Little jug	<i>Roman silver distribution</i>	Sample	Settlement	Country	Reference	Object	
		<i>Pre-roman silver distribution</i>	MS042	Museum	Germany	Westner et al., 2020	Denarius (Rome mint)	
			Gr2		Turkey	Desaulty et al., 2011	Silver coin (Ionia Miletos mint)	
			G35	Gatas/Gatar	Spain	OXALID	Silver earring	
			4344/11	Acinipo	Spain	Renzi et al., 2016	Silver bead	
Sample Inv. 1185	Description Sling bullet	<i>Roman, non-local lead items</i>	Sample	Settlement	Country	Reference	Object	
			CH14	Cerro de la Alegria	Spain	Cuchf-Oterino et al., 2021	Loom weight	
			CH18	Ripoll-Saso	Spain	Cuchf-Oterino et al., 2021	Loom weight	
			43-10-00-12-24	Vagnari	Italy	Carroll et al., 2021	Cut sheet	
			P6984	Vagnari	Italy	Carroll et al., 2021	Sieve fragment	
			90.3-260 (3)	Cartaghe - Roman Circus	Tunisia	Skaggs et al., 2012	Curse tablet	
			90.3-260 (2)	Cartaghe - Roman Circus	Tunisia	Skaggs et al., 2012	Curse tablet	
		<i>Roman silver distribution</i>	CMP.1.09.E36.6.15	Castromaior	Spain	Orejas Saco del Valle et al., 2015	Denarius (Hispania)	
			CMP.1.09.E36.6.13	Castromaior	Spain	Orejas Saco del Valle et al., 2015	Denarius (Rome mint)	
		<i>Pre-roman silver distribution</i>	MS027	Museum	Italy	Westner et al., 2020	Denarius (Rome mint)	
			CE08310	Laurita - Cerro de San Cristóbal	Spain	Murillo-Barroso et al., 2016	Silver scarab ring	
			A/CE09606	Toscanos	Spain	Murillo-Barroso et al., 2016	Silver fibula?	
Sample Inv. 1554	Description Spoon		Sample No matches	Settlement	Country	Reference	Object	

5.3. Comparison with other mathematical methods

The AMALIA algorithm compares satisfactorily with recently published numerical methods used to assign provenance. For example, DeCeuster and Degryse (2020) provide a “match-no match” statistical method based on kernel density estimates (KDE) that allowed them to assign potential provenances for two well-studied Roman lead ingots from the Bajo de Dentro wreck (off Cartagena, Spain) and the Haltern military camp (North Rhine-Westphalia, Germany). The KDE method assigned the lead employed in the manufacture of these ingots to local sources from Spain and Siegerland (Germany), respectively. By using the AMALIA algorithm (see Appendix S3 in Supporting Information) we can better indicate that the Spanish ingot was most probably produced from galena extracted from some of the Mazarrón mines (Cartagena mining district) or alternatively from some nearby mines in Almería (Lorca-Sierra Almagrera). Similarly, the German ingot is identical to galena ores from the Lüderick mine (Bensberg district, North Rhine-Westphalia) and the Victoria mine (Littfeld, Siegerland), as well as to one remote mine from Morocco. The Lüderick mine was exploited since Roman times, so it is the best potential source for the German ingot, as previously indicated by Bode et al. (2009).

We have also tested the Euclidean distances method of Birch et al. (2020) in our case study and with our own database. As might be expected, because both methods are based on distances in a geometric space, most matches of the AMALIA algorithm also have the smallest

Euclidean distances (Table S2). Some discrepancies arise however because the Euclidean distance of Birch et al. is calculated as a magnitude without the vector information (the direction of the magnitude in three-dimensional space). In other words, the Euclidean distance considers the magnitudes along any of the three axes equally (e.g., $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$), whereas AMALIA establishes that two samples are at zero distance (are analytically indistinguishable) at different magnitudes along each axis. Graphically, for any analysis represented as a unidimensional spot, the threshold value of the Euclidean distance towards comparable, neighboring spots would draw a sphere around the first spot, the Euclidean distance being the radius of the sphere. In the case of the AMALIA algorithm, the zero distances represent the three radii of an ellipsoid around the spot.

5.4. Fuente Spitz: An archaeometric and archaeological interpretation

The homogeneity of the isotopic signatures of all the objects analyzed (with the exception of samples Inv. 1554 and Inv. 1185) from Fuente Spitz, together with the strong match between them and the ores from La Carolina and Linares (Table 1), show that the foundry was established to process ores exclusively from these local mining districts. The recycling of previously existing lead products, at least of those that were derived from lead mining areas with different isotopic readings, was not significant. If this were the case, more heterogeneous isotopic readings should be expected, as happened with the *grandes plomos* from

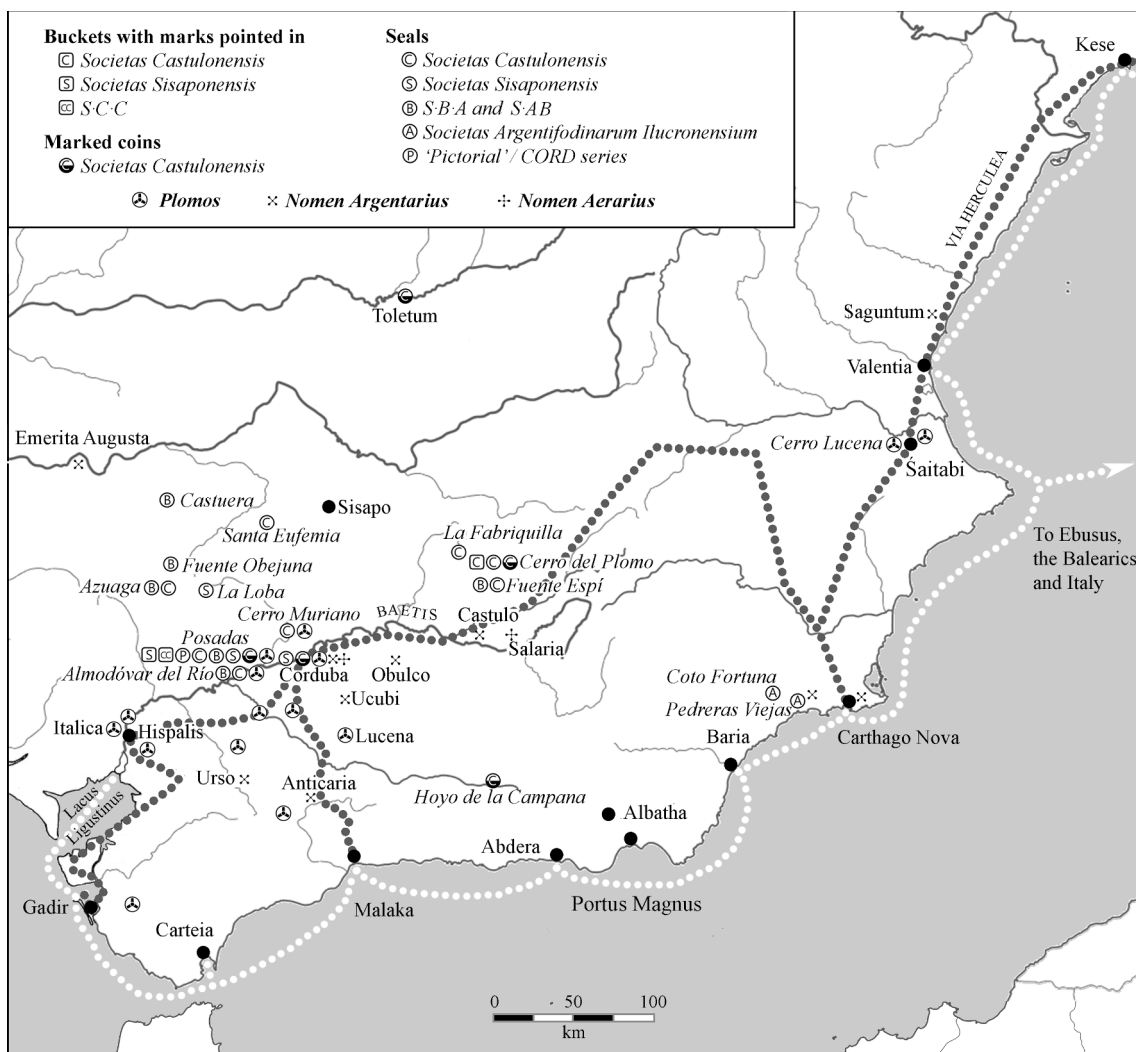


Fig. 4. Evidence for the anonymous mining societates of Baetica (Stannard et al., 2021).

Baetica (Fig. 4). Their lead is attributed in some cases to the south-east of Spain (Cartagena-Mazarrón mines) and in others as a mixture of Cartagena-Mazarrón lead with that from Linares, Ossa Morena and the Alcuía Valley (Stannard et al., 2019). The only sample at Fuente Spitz that allows for such recycling practices is Inv. 1554 for which there is no match in our database. But this could also be a consequence of mixing different ores at the foundry.

However, in order to better understand the exact provenance of the ores, how local those mines were, and the distribution of the products manufactured at Fuente Spitz, we conducted a second data treatment using AMALIA. The goal was to compare the isotope signatures of the 29 objects from Fuente Spitz with the archaeological objects in the MedConTaCCT databank as well as two newly analyzed lead seals from the near Cerro del Plomo smelting site (Table 1: Inv. 1946 and Inv. 1954). The test clearly shows a complete analytical affinity between the objects recovered at Fuente Spitz and the lead seals at Cerro del Plomo and several ingots found at the Cabrera 5 wreck (Table 3). Taking into account the proximity between the two foundries and in relation to the El Centenillo mine, with which our samples analyzed also match isotopically, it is quite likely that both smelters were mostly supplied with ore from this mine. This proves that proximity to the mining exploitations, which means low transportation costs, was a key element in selecting the location of foundries like Fuente Spitz and Cerro del Plomo. Our results also indicate that foundries such as the ones discussed here did not process the ores of whole mining regions, but instead those from one or a few nearby mines.

It has been suggested that Roman mining companies not only operated in different mining districts but also controlled the entire production process, from mineral extraction to metal production. While this seems to be true in many cases (e.g. all the inscribed objects recovered from Cerro del Plomo belong to the *societas Castulonensis* or *Cordubensis* (-S•C-, see Fig. 4), the fact that seals inscribed S•C and S•AB (the last, abbreviation of *Societas A(rgentifodinarum) B(---)*; Antolinos Marín et al., 2013) have been recorded in Fuente Spitz may indicate that alternative arrangements were also possible. Lead seals were used to guarantee that the bags of ore were not tampered with during their transfer from the mines to the foundries (Hill and Sandars, 1911), or alternatively as quality trading marks attached to a label (Stannard et al., 2021). If, as isotopic data suggest, the ores reaching Fuente Spitz and Cerro del Plomo came from the exploitation of the El Centenillo mine, the fact that seals from at least two *societates* (S•C and S•AB) were recovered at Fuente Spitz indicates that these two companies worked the same mine. The dominance of the S•C marks in the region and especially in the foundries of Cerro del Plomo and Fuente Spitz (Fig. 4) could suggest that only certain *societates* owned foundries and controlled the entire production process (S•C in this case), while others, such as S•AB, did not, and either sold their ores to larger companies that owned foundries and smelting facilities or rented them to process their ores. This hypothesis would explain the matching isotopic results as well as why the seals marked as S•AB are less common and only appear in Fuente Spitz and not in Cerro del Plomo. Unfortunately, while this hypothesis is attractive, we need to be cautious and consider it just as a possibility. The lack of archaeological context and chronological adscription of the seals at Fuente Spitz cannot rule out the possibility that the mining company exploiting El Centenillo also owned Fuente Spitz and just changed ownership over time from S•C to S•AB (or vice versa).

The isotopic results together with the diverse typology and purpose of the objects analyzed show that foundries like the Fuente Spitz and Cerro del Plomo produced lead ingots to be exported overseas, but also manufactured lead objects to be used/sold locally and regionally. The production of ingots is corroborated by the isotopic match between the objects at Fuente Spitz and Cerro del Plomo and the lead ingots nos. 12981, 12988, 12813 and 12999 from the Cabrera 5 wreck (Table 3). The existence of at least one ingot (although three are mentioned in the literature) among the objects recovered at Fuente Spitz also supports this

idea. Unfortunately, the ingots mentioned by Gimeno Pascual and Ramos Miguel (2021) are not in the Museum of la Carolina and therefore we were unable to analyze them. The hypothesis that suggests that foundries such as Fuente Spitz and Cerro del Plomo also manufactured all sorts of lead objects for their commercialization is also strongly supported by our isotopic results, as well as by epigraphic evidence.

The remains of an inscribed lead pipe with the inscription [---] *usculi plumbari • ex offf(icina?)* [---] found in the surroundings of Fuente Spitz around 1987 (Gimeno Pascual and Ramos Miguel, 2021), epigraphically attest a *plumbarius* in the area. This means that a manufacturer of lead objects, which had among its specializations the production of *fistulae aquariae* (Bruun, 1991), can be located at or near Fuente Spitz. As for the isotopic evidence, AMALIA has identified the analytical concordance between several objects in the Fuente Spitz collection (pipes, sarcophagi, seals, plumb bobs) and various lead sling bullets from Ategua (Corduba) and Cerro de las Balas (Seville), dated around 46–45 BCE in the context of the civil war between Pompey and Caesar (Müller et al., 2015), as well as sling bullets employed in the 57 BCE siege of the *oppidum* of Thuin, Belgium (Paridaens et al., 2020). These sling bullets also match isotopically with some galena ores from mines near La Carolina-El Centenillo (Los Guindos, El Sinapismo) and Linares. The archaeological dating provided for all these matching sling bullets in Corduba, Seville and Belgium fits well the period of activity of Fuente Spitz (1st c. BCE – end of the 1st c. CE) and Cerro del Plomo (1st. c. BCE – mid 1st c. CE) (Domergue, 1971) and may well provide evidence of the regional and long distant commercialization circuits that these foundries were supplying.

Other than the distribution of metallic lead, it is also clear that Romans profited the silver extracted at the local mines of La Carolina and sent the precious metal to the centers of power for coinage minting and the fabrication of luxury items. The exploitation of silver at these mines began before the Roman conquest, as is witnessed by the circulation of silver objects that match the lead isotope signature of La Carolina mines across southern Spain since the Bronze Age (Bartelheim et al., 2012). Silver origin at La Carolina-El Centenillo is clear for those artifacts with a great number of matches with the AMALIA algorithm. In addition, there is a large group of silver objects with few matches. We have only tested them with the group of lead objects from Fuente Spitz and their origin remains unclear. A mix of silver of La Carolina with volumetrically minor silver from an unknown, exotic source may explain the poor match with the lead objects from La Carolina. Clearly a thorough study that is out of the scope of this work is needed to shed more light on this topic. Additionally, the single match with an ancient Greek coin from Ionia Miletos should be considered an analytical coincidence with no archaeological ground. Especially because we have found a large number of matches between the lead objects from Fuente Spitz and some galena from Chios, Greece (Wagner et al., 1983–1984; Stos-Gale and Gale, 2009: OXALID database).

6. Conclusions

This article is the first to present the methodology employed by the *Mediterranean Connectivity, Trade and Commercial Circuits in the Roman West* (MedConTaCCT) project (<https://medcontactct.uvic.ca/>), including A Matching Algorithm for Lead Isotope Analyses (AMALIA) that correlates raw materials and archaeological artifacts. This study shows that the AMALIA algorithm is effective and yield accurate results, and should become an important tool in metal provenance studies. It also emphasizes the need to carefully evaluate the accuracy of isotopic results obtained using TIMS on a case-by-case basis. From an archaeological perspective, the archaeometric results provide a more detailed and nuanced interpretation of how foundries and smelting sites such as Fuente Spitz and Cerro del Plomo functioned, demonstrating that these workshops processed galena ores from mines of La Carolina and Linares mining districts to produce a variety of manufactured lead objects, such as the sling bullets recovered in Corduba, Seville and Belgium, as well as

lead ingots, including some of those recovered from the Cabrera 5 wreck (Colls et al., 1986; Domergue, 2004). The objects produced at Fuente Spitz and Cerro del Plomo were distributed regionally as well as overseas to distant Roman provinces.

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CRediT authorship contribution statement

Javier Rodríguez: Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Data curation, Writing – original draft, Writing – review & editing. **Alejandro G. Sinner:** Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing – original draft, Writing – review & editing, Funding acquisition. **David Martínez-Chico:** Investigation, Visualization, Writing – original draft, Writing – review & editing. **José Francisco Santos Zalduegui:** Methodology, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary material

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