Lead isotopes in Celtiberian denarii from *Turiasu* and Roman asses minted in cities of the *Conventus Caesaraugustanus* (Hither Spain)

José Antonio Cuchí[1], Jesús Martín-Gil[2], Isidro Aguilera Aragón[3] and Pablo Martín-Ramos[1,*]

[1] EPS, Universidad de Zaragoza. Carretera de Cuarte s/n, 22071 Huesca, Spain
[2] Laboratory of Heritage Conservation (LICOPCYL), ETSIIAA, Universidad de Valladolid, Avenida Madrid 57, 34004 Palencia, Spain
[3] Museo de Zaragoza, Plaza los Sitios, 6, 50001 Zaragoza, Spain

[*] Corresponding author: pmr@unizar.es

Abstract

Isotope analysis is a flexible and powerful tool for provenance studies of archaeological objects. In the present study, lead isotopic analysis was used in order to gain insight into the origin of the mineral sources of coins minted in the Celtiberian mint of *Turiasu* and in six *coloniae* and *municipia* of the *Conventus Caesaraugustanus* (Hither Spain) under Augustus and Tiberius (27 BC – 37 AD). In the case of *Turiasu* denarii, *argentum* from the Iberian Range mines was used, and in the Roman asses coined in *civitates* of the middle Ebro River, the copper minerals had a diverse origin (Pyrenean, from the Iberian Range, from the southeastern Iberian Peninsula and from the Central Iberian Zone). Differences in ore provenance and in bulk composition (investigated through X-ray fluorescence) were not only detected among the early Augustan, late Augustan and early Tiberian periods, but also among the mints under study. Moreover, differences were also detected among the mints of the *Conventus Caesaraugustanus* and those that supplied the official mint of Rome, evidencing that the former enjoyed autonomy in terms of the choice of ore origin and processing.

Keywords: archaeometry; Cu; isotopic analysis; Pb; Roman coins

1. Introduction

Linking metal archaeological objects and the source of the mineral used in their manufacture is of interest to gain insight into the sources of raw materials and how they were marketed in a specific geographic area at a certain time (Rehren and Pernicka, 2008, Chiarantini, et al., 2018, Soares, et al., 2020). In a first attempt to understand this problem in the middle basin of the Ebro River (Zaragoza, Spain), between the 1st century BC and 1st century AD, the minting of 3 Celtiberian denarii from *Turiaso* (Roman Tarazona) and 16 asses and 1 semis from other 6 mints from the *Conventus Ceasaugustanus* (Figure 1a) −an administrative subdivision of the Roman province of Hispania Tarraconense− was chosen.

The selection of the mints was made on the basis of their significance within the Roman Empire commercial networks. *Turiasu* (*Turiaso*, Tarazona, Zaragoza) was a Celtiberian city that, between 140 and 70 BC, coined one of the largest productions of silver denarii of the republican period in Roman Hispania, characterized by the use of *ka-s-tu* signs on the obverse. This production was disproportionate for such a small settlement and it is probably related to tax payments or perhaps financing armies acting nearby. An interesting problem is to elucidate the degree of Roman intervention in this process, since the minting seems to be closely related to their presence (Gozalbes Fernández de Palencia, 2009).

Another important Celtiberian mint was *Bilbilis* (Calatayud, Zaragoza), later renamed *Municipium Augusta Bilbilis* by Augustus, which issued asses and semises with the Latin legend of *Bilbilis Italica* till Caligula.

The primitive Celtiberian mint of *Kelse* (Velilla de Ebro, Zaragoza) gave way, between the years 44 and 36 BC, to the *Colonia Victrix Iulia Lepida,* subsequently renamed (in 36 BC) as *Colonia Victrix Iulia Celsa.* During the period of August and Tiberius (27 BC–37 AD), it continued its coinage of typically military values (asses) with the imperial symbols (bust and Civic Crown) on the obverse and the characteristic bull of the Ebro region with the names of the monetary magistrates on the reverse. The coinage was abundant.

Municipium Calagurris Iulia Nassica (former *Kalakorikos*, nowadays Calahorra, La Rioja) issued in imperial times abundant asses and semises, playing an important role in supplying the army. The semis with the front head of the bull deserves special mention.

The *Caesar Augusta colony* (Zaragoza) was founded during the reign of Augustus (around 4 BC) with veterans of the IV, VI and X legions where the Iberian *oppidum* of *Saltuie* used to be. As a mint, it became the first in number of coin dies in all of Hispania. Its iconography was military and, in some asses, the foundational yoke appeared.

Another Celtiberian mint that resumed its activity in the times of Tiberius (14–37 AD) was *Kaiskata*, converted into *Municipium Cascantum* (Cascante, Navarre), which also issued asses and semises.

Of particular interest is *Graccurris* (Alfaro, La Rioja), a *municipium* founded by Tiberius Sempronius Gracchus in 179 BC to settle the defeated local population after the first Celtiberian war, on the ruins of the city of *Ilurci*, which also issued asses and semises with the bull for the units or the bull's head for the divisors.

The analysis of lead isotopes is the most common technological resource in archaeometry for tracing sources of raw materials, both for metals and alloys (Stos-Gale, et al., 1995, Stos-Gale and Gale, 2009, Baker, et al., 2006, Holmqvist, et al., 2019). Hence, this technique was adopted to try to identify the mineral origin of the material used in each coin, based on comparisons of the isotopic ratios of the coins with those of primary ores (reported by Subías *et al*. (Fanlo, et al., 1998, Subías, et al., 2015, Subías, et al., 2010, Subías, et al., 1997) and García-Sansegundo, et al. (2014) for Pyrenean and Iberian Range mineral sources, and by Klein, et al. (2009) for mineral sources of the southern Iberian Peninsula).

The results should contribute to expand the body of knowledge on Roman coins in the time of Augustus and Tiberius (27 BC – 37 AD), complementing the comprehensive work on aes (but not on mints) by Klein, et al. (2004), the study on 6 silver coins from Ampurias and 5 coins from Mas Castellar de Pontos (Montero Ruiz, et al., 2008), and the study on the provenance of the metals of coins of the NE Iberian Peninsula by Montero-Ruiz, et al. (2011) (which covers 13 coins from the mints of *Iltirtasalirustin*, *Iltirtasalirban*, *Bolskan*, *Kese*, *Iltirkesken*, *Iltirta*, *Ilerda* and *Kelse*, part of the Museum of Lérida collection). The former work, on 241 copper-based coins from the Museo Nazionale Romano collection, minted at the official mint of Rome, found good matches with mineral deposits from the SW Iberian Peninsula (Rio Tinto) and the SE Iberian Peninsula (Almeria and Murcia) districts, with temporal changes in the mixing ratios and mixed ore bodies. In the two studies by Montero-Ruiz *et al.*, a high frequency of recycling and a diversity of sources was found, with a predominance of SE Iberian Peninsula (Murcia area) raw material provenance. Nonetheless, the usage of an unknown source is suggested for the silver coins from *Bolskan* and the bronze coins from *Iltirta*. In this work, alternative ore resources (from the Iberian Ranges and Pyrenees) are also considered, investigating if these local ores could have been exploited in Roman times.

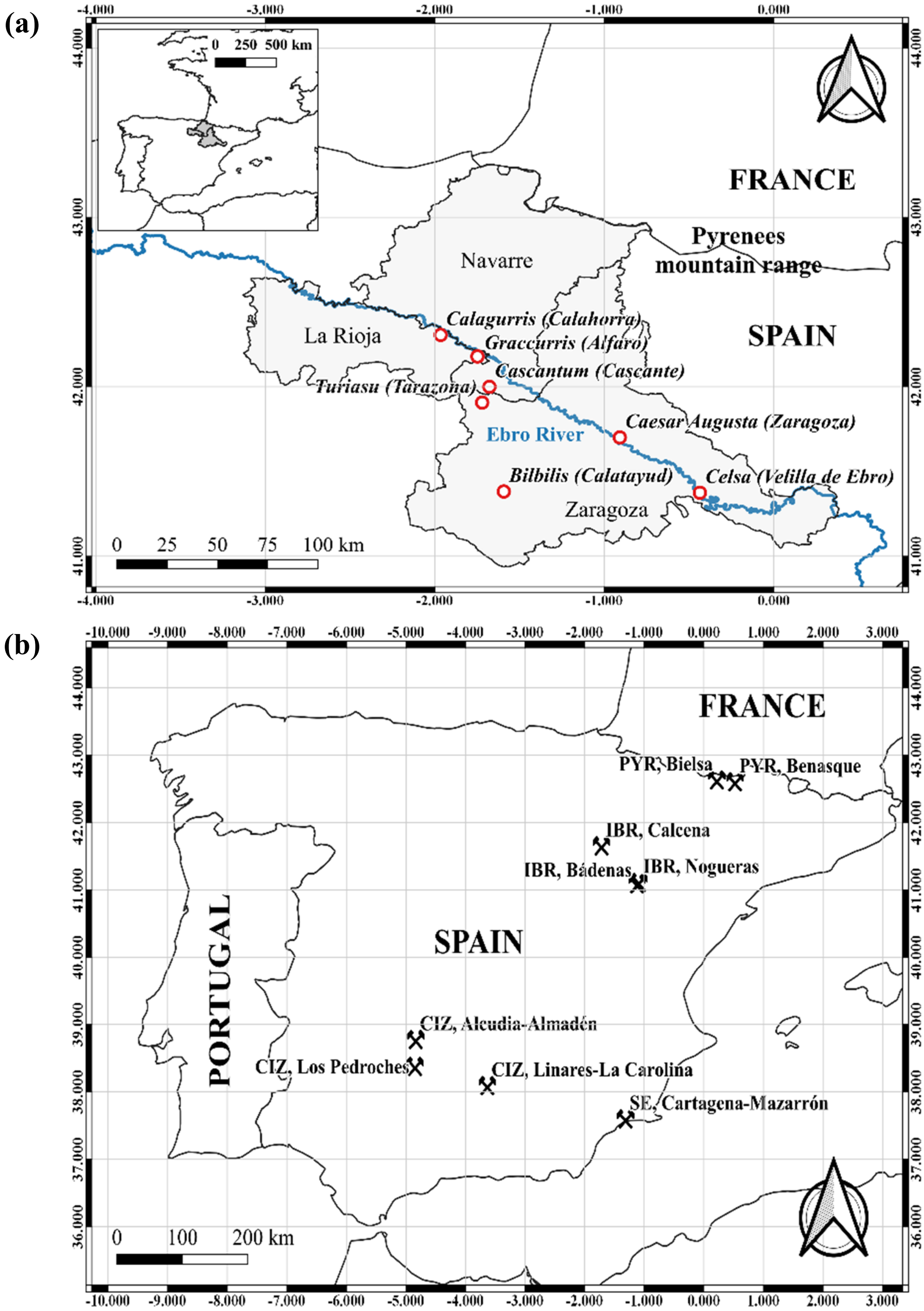

Figure 1. **(a)** Location of the six mints that issued the 20 Celtiberian and Roman coins under study. Modern names of the cities are indicated between parentheses. *Inset*: Location of La Rioja, Navarre and Zaragoza modern provinces in Spain. **(b)** Location of ores sources used in the provenance analysis.

2. Materials and methods

2.1. *Studied samples*

The coins under study (Figure S1) are part of Museo de León collections (León, Spain). The coins were sampled by drilling to obtain fresh and chemically representative metal. Drilling chips from the first few micrometers were discarded to avoid surficial material potentially affected by, for instance, corrosion processes or conservation treatment. The rest of the drilled material was first subjected to non-destructive XRF characterization and then it was chemically dissolved and measured for its Pb isotope signature.

2.2. *Chemical analysis*

Major and minor elements were determined by using a Niton XL3t GOLDD+ XRF Analyzer (ThermoFisher Scientific), using the TestAll™Geo mode for the analysis of major and trace elements (i.e. an overall composition of the sample). X-ray tube: Au anode, 50 kV, 200 µA; 3 mm small-spot collimation. Data were processed using the Niton Data Transfer (NDT™) PC software suite.

2.3. *Lead isotopic analyses*

The lead isotopic analyses were carried out at Laboratorio de Técnicas Instrumentales (LTI), Universidad de Valladolid (Valladolid, Spain), by Multicollector-Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS), according to the procedure reported by Huelga-Suarez, et al. (2014). To digest the samples, they were deposited in 15 mL PFA vials, to which 8 mL of 3:1 HCl/HNO_3 was added. The closed vials were heated at 110 °C for 24 hours and, after evaporation to dryness, 2 mL of 1M HNO_3 were added. Then they were heated at 110 °C for 30 minutes, leaving the samples prepared for the chemical separation of lead from their matrix. This was done using the PbSpec selective resin: 100 mL of Milli-Q water was added to 15 g of resin, the supernatant was replaced by Milli-Q water (twice) and the resin was loaded on a Bio-Rad column of polypropylene in order to obtain a base of 0.5 mL of resin. The separation columns were washed before use by successive dives (24 hours each) in sub-boiling baths of 10% HCl (v/v), 10% HNO_3 (v/v) and Milli-Q water. Once the resin was loaded, 2 mL of Milli-Q water was added to eliminate the possible residual lead. Immediately afterwards, 1 mL of 1M HNO_3 was used for conditioning the resin and 1 mL of sample was added in 1M HNO_3 medium. Next, the matrix of the sample was removed by using 6 mL of 0.14M HNO_3. Lead elution was performed with 5 mL of 0.05M ammonium oxalate [$(NH_4)_2C_2O_4 \cdot H_2O$], a solution that was subsequently brought to 110 °C until dryness. The addition of 4 mL of *aqua regia* ensured the digestion of the possible organic content introduced by the ammonium oxalate. After heating at 110 °C for 24 hours, the samples were evaporated again to dryness and, finally, they were re-dissolved in 2 mL of 0.42M HNO_3.

For the measurement of lead isotopic relationships by MC-ICP-MS, the pure lead fractions obtained were diluted with 0.42M HNO_3, adjusting their concentration so that they did not saturate the equipment detectors and, subsequently, they were introduced into the MC-ICP-MS (Neptune, Thermo Scientific) following the "sample standard bracketing" sequence. For this purpose, a solution of 200 $\mu g \cdot L^{-1}$ of the certified reference material in isotopic lead composition NIST SRM 981 was used. In addition, NIST SRM 997 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 was performed via cluster analysis in SPSS software (IBM, Armonk, NY, USA). Lead isotopic analyses reported by Subías *et al*. (Fanlo, et al., 1998, Subías, et al., 2015, Subías, et al., 2010, Subías, et al., 1997) and by García-Sansegundo, et al. (2014) for Pyrenean and Iberian Range mineral sources, and by Klein, et al. (2009) for mineral sources of the southern Iberian Peninsula, were taken as a reference (Figure 1b). The starting point was a hierarchical cluster analysis 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 coins.

3. Results

3.1. *Typology and chemical composition of the coins under study, grouped according to mints*

The typological descriptions, physical characteristics and elemental analyses of each of the examined coins are summarized in Table 1.

From the elemental analysis results (Table 1), it may be noted that the denarii from *Turiasu* displayed homogeneous silver contents in the upper limit of the range reported in the literature (ca. 85-95%), comparable to the purity of coetaneous republican denarii, with silver contents of approximately 97% (Gozalbes Fernández de Palencia, 2003). Such homogeneity and high purity were generally associated with payments to the legions and other expenses related to the Roman intendancy (Gozalbes Fernández de Palencia, 2003).

Regarding the Augustus bronzes minted in his early period (16-6 BC), they had very different lead contents: high in *Bilbilis* (coin 145) and low in *Celsa* (203, 200 and 201). However, in subsequent coin emissions from *Calagurris* (159, 163, 170, 158 and 160) there was greater uniformity and they were leaded bronzes with a 3.6 wt% average Pb content.

In the early Tiberian period, while *Caesar Augusta* coined binary bronze, with very low lead content (150 and 152), *Calagurris* (174 and 175) coined leaded bronzes, containing up to 7.6 wt% Pb. *Cascantum* and *Gracurris* issued bronzes with intermediate values (2.1-3.8 wt% Pb).

Table 1. Elemental analysis results, typology and mint details for the 20 coins under study. Coins are organized by mint.

Code	Elemental analysis (wt%)*	Description and typology	Mint
137 (Box 4) 1991/6/2/19	Ag: 96.2 % Cu: 2.8% Pb: 0.6%	Iberian denarius. Manly head right, framed by Iberian signs - Horseman holding spear riding right. (Vives: 51-7; GMI: 352). 2.98 g; 19.7 mm; 11 h	*Turiasu* (c.100-72 BC)
138 (Box 3) 1991/6/2/16	Ag: 96.2% Cu: 3.03% Pb: 0.5%	Iberian denarius. Manly head right, framed by Iberian signs - Horseman holding spear riding right. (Vives: 51-7; GMI: 352). 2.8 g; 19.2 mm; 12 h	*Turiasu* (c.100-72 BC)
139 (Box 3) 2275	Ag: 96.3% Cu: 2.1% Pb: 0.3%	Iberian denarius. Manly head right, framed by Iberian signs - Horseman holding spear riding right. (You live: 51-7; GMI: 352). 2.2 g; 17.2 mm; 12 h	*Turiasu* (c.100-72 BC)
145 (Box 3) 2127	Cu: 89.0% Pb: 5.1% Sn: 4.7% Zn: 0.6% Fe: 0.3% Ni: 0.2%	As - Imperial – Laureate head of Augustus right, overstruck with eagle head. AVGVSTS (*divi f.*) PATER PATRIAE MVN AVGVSTA BILBILIS L COR CALDO M SEMP RVTILO II VIR. Laurea (Vives: 139-4; GMI: 456; RPC: 395). 13 g, 29.3 mm; 9 h; 6.02 g·cm^{-3}	*Bilbilis* (16-6 BC)
203 (Box 3) 2105	Cu: 96.3% Pb: 0.95% Sn: 1.5% Zn: 0.7% Fe: 0.3%	As - Imperial – Laureate head of Augustus right. Illegible. C V I CEL CN DO (mit c) POMPEI II V(ir). Bull standing right. Overstruck on reverse: R within a circle (Vives: 161-8; GMI: 444; RPC: 278). 12.65 g; 29.1 mm; 3 h; 6.43 g·cm^{-3}	*Celsa* (16-6 BC)
200 (Box 3) 2160	Cu: 91.7% Pb: 3.5% Sn: 3.5% Zn: 0.7% Fe: 0.2%	As - Imperial - Laureate head of Augustus right. AVGVSTVS DIVI F C V I CEL L BAGGIO MAN FESTO II VIR Bull standing right. (Vives: 161-2; GMI: 440; RPC: 273) 11.4 g; 26.2 mm; 6 h; 6.65 g·cm^{-3}	*Celsa* (16-6 BC)
201 (Box 3) 2264	Cu: 91.8% Pb: 3.4% Sn: 3.6% Zn: 0.8% Fe: 0.2%	As - Imperial - Head of Augustus right. Illegible. Bull standing right. (Vives: 161-2; GMI: 440; RPC: 273) 9.17 g; 29 mm; 12 h	*Celsa* (16-6 BC)
159 (Box 3)	Cu: 96.8% Pb: 0.7%	As - Imperial - Head of Augustus right. MVN CAL IMP AVGVS	*Calagurris* (AD 10-12)

2087	Sn: 1.3% Zn: 0.6% Fe: 0.2%	L BAEBIO P ANTESTIO II/VIR Bull standing right. (Vives: 158-3; RCP: 439) 14.2 g; 28.7 mm; 3 h; 6.73 $g\cdot cm^{-3}$	
163 (Box 3) 2235	Cu: 88.7% Pb: 4.9% Sn: 3.0% Zn: 0.9% Faith: 1.7%	Semis - Imperial - Laureate head of Augustus right. MVN CAL AVGVS(*tus*) L PRISCO C BROCCHIO II VIR Bull's head facing front (Vives: 158-12; GMI: 695; RPC: 442). 5.1 g; 20.6 mm; 7 h; 8.48 $g\cdot cm^{-3}$	*Calagurris* (AD 10-12)
170 (Box 4) 2331	Cu: 92.0% Pb: 2.6% Sn: 3.8% Zn: 0.9% Fe: 0.2%	As - Imperial - Laureate head of Augustus right. (*m cal*) L VALENTINO L NOVO II VIR Standing bull. Overstruck on reverse: B (Vives: 159-2; GMI: 690; RPC: 445) 11 g; 29 mm; 12 h; 5.42 $g\cdot cm^{-3}$	*Calagurris* (AD 10-12)
158 (Box 3) 1988/1/47	Cu: 87.3% Pb: 5.5% Sn: 5.2% Zn: 1.0% Fe: 0.3%	Ace - Imperial - Head of Augustus right. MVN CAL IIVIR (*q aem c post mil*) AEMMRS L IVNIS Bull standing right. (Vives: 157-7; GMI: 685; RPC: 436) 11.1 g; 28.5 mm; 6 h	*Calagurris* (AD 10-12)
160 (Box 3) 2068	Cu: 88.9% Pb: 4.2% Sn: 3.0% Zn: 0.9% Fe: 1.7%	As - Imperial - Laureate head of Augustus right. MVN CAL IVL AVGVSTVS L BAEB PRISCO C CRAN BROC II VIR Bull standing right. (Vives: 158-9; GMI: 688; RPC: 441) 12.7 g; 27.2 mm; 12 h; 8.75 $g\cdot cm^{-3}$	*Calagurris* (AD 10-12)
174 (Box 4) 1988/1/50	Cu: 86.3% Pb: 7.6% Sn: 4.1% Zn: 0.6% Fe: 0.9%	As - Imperial - Head of Tiberius unrecognizable (*ti augustus*) DIVI AVG(*usti f imp caesar*) M C I L FVL SPARSO L SATVRNINO II VIR Bull standing right. (Vives: 159-5; GMI: 692; RPC: 448) 12.9 g; 28.2 mm; 2 h	*Calagurris* (AD 15-16)
175 (Box 3) 1988/1/49	Cu: 87.9% Pb: 6.5% Sn: 4.0% Zn: 0.6% Fe: 0.8%	As - Imperial - Laureate head of Tiberius right. (*ti august*)VS DIVI F AVGVSTI F I(*mp caesar*). Eagle head overstruck on obverse. M C I L FVL SPARSO L SATVRNINO II VIR Bull standing right. (Vives: 159-5; GMI: 692; RPC: 448) 11.7 g; 26.4 mm; 9 h	*Calagurris* (AD 15-16)
181 (Box 3) 2193	Cu: 95.0% Pb: 1.4% Sn: 2.1% Zn: 0.7% Fe: 0.3%	As - Imperial - Laureate head of Tiberius right. TI CAESAR DIVI AVG (*f aug*)VSTVS. Eagle head overstruck M C (i) C CELERE C RECTO II VIR Bull standing right. (Vives: 159-6; GMI: 694; RPC: 450) 12.7 g; 27.3 mm; 6 h; 6.34 $g\cdot cm^{-3}$	*Calagurris* (AD 15-16)
152 (Box 3) 2022	Cu: 95.6% Pb: 0.1% Sn: 3.1% Zn: 0.6% Fe: 0.4%	As - Imperial - Head of Agrippa wearing rostral crown left. M AGGRIPPA L F COS III TITVLO ET MONTANO II VIR Foundational yoke right. Overstruck: CCA (Vives: 153-2) 11.1 g; 25.8 mm; 9 h	*Caesar Augusta* (AD 15-16)
150 (Box 3) 1988/1/45	Cu: 96.2% Pb: 0.1% Sn: 2.8% Zn: 0.4% Fe: 0.3%	As - Imperial - Laureate head of Tiberius right. (*ti caesar divi aug f augustus*) CCA T CAECILIO LEPIDO C AVFIDIO GEMELLO II VIR. Bull looking right. (Vives: 152-4; RCP: 367) 12.5 g; 27.2 mm; 8 h	*Caesar Augusta* (AD 22/3-37)

192 (Box 4) 1988/1/53	Cu: 90.8% Pb: 3.8% Sn: 3.9% Zn: 0.7% Fe: 0.7%	As - Imperial – Laureate head of Tiberius right. (*ti caesar*) DIVI AVG F AVGVSTVS. MVNICIP CASCANTVM Bull standing right. Overstruck eagle's head on obverse. (Vives: 161-2; RPC: 427) 7.26 g; 25.5 mm; 9 h	*Cascantum* (AD 22/3-37)
194 (Box 3) 2050	Cu: 91.2% Pb: 3.1% Sn: 3.5% Zn: 0.6% Fe: 0.3%	As - Imperial – Laureate head of Tiberius right. (*ti cae*)SAR DIVI AVG F AVGVS(*tus*) MVNICIP CASCA(*antum*) Bull standing right. Overstruck eagle's head on obverse. (Vives: 161-2; RPC: 427) 6.48 g; 29 mm; 6 h; 9 $g \cdot cm^{-3}$	*Cascantum* (AD 22/3-37)
228 (Box 3) 2341	Cu: 93.6% Pb: 2.1% Sn: 2.0% Zn: 1.0% Fe: 0.6%	As - Imperial - Laureate head of Tiberius right. (*ti cae*)SAR (*divi*) AVG F A(*ugustus imp*) MVNICIP GRACVRRIS. Bull standing right. (Vives: 163-1; GMI: 1062; RPC: 429) 9 g; 27.2 mm; 6 h; 5.68 $g \cdot cm^{-3}$	*Graccurris* (AD 22/3-37)

Vives = Vives y Escudero (1926); GMI = Guadán (1980); RPC = Roman Provincial Coinage (Burnett, et al., 1998)
[*] Trace elements are not shown.

3.2. *Probable provenances for the coins under study*

The provenance of the metals used for coinage in the various mints was assigned using as criterion the lead isotopic ratios of representative mineral sources reported in the literature (Table 2), including Pb–Zn, Pb–Zn–(Ag) and Pb-Zn-Cu-Ag deposits, and copper ores. According to these data, and as shown in Figure 2, Figure 3 and Table 3, it is highly probable that the denarii from *Turiaso* (in which Pb is inherited from smelting) were minted with argentiferous galena from the Iberian Ranges, IBR.

Regarding the lead- and tin-rich copper coins from the Roman period minted in the *Conventus Caesaraugustanus*, clear temporal differences were observed. According to the cluster analysis, in the early Augustan period (16-6 BC), the raw materials would have originated from mineral sources in the Pyrenees and in the Iberian Range, which were the closest mining areas, whereas in the late Augustan period (AD 10-12) *Calagurris* mint kept using mineral sources with a Pyrenean origin, but also used mineral sources from the SE of the Iberian Peninsula (*Hispania Citerior*). These two sources were again used in the early Tiberian period (*Calagurris* used Pyrenean mineral sources and *Caesar Augusta* used Cartagena-Mazarrón ores), while in the late coinage of this period (*Cascantvm* and *Gracurris* mints) ores from the Central Iberian Zone (Los Pedroches/Linares-La Carolina/Alcudia-Almadén) were used together with the local resources from the Pyrenees (instead of ores from the SE of the Iberian Peninsula).

Table 2. Data on isotopic ratios in lead ores of the South Central Pyrenees (Huesca province), Central Iberian Zone (Córdoba, Ciudad Real and Jaén provinces), Southern Iberian Peninsula (Murcia province), and Iberian Range mineral sources (Teruel and Zaragoza provinces). Values between parentheses indicate uncertainty (2σ).

Mineral source	**Pb^{206}/Pb^{204}**	**Pb^{207}/Pb^{204}**	**Pb^{208}/Pb^{204}**	**Pb^{207}/Pb^{206}**	**Pb^{208}/Pb^{206}**	**Ref.**
Benasque, Eriste	18.4680 (0.0108)	15.7099 (0.0117)	38.5836 (0.0339)	0.8507 (0.0002)	2.0892 (0.0008)	(Martín Ramos, et al., 2019)
Bielsa-Parzán, *Ana*	18.4509 (0.0216)	15.6815 (0.0197)	38.6815 (0.0546)	0.8499 (0.0003)	2.0965 (0.0011)	
Bielsa-Parzán, *P4-7*	18.488	15.753	38.563	0.852	2.086	(Subías, et al., 2015)
Benasque *1*	18.087 (0.0007)	15.690 (0.0006)	38.299 (0.0017)	0.867	2.117	(García-Sansegundo, et al., 2014)
Benasque *2*	18.094 (0.0009)	15.692 (0.0009)	38.321 (0.0023)	0.867	2.118	
CIZ, Los Pedroches	18.2468 (0.00030)	15.6135 (0.00033)	38.3609 (0.00096)	0.8557 (0.00001)	2.1024 (0.00003)	(Klein, et al., 2009)
CIZ, Linares-La Carolina	18.2419 (0.00035)	15.6125 (0.00034)	38.3470 (0.00099)	0.8559 (0.00001)	2.1018 (0.00002)	
CIZ, Alcudia-Almadén	18.1855 (0.00056)	15.5238 (0.00060)	38.3052 (0.00144)	0.8591 (0.00001)	2.1063 (0.00002)	
SE, SVP, Cartagena-Maz.	18.7586 (0.00046)	15.6715 (0.00034)	39.0820 (0.00059)	0.8354 (0.00001)	2.0834 (0.00000)	
IBR, Bádenas	18.4780	15.7690	38.9910	0.8534	2.1101	(Subías, et al., 2010, Subias Pérez, et al., 1994)
IBR, Cuca-Alta	18.3220	15.6990	38.6810	0.8568	2.1112	
IBR, Calcena, *Valdeplata*	18.4140	15.7570	38.908	0.8557	2.1130	
IBR, Nogueras	18.2770	15.7000	38.757	0.8590	2.1205	

CIZ = Central Iberian Zone; SE = Southeast; SVP = Southern Volcanic Zone; IBR = Iberian Range

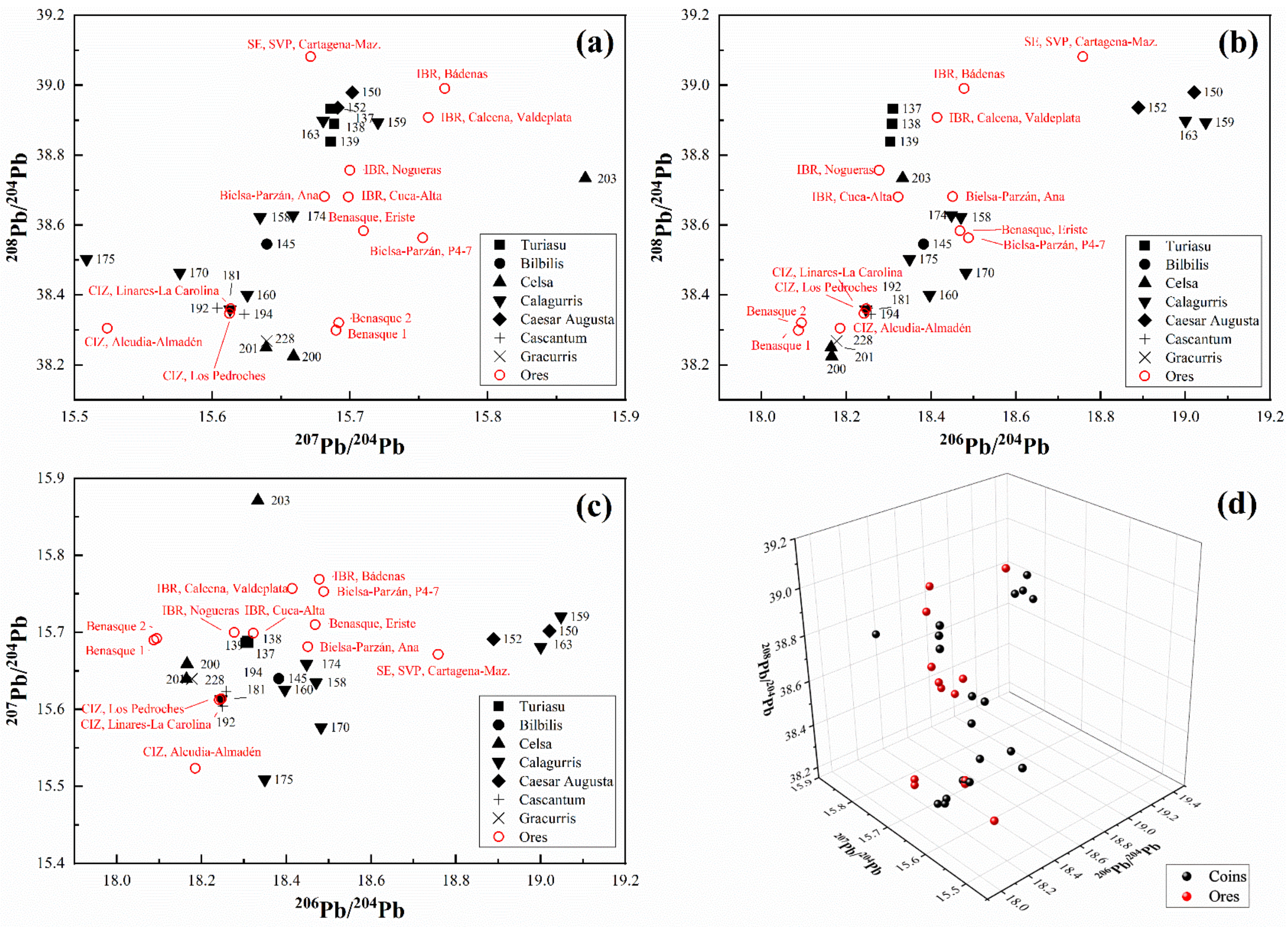

Figure 2. (**a-c**) Binary projections and (**d**) ternary plot, using ^{204}Pb normalization, of Pb isotope compositions of the coins under study and ores reported in the literature.

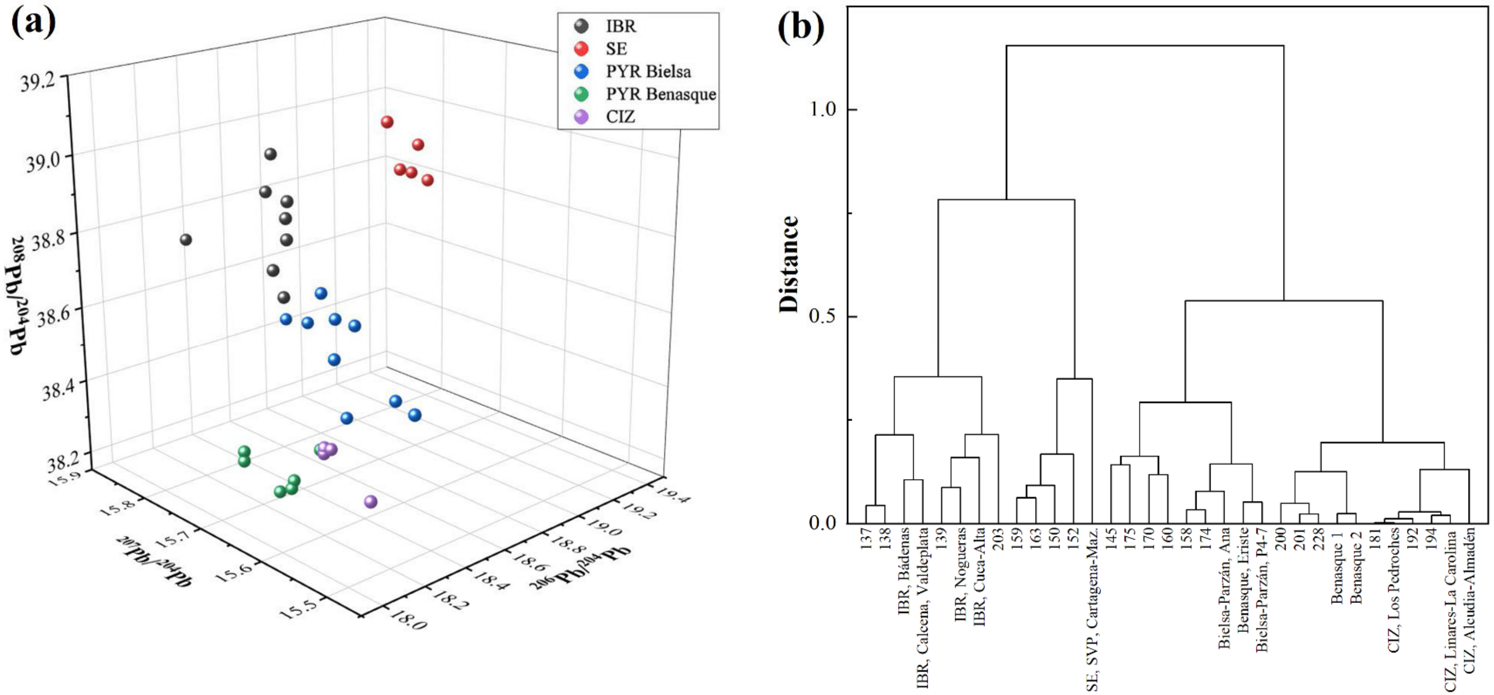

Figure 3. Cluster analysis applied to Pb isotope compositions of the coins under study and ores reported in the literature: (**a**) 3D plot, (**b**) hierarchical cluster analysis dendrogram.

Table 3. Suggested origin of the raw material used in the minting of the coins (organized by period), according to cluster analysis results, using as a criterion the lead isotopic ratios.

Code	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{206}Pb$	Mint	Period	Suggested origin
137	18.3101	15.6862	38.9328	0.8567	2.1263	*Turiasu*	Iberian	Iberian Range
138	18.3087	15.6887	38.8895	0.8569	2.1241	*Turiasu*	Iberian	Iberian Range
139	18.3035	15.6861	38.8382	0.8570	2.1219	*Turiasu*	Iberian	Iberian Range
145	18.3823	15.6397	38.5458	0.8508	2.0969	*Bilbilis*	Early AVG	Pyrenees, Bielsa-Parzán
203	18.3330	15.8709	38.7339	0.8657	2.1128	*Celsa*	Early AVG	Iberian Range / Unknown source*
200	18.1660	15.6591	38.2249	0.8620	2.1042	*Celsa*	Early AVG	Pyrenees, Benasque
201	18.1641	15.6393	38.2500	0.8610	2.1058	*Celsa*	Early AVG	Pyrenees, Benasque
159	19.0480	15.7203	38.8941	0.8253	2.0419	*Calagurris*	Late AVG	SE, SVP, Cartagena-Mazarrón
163	19.0001	15.6807	38.8987	0.8253	2.0473	*Calagurris*	Late AVG	SE, SVP, Cartagena-Mazarrón
170	18.4821	15.5766	38.4631	0.8428	2.0811	*Calagurris*	Late AVG	Pyrenees, Bielsa-Parzán
158	18.4701	15.6349	38.6226	0.8465	2.0911	*Calagurris*	Late AVG	Pyrenees, Bielsa-Parzán
160	18.3962	15.6256	38.3998	0.8494	2.0874	*Calagurris*	Late AVG	Pyrenees, Bielsa-Parzán
174	18.4480	15.6587	38.6283	0.8488	2.0939	*Calagurris*	Early TI	Pyrenees, Bielsa-Parzán
175	18.3493	15.5088	38.5023	0.8452	2.0983	*Calagurris*	Early TI	Pyrenees, Bielsa-Parzán
152	18.8890	15.6911	38.9359	0.8307	2.0613	*Caesar Augusta*	Early TI	SE, SVP, Cartagena-Mazarrón
150	19.0210	15.7018	38.9797	0.8255	2.0493	*Caesar Augusta*	Early TI	SE, SVP, Cartagena-Mazarrón
181	18.2461	15.6131	38.3588	0.8557	2.1023	*Calagurris*	Late TI	CIZ
192	18.2499	15.6037	38.3631	0.8550	2.1021	*Cascantum*	Late TI	CIZ
194	18.2580	15.6234	38.3454	0.8557	2.1002	*Cascantum*	Late TI	Pyrenees, Benasque
228	18.1770	15.6395	38.2680	0.8604	2.1053	*Graccurris*	Late TI	Pyrenees, Benasque

AVG = Augustus, TI = Tiberius, CIZ = Central Iberian Zone, SE = Southeast, SVP = Southern Volcanic Zone.

* The assignment of coin 203 to mineral sources from the Iberian Ranges is dubious, and it could likely be from a source not reported in Table 2.

3.3. *Results according to chronology*

In accordance with Klein, et al. (2004) hypotheses, the primitive Augustan asses (16-6 BC), such as coins 145, 203, 200 and 201, exhibited $^{208}Pb/^{206}Pb$ ratio values > 2.095, while the late ones (10-12 BC), such as coins 170, 158, and 160, presented values of $^{208}Pb/^{206}Pb$ of around 2.09 (Table 3). As for the Tiberius asses, and also in agreement with Klein, et al. (2004), the oldest samples (15-16 BC) were characterized by $^{207}Pb/^{206}Pb$ ratios < 0.85 and $^{208}Pb/^{206}Pb$ < 2.095, while the ones that were coined later on (22/3-37 BC) presented $^{207}Pb/^{206}Pb$ > 0.85 and $^{208}Pb/^{206}Pb$ > 2.095.

4. Discussion

4.1. *Silver coins*

Regarding the three denarii from *Turiasu*, the suggested provenance from the Iberian Ranges supports the hypothesis of Rovira Lloréns, et al. (2012). It is worth noting that this was the nearest raw material source (for instance, *Valdeplata* mines are only 26 km far from *Turiaso* as the crow flies). According to Sanz Pérez (2003), the access to Valdeplata metallogenic resources would explain why the *ka-s-tu* type denarii from *Turiaso* reached much higher numbers in their issues than the rest of the neighboring Celtiberian mints.

4.2. *Bronze coins*

It is worth noting that lead isotope analysis technique does not provide absolute assurance in the correlations because, in some cases, it is affected by the mixing of ores or metals from various origins and from the melting and re-melting of pieces from diverse origins. Further, lead is a ubiquitous component of copper-based coins that can be both of natural and anthropogenic origin (i.e., it can be a residue of the ore(s) or can be deliberately added as a cheap 'filler'). Given that universally valid threshold values are virtually impossible to define due to the natural variation of Pb contents in ores (Westner, et al., 2020), the interpretation of Pb isotope data presented above to assess the possible provenance of the copper raw material in bronze coins should be taken with caution.

In the assignments made in this study, an unequivocal match between the sources presented in Table 2 and the objects was only attained for the group formed by samples 192, 194 and 181, that clearly matched CIZ.

In the case of coins 150, 152, 159 and 163, tentatively associated with SE Iberian Peninsula sources, a good overlap was not observed. Alternative raw material sources may thus be considered: for instance, the signature of Kalavasos (Cyprus) lead isotope data, with $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ ratios in the 0.82-0.83 and 2.04-2.06 range, respectively, could be a better match (Stos-Gale, et al., 1997). Given that during the late Augustan period and continuing with Tiberius, copper for coinage from Cyprus has been documented (Klein, et al., 2004), this hypothesis may be plausible.

For samples 200, 201 and 228, for which the cluster analysis suggested Benasque (Pyrenees) as the most probable origin, their distribution on $^{207}Pb/^{204}Pb$ vs $^{206}Pb/^{204}Pb$ is somewhat compatible with a mixing pattern between Benasque and CIZ, but $^{208}Pb/^{204}Pb$ values are not, with systematically lower values in the samples.

Samples 145, 158, 160 and 174, that are attributed to the source of Bielsa-Parzán in the Pyrenees, can be an intermediate mixture of Pyrenean and CIZ deposits, as they lie in the middle of a mixing line between these two sources. Sample 175 has lower $^{207}Pb/^{204}Pb$ than any source presented here, and could be a possible singleton. Sample 170 does not show any good compatibility with any source either. This does not mean that the hypothesis that mixings occurred should be ruled out, but no clear pattern at two end-members can be identified at this point.

Even if there is some degree of speculation in the assignments obtained from the cluster analysis (due not only to mixing, but also to large variations in signature within the same deposit, differences in the $^{208}Pb/^{204}Pb$ may result from the equipment used −depending on whether it is a Q-ICP-MS or a MC-ICP-MS (Montero Ruiz, 2018)−, etc.), the results suggest that the mints in

Conventus Caesaraugustanus made use of nearby ore deposits, either alone or mixed with ores from the southern Iberian Peninsula (Hunt Ortiz, 2007), which is a reasonable assumption. These results would be consistent with those obtained for 38 bronze coins selected by Resano, et al. (2010) to cover the period II BC to 54 AD from *Bilbilis* mint. Although in that work only ratios over ^{206}Pb were reported (which were used in archaeometry in early days of Pb isotopes), the origin of the raw material for many of the 'group I' *Bilbilis* coins may also be tentatively assigned to Pyrenean ores (Figure S2).

From the analyses of 241 well dated coins from the official mint in Rome found in the River Tiber, Klein *et al*. (Klein, et al., 2004, Klein and von Kaenel, 2000) reached the conclusion that, during the Augustan period, the main copper supply came from Sardinia and southern Spain, and that late Tiberian asses came from southwestern Spain. The results presented herein would partly match those by Klein: late Augustan and early Tiberian asses would have used mineral that came from southeastern Spain, but not for the late Tiberian asses, which in our case apparently came from the Central Iberian Zone (Los Pedroches-Linares-La Carolina/Alcudia-Almadén).

5. Conclusions

Provincial coin-issues, which had a civic and local purpose, coexisted with the imperial issues and would have been necessary to ensure sufficient monetary flow to guarantee transactions with the rest of the Empire. In this case of study, the composition and provenance of 3 Iberian denarii and 17 bronze coins from the *civitates* of the middle Ebro River (Hispania) have been investigated. A high homogeneity in the composition of the Celtiberian silver coins from *Turiasu* was observed using XRF analyses, with high silver contents (96%) comparable to the purity of coetaneous republican denarii. With regard to the six mints of the *Conventus Caesaraugustanus*, noticeable differences in lead contents were found among Augustus bronzes issued in his early period depending on the mint, while greater uniformity was found in subsequent coin emissions. Differences in elemental composition from one mint to another were also found in the early Tiberian period. In relation to the ore source provenance, MC-ICP-MS results showed that Celtiberian denarii used *argentum* from the Iberian Range mines. As for the Augustan and Tiberian periods coinage, the raw material for 11 of the 17 bronze coins came −at least in part− from nearby ore deposits (in the Pyrenees and the Iberian Range), while for the rest a southeastern Iberian Peninsula/Cyprus (late Augustan and early Tiberian periods) or Central Iberian Zone (late Tiberian period) provenance may be suggested. The differences detected with the official mint of Rome suggest that the mints of the *Conventus Caesaraugustanus* would have enjoyed a certain degree of autonomy in both the pattern of ore processing and the choice of ore supply, and that they generally opted for the nearest ores deposits. Further studies on a larger data set are needed to confirm this hypothesis.

Acknowledgements

The financial support of Instituto de Estudios Altoaragoneses (Diputación de Huesca) is gratefully acknowledged.

Declaration of interest

The authors declare no conflict of interest.

References

Rehren, T.H., Pernicka, E., 2008. Coins, artefacts and isotopes—Archaeometallurgy and archaeometry, Archaeometry 50, 232-248.

Chiarantini, L., Benvenuti, M., Costagliola, P., Dini, A., Firmati, M., Guideri, S., Villa, I.M., Corretti, A., 2018. Copper metallurgy in ancient Etruria (southern Tuscany, Italy) at the Bronze-Iron Age transition: a lead isotope provenance study, Journal of Archaeological Science: Reports 19, 11-23.

Soares, A.M.M., Valério, P., Gomes, S.S., Mataloto, R., Soares, S.M., Silva, R.J.C., Soares, R.M., 2020. A first appraisal on copper sources for Chalcolithic settlements in southern Portugal using Pb isotope analysis, Journal of Archaeological Science: Reports 33.

Gozalbes Fernández de Palencia, M., 2009. La ceca de Turiazu: monedas celtibéricas en la Hispania republicana, Diputación de Valencia, Valencia, Spain.

Stos-Gale, Z., Gale, N.H., Houghton, J., Speakman, R., 1995. Lead isotope data from the Isotrace Laboratory, Oxford: Archaeometry data base 1, ores from the Western Mediterranean, Archaeometry 37, 407-415.

Stos-Gale, Z.A., Gale, N.H., 2009. Metal provenancing using isotopes and the Oxford archaeological lead isotope database (OXALID), Archaeological and Anthropological Sciences 1, 195-213.

Baker, J., Stos, S., Waight, T., 2006. Lead isotope analysis of archaeological metals by Multiple-Collector Inductively Coupled Plasma Mass Spectrometry, Archaeometry 48, 45-56.

Holmqvist, E., Wessman, A., Mänttäri, I., Lahaye, Y., 2019. Lead isotope and geochemical analyses of copper-based metal artefacts from the Iron Age water burial in Levänluhta, Western Finland, Journal of Archaeological Science: Reports 26.

Fanlo, I., Touray, J.C., Subías, I., Fernández-Nieto, C., 1998. Geochemical patterns of a sheared fluorite vein, Parzan, Spanish Central Pyrenees, Mineralium Deposita 33, 620-632.

Subías, I., Fanlo, I., Billström, K., 2015. Ore-forming timing of polymetallic-fluorite low temperature veins from Central Pyrenees: A Pb, Nd and Sr isotope perspective, Ore Geology Reviews 70, 241-251.

Subías, I., Fanlo, I., Mateo, E., Billström, K., Recio, C., 2010. Isotopic studies of Pb–Zn–(Ag) and barite Alpine vein deposits in the Iberian Range (NE Spain), Geochemistry 70, 149-158.

Subías, I., Recio, C., Fanlo, I., Fernández-Nieto, C., 1997. Stable isotope composition of F-Pb-Zn mineralization in the Valle de Tena (Spanish Central Pyrenees), Mineralium Deposita 32, 180-188.

García-Sansegundo, J., Martin-Izard, A., Gavaldà, J., 2014. Structural control and geological significance of the Zn–Pb ores formed in the Benasque Pass area (Central Pyrenees) during the post-late Ordovician extensional event of the Gondwana margin, Ore Geology Reviews 56, 516-527.

Klein, S., Domergue, C., Lahaye, Y., Brey, G., Von Kaenel, H., 2009. The lead and copper isotopic composition of copper ores from the Sierra Morena (Spain), Journal of Iberian Geology 35, 59-68.

Klein, S., Lahaye, Y., Brey, G.P., von Kaenel, H.M., 2004. The early roman imperial AES coinage II: Tracing the copper sources by analysis of lead and copper isotopes-copper coins of Augustus and Tiberius, Archaeometry 46, 469-480.

Montero Ruiz, I., Santos Retolaza, M., Castanyer, P., Hunt Ortiz, M., Pons, E., Rovira Hortala, M., Rovira Llorens, S., 2008. Estudio de procedencia de metal en monedas prerromanas, V Simposio Internacional sobre Minería y Metalurgia Históricas en el Suroeste Europeo, Universidad de Léon, León, pp. 315-328.

Montero-Ruiz, I., Pérez Almoguera, A., Rafel Fontanals, N., 2011. Sobre la procedencia de los metales de las primeras monedas del NE ibérico. Aplicación de análisis de isótopos de plomo, in: García Bellido, M.P., Callegarin, L., Jiménez, A. (Eds.), Barter, Money and coinage in the ancient mediterranean (10th-1st Century BC), Consejo Superior de Investigaciones Científicas, Madrid, Spain, pp. 203-212.

Huelga-Suarez, G., Moldovan, M., Suárez Fernández, M., de Blas Cortina, M.Á., García Alonso, J.I., 2014. Isotopic composition of lead in copper ores and a copper artefact from the La Profunda mine (León, Spain), Archaeometry 56, 651-664.

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.

Gozalbes Fernández de Palencia, M., 2003. Desarrollo y contexto de las emisiones de "Turiazu", Kalathos: Revista del seminario de arqueología y etnología turolense, 251-270.

Vives y Escudero, A., 1926. La moneda hispánica, Talleres tipográficos de la "Editorial Reus", Madrid, Spain.
Guadán, A.M.d., 1980. La moneda ibérica: Catálogo de numismática ibérica e ibero-romana, Cuadernos de Numismática, Madrid, Spain.
Burnett, A., Amandry, M., Ripollés Alegre, P.P., Butcher, M.S., 1998. Roman provincial coinage, British Museum Press; Bibliothèque Nationale de France, London; Paris.
Sanz Pérez, E., 2003. La minería de plata en la Celtiberia: Una aproximación, Cuadernos de estudios borjanos, 15-50.
Rovira Lloréns, S., Burillo Mozota, F., López, R., Ibáñez, J., Domergue, C., 2012. Metalurgia y explotación de recursos minerales en el entorno de la ciudad-estado celtibérica de Segeda I (Mara, Zaragoza), Minería y metalurgia antiguas: visiones y revisiones: homenaje a Claude Domergue, Madrid : Casa de Velázquez, 2012, pp. 27-42.
Martín Ramos, P., Moldovan, M., Martín Gil, J., Oliván, M., Cuchí Oterino, J.A., 2019. Examen de muestras minerales de las minas de Bielsa y Eriste, Lucas Mallada 21.
Subias Pérez, I., López Ciriano, A., Fanlo González, I., Fernández Nieto, C., 1994. La mineralización de Pb-An-Cu-Ag de Valdeplata (Calcena, Zaragoza), Boletín de la Sociedad Española de Mineralogía 17, 95-102.
Westner, K.J., Kemmers, F., Klein, S., 2020. A novel combined approach for compositional and Pb isotope data of (leaded) copper-based alloys: bronze coinage in Magna Graecia and Rome (5th to 2nd centuries BCE), Journal of Archaeological Science 121.
Stos-Gale, Z.A., Maliotis, G., Gale, N.H., Annetts, N., 1997. Lead Isotope Characteristics of the Cyprus Copper Ore Deposits Applied to Provenance Studies of Copper Oxhide Ingots, Archaeometry 39, 83-123.
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.
Hunt Ortiz, M.A., 2007. Análisis de isótopos de plomo aplicados a la investigación de la producción y distribución metálica romana hispánica, Sautuola: Revista del Instituto de Prehistoria y Arqueología Sautuola 13, 45-58.
Resano, M., Marzo, M.P., Alloza, R., Saénz, C., Vanhaecke, F., Yang, L., Willie, S., Sturgeon, R.E., 2010. Laser ablation single-collector inductively coupled plasma mass spectrometry for lead isotopic analysis to investigate evolution of the Bilbilis mint, Anal. Chim. Acta 677, 55-63.
Klein, S., von Kaenel, H., 2000. The early Roman Imperial aes coinage I: Metal analysis and numismatic studies, Schweizerische Numismatische Rundschau 79, 53-106.