

## Semi-refined silver for the silversmiths of the Iron Age Mediterranean: A mechanism for the elusiveness of Iberian silver

*Plata semirrefinada para los plateros de la Edad del Hierro en el Mediterráneo: un mecanismo para identificar la plata ibérica*

Jonathan R. Wood<sup>a</sup> and Ignacio Montero-Ruiz<sup>b</sup>

### ABSTRACT

A fragment of a silver ingot recovered from the Phoenician settlement of La Rebanadilla, near Malaga, in south-east Iberia has been investigated using lead isotope and compositional analyses. The ingot, which was found at the lowest levels of the site, potentially dates from 11<sup>th</sup>-9<sup>th</sup> century BC, placing it alongside the hoards of hacksilver found in the southern Levant in terms of chronology. The Pb crustal age (from lead isotope data) and compositional data support that the ingot derives from Hercynian-age ores with high bismuth concentrations. This signature is consistent with the Pyritic belt of south-west Iberia, particularly around the ancient mining areas of Riotinto. It is proposed that the silver for this ingot was extracted from jarosite ores at Riotinto, where it was coarsely refined through cupellation into an ingot still retaining high levels of lead, before being transported to La Rebanadilla, which was a potential point of departure back to the Phoenician homeland. The significance of transporting silver in a form which would have required further refining is discussed in relation to the movement of silver by the Phoenicians in the Iron Age Mediterranean. A new mechanism is proposed to explain the elusive nature of Iberian silver in the archaeological record.

### RESUMEN

*Un fragmento de lingote de plata descubierto recientemente en el yacimiento fenicio de La Rebanadilla (Málaga) ha sido investigado mediante isótopos de plomo y análisis elemental. El lingote recuperado en los niveles inferiores del yacimiento, se fecha potencialmente entre fines del siglo XI y el IX a. C., situándose en cronología similar a algunos depósitos de hacksilver del área del levante mediterráneo. La edad de la corteza calculada a partir de los isótopos de plomo y la composición señalan que el lingote fue obtenido*

*de minerales de Edad Hercínica con concentraciones altas de bismuto. Esta signatura es compatible con la de la Faja Pirítica del suroeste de la península ibérica, en particular con la de las antiguas minas de la zona de Riotinto. Se propone que la plata de este lingote fue obtenida de las jarositas argentíferas de Riotinto, donde sufrió solo un primer refinado mediante copelación, conservando un alto contenido en plomo antes de ser comercializado hacia La Rebanadilla, que pudo ser un lugar potencial para su transporte hacia los territorios fenicios en el Mediterráneo oriental. Las implicaciones del transporte de plata sin refinar son discutidas en relación al comercio de la plata por los fenicios durante la Edad del Hierro en el Mediterráneo y la dificultad de identificar la plata ibérica en el registro arqueológico.*

**Key words:** Iberia; Phoenician; Ingot; La Rebanadilla; Composition; Lead isotopes.

**Palabras clave:** Península ibérica; Fenicios; Lingote; La Rebanadilla; Análisis de composición; Isótopos de plomo.

### BACKGROUND

The amount of ancient silver production slag at Riotinto is in the region of 6-15 million tonnes (Rothenberg and Blanco-Freijeiro 1981; Domergue 1990; Anguilano 2012) with Salkield calculating about 9 million tonnes (Salkield 1987: 13-14). Williams states that Roman and 'Ancient' miners could have exploited about 2 million tonnes of silver-ore (Williams 1950). Other authors propose up to 3 million tonnes, e.g. Dutrizac *et al.* (1985: 28). Assuming that ancient silver-ore had approximately the same amount of silver

<sup>a</sup> UCL Institute of Archaeology, 31-34 Gordon Square, Kings Cross, London WC1H 0PY, United Kingdom. E-mail: [uczljrw@ucl.ac.uk](mailto:uczljrw@ucl.ac.uk)  
<https://orcid.org/0000-0001-6630-6916>

<sup>b</sup> Instituto de Historia-CSIC. C/ Albasanz 26-28. 28037 Madrid, Spain. E-mail: [ignacio.montero@cchs.csic.es](mailto:ignacio.montero@cchs.csic.es) <https://orcid.org/0000-0003-0897-1031>  
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as modern ores, *i.e.* 0.2 % silver (range: 0.016-0.68 % Salkield 1987: 14), this suggests that 4000-6000 tonnes (*i.e.* 4-6 million kilograms) of silver metal could have been produced at Riotinto alone.

Even though this is a very approximate calculation, and accepting that Roman exploitation of silver in south-west Iberia was considerable, what it demonstrates is that potentially a lot of silver was extracted in the Orientalising period (c. 8<sup>th</sup>-6<sup>th</sup> centuries BC) or earlier, as evidenced by the Phoenician slag levels at mining sites such as Corta Lago (Rothenberg and Blanco-Freijeiro 1981; Anguilano 2012). With only a few tens of silver objects recovered in Iberia (Murillo-Barroso *et al.* 2016) from this time, this suggests that most of the silver moved East. In fact, the circulation of “[silver], which had provided a standard of exchange (and, in a growing number of circumstances, a medium of exchange) in the Near East since at least the end of the 3<sup>rd</sup> millennium” (Sherratt 2016: 297) potentially resulted in a growing network of commercial traders and routes. Essentially, a net increase in silver circulation, as silver from Iberia fed into the system, would have potentially facilitated entrepreneurial trade. This would have made the middle of the 1<sup>st</sup> millennium BC in the eastern Mediterranean and the Near East look very different to it how had been less than a millennium earlier (Sherratt 2016). Understanding the sources and movement of silver is therefore prerequisite in order to appreciate the subsequent movements and interactions of the 1<sup>st</sup> millennium BC Mediterranean. The problems in addressing this task, however, are put into perspective when it is appreciated that silver with Iberian isotopic and compositional signatures is very difficult to locate in the archaeological record of the Iron Age Mediterranean.

## LA REBANADILLA

At present, the site of La Rebanadilla in south-east Iberia (Fig. 1), where a fragment of a silver ingot was recovered, is known through a brief preliminary report, which describes the main phases of occupation and the finds (Arancibia Román *et al.* 2011: 130-132; Sánchez *et al.* 2011; Pappa 2012: 36-37). It has been described as a Phoenician settlement (Sánchez-Moreno *et al.* 2012: 67-85). In the Phoenician period, the settlement would have been located on an islet in the Guadalhorce estuary—prior to its sedimentary infilling. Four phases of occupation have been identified with only the earliest phase (IV) yielding evidence for metallurgical activities. With the discovery of kilns and tuyères, the last phase (I) appears commensurate with a transformation into an area of industrial activities, perhaps as



Fig. 1. The Iberian Peninsula and the north-western African coastline showing the sites of La Rebanadilla, Huelva, Las Arenillas and La Fonteta.

a satellite production centre of the large nearby colony of Malaga (Arancibia Román *et al.* 2011: 130-132).

Radiocarbon determinations are available for phases IV and I obtained by different methods (Arancibia Román *et al.* 2011: 130-132; Pappa 2012: 36-37). The earliest phase produced two calibrated date ranges of 1040-840 cal BC and 1010-830 cal BC at 2 $\sigma$  from two measurements. The last phase is anchored by another set, calibrated at 2 $\sigma$  as 920-800 cal BC and 890-870 BC/850-780 cal BC. All this points to a brief occupation of the site. Therefore, according to the present radiometric results, the earliest possible date is between 1040/1010 cal BC and the latest possible is 800/780 cal BC.

Despite the uncertainty in the chronology, the silver ingot (Fig. 2) was recovered from the lowest levels of the site. It should be noted that there is no evidence of silver metallurgy having been conducted at La Rebanadilla, such as galena ore, cupels or litharge, only copper slags and objects, lead droplets and some tuyères (Murillo-Barroso 2013; Renzi 2013). Furthermore, as with the central Huelva finds in 1998 (Plaza de las Monjas-Méndez Núñez St) (González de Canales *et al.* 2008), which pushed dates for a Phoenician presence in Iberia back to the 10<sup>th</sup> or early 9<sup>th</sup> centuries BC, Attic skyphoi were also found in association with Phoenician material at La Rebanadilla (Arancibia Román *et al.* 2011: 130-132; Pappa 2012: 36-37; Sánchez-Moreno *et al.* 2012). This could suggest that the Phoenicians' route to Iberia sometimes traversed the central Aegean, which could clearly have repercussions on transmission models of silver exploited in Iberia and transported across the Mediterranean.

The absence of silver metallurgy at La Rebanadilla suggests that the silver ingot was not produced at this site. This would lend support to the view that when the



Fig. 2. La Rebanadilla silver ingot fragment. Cut in antiquity (photo by I. Montero-Ruiz).

Phoenicians arrived in the Iberian Peninsula, they did not exploit the argentiferous lead sources (*i.e.* galena) in the south-east which were later prodigiously mined by the Carthaginians and the Romans (Domergue 1990; Antolinos 2003). Instead, they went to the south-west and exploited the jarositic ores of the Huelva region around Riotinto. In fact, even the galena ores recovered at the 8<sup>th</sup>-6<sup>th</sup> century BC Phoenician site of La Fonteta (Alicante) (Renzi *et al.* 2009) and the c. 6<sup>th</sup> century BC Phoenician shipwreck (Bajo de la Campana) (Polzer 2014), which have been provenanced to the mines of Gador, Almeria in south-eastern Iberia, were found to be devoid of silver, suggesting that non-argentiferous galena was mined, solely for lead. This hints at the possibility that the Phoenicians acquired the knowledge how to extract silver from jarosite prior to any attempts to extract silver from argentiferous lead ores.

## EXPERIMENTAL

The silver ingot recovered from La Rebanadilla, near Malaga, Iberia (Fig. 2) was found to have high levels of lead (over 18 % Pb) using portable x-ray fluorescence spectroscopy (pXRF)<sup>1</sup>, indicating strongly that the silver had undergone cupellation. As an ingot, this suggests the silver is less likely to have been mixed with silver from different locations and that its composition should therefore reflect the signature of its ore source, or that of the lead added to extract the silver. Levels of lead in

silver have been used to differentiate cupellated silver and native silver, with low lead levels suggesting a native silver source (Craddock 2014; Murillo-Barroso *et al.* 2014; Bachmann 1993). However, the efficiency of cupellation, in particular the number of times silver is refined by cupellation, is also reflected in the lead concentration as well as the presence of other metals (*e.g.* bismuth) which can contaminate the silver (L'Heritier *et al.* 2015). An ingot with a high concentration of lead, therefore, reflects low levels of refining, perhaps being cupellated once in a simple scorifier.

In the current investigation, compositional analyses were conducted on the La Rebanadilla ingot using an electron probe microanalyser (EPMA) to complement the preliminary pXRF measurements, in order to provide the range of elements to ascertain the probable provenance of the ore from which the ingot derived.

A fragment of the ingot was cut. The silver sample was mounted in epoxy resin, polished down to 1  $\mu\text{m}$  using alumina paste and observed under the optical microscope throughout the polishing process. Four silver reference materials were used for calibration purposes: MBH131XPag1 is a pure silver standard (99.9 % Ag) with trace elements, including gold (120 ppm). AGA1, AGA2 and AGA3 have major and minor elements including gold, as well as trace elements (Tab. 1). AGA2 was used only as an internal calibrant during measurements.

A JEOL JXA-8100 electron probe microanalyser with a wavelength dispersive X-ray spectrometer (WDS) was run with a 20 kV accelerating voltage and a probe current of  $5 \times 10^{-8}$   $\mu\text{A}$ . Samples were examined at a working distance of 11 mm. The system was calibrated at  $\times 1000$  magnification (about  $100 \mu\text{m} \times 100 \mu\text{m}$ ) by curve-fitting to three known standards (AGA1, AGA3 and MBH131XPag1) for each of the following elements: Ag, Au, Zn, Cu, Pb, Sn, Sb and Bi. Emission lines were chosen to minimise overlapping peaks, and peak/background acquisition times (in seconds) reflected the absolute concentrations present: Ag (La) 30/10, Au (Ma) 60/20, Zn (Ka) 60/20, Cu (Ka) 30/10, Sn (La) 60/20, Pb (Ma) 60/20, Sb (La) 60/20 and Bi (Ma) 60/20. Nine areas were measured on each standard. Linear and quadratic fits were compared for each element during the calibration set up. A linear regression was adopted for all elements as differences between the two fits were found to be negligible.

Errors on each element were determined measuring an internal standard (AGA2) before and after all sample measurements. Some elements show appreciably different concentrations (*e.g.* Cu) between pXRF and EPMA (see Tab. 2). Although this may be a consequence of issues surrounding corrosion that can affect surface techniques like pXRF or overlapping X-ray peaks affected by the large amounts of lead in this

<sup>1</sup> INNOV-X Alpha with X-ray tube and silver anode was used. Detailed information about the equipment, analytical procedures and calibration can be found in Rovira Llorens and Montero Ruiz (2018).

Element %	Ag	Cu	Pb	Au	Zn	Sn	Sb	Bi
AGA1	77.372	19.95	0.207	1.48	0.211	0.291	0.050	0.194
AGA2	86.968	10.00	1.02	0.507	0.502	0.520	0.192	0.113
AGA3	90.546	4.91	1.89	0.258	0.816	0.921	0.459	0.048
MBH131XPAg1	99.9	0.0075	0.004	0.012	0.005	0.004	0.005	0.004

Tab. 1. Compositions of the silver reference materials in weight percent.

La Rebanadilla (ED-XRF) – PA20883												
%	Fe	Ni	Cu	Zn	As	Ag	Sn	Sb	Au	Pb	Bi	other elements
mean	1.34	bdl	0.35	bdl	bdl	77.01	bdl	bdl	bdl	18.98	1.95	bdl
La Rebanadilla (ICP-MS) - Renzi and Montero-Ruiz (unpublished)												
			<sup>208</sup> Pb/ <sup>206</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb					Pb crustal age (Ma)
mean			2.106	0.858	18.193	15.609	38.303					371
st. dev.			0.00056	0.00016	0.01423	0.01283	0.03398					
La Rebanadilla (EPMA)												
%	Au	Ag	Zn	Cu	Sn	Pb	Sb	Bi	Totals			
mean (5)	0.119	78.712	0.107	1.346	bdl	16.065	0.071	3.889	100.309			
st. dev.	0.005	1.099	0.017	0.163	bdl	1.670	0.017	2.570	1.172			

Tab. 2. Composition of the La Rebanadilla ingot (PA20883) by Energy Dispersive X-Ray Fluorescence Spectroscopy (ED-XRF) and electron probe microanalyser (EPMA) and the Lead Isotope Analysis Analysis. The Pb crustal age was calculated from the parameters of Desaulty *et al.* 2011. bdl denotes below the detection limit: For ED-XRF this is estimated at 0.1% for all the elements except Au and Sb which are 0.15%.

semi-refined ingot (both of which are less of a problem for EPMA on sectioned material), it is also possible that the inhomogeneity of copper in the AGA reference materials used to calibrate the EPMA increased the errors for this element.

## RESULTS

pXRF measurements from Renzi and Montero-Ruiz (unpublished) are presented in table 2 for the La Rebanadilla ingot. The sample was described as a Ag-Pb cake, reflecting the high levels of lead in this semi-refined ingot. LIA values were used to calculate the Pb crustal age in millions of years (Ma) from the two-stage evolution model (see Wood *et al.* 2017, 2019; Desaulty *et al.* 2011). EPMA was conducted as it permitted measurement of elements not detected by pXRF, e.g. Au (Tab. 2).

Figure 3 shows the inhomogeneity of the La Rebanadilla ingot. Gold and lead have migrated to the grain boundaries. Although this makes it difficult to determine a representative composition due to the inhomogeneity of this semi-refined ingot, the fact that gold is completely soluble in silver means that further

cupellation (or even melting) will result in the gold returning to the silver as the lead oxidises to form litharge. In other words, gold in silver will reflect the ore source from where the silver derives.

Although the inhomogeneity of the La Rebanadilla ingot appears to be primarily an inconvenience in determining a representative composition which can be compared with silver from other locations, it provides important information on what the Phoenicians traded as a raw material. In effect, the ED-XRF and the EPMA results show that this ingot has about 77-79 % silver, 16-19 % Pb and 2-4 % Bi. Copper was probably not a deliberate addition, ranging between 0.35-1.35 % Cu, and thereby derived from the ore along with iron, antimony, zinc and gold. This suggests that jarosite ores were smelted with excess lead to produce argentiferous lead which then underwent cupellation (*i.e.* the lead was oxidised to litharge resulting in separation of the silver). The fact that so much lead remains in the ingot, suggests that this process was not conducted to produce high quality silver, such as the silver found in silver objects (*i.e.* generally > 95 % Ag). In other words, the traded raw material which came out of Iberia was effectively a coarse silver-lead alloy which only later in the *chaîne opératoire*

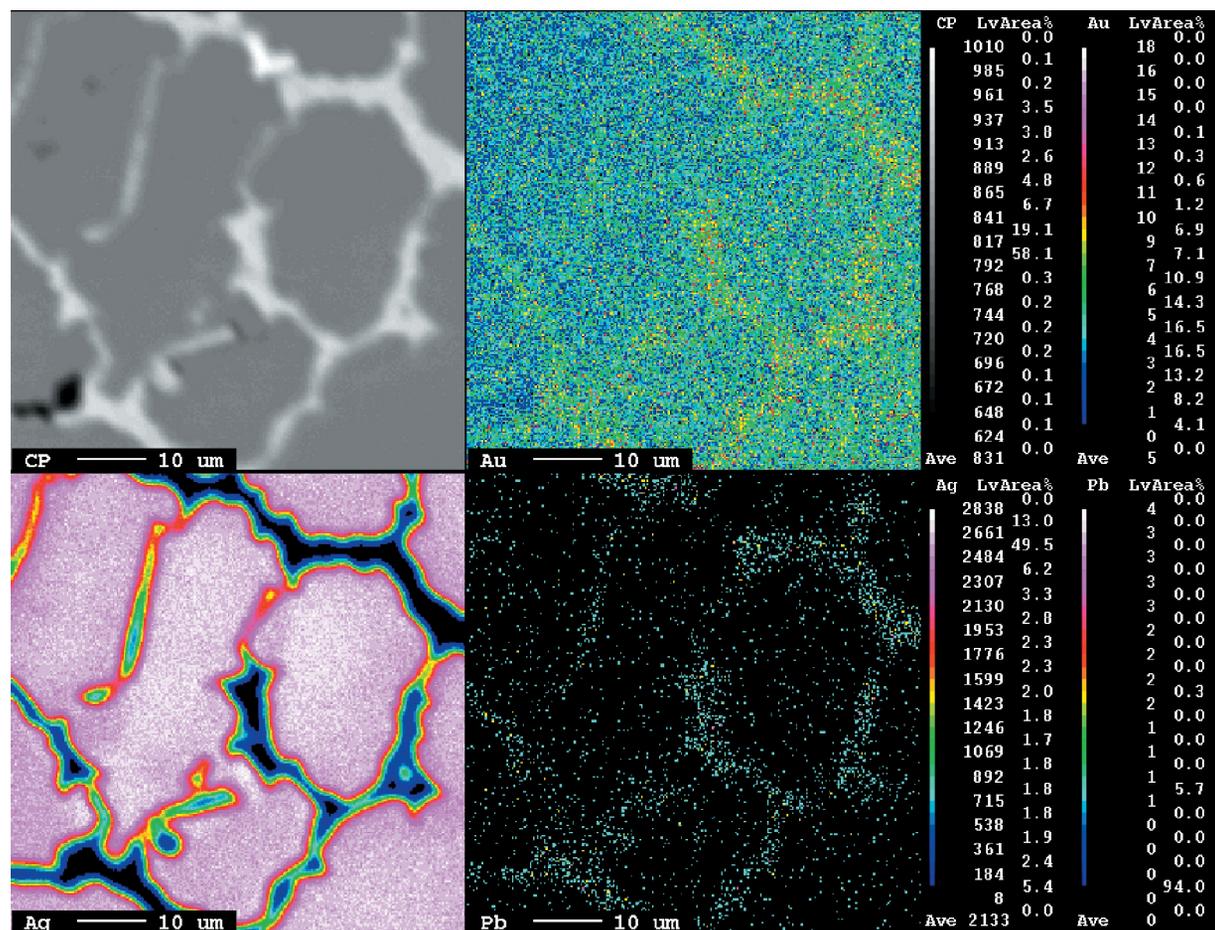


Fig. 3. Electron Probe Microanalyser (EPMA) map for the La Rebanadilla silver ingot, showing phase segregation of both lead and gold at the grain boundaries. This inhomogeneity makes it difficult to provide a representative composition as different magnifications would result in different compositional signatures. Further cupellation would result in lead being removed from the silver (as it oxidises to litharge), but the gold would return to the silver as it forms a solid solution across the full range of concentrations (in colour in the electronic version).

toire was purified through further cupellation, and often deliberately alloyed with copper to produce objects.

The high bismuth and low gold concentrations of the ingot from La Rebanadilla (3.889 % Bi; 0.119 % Au) are consistent with the ores of the Pyritic belt, especially around Riotinto in south-west Iberia (Gale *et al.* 1980), which is further supported by the ingot's Hercynian age (371Ma) (Desaulty *et al.* 2011; Albarède *et al.* 2012). The lead isotope signature (and thereby the crustal age) could have derived from other sources of Iberian lead. La Fonteta, for example, has galena with Hercynian ages which probably derived from Gador, Almería (Renzi *et al.* 2009). However, as mentioned above, this galena is not argentiferous, and could therefore have been only used as a silver collector. This does not automatically mean that the lead isotope signature for the ingot from La Rebanadilla derived from

Riotinto. Nevertheless, there is galena at Riotinto from the altered porphyry wall rocks along the surface of the North, South and San Dionisio lodes, which are scored with ancient tool marks (Allan 1970). Furthermore, a workshop at Monte Romero (Huelva) suggest that local ores were used, perhaps mixed with lead from further afield (Stos-Gale 2001; Kassianidou 2003). Essentially, the ingot from La Rebanadilla has an Iberian compositional and LIA signature (and Hercynian crustal age) consistent with south-west Iberia.

## DISCUSSION

In terms of archaeological significance, the La Rebanadilla ingot provides support for the movement of silver from Iberia to the southern Levant in the Early

	Ref. no.	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{209}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	Pb crustal age (Ma)	Au%	Ag%	Cu (%)	Pb (%)	Au/Ag x100
La Rebanadilla	UE1200	2.106	0.858	18.193	15.609	38.303	371	0.119	78.712	1.346	16.065	0.15
Tell Keisan	KSN001 / IAA#79-549	2.102	0.858	18.344	15.741	38.557	411	<0.2	92.96	6.75	0.29	<0.22
Tell Keisan	KSN006 / IAA#79-549	2.101	0.858	18.289	15.695	38.428	399	0.30	80.70	19.00	<0.2	0.37
Eshtemoa	EST013	2.111	0.858	18.212	15.620	38.453	370	<0.2	99.37	0.63	<0.2 High Bi	<0.20
Eshtemoa	EST008	2.103	0.857	18.212	15.611	38.296	361	0.15	98.66	0.00	1.18	0.15
Eshtemoa	EST009	2.108	0.858	18.176	15.601	38.312	374	0.19	99.25	0.56	<0.2	0.19
Eshtemoa	EST005	2.106	0.858	18.207	15.616	38.341	369	0.18	99.40	0.42	<0.2	0.18
Eshtemoa	EST006	2.101	0.856	18.246	15.618	38.342	344	<0.2	95.77	3.79	0.44	<0.21
Eshtemoa	EST002	2.106	0.857	18.238	15.630	38.407	363	<0.2	98.00	0.33	1.67	<0.20
Eshtemoa	EST012	2.103	0.855	18.192	15.558	38.261	315	0.28	98.54	0.47	0.71	0.28
Ein Hofez	HFZ002 /2246	2.101	0.854	18.297	15.623	38.441	314	0.22	98.23	0.23	1.32	0.22
Ein Hofez	HFZ003 /2246	2.106	0.857	18.262	15.654	38.459	372	<0.2	98.84	0.39	0.76	<0.20
Ein Hofez	HFZ004 /2246	2.103	0.856	18.257	15.625	38.391	344	0.24	97.33	0.30	2.13	0.25
Ein Hofez	HFZ005 /2247	2.105	0.857	18.271	15.657	38.453	370	<0.2	100.00	0.00	<0.2	<0.20
Ein Hofez	2HFZ002 /2247	2.100	0.855	18.239	15.601	38.293	330	<0.2	97.15	0.41	2.44	<0.21
Ein Hofez	2HFZ006 /2247	2.103	0.854	18.297	15.619	38.478	309	0.16	97.83	0.54	1.48	0.16
Tel Dor	DOR004 / 97.3320/1	20.964	0.852	18.499	15.761	38.782	327	<0.2	99.79	0.21	<0.2	<0.2

Tab. 3. Comparison of Lead Isotope Analysis values, Pb crustal ages (Ma) and Au/Ag ratios in silver from La Rebanadilla ingot and silver in the hoards of the southern Levant (Tell Keisan, Eshtemoa, Ein Hofez and Tel Dor. LIA data and compositional analysis of southern Levantine silver from Energy Dispersive X-Ray Fluorescence Spectroscopy (ED-XRF) (Oxalid 2019). The Pb crustal age was calculated from the parameters of Desauty *et al.* (2011).

	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	Pb crustal age (Ma)
La Rebanadilla ingot	2.106	0.858	18.193	15.609	38.303	371
Las Arenillas ingot	2.103	0.859	18.202	15.643	38.275	403
Miletus coin	2.111	0.858	18.212	15.620	38.453	370
500 BC	2.103	0.856	18.274	15.645	38.436	354
Eshtemoa (c. 11 <sup>th</sup> -9 <sup>th</sup> or 10 <sup>th</sup> -8 <sup>th</sup> centuries BC)	2.111	0.858	18.212	15.620	38.453	370
Aeginetan coin 550-485 BC	2.101	0.855	18.284	15.638	38.423	340
Athenian coin 393-300 BC	2.064	0.833	18.858	15.701	38.914	8

Tab. 4. Comparison of Lead Isotope Analysis values and Pb crustal ages (Ma) in silver from La Rebanadilla ingot, a silver fragment from Las Arenillas, silver coins from Miletus, Athens (Desaulty *et al.* 2011), Aegina (Gale *et al.* 1980) and Eshtemoa (Oxalid 2019). The Pb crustal age was calculated from the parameters of Desaulty *et al.* (2011).

Iron Age. The crustal age of the silver in this ingot (371 Ma) and the ratio of  $\text{Au}/\text{Ag} \times 100$  of 0.15 determined from EPMA, coupled with its potential late 11<sup>th</sup>-9<sup>th</sup> century BC chronology based on radiocarbon (or mid-9<sup>th</sup> century BC using archaeological arguments) suggests that this ingot is very similar to pieces of hacksilver found in some of the southern Levantine hoards (Wood *et al.* 2019). Table 3 shows that LIA values, Pb crustal ages and gold/silver fractions for silver from the southern Levantine hoards of Ein Hofez (c. 10<sup>th</sup>-9<sup>th</sup> centuries BC), Eshtemoa (c. 11<sup>th</sup>-9<sup>th</sup> or 10<sup>th</sup>-8<sup>th</sup> centuries BC), Tell Keisan (late 11<sup>th</sup> century BC) and Tel Dor (11<sup>th</sup>-10<sup>th</sup> century BC), are very similar those of the silver from the ingot at La Rebanadilla (see Wood *et al.* 2019 for more details on these hoards). Furthermore, although the ingot was recovered from La Rebanadilla in south-east Iberia it has a crustal age more consistent with the Huelva region, *i.e.* a Hercynian (250-400 Ma) rather than a Betic (Alpine) (<90 Ma) orogeny (see Desaulty *et al.* 2011; Albarède *et al.* 2012) found in the vicinity of La Rebanadilla. This supports that the ingot travelled to La Rebanadilla from Huelva, and that this type of ingot could have travelled further east.

A Huelva provenance for the La Rebanadilla ingot is further supported from its bismuth levels. Although there is high variation in the concentration of Bi in the sample (Tab. 2), its presence in significant amounts (3-4 % Bi) suggests an ore source with an appreciable bismuth content. Riotinto ores have been found to have high bismuth levels (Bi: trace - 0.25 %), and refined silver found on the slag heaps has levels of Bi of 0.42 % (Salkield 1982: 137-147). A piece of semi-refined silver found at the site of Las Arenillas (site RT103: also known as Cerro del Moro, a prominent steep hill next to the small mining town of Nerva, about 3km from the main slag heaps of the Riotinto mine – recovered by Beno Rothenberg for the Institute of Archaeology, UCL) has high levels of bismuth (15.4 %

from Craddock *et al.* 1987, and around 22 % from the EPMA conducted by the current authors) and a crustal age commensurate with Riotinto (403 Ma) (see Tab. 4).

The high concentration of lead (over 16 %) in the ingot from La Rebanadilla suggests that the silver was refined using cupellation. The higher levels of lead (*i.e.* >1 %) in the silver hoards of the southern Levant suggest that at least some of this silver was cupellated<sup>2</sup>. Nonetheless, all the silver objects in table 3 are consistent with silver deriving from south-west Iberia. The purity of the silver in the southern Levant silver, often above 95 % Ag (Tab. 3), suggests that any silver received in the form of semi-refined ingots must have undergone further refinement and, in some instances, alloyed subsequently with copper before being made into objects, *i.e.* some silver appears to have higher levels of copper than would be expected to survive the refining process from copper naturally occurring in the ores.

There appears to be continuity in the practice of producing semi-refined ingots. A silver ingot from the 7<sup>th</sup> century BC site of El Risco in the province of Cáceres also exhibits high levels of lead (25-34 % Pb from XRF), suggesting that this practice continued into the Orientalising period (c. 8<sup>th</sup>-6<sup>th</sup> centuries BC) (Murrillo-Barroso 2013: 225-226). Hunt Ortiz (2003: 208) published the composition of an ingot from Castrejones (a site in the Aznalcóllar mining district c. 8<sup>th</sup> century BC) with 25.3 % Pb (from atomic absorption spectroscopy), with an external concentration of 23.47%Pb and internal concentration of 16.87 % Pb (from PIXE) (the difference potentially reflecting lead migration during post-deposition). A silver ‘drop’ from Huelva was found to have 40.77 % Pb (from SEM-EDS) (González de Canales *et al.* 2004). Moreover, a silver

<sup>2</sup> Even low lead levels could indicate multiple cupellation operations, although it could also indicate non-cupellated silver from native silver or dry silver ores (Patterson 1971; Montero-Ruiz *et al.* 1995; Craddock 2014).

ingot from Cerro del Villar, a Phoenician site near Malaga which was inhabited after La Rebanadilla, had 5.17 % Pb (from SEM-EDS) (Renzi 2013: 203). These high lead levels in silver ingots not only indicate that it was common to produce semi-refined silver but that the Phoenicians were involved in this practice.

The repercussions of transporting ingots, which were effectively silver-lead alloys, is that anyone who procured this silver, such as the Phoenicians in the Levant, must have known how to conduct cupellation as a refining method and alloy silver with copper in order to produce silver with the desired aesthetic and mechanical properties. This strongly suggests two forms of knowledge were present among the Phoenicians who dealt with silver: knowledge of how to prospect, mine, and smelt argentiferous ores and coarsely refine the argentiferous lead to make ingots, and knowledge of how to further refine, alloy and work silver into objects. These forms of knowledge in the Iron Age were probably quite separate, with each group being specialists in their respective fields. In other words, those who produced the ingots were probably adept at prospecting, mining, smelting and producing large quantities of semi-refined silver, while those who acquired and worked with the ingots were silversmiths.

The association of Phoenician and Greek material at La Rebanadilla (as with the central Huelva finds) could suggest that the recipients of such silver ingots were not only Levantine silversmiths, but also those resident around the Aegean. In fact, the amounts of Greek and Phoenician material found in association at La Rebanadilla (*i.e.* Attic skyphoi) and at the site in central Huelva (with the 8000 sherds catalogued as: Phoenician —over 3000—, Greek —with two types of Euboean pottery —33—, Cypriot —8—, Sardinian —30— and Italian —2—), could support the clear indication of apparently peaceful Phoenician contact with Euboea and Attica at the time (Kourou 2012). As discussed by Hodos (2009), this would be in contrast to the often depicted Greek-Phoenician rivalry. Furthermore, in addition to the Middle Geometric II (c. 800-750 BC) material, there is increasing amounts of evidence, from the identification of the Late Helladic IIIC (1153-1070 BC) and Late Minoan IIIC (1190-1070 BC) pottery at Huelva (Palos de la Frontera Street), that the movement between the Western Mediterranean and the Eastern Mediterranean, Phoenicia, Cyprus and Crete was active in the first half of the 12<sup>th</sup> century BC (Gómez Toscano and Mederos Martín 2018).

## IBERIAN SILVER IN GREECE

The presence of Greek pottery at the harbour site of La Rebanadilla could suggest that the amounts of

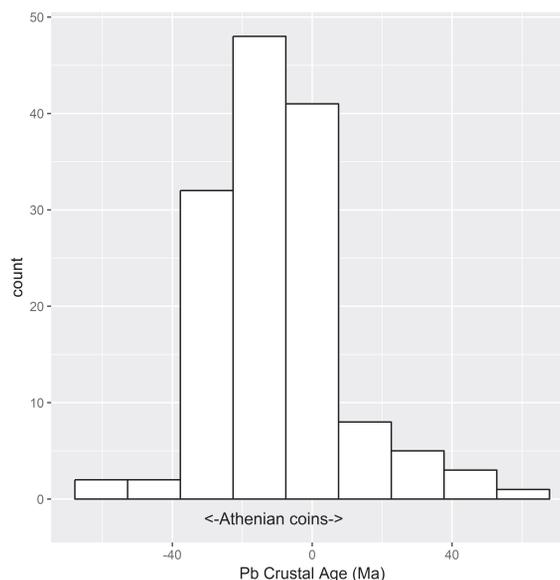


Fig. 4. Histogram of the Pb crustal age (Ma) for ores from Laurion, as calculated from the lead isotopes from the Greek ores database (Oxalid 2019) using the two-stage evolution model with parameters from Desaulty *et al.* (2011). The range of calculated Pb crustal ages for Athenian coins from the Asyut hoard (Gale *et al.* 1980) is also presented which shows that these coins are commensurate with silver mined from Laurion.

silver which were mined during the ‘Orientalising’ period (c. 8<sup>th</sup>-6<sup>th</sup> centuries BC) or earlier in Iberia, which is largely unaccounted for, may have not only ended up in hoards in the southern Levant and the Near East but also around the Aegean. There are some indications which support this view. For example, a Greek silver coin recovered at Ionia Miletus (c. 500 BC) has an isotopic signature (Desaulty *et al.* 2011) which is very close to the silver ingot recovered at La Rebanadilla and the silver fragment recovered at Las Arenillas near the Riotinto mining sites (Tab. 4). It is also very different to that of Athenian coins which were produced from silver mined at Laurion in Attica, which generally have low Pb crustal ages (see Wood *et al.* 2017, 2019) (Tab. 4). Figure 4 shows that the Pb crustal ages of Laurion ores lie between -30 and +55 Ma and a range of ages of Athenian coins from the Asyut hoard (Gale *et al.* 1980).

This Greek coin from Miletus (c. 500 BC) appears to be rare example of Iberian silver in the Aegean. However, the movement of semi-refined ingots perhaps provides an explanation for paucity of silver recognised as deriving from Iberia in the archaeological record (something which needs to be reconciled with the aforementioned massive silver slag heaps at Iberian mining sites, such as Riotinto).

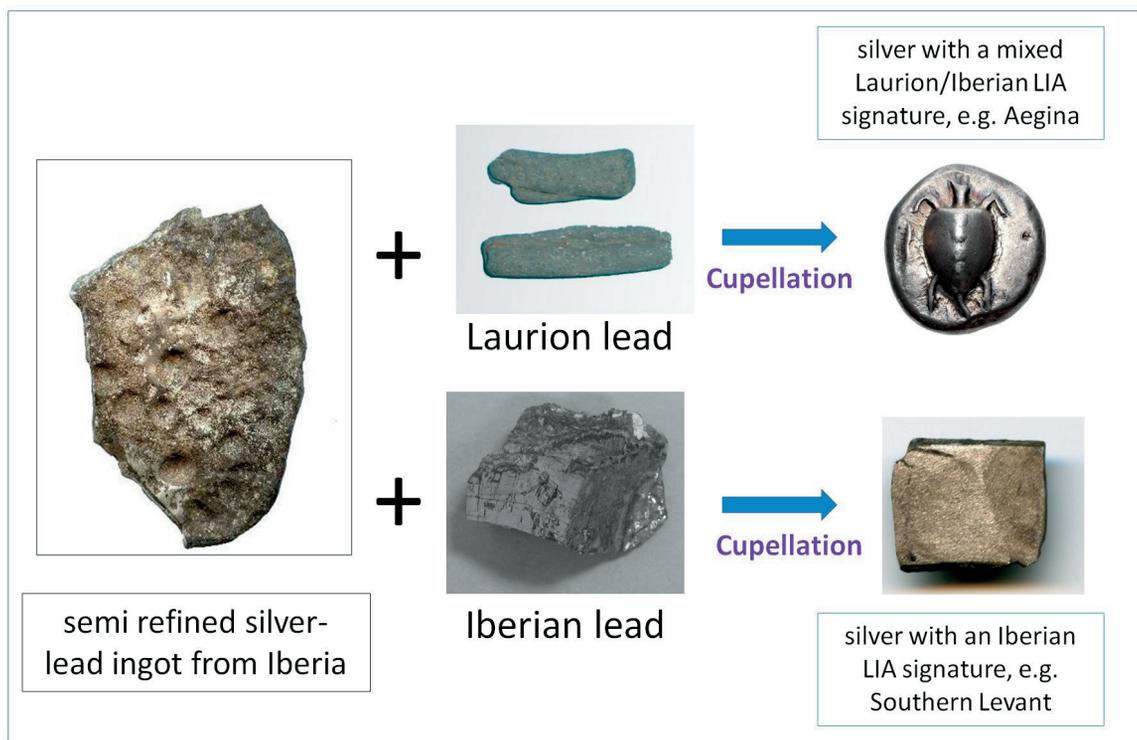


Fig. 5. Proposed mechanism behind the elusive nature of Iberian silver in the archaeological record: Silver in the form of semi-refined silver-lead ingots was transported from Iberia to silversmiths at various locations in the Central and East Mediterranean and refined using lead available to the local silversmiths. In Greece, Iberian silver-lead ingots could have been refined with lead from Laurion, resulting in silver artefacts with mixed lead isotopic signatures. In the southern Levant (an area with no lead sources of its own), Iberian lead was potentially conveyed along with the silver-lead ingots for silversmiths, thereby maintaining the integrity of the Iberian lead isotope signatures in some of the hacksilver recovered in the southern Levantine hoards (in colour in the electronic version).

A possible mechanism could be as follows: Silver in the form of semi-refined ingots (*i.e.* silver-lead alloys) from south-west Iberia was transported across the Mediterranean, probably conveyed by Phoenicians. This silver had been extracted and coarsely refined by the prospectors, miners and smelters working in Iberia (again, probably Phoenicians or indigenous people directed by Phoenicians). The silver ingots they produced were supplied to silversmiths in the Levant and other silversmiths *en route*, including those working around the Aegean. These ingots required further refining before they were fashioned into objects. This required cupellation, and therefore required access to lead to act as a silver collector. Unlike Greece, with its lead mines at Laurion on Attica, the Levant is poor in mineral resources, including lead. This could suggest that whereas the Greeks used their own local lead to refine the silver ingots they procured, the Phoenicians could have supplied the silversmiths in the areas of the Levant with lead from Iberia as well as with silver ingots.

This would result in two types of isotopic and compositional signature (Fig. 5): In Greece (and at any other location *en route* where silver was transported), there would be dilution of the Iberian compositional and isotopic signatures as lead from local sources (*e.g.* from Laurion on Attica in Greece) was added to refine the ingots; In the Levant, an Iberian signature would be maintained (such as the silver in table 3 from the southern Levant) as the lead used by the silversmiths derived from different parts of Iberia (Murillo-Barroso *et al.* 2016). In fact, one silver piece from Eshtemoa (EST013) was recorded as having high levels of bismuth (Oxalid 2019), which along with its LIA values further supports a south-west Iberian provenance (Tabs. 3 and 4). In essence, the silver-lead ingots were probably considered as the raw material used by silversmiths to produce objects. It should be noted that the refining required to get high purity silver and its associated dilution of signatures would have taken place prior to any subsequent dilution due to recycling, alloying and mixing of silver (Wood *et al.* 2019).

In effect, this would indicate that it would be rare to find silver with a pure Iberian signature<sup>3</sup> in any location where there were sufficient local lead sources to conduct cupellation. As lead sources are common around the Mediterranean, this suggests that Iberian silver would be rendered almost invisible in terms of its compositional and isotopic signatures. Put another way, the Levant may be one of the only locations where silver with an intact Iberian signature is maintained, apart from occasional finds such as the coin from Miletus (Tab. 4).

This has significant repercussions. First, it could suggest that before Laurion was fully recognised for its argentiferous galena sources, Iberian silver may have been supplied to areas around the Mediterranean during the Iron Age. This would appear to contradict the view that Laurion was exploited for silver from the Late Bronze Age (Gale and Stos-Gale 1981; Gale *et al.* 1980; Shepherd 1993: 75; Kassianidou and Knapp 2005: 220). However, refining Iberian silver using lead from local ores (*e.g.* such as lead ores from Laurion), would be very difficult to distinguish from silver deriving from local argentiferous lead sources, especially using techniques based solely on analysing lead isotopes. Essentially, the use of local lead to refine coarsely cupellated Iberian silver would always result in mixed signatures - signatures which would produce Pb crustal ages which lie between the end members of the sources of lead.

## ISLAND OF AEGINA

A possible example of such mixed signatures may be found in the silver from the Greek island of Aegina, an island which had no significant mineral resources and certainly no silver sources of its own (Gale *et al.* 1980). The histogram in figure 6 shows the range of Pb crustal ages (Ma) calculated from the LIA (Gale *et al.* 1980) of 38 Aeginetan silver coins (*c.* 550-485 BC) recovered in the Asyut hoard in Egypt. Coins with low Pb crustal ages (*i.e.* the dark bars) have ages consistent with the ores at Laurion (and Athenian coins) (see Fig. 4). 26 coins have Pb crustal ages which are not only higher than expected from silver deriving from Laurion but also cover a wide range of Pb crustal ages.

The provenance of Aeginetan coins and silver is usually discussed in relation to silver from Laurion, the Aegean island of Siphnos and unknown sources.

<sup>3</sup> The arrival of different lead resources into south-west Iberia for the cupellation process makes it difficult to determine exactly from where the lead derived which was used as a silver collector to produce lead-silver ingots. Nevertheless, older crustal ages are indicative of an Iberian source and, as these are absent in Eastern Mediterranean lead sources, this suggests that this signature is associated with silver extracted using Iberian lead.

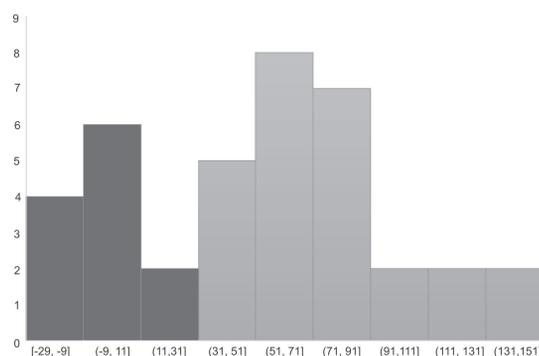


Fig. 6. A histogram showing the range of Pb crustal ages in 38 Aeginetan silver coins (*c.* 550-485 BC) recovered from the Asyut hoard in Egypt. The low crustal ages (dark) are consistent with galena ores from Laurion (and Athenian coins). The higher ages reflect silver deriving from older sources. One Aeginetan coin has a Pb crustal age of 340 Ma (not shown due to scale).

Laurion silver tends to have low levels of gold (*i.e.* <0.1 % Au) (see Gale *et al.* 1980; Pernicka 1981; Craddock 1995: 213; Meyers 2003), as well as low bismuth and antimony (Gale *et al.* 1980), reflecting its galena source. However, the appreciable levels of bismuth (mean: 0.165 % Bi; range: 0.011-0.67 % Bi) and gold (mean: 0.29 %; range: 0.012-1.58 %) in many of these Aeginetan coins, and the generally low levels of gold in silver on Siphnos (between 0.005-0.05 % Au, potentially between 0.01-0.2 % Au and rarely up to 1 % Au in extracted silver - see Gale *et al.* 1980: 39) is difficult to reconcile with all of these coins being minted from either Laurion or Siphnian silver. Further doubt is raised when it is noted that at least 17 coins fall within neither the Laurion nor Siphnian LIA fields (Gale *et al.* 1980: 42, figs. 10 and 12).

The LIA plot in figure 7 shows that the Aeginetan coins could lie on a trajectory between Athenian coins (considered to have been minted from Laurion silver) and Iberian ores (the ores of Riotinto, Gador etc.). This interaction would be expected of mixing lead from these locations, *i.e.* silver-lead ingots from Iberia mixed with lead from Laurion.

Figure 8 shows the same data, incorporating the gold to silver fraction in the Aeginetan coins, plotted against their Pb Crustal age (Ma) calculated from their lead isotope ratios (see Wood *et al.* 2017). What is immediately apparent is that many of the coins have Au/Ag levels which are higher than expected from silver deriving from Laurion or Siphnian ores (which generally have less than 0.1 % Au). The gold concentrations in silver deriving from ores at Riotinto range from 0.3-16 % Au, with yellow ores having the highest (Au/Ag ×100

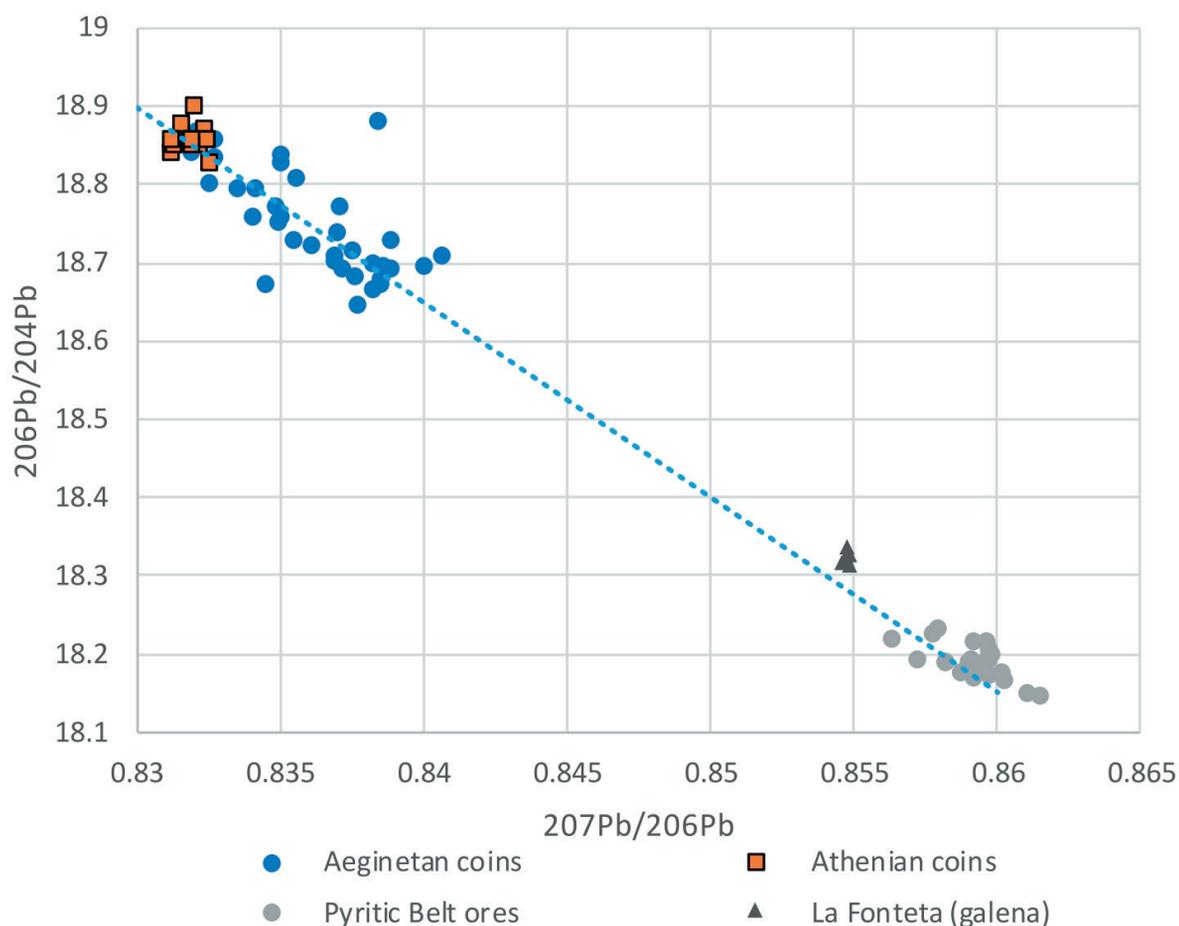


Fig. 7. Lead Isotope Analysis plot of Athenian coins (considered to derive from ores at Laurion), Aeginetan coins (data from Gale *et al.* 1980), ores from the Pyritic belt in south-west Iberia (data from Marcoux 1998 and Pomiès *et al.* 1998) and galena ores from La Fonteta (Renzi *et al.* 2009). The hand drawn line is presented to indicate that some of the Aeginetan silver coins could lie on a trajectory to Iberian ores (in colour in the electronic version).

= 19.38) and grey-green ores having the lowest ( $\text{Au/Ag} \times 100 = 0.32$ ) levels of gold (Gale *et al.* 1980: tab. 1). This suggests that gold from a different silver source has entered the system, potentially from Iberian silver being mixed with Laurion lead. It is not claimed here that Aegina only received silver from Iberia. It is probable that the people of this silver-free island procured silver from wherever they could. In fact, six of the 38 Aeginetan coins have high levels of iridium, which possibly indicates a Near East origin for the silver (Wood *et al.* 2017). Nonetheless, this silver may still have been refined with lead from Laurion.

There are possibly several interactions in figure 8 which support that silver with high gold levels from older silver ores was mixed with younger lead. There is some semblance of a linear relationship on the  $\text{Au/Ag} \times 100$  vs Pb crustal age (Ma) plot which allows

investigation. Note that a linear regression was not applied to the data as it is unknown, a priori, which silver is mixed. However, extrapolating this hand drawn line to 350 Ma (approximately the Pb crustal age of Iberian silver – Tabs. 3 and 4) would result in an  $\text{Au/Ag} \times 100$  level of about 5. This is clearly within the range of  $\text{Au/Ag}$  levels for Iberian silver from Riotinto ores (Gale *et al.* 1980: table 1). It is also similar to some silver recovered in the southern Levant hoards which have Pb crustal ages ranging between 300-500 Ma and high  $\text{Au/Ag} \times 100$  values (Tab. 5).

In effect, figures 6, 7 and 8 are consistent with Aeginetan silver coins minted from Iberian silver refined with lead from Laurion. Furthermore, one of the Aeginetan coins (reverse style: five segments IIIa; sample 444) analysed by Gale *et al.* (1980) is highly consistent with the ingot from La Rebanadilla (Tab. 4),

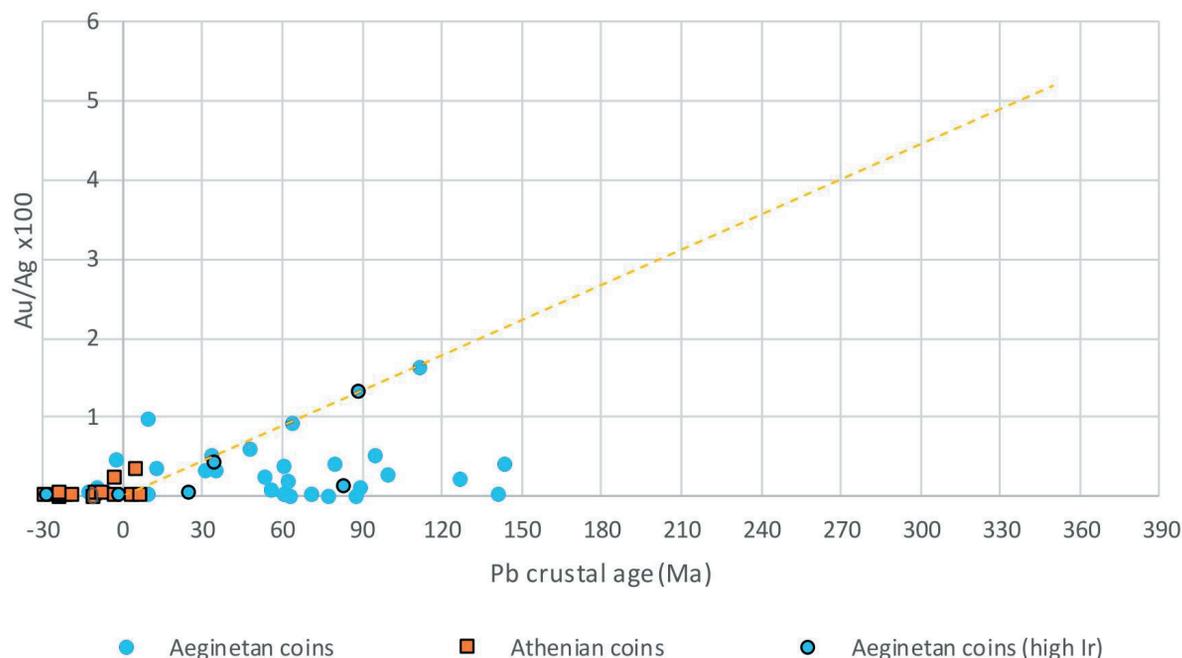


Fig. 8. Fraction of gold in silver vs. the Pb crustal age for Aeginetan and Athenian coins. The Pb crustal age was calculated from the lead isotope values (Gale *et al.* 1980) using the parameters of Desaulty *et al.* (2011). Most coins from Aegina have Au/Ag levels in excess of that found in the ores of Laurion or Siphnos, suggesting a different silver source. The silver in coins with high iridium levels may have derived from the Near East (Wood *et al.* 2017). The range of crustal ages and the semblance of linearity are suggestive of silver from Iberia mixed with lead from Laurion (in colour in the electronic version).

suggesting that some ‘pure’ Iberian silver (*i.e.* silver which was not further cupellated or had been cupellated with Iberian lead) was also available to be minted on Aegina. Although no compositional data was available for this coin, its crustal age (340 Ma) suggests that Iberian silver was available on Aegina in the late 6<sup>th</sup> and early 5<sup>th</sup> centuries BC. This could resurrect the view that Iberia was an important source for Aeginetan

silver (Dayton 1978), a view that perhaps should re-surface as previous arguments against this have centred on the claims that *Greek* trade (*i.e.* not necessarily Levantine trade) was interrupted because “the defeat of the Phocaeans in *c.* 540 BC by the Carthaginians and Etruscans virtually closed off the western Mediterranean” (Gale *et al.* 1980: 43).

In essence, the re-evaluation presented here would suggest that Iberia was a major silver supplier prior to the Classical period, with the recipients of this silver providing their own lead to further refine the semi-refined ingots they procured. Moreover, this mechanism, which focusses on the movement of semi-refined silver ingots, would perhaps go some way toward finding an explanation for the dearth of silver with an intact Iberian signature in 1<sup>st</sup> millennium BC contexts.

## CONCLUSIONS

The recovery of a silver ingot from the Phoenician settlement of La Rebanadilla, a site with no evidence of silver metallurgy debris (*i.e.* ore, cupels or litharge), supports the proposition that the Phoenicians did not exploit the argentiferous galena ores of south-east Iberia.

Find location	Chronology	Pb crustal age (Ma)	Au/Ag x 100
Shechem	LBA/IA or 1000-200 BC	384	4.83
		374	4.20
		355	5.76
Tel Dor	11 <sup>th</sup> -10 <sup>th</sup> centuries BC	311	1.94
Tell Keisan	Second half of 11 <sup>th</sup> century BC	312	2.69
Ashkelon	Late 12 <sup>th</sup> century BC / 1100BC	316	4.34
		469	2.83

Tab. 5. Silver from the hoards of the southern Levant with high levels of gold and high Pb crustal ages.

Bismuth's presence in the La Rebanadilla ingot is indicative of a bismuth-rich silver ore, which coupled with the Hercynian crustal age (371 Ma) suggests that both the ore and the lead used for cupellation potentially came from the Pyritic belt in south-west Iberia before being transported to La Rebanadilla. The Au/Ag ratio in the La Rebanadilla ingot, along with its crustal age, is consistent with some of the pieces of silver found in the hoards of the southern Levant. This is consistent with movement of silver ingots from south-west to south-east Iberia before travelling across the Mediterranean. Although the chronology of the La Rebanadilla site is not precise, the fact that the ingot was found at the lowest levels could suggest that the La Rebanadilla ingot was potentially part of the same movement of silver which resulted in the subsequent deposition of silver in the hoards of the southern Levant, *i.e.* Ein Hofez (c. 10<sup>th</sup>-9<sup>th</sup> centuries BC), Eshtemoa (c. 11<sup>th</sup>-9<sup>th</sup> or 10<sup>th</sup>-8<sup>th</sup> centuries BC), Tell Keisan (late 11<sup>th</sup> century BC) and Tel Dor (11<sup>th</sup>-10<sup>th</sup> century BC)<sup>4</sup>. However, before any of this silver was deposited, the ingot it derived from was refined through further cupellation and sometimes alloyed with copper, potentially by silversmiths who may not have had any working knowledge of prospecting, mining or smelting argentiferous ores.

With Greek material also forming part of the assemblages at La Rebanadilla and in central Huelva, with the latter contributing to the dating of the Phoenician material found with it to the 10<sup>th</sup> or early 9<sup>th</sup> centuries BC (González de Canales *et al.* 2008: 631-655), it is conceivable that some of the Iberian silver transported east ended up elsewhere *en route*, potentially in the hands of silversmiths around the Aegean as well as the Levant. The use of local lead by the procurers of these ingots to refine the silver before fashioning it into objects could explain the dearth of silver with intact Iberian compositional and LIA signatures outside of Iberia and areas such as the southern Levant. In effect, analysis of silver recovered around the Mediterranean, including the Aegean, may reveal where the large amounts of silver extracted from Iberia ended up, as well as providing a useful metric to assess how these areas were affected by their contact with Iberian silver.

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<sup>4</sup> A recent paper by Eshel *et al.* 2019 offers a shorter chronology to these hoards (10<sup>th</sup>-8<sup>th</sup> century BC) but essentially identified Iberian silver at least from the 9<sup>th</sup> century BC, interpreting the use of Sardinian resources for earlier dates.

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