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RESEARCH

An insight from the sea: provenance studies on Roman lead artefacts from the Arade River, Portimão (Portugal)

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Abstract

In the present study, 37 lead artefacts were characterised to identify possible lead sources allowing to establish trade fluxes concerning food and textile products during Roman times. These artefacts were uncovered by dredging works at the Arade River estuary (Portimão). The city of Portimão (Lusitania province) was an important harbour, where several fish-processing factories were installed, and Arade River provides major access to the hinterland, both displaying an important commercial activity during the Late Antiquity. The methodology includes the typological and chemical (elemental and Pb isotopes) characterisation of artefacts. Samples were divided into the following: (i) rectangular plaques with decorations in relief such as tridents, fishes, or palms leaf, an iconography known to be displayed in some African amphora handles; (ii) small plaques with one perforation and incised Roman numerals, probably related with textile products; and (iii) fishing net weights, smooth plaques of unknown functionality, and a small rectangular prismatic plaque, perhaps an ingot. Elemental analysis was performed by ICP-MS, and results were interpreted by multivariate statistical analysis, which suggested different processes to obtain raw materials, namely lead obtained by the reduction of litharge or smelting of silver-poor galena. Cluster analysis grouped most of samples with motif depictions, which were further analysed by MC-ICP-MS to determine Pb isotope ratios. The possible sources of lead were identified by combining archaeological data with the nearest Euclidean neighbours using a large database comprising the Iberian Peninsula and Mediterranean region. The Pb isotope signatures suggested lead sources located not only in the Iberian Peninsula but also in North Africa, evidencing a long-distance trade between those Roman provinces.

Keywords Lead labels · Elemental composition · Lead isotopes · Provenance · Roman trade networks · Portimão

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Introduction

The Mediterranean and Atlantic coasts of the Iberian Peninsula were systematically and intensively sailed during the Iron Age, being an important part of Phoenician trade routes to exchange goods and raw materials, in particular metals (Murillo-Barroso et al. 2016; Arruda 2020). Later on, with the romanization, the contacts of the Iberian Peninsula with the Mediterranean were reinforced to satisfy the growing Roman demand for metals since the Italic Peninsula was poor in mineral resources (Domergue and Rico 2014). Romans arrived in the Iberian Peninsula at the beginning of the Second Punic War (218 BC), which finished with the Carthaginians defeat in 206 BC, giving rise to the romanization of the peninsula (Rodà de Llanza 2009). The geographic location of the Iberian Peninsula in the western Mediterranean area easily implemented contacts with

Roman provinces, including North African ones, supplying local communities with raw materials and foodstuffs and, at the same time, stimulating the growth of social, cultural, and economic Roman models (Mantas 2004; Bombico 2015).

The Iberian Peninsula was an important metalliferous region, and as expected, the Roman expansion in the II century BC started with the demand for ore deposits rich in lead, silver, copper, tin, and gold (Blázquez Martinez 1989; Cano 2003; Rodà de Llanza 2009). Trade networks were developed including well-defined and organised terrestrial and maritime routes (Domergue and Rico 2014). Galena (PbS), one of the most abundant ores in the Iberian Peninsula, was the primary source of lead and an important source of silver (argentiferous galena) being mainly used by Romans for coinage (Cano 2003). For instance, argentiferous galena from Cartagena-Mazarrón region was the main source of lead and silver, being largely used during the Roman Republic Period (Domergue 2008; Rodà de Llanza 2005). As a consequence of the cupellation process applied in silver metallurgy, lead was used on a large scale and became of high economic importance, not only in the silver production but also in the manufacture of lead artefacts, such as weapons (sling bullets), water pipes, and shipbuilding materials. At the end of the Republic, the mining district of Sierra Morena became an important source of lead and silver due to the gradual Roman conquest of the Iberian Peninsula (Domergue 2008; Domergue et al. 2012, 2016). Later, within the Roman Empire, the Iberian Pyrite Belt was intensively mined to produce silver and copper (Delgado Domínguez 2006). In this period, lead had an enhanced use for the manufacture of pipes used in hydraulic systems of public and private buildings (Boni et al. 2000; Gomes et al. 2016). For instance, the rebuilding of Carthage during the II–III century AD (Africa Proconsularis) required a significant use of lead resources, thus becoming the third largest city of the Roman Empire. During this time, ore deposits in North Africa, namely in present-day Tunisia, were extensively used and there is evidence of metal trade between the Iberian Peninsula and North African regions (Fenn et al. 2009; Skaggs et al. 2012).

Regarding traded goods, lead ingots were carried together with amphorae containing distinct foodstuffs, as it has been recorded in numerous shipwrecks and archaeological underwater finds in maritime and fluvial contexts along the Mediterranean Sea and the Atlantic coastline of the Lusitania province (Parker 1992; Trincherini et al. 2001; Domergue and Rico 2014; Bombico 2015; Quevedo and Bombico 2016; Domergue et al. 2016; Bode et al. 2021). In the Iberian Peninsula, examples of wealthy wine producers in the Tarraconensis province are known (Cervantes 2020), whilst the Guadalquivir valley, located in the Roman province of Baetica, became a significant supplier of olive oil (Blázquez Martínez 1980; Blázquez Martínez and Remesal Rodríguez 1983). In the southwestern end of Lusitania province (Algarve region), several kilns for amphorae manufacture have been recorded, which are closely related to local fish-processing factories, namely to the production of garum (a fish sauce) and salted fish. Also, the development of salt exploitation linked to fishing activities made the fish processing one of the most important industries in Lusitania during the Late Antiquity (Fabião 2009; Bombico 2015; Bernardes and Viegas 2016). From the 3rd century on, North African Roman provinces became important suppliers of wine, olive oil, and fish products, as evidenced by numerous African amphorae found, sometimes together with Lusitanian amphorae, in shipwrecks and inland sites (Parker 1992; Bonifay 2004; Slim et al. 2007; Bombico 2016). Moreover, the products stored in these containers or their provenance could sometimes be identified from lead labels fixed to the amphora handle (Lequément 1975; Slim et al. 2007, Bonifay 2021). As mentioned by Lequément (1975), these lead labels wrapped around handles are strongly associated to African amphorae, as evidenced in Annaba Roman shipwreck—on the contrary, the lead plaque from Pampelonne wreck found inside one amphora was probably just a fishing net weight (Lequément 1976). The lead labels from the Anaba wreck presented some inscriptions mentioning the officina or ex-officina, whilst others bear just iconographic motives with no epigraphic inscriptions (Lequément 1975).

The Latin officina word has actually some polysemy, as it can be used both for potteries (e.g. in stamps from Hispanic Sigillata or Roman African lamps, Bonifay 2004) and for fish sauce production units, as one can see in the well-known mosaic at the house of Aulus Umbricius Scaurus at Pompei (Etienne and Mayet 1998). In the case of the lead amphora labels, the hypothesis that relates these officinae to fish-salted products factories (cetariae) seems more credible (Lequément 1975), as lead labels are removable allowing to identify the content production and distribution. The representation of a trident on some of these labels, also known from those found at Portmán, near Cartagena in the Spanish coast (Quevedo and Fernández-Diaz 2020), reinforced the idea of a relation with fisheries and so with fish-salted products.

Roman lead amphora labels are scarce in the archaeological record, probably due to the easy reuse of this metal. The present work is the first analytical study, as far as we know, concerning such labels in use during Late Antiquity. It seems clear that the use of such labels was just a Roman African tradition, not frequent and with a short time lap, as suggested by Lequément (1975), despite different comments by Quevedo and Fernández-Diaz (2020) in a recent publication of labels from Portmán. On chronology, the lead labels from Portimão have no actual relevance, as we are dealing with artefacts without specific context. On probable origin, the study can give important answers, even bearing in mind all the possible recycling of the metal, as was also pointed by Quevedo and Fernández-Diaz (2020). So, the study of the lead artifact collection uncovered by dredging works at the mouth of the Arade River, Portimão (Portugal), constitutes the aim of this research. Portimão, located at the southern Portuguese coast of Algarve (southwestern Iberian Peninsula) in the Roman Lusitania province (Fig. 1), is often related to a certain Portus Hannibalis, a town mentioned by the Roman Baetican Geographer Pomponius Mela (mid I century AD) in his Chorography (3.7), but actually no signs of that an ancient town has ever been found.

At Portimão, some Roman fish salted factories (cetariae) are known since the late XIX century (Veiga 1905) and others recently excavated, but not yet published. The presence of these cetariae and several other Roman sites in the area strongly suggests that the Arade River estuary played an important role in ancient cultural and commercial exchanges between Mediterranean and Atlantic regions (Teichner 1995, 1997; Tavares da Silva et al. 1987; Diogo et al. 2000; Fonseca 2015; Fonseca et al. 2018). The Arade River was also a relevant path to access the rich hinterland where several ancient sites are also known, namely Cerro da Rocha Branca (Silves) or the small island, Ilhéu do Rosário, with a large diachronic occupation, from the Iron Age to Late Antiquity (Gomes and Beirão 1986; Gomes 1993; Arruda 1999–2000). So, being the place of that Portus Hannibalis or not, Portimão should have had some port facilities during Late Antiquity and should have displayed an important commercial activity, evidenced by numerous archaeological underwater finds uncovered during modern dredging works.

The Roman artefact collection from the Portimão Museum collected during dredging works includes amphora fragments from many provenances, including Africa Proconsularis, widely used in fish product transport. The collection includes also miscellaneous metallic artefacts with special emphasis on Roman lead plaques, rectangular in shape and with variable sizes (33–129 mm length, 32–66 mm width, and few millimetres of thickness). Some of these plaques display characteristic motifs in relief, similar to those of amphorae handle labels from other Roman locations (Lequément 1975; Quevedo and Fernández-Diaz 2020), whilst others show incised inscriptions generally used in textile products (Hidalgo et al. 2016). There are also examples of lead plaques without decorative motives and, therefore, of unknown chronology, although the ones believed to be fishing net weights are quite similar to counterparts belonging to Roman lead collections from Algarve (Tavares da Silva et al. 1992).

The present study includes a typological characterisation of lead artefacts, and elemental and Pb isotope analysis was performed by accurate analytical techniques, Q-ICPMS and MC-ICPMS, respectively. Based on the historical and typological data and combining concentrations of trace and minor elements with Pb isotopic ratios, it was possible

Fig. 1 Geographical location of the Arade estuary (Portimão), Roman sites, and shipwrecks cited in text and the approximate location of the Ossa-Morena Zone (OMZ) and Iberian Pyrite Belt (IPB) in the southwestern Iberian Peninsula (map source: Freeworldmaps.net, adapted)

to identify probable lead sources and to establish trade routes of products transported in amphorae during the Late Antiquity.

Archaeological collection

In the present study, 37 lead plaques from Portimão Museum collections were analysed. Selected artefacts could be divided into different groups in compliance with presence or absence of decorations or inscriptions, namely labels with depiction of motifs in relief and labels with one perforation incised inscriptions (Table 1). Other types comprise simple undecorated artefacts, namely fishing net weights, smooth plaques of unknown functionality, and a possible small ingot (Table 2).

The first group is composed of 18 rectangular plaques with variable sizes (39–129 mm length, 28–67 mm width, and 40–199 g weight for those complete) containing depicted motifs in relief, including a trident composed of three vertical spikes, a fish, a wheat cob or palm leaf, and geometric rosettes (Fig. 2). In this particular set of artefacts, the iconographic motifs are depicted on just one side, with only one exception (2002/123, see Table 1) and most plaques show folding traces suggesting that would have been wrapped up crosswise around a cylindrical surface, an amphora handle, for instance. Dredging works also collected amphorae fragments of different typologies, which suggest the existence of Roman shipwrecks or, at least, an intensive trade in the estuary of the Arade River since the II century BC until the V century AD. All these finds suggest a frequent sailing activity including the transport of amphorae both from abroad, but also those produced in Algarve, particularly those made for fish-processed foodstuffs (Tavares da Silva et al. 1987; Diogo et al. 2000). Moreover, in the Portimão Museum, there are numerous examples of North African amphorae recovered at the Arade River. Not far from Portimão, the Roman villa of Montemar (Freitas and Almeida in press) and Vale da Arrancada (Fabião et al. 2016; Viegas 2019) show abundant North African ceramics dated to the II century AD until the first half of the VI century AD.

On the other hand, it must be noted that there is similarity of sizes and iconographic motifs of our lead plaques with Roman lead labels found in Annaba shipwreck and Portmán villa (Fig. 2) (Quevedo and Fernández-Diaz 2020). The labels from Annaba shipwreck show an iconography indicating a specific activity related with fish-processing factories. This fact is evidenced by a trident

Table 1 Lead artefacts with iconography recovered in Arade River, Portimão (dimensions in mm and weight in g)

Type	Reference	Description	Length	Width	Weight	LIA
Plaques with depiction of motifs in relief	K165	Depiction of an ear of corn or palm and a zigzag element associated to an X at one end	129	67	199	No
	K166	Depiction of two rosettes with petals inscribed in circles	105	52	93	Yes
	K168	Depiction of a fish	86	42	64	Yes
	K187	Depiction of an ear of corn or palm	106	38	65	Yes
	K188	Depiction of a trident	116	42	83	Yes
	K451	Depiction of an ear of corn or palm	76	51	87	Yes
	K452	Depiction of an ear of corn or palm	89	28	40	Yes
	K681	Depiction of a trident	105	47	82	Yes
	K682A	Undetermined depiction	69	-	77	No
	K682B	Depiction of an ear of corn or palm	94	57	107	No
	K683	Undetermined depiction	101	60	85	No
	K684	Depiction of a trident (with intentional perforation suggesting a reuse)	41	32	22	No
	K685	Depiction of a rosette with six petals inscribed in a circle and bounded by four small rosettes	73	52	81	No
	K686	Depiction an ear of corn or palm	64	31	26	Yes
	K687	Depiction of a trident	95	49	87	Yes
	2002/116	Depiction of a trident	92	53	76	No
	H190	Slightly rolled up plaque with an X in relief	39	33	24	No
	2002/123	Deformed plaque with yew branches randomly distributed in both sides	92	67	79	No
Plaques with one perforation and incised inscriptions	K280	Face a: VI XXI; Face b: IXIX	46	20	7	No
	K282	Face a: CVI; Face b: (…) X	32	16	3	Yes

LIA samples selected for MC-ICP-MS analysis regarding Pb isotope ratios

Table 2 Lead plaques without iconography recovered in Arade River, Portimão (dimensions in mm and weight in g)

Type	Reference	Description	Length	Width	Weight
Fishing net weights	G252	Rolled longitudinally (overlapping edges)	62	32	67
	H221	Rolled longitudinally (overlapping edges)	25	14	11
Unknown functionality	G229	Rectangular plaque	45	29	40
	G230	Rectangular trend plaque	45	29	37
	G231	Rectangular folded plaque	56	52	76
	G232	Rectangular plaque with an edge rolled up and one perforation at the top end	45	65	88
	G250	Rectangular plaque rolled up and folded at the edges	41	27	18
	G251	Rectangular plaque slightly rolled up	58	32	28
	G271	Rectangular plaque (fragment)	25	24	11
	G272	Rectangular folded plaque	17	18	14
	G273	Rectangular plaque rolled up with folded edges	11	13	3
	G343	Rectangular trended plaque with folded corners	71	41	21
	G362	Rectangular rolled up plaque	53	50	113
	G397	Rectangular trended plaque with rolled up edges	34	14	15
	H224	Rectangular trended plaque with two perforations (one containing a copper-based nail)	84	58	68
	2016-017/0018	Rectangular trended plaque	70	62	84
Possible ingot	2016-017/0031	Small rectangular prism	106	55	560

depiction, a usual symbol of the god Neptune often portrayed in Roman art, particularly in North African mosaics (Lequément 1975; Quevedo and Fernández-Diaz 2020). At the Roman villa of Portmán, two lead labels with a trident depiction and a high proportion of African ceramic containers were found. Also, at the Isla del Fraile (Águilas–Murcia, Spain), a late-African amphora labelled with a palm-shaped graffiti containing fish sauces was also recovered (Quevedo and Fernández-Diaz 2020).

The variety of depicted motives suggests different officinae origins, as one may think that these differences aim to indicated those different origins. Unfortunately, almost all are anepigraphic motives, as those from the Roman villa of Portmán and even those that have just one trident show a different image (Quevedo and Fernández-Diaz 2020). So, it is unknown if they came from the same officina represented by the trident or from different officinae using the trident as label mark. The only label with an epigraphic inscription is unfortunately distorted (K683), although the two-line inscription with a possible iconographic motif has some resemblance with an epigraphic label from Officina Libertorum showing a two-line inscription surrounding a palm leaf with crown (Lequément 1975). The distorted iconographic motif of K683 suggests the presence of the palm leaf, by one horizontal line between the two lines inscribed, and also some semicircular lines to the left. But this is not a definite interpretation; for sure, we just may say that the label features other usual North African Roman lead labels already published (Lequément 1975).

The second set is constituted by a different kind of label, in this case two well cut small rectangular plaques (32–46 mm length, 16–20 mm width, and 3–7 g weight, Fig. 3), with a circular perforation in one extremity, whose purpose was probably to pass a wire or a thread for the attachment of the identification label. Inscriptions are present in both sides of these labels (K280 and K282) and consist in fine and superficial incisions. The inscriptions show Roman numerals that probably reflect weight, amount, or a product price. This type of label is thought to be associated with textiles and fall into a very specific chronological period comprising the reign of Emperor Tiberius (14–37 AD) or shortly before (Hidalgo et al. 2016). Nevertheless, a recent study concerning a similar collection of incised labels recovered in a Roman pottery centre, close to the Portuguese coast at Peniche (Portuguese Estremadura), suggested that such artefacts could also be amphora labels, in this instance with a chronology from the 2nd or the beginning of the III century AD (Cardoso et al. 2018). Despite the context, the shape, dimension, and inscribed signs (just numerals) suggest other uses than labelling amphorae.

The third group is constituted, as mentioned before, by two fishing net weights (25–62 mm length, 14–32 mm width, and 11–67 g weight) rolled longitudinally with overlapping ends (Fig. 3—G252 and H221). This group includes also 14 plaques with distinct dimensions and without decoration or inscriptions. Some of them are deformed fragments, perhaps associated to the production of fishing net weights or even labels like those of

Fig. 2 Lead labels with motifs in relief recovered at the Arade River (Portimão) compared with those found in Annaba shipwreck (1) and at the Roman Portmán villa (2) (Quevedo and Fernández-Diaz 2020)

the first groups (Fig. 3—G231 and G232), but their precise functionality and chronology are unknown. The third group is completed by a rectangular prismatic artefact (2016-017/0031: 106 mm length, 55 mm width, and 560 g weight, Fig. 3) perhaps an ingot, although smaller in size than Roman lead ingots found in shipwrecks (Domergue et al. 2012, 2016).

Analytical methods

Sample preparation and artefact conservation

Due to the archaeological and museological relevance of those lead artefacts, a small amount (< 50 mg) was cut

Fig. 3 Lead labels with perforation and inscription (K280 and K282), fishing net weights (G252 and H221), examples of plaques of unknown functionality (G231 and G232) and the possible ingot (2016-017/0031)

on an area previously cleaned from corrosion products. After sampling, the affected area of artefacts was restored to avoid the increase of corrosion processes. The conservation treatment consisted in the application of a benzotriazole corrosion inhibitor (3% m/v in acetone) followed by a Paraloid B-72 acrylic polymer (10% m/v in acetone). Afterwards, a mixture of pigments dissolved in the acrylic polymer solution was applied to replicate the coloration of the corrosion products.

Each sample (40–50 mg) was transferred to a polypropylene tube, where it was dissolved with 25 mL of 20% HNO_3 solution and heated for 90 min at 35 °C in an ultrasonic bath. The dissolution volume of HNO_3 was adjusted according to the sample mass.

Elemental analysis

Analytical measurements and the sample preparation concerning a set of 37 lead artefacts mentioned above were performed in a clean room, Class 5. All material used in the laboratory was acid resistant, as TEFLON and PFA. Bi-distilled HNO_3 and ultra-pure water were used in sample dissolution and dilution, as well as in the preparation of standard and reference materials.

Each sample solution was diluted to be performed elemental analysis. Due to the wide variation range on the minor and trace elements contents, analytical measurements implied variable calibration ranges and dilutions. An aliquot of each sample solution was diluted (1:10) for the determination of elements present in trace amounts (Ni, Cu, As, Ag, Sn, Sb, and Bi). For elements present as minor contents, usually Sb and Sn, the sample solution was diluted (1:100 or 1:200). The certified reference material BCR-288 (lead containing added impurities) was used to quality control of measurements (Table 3). The analytical preparation used in reference material is the same procedure used in samples.

Limits of detection and quantification (LOD and LOQ—Table 3) were determined based on the low-range concentrations of the multi-element standard used for each external calibration curve, according to the Validation of Analytical Procedure Methodology Guidelines reported by the International Conference on Harmonisation Expert Working Group (ICH 1996). Further details are described in a previously published work (Gomes et al. 2016; 2018).

Instrumentation

Measurements for elemental analysis were carried out by inductively coupled plasma mass spectrometry with a

Table 3 Isotopes monitored, limits of detection (LOD), limits of quantification (LOQ), and BCR-288 (mean and standard deviation for five determinations) performed by Q-ICPMS (values in mg kg^{-1})

Isotope	^{60}Ni	^{63}Cu	^{75}As	^{107}Ag	^{118}Sn	^{123}Sb	^{209}Bi
LOD	1.04	0.72	1.03	0.34	0.62	2.16	0.32
LOQ	3.14	2.17	3.12	1.04	1.88	6.54	0.98
BCR 288							
Obtained results	5 ± 0.42	22 ± 2.07	66 ± 7.69	35 ± 4.59	30 ± 2.12	32 ± 2.26	207 ± 31
Certified	4.57	19.3	55.7	30.5	30.6	32.5	215.8

Quadrupole mass filter (ICP-QMS), ELAN DRC-e (Axial Field Technology) from PerkinElmer Sciex, provided with an automated sample introduction system, a concentric nebulizer and a cyclonic spray chamber. Sampler and skimmer cones are made of Ni. A complete description of this instrumentation and the operating conditions are detailed elsewhere (Gomes et al. 2018).

Samples were sent ready for Pb isotope analysis that were performed by a Thermo NEPTUNE MC-ICP-MS located at the Geochronology and Geochemistry SGIker Facility of the University of the Basque Country UPV/EHU (Spain).

Multivariate statistical analysis

Multivariate analysis of the elemental composition data set, considering seven variables (Ni, Cu, As, Ag, Sn, Sb, and Bi) and 37 samples, was carried out with STATISTICA (v.13.5.0.7) software package. The applied statistical method was factor analysis using a varimax rotation, the common orthogonal rotation method. Factor analysis is used to simplify data minimising number of variables that have high loadings on each factor and facilitating the interpretation (Marques de Sá et al. 2014). The hierarchical clustering was performed on the normalised data using the method of Ward, which analyses the variance in order to evaluate distances between clusters and identify groups with similar concentration patterns (Templ et al. 2008).

Pb isotope ratios

A total of ten samples were selected for lead isotope analysis: nine from rectangular plaques with iconographic depictions and one sample from a small plaque with a perforation and inscription (Table 1). The selection of samples was based on typological features, particularly artefacts displaying iconographic motifs. Samples previously dissolved in 20% HNO_3 were separated in aliquots of approximately 6 mL for further isotope ratio determination. Analyses were carried out at the Geochronology and Geochemistry SGIker Facility of the University of the Basque Country UPV/EHU (Spain). Mass fractionation of Pb isotopes was corrected with the addition to standards and unknowns of thallium isotopic reference material NBS-997 and using the ratio of $^{205}Tl/^{203}Tl = 2.3889$. Reference material NBS 981 Pb was analysed to measure the accuracy of the analytical procedure. The overall analytical procedure is detailed described in (Rodríguez et al. 2020).

Euclidean distances (*d*) of measured Pb isotopes in 10 plaques recovered from Arade River were determined with the lead mining regions Pb isotope database from the Iberian Peninsula and Mediterranean region (Birch et al. 2020).

Results

Elemental analysis

Results on quantification of minor and trace elements of lead samples are presented in Table 4. Variation ranges, mean values, and standard deviation of determined elements are displayed in Fig. 4.

All lead artefacts analysed contain Ni, As, Ag, and Bi in trace concentrations (< 1000 mg kg^{-1}), whilst Sb and Cu are also present as minor contents (≥ 1000 mg kg^{-1}) in some instances. Only two samples contain Cu as a trace element, namely K280 (1007 mg kg^{-1}; with an incised inscription) and K682B (1145 mg kg^{-1}; with an ear of corn or palm). Likewise, Sb is present in most samples with a trace content, with the exception of K682B, one of previous plaques, and G362, a plaque of unknown functionality. Ni is the trace element that is present in the lowest concentration, whilst Cu, Ag, and Bi were present in all samples. The sample G362 contains the highest content of Ag (218 mg kg^{-1}) and the lowest content of Bi (2.5 mg kg^{-1}). Moreover, it has an exceptionally higher Sb content (6601 mg kg^{-1}). With regard to Sn contents, the set of determined values presents a significant behaviour, dividing the collection among plaques without any kind of motif depictions or incisions (third group mentioned above) and plaques with depictions or inscriptions (first and second groups). A similar behaviour occurs with the As content since the third group is mostly composed of artefacts having the lowest As and Sn contents, whilst the first and second groups contain the highest values of these elements. Among these, the rectangular plaque with an X in relief, H190, has the highest As content (972 mg kg^{-1}, outlier in Fig. 4) and one of the uppermost values for Sn (3168 mg kg^{-1}).

Multivariate analysis

Factor and cluster analysis were performed using the elemental contents (Ni, Cu, As, Ag, Sn, Sb, and Bi) of 37 lead artefacts. Factor loadings obtained by factor analysis are presented in Table 5, showing that three extracted factors account for 65% of the total variance in the original data matrix. Factor 1 is related with the distribution of Ag and Sb contents, exhibiting a high positive correlation for Ag (0.775) and Sb (0.844). Factor 2 accounts for the As and Sn distribution, representing also a strong positive correlation for As (0.870) and Sn (0.816). Factor 1 and Factor 2 show a comparable importance, explaining 24% and 23% of the total variance, respectively. The dominant variables of Factor 3 are Ni and Bi contents, accounting for 18% of

Table 4 Results of minor and trace elements content of lead artefacts from the Arade River, Portimão, performed by Q-ICPMS

Group	Reference	Ni	Cu	As	Ag	Sn	Sb	Bi
1	K165	< 3.14	357	127	96	1364	32	35
	K166	3.5	219	39	154	1112	23	25
	K168	22	284	178	64	3329	99	21
	K187	< 3.14	422	111	94	391	32	17
	K188	< 3.14	115	111	80	3388	7.3	22
	K451	< 3.14	274	58	62	1748	10	17
	K452	4.7	328	78	110	4036	86	40
	K681	< 3.14	359	116	70	1760	141	41
	K682A	5.1	280	104	66	1476	40	53
	K682B	7.6	1145	524	55	1601	1765	63
	K685	< 3.14	312	42	120	1290	37	262
	K683	23	246	80	72	1290	33	16
	K684	3.14	282	302	73	4973	91	21
	K686	< 3.14	395	69	104	2442	25	23
	K687	< 3.14	282	42	63	2610	19	19
	2002/116	3.9	333	66	58	402	44	15
	H190	3.14	250	972	64	3168	49	13
	2002/123	n.d.	364	29	153	981	156	58
2	K280	8.6	1007	64	113	708	512	114
	K282	13	897	35	96	155	382	33
3	G252	n.d.	2.9	n.d.	7.7	n.d.	n.d.	53
	H221	3.5	402	31	81	125	352	58
	G229	37	59	n.d.	107	3.0	38	3.9
	G230	40	59	n.d.	108	3.1	39	3.1
	G231	< 3.14	206	34	80	n.d.	290	1.7
	G232	3.5	438	5.1	57	n.d.	525	49
	G250	n.d.	4.3	n.d.	14	n.d.	n.d.	166
	G251	8	376	n.d.	39	116	60	50
	G271	12	94	n.d.	62	n.d.	41	2.0
	G272	< 3.14	233	n.d.	43	42	44	178
	G273	< 3.14	124	9	39	n.d.	576	209
	G343	< 3.14	384	43	50	1089	388	517
	G362	5.1	298	20	218	89	6601	2.5
	G397	< 3.14	58	n.d.	169	112	8	1.8
	H224	10	245	n.d.	19	n.d.	< 6.54	13
	2016-017/0018	< 3.14	6.0	n.d.	18	2169	7.4	63
	2016-017/0031	4.29	295	n.d.	47	1695	59	146
Min–max		*n.d.–40*	*2.9–1145*	*n.d.–972*	*7.7–218*	*n.d.–4973*	*n.d.–6601*	*1.7–517*

Single determination expressed in mg kg^{-1}

1 plaques with depiction of motifs in relief; *2* plaques with perforation and incised inscriptions; *3* fishing net weights, unknown functionality, and "ingot"; *n.d.* not detected

the total variance, despite being highly negatively correlated (− 0.708 and 0.779, respectively).

Cluster analysis applying the Ward method and complete linkage (Euclidean distance) allowed the identification of groups of artefacts with similar composition (Fig. 5).

Dendrogram defines two main groups (A and B), which are as expected mostly distinguished by As and Sn contents, elements that were also found to be positively correlated in factor analysis (Factor 2). Group A contains samples with lower As and Sn contents (often below of the detection limit) namely the majority of plaques without any kind of depictions. Exceptions include two small plaques with perforation and incised inscriptions (K280 and K282) and two plaques with iconography in relief (K187 and 2002/116). There is also an outlier (G362, the only sample of sub-group A2) exhibiting the highest Ag and Sb contents (Factor 1). Group

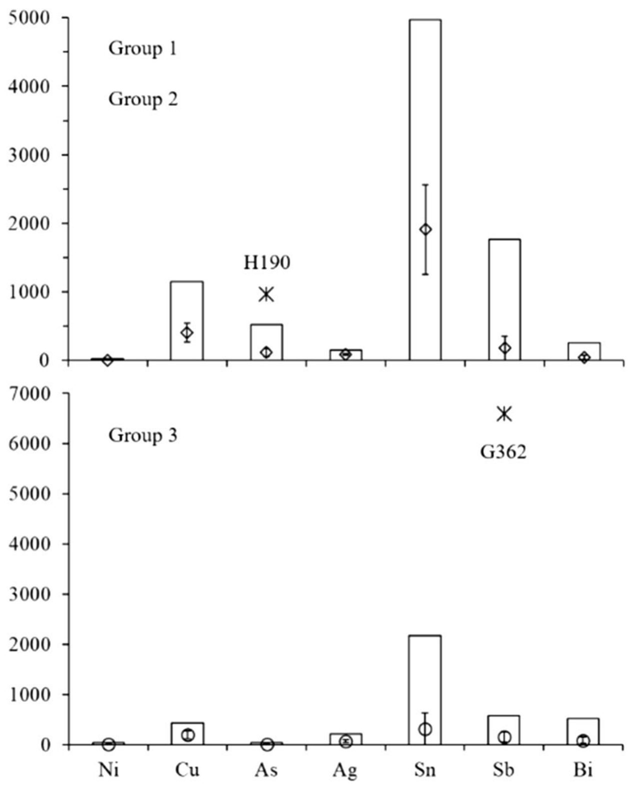

Fig. 4 Concentration range, mean values, and standard deviations obtained for minor and trace elements identified in lead artefacts recovered in the Arade River (Portimão, Portugal): Group 1, plaques with depiction of motifs in relief; Group 2, plaques with perforation and incised inscriptions; Group 3, fishing net weights, plaques of unknown functionality and a possible ingot

Table 5 Factor loadings of elemental contents determined in lead artefacts from the Arade River, Portimão (7 variables; 37 samples, method extraction: principal components; varimax normalised; bold: factor loadings with absolute value higher than 0.7)

Variable	Factor 1	Factor 2	Factor 3
Ni	− 0.033	− 0.189	**− 0.708**
Cu	0.497	0.320	0.363
As	0.073	**0.870**	0.037
Ag	**0.775**	− 0.055	− 0.302
Sn	− 0.147	**0.816**	− 0.026
Sb	**0.844**	− 0.107	0.049
Bi	− 0.150	− 0.259	**0.779**
Total variance (%)	24	23	18
Cumulative (%)	24	47	65

B includes the majority of rectangular plaques with motifs in relief, in addition to the small "ingot" (2016-017/0031) and two plaques without any kind of depiction (G343 and 2016-017/0018). These last three samples show high-Sn contents, compatible with a series of values determined for plaques with iconographic motifs.

Pb isotope analysis

The Pb isotope analysis was carried out in artefacts with decorative motifs, which were chosen to be representative of the different sub-groups obtained by hierarchical cluster analysis. Selected artefacts comprise 9 labels with iconographic reliefs and one small label with perforation and incised inscription. Analysed samples and NIST 981 lead isotopic ratios, as well as the respective standard deviations, are listed in Table 6.

Plot of Pb isotopic ratios determined for Roman label samples in diagrams $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ vs. $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ shows a linear distribution as seen in Fig. 6, where it is possible to identify distinct clusters. The main group is composed of samples K168, K187, K188, K451, K452, K681, K686, and K687 ($^{206}Pb/^{204}Pb$ 18.6481–18.7205, $^{207}Pb/^{204}Pb$ 15.6707–15.6785, and $^{208}Pb/^{204}Pb$ 38.7555–38.8243). Additionally, there are two samples displaying different Pb isotope ratios, namely the K685 and K282.

Discussion

Elemental results of lead artefacts demonstrate a compositional variability (Fig. 4) that highlights two large groups (Fig. 5) based on As and Sn contents (Table 5, Factor 2). Most plaques without motifs in relief including fishing net weights and two labels with incised inscriptions present low contents of As and Sn (Group A—Fig. 5), most of them close to detection limits (1.03 mg kg^{-1} and 0.62 mg kg^{-1}, respectively). On the contrary, all plaques with motifs in relief contain higher As contents (29–972 mg kg^{-1}) and variable Sn concentrations (most of them rather high: 981–4973 mg kg^{-1}), thus being included in Group B. Exceptions are the small "ingot" (2016-017/0031) and a plaque of unknown functionality (2016-017/0018) containing high-Sn contents (1695 and 2169 mg kg^{-1}, respectively), whilst As is below the detection limit.

Regarding other trace elements, it must be considered that, according to several authors, lead artefacts with Ag contents up to 100 mg kg^{-1} point to a raw material obtained by litharge reduction (Ham-Meert et al. 2019; Montero-Ruiz et al. 2008; Kuleff et al. 2006; Craddock 1995). For lead with Ag concentrations ranging from 100 to 400 mg kg^{-1}, the raw material seems to be associated with the smelting of silver-poor galena (Montero-Ruiz et al. 2008; 2009a; Kuleff et al. 2006). The high positive correlation between Ag and Sb (Factor 1) might indicate a probable association with silver-poor galena enriched with Sb as the probable lead source. Such correlation is well illustrated in dendrogram by sample G362, containing the highest Ag (218 mg kg^{-1}) and Sb (6601 mg kg^{-1}) contents. As an example, the use of

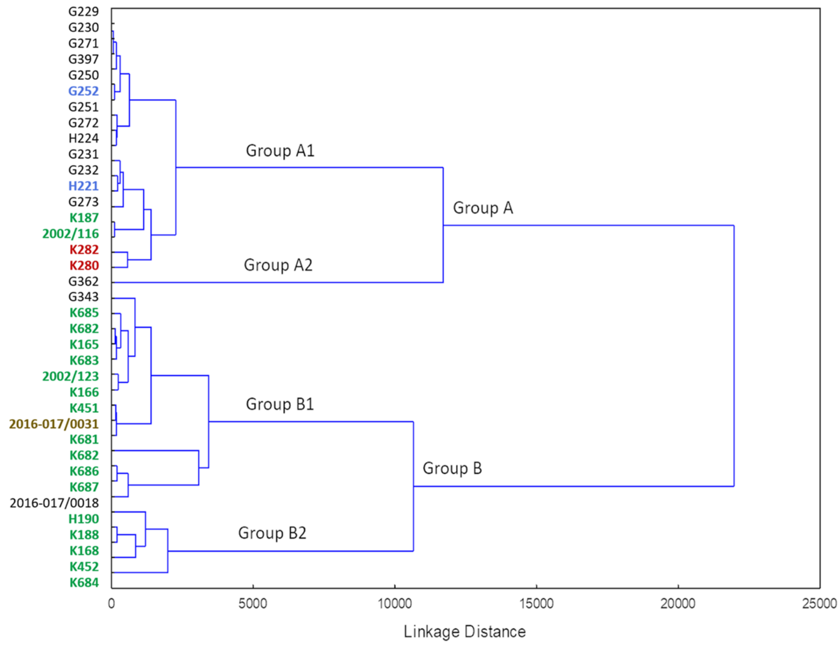

Fig. 5 Dendrogram of the hierarchical cluster analysis (Ward's method and Euclidean distances) for minor and trace elements of lead artefacts recovered in the Arade River (Portimão, Algarve) (black samples are plaques of unknown functionality, green samples are rectangular plaques with iconography in relief, red samples are small rectangular plaques with perforation and inscription, blue samples are fishing net weights, and the brown sample is a possible ingot)

Table 6 Results of Pb isotopic ratios $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$, $^{207}Pb/^{206}Pb$, and $^{208}Pb/^{206}Pb$ (± 2SE) of nine lead rectangular plaques with motifs depicted in relief and a small plaque with perforation and inscription from the Arade River, Portimão, performed by MC-ICP-MS (SGIker–UPV/EHU/ERDF, EU). Certified Reference Material NIST 981 was used as mass bias correction solution for isotope ratio analysis (mean and standard deviations for two determinations)

	Reference	$^{206}Pb/^{204}Pb$	2SE	$^{207}Pb/^{204}Pb$	2SE	$^{208}Pb/^{204}Pb$	2SE	$^{207}Pb/^{206}Pb$	2SE	$^{208}Pb/^{206}Pb$	2SE
Group	K168	18.6814	0.0010	15.6740	0.0009	38.7851	0.0023	0.83902	0.00001	2.07614	0.00004
	K187	18.7205	0.0008	15.6775	0.0008	38.8243	0.0020	0.83745	0.00001	2.07389	0.00004
	K188	18.7046	0.0009	15.6765	0.0009	38.8091	0.0023	0.83811	0.00001	2.07485	0.00005
	K451	18.7010	0.0008	15.6756	0.0007	38.8022	0.0019	0.83822	0.00001	2.07488	0.00004
	K452	18.6481	0.0009	15.6741	0.0008	38.7732	0.0023	0.84052	0.00001	2.07921	0.00005
	K681	18.6503	0.0009	15.6707	0.0008	38.7555	0.0022	0.84024	0.00001	2.07800	0.00005
	K686	18.7068	0.0009	15.6785	0.0009	38.8110	0.0023	0.83812	0.00001	2.07470	0.00004
	K687	18.7004	0.0010	15.6762	0.0009	38.8032	0.0026	0.83828	0.00001	2.07499	0.00005
	K685	18.4067	0.0009	15.6530	0.0008	38.5738	0.0021	0.85039	0.00001	2.09564	0.00004
	K282	18.1823	0.0008	15.6063	0.0008	38.4041	0.0022	0.85832	0.00001	2.11217	0.00005
	NIST 981	$^{206}Pb/^{204}Pb$	2SD	$^{207}Pb/^{204}Pb$	2SD	$^{208}Pb/^{204}Pb$	2SD	$^{207}Pb/^{206}Pb$	2SD	$^{208}Pb/^{206}Pb$	2SD
	$n = 2$	16.9439	0.0016	15.5013	0.0009	36.7289	0.0019	0.91486	0.00003	2.16768	0.00009

a silver-poor galena enriched in Sb was also suggested for two shapeless lead fragments from the Roman Republican site of Monte dos Castelinhos (Vila Franca de Xira) (Gomes et al. 2018). The high negative correlation between Ni and Bi (Factor 3) indicates a diverse lead source. Most samples present Ni in very low concentrations, often close to the limit of quantification (3.14 mg kg^{-1}), because lead ores do not usually contain measurable Ni concentrations (Kuleff et al. 2006). Besides, low-Bi concentrations in some samples suggest a lead produced from litharge reduction (desilvered lead), since Bi remains preferentially with Ag in molten lead during the silver cupellation process (Craddock 1995; Gale and Stos-Gale 1981), whilst Sn and Sb oxidise at the beginning of the reaction (L'Héritier et al. 2015). Sample G362 presents a different composition containing low Bi and the highest Ag e Sb contents suggesting, as mentioned above, lead obtained by reduction of silver-poor galena (Montero-Ruiz et al. 2008).

Concerning the Sn content an important question arises: what do these Sn contents in our samples mean? A high-Sn content (> 100 mg kg^{-1}) present in lead artefacts is usually attributed to the use of scrap lead containing leftovers of tin solders, i.e. recycled lead (Wyttenbach and Schubiger 1973; Asderaki and Rehren 2006). The lead recycling became very common with the decline of the Roman Empire, owing to the destruction of public and private structures and interruption

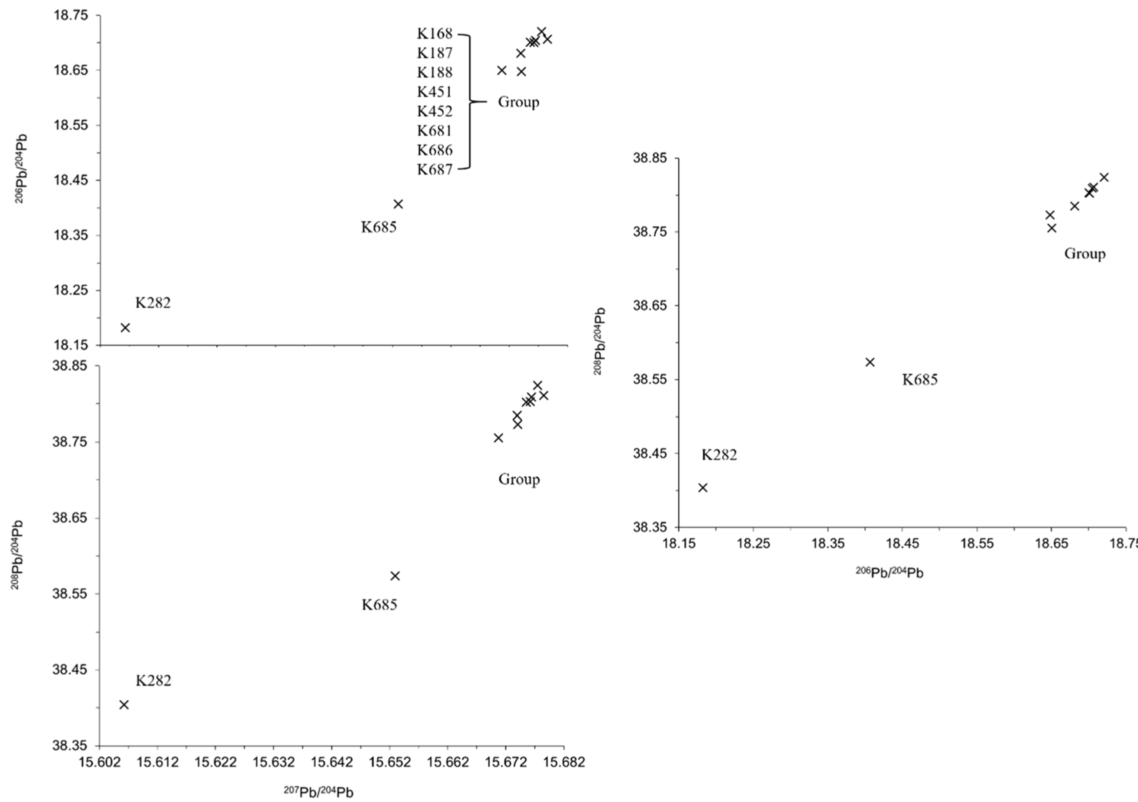

Fig. 6 Pb isotope diagram with ^{206}Pb/^{204}Pb and ^{208}Pb/^{204}Pb versus ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb versus ^{206}Pb/^{204}Pb for 10 Roman lead labels with iconography recovered in the Arade River (Portimão, Algarve). Analytical errors smaller than symbols

of the metal supply (Pernicka 2014). Emblematic examples are water pipes from aqueducts, public baths, and private houses, where the lead was actively sought and reused in huge amounts (Boni et al. 2000; Gomes et al. 2016). However, as mentioned previously, Sn contained in the initial Pb-Ag bullion oxidise at the beginning of the cupellation process and, consequently, the Sn content in the lead metal obtained from the reduction of litharge should not deviate too much from that existing in the Pb-Ag bullion (L'Héritier et al. 2015). Thereby, in some mining areas, namely at the Molar-Bellmunt-Falset mining district (Catalonian region), argentiferous galenas, and lead artefacts obtained from these ores, present high-Sn contents reaching 2000–4000 mg kg^{-1} (Montero-Ruiz et al. 2008). Another example comes from the Iberian Pyrite Belt characterised by polymetallic massive sulphide deposits. Due to the weathering suffered by these deposits over geologic time, various minerals and polymetallic mineral mixtures were formed, namely jarosites. Specific minerals such as argentiferous jarosites and plumbojarosites were widely exploited during the Roman Empire for silver production. These polymetallic complexes usually have Sn as a host metal or tin ores mixed with them (Craddock et al. 1987; Hunt-Ortiz 2003; Anguilano 2012). Palmer (1927) refers a sample of argentiferous lead found in a dump of silver slag containing 1.21% Sn, and Salkield (1987) mentions elemental analysis showing that jarosites from Rio Tinto mines have Sn contents ranging from 0.05 to 1.7%. As stated by L'Héritier et al. (2015) in the cupellation process "the trace elements contained in the initial lead bullion are separated according to their affinity with oxygen: noble elements tend to stay in the silver button, whereas elements which oxidise end up in litharge and therefore in the resulting metallic lead obtained after litharge resmelting". Other examples from RioTinto mines evidence a Sn content of 0.2% in refined lead (Craddock et al. 1985), or Sn ranging 0.5–4.3% in *speiss* from slag heaps (Craddock et al. 1987). A study by Anguilano (2012) concerning "The Roman lead silver smelting at RioTinto – the case study of Corta Lago" analysed lots of pre-Roman and Roman slags (tapped and plate slags) and two litharge samples (Roman Imperial period), whose results show SnO_2 contents up to 1000–4000 mg kg^{-1} in the slags, whilst the two litharge samples had a content of 400 mg kg^{-1} and 1700 mg kg^{-1}, respectively. In addition, the Sn content determined in prehistoric lead artefacts from Aegean show high Sn values ranging from 0.03 to 0.2% (Pernicka et al. 1982). On the other hand, it must be noted that the Sn content of silver-poor galena is usually very low, as happens, for instance, in the Braçal Mining complex (Central-Western Portugal) with average 50 mg kg^{-1} (Marques de Sá and Noronha 2011).

As a consequence, it becomes difficult to differentiate the origin of high-Sn contents in lead artefacts, as it could result from the recycling of scrap lead with a Pb-Sn solder or from ores with a high-Sn content. However, this distinction is important, if Pb isotopic ratios are used to determine the origin of lead used in the manufacture of the artefact. In any case, the *a priori* inference that lead samples with a Sn content above 100 mg kg^{-1} results from a recycled metal should be questioned.

As mentioned above, two distinct compositional groups (A and B) are probably associated to different sources of lead and/or to distinct metallurgical processes applied in the manufacture of these artefacts. A different chronology may be ascribed for the two groups, since the analysed artefacts come from the bottom of the estuary, where materials of different chronology can be found. Labels with depicted motifs in relief or with incised inscription are certainly ascribed to the Roman times, but the remaining lead artefacts can be attributed to a different chronology and/or to a different provenance.

Trace element contents suggest the use of lead from distinct sources (e.g. silver-poor galena and litharge reduction), whilst Pb isotopic ratios point to the use of lead ores with different geological ages (e.g. $^{206}Pb/^{204}Pb$ from 18.1823 to 18.7205, a high variation in first decimal place). Moreover, the Pb isotope ratios distribution of lead labels (Fig. 6) allowed the identification of a main Group having different isotopic signatures and two single samples with diverse isotopic ratios. The main group is composed of labels with distinct motifs in relief, such as a fish (K168), palms (K187, K451, K452, K686) and tridents (K188, K681, K687). The two isolated samples are the label K685 with a rosette and the small plaque K282 with a perforation and incised inscription.

In an attempt to determine a provenance of the analysed samples, Pb isotope signatures will be plot with published data of Pb isotopic ratios of probable lead sources in the Iberian Peninsula and Mediterranean regions. The Pb isotope database used in this study resulted in the data compilation covering lead ores from the Iberian Peninsula (García de Madinabeitia et al. 2021; Milot et al. 2021) and Mediterranean regions (Blichert-Toft et al. 2016; Killick et al. 2020). Euclidean distances were calculated according to the formula given by Birch and collaborators (2020) and are presented in additional supporting information S1 for Euclidean neighbours of each sample.

Concerning lead sources of the Iberian Peninsula, the Pb isotope signature from several mining districts was considered: (i) Cartagena-Mazarrón, which was the main supplier of lead and silver during the Roman Republic (Domergue 2008; Rodà de Llanza 2009); (ii) Ossa Morena Zone and Sierra Morena, two important areas during the beginning of the Roman Empire, when the lead and silver production declined in Cartagena region (Domergue 2008; Rodà de Llanza 2009); (iii) the Iberian Pyrite Belt, a major metallogenic province in Ancient Europe, composed of large massive sulphide bodies, where argentiferous jarosites from the gossan zone were mined during the Empire to obtain copper and silver (Domergue 2008; Delgado Dominguez 2006); (iv) Almeria, an important region during the end of Republic and the beginning of the I century AD (Domergue 2008; Arboledas Martinez 2010); (v) Catalonian Coast ranges (Montero-Ruiz et al. 2009b); (vi) other Portuguese mineralizations mined during the middle of the I century AD (Martins 2011); and (vii) Basque-Cantabrian basin and West-Asturian Leonese Zone (Blàquez Martinez 1989; Rodà de Llanza 2005). Diagrams show the comparison of Pb isotope signatures of Arade River samples with those lead sources having *d* values < 0.05 (Fig. 7).

Euclidean distances obtained for the majority of lead labels of the main Group point to the Betic Cordillera, namely some mining regions in Baleares (see Euclidean neighbours of K168, K188, K451, K686 and K687 in additional supporting information S1) and Almeria (idem for K187) as the possible source of raw materials. Exceptions are labels K452 and K681, which are mostly associated with sources from Basque-Cantabria basin. The sample K685 displays a different provenance showing the lowest Euclidian distances with the Pb isotopic field of Northern branch of the Iberian Massif. On the contrary, the different isotopic pattern of sample K282 matches the Pb isotopic field of Ossa Morena Zone, highlighted by the lowest determined Euclidean distance ($d = 0.005$).

Considering the archaeological evidence of important trade, particularly of salted fish-processed products (*garum*, for instance) between the Iberian Peninsula and other Roman provinces, namely those in North Africa during the Late Roman Empire (Lequément 1975; Tavares da Silva et al. 1992; Quevedo and Fernández-Diaz 2020), it seems also important to investigate the lead sources from those Mediterranean regions (Fig. 8). In this respect, Sardinia/Italy, Greece, France, and Turkey were important suppliers of lead and silver ores, besides other metals of economic interest, during the Roman Republic (Domergue 2008).

In general, samples of the main Group seem to be associated to mining regions in Greece, Italy, Turkey, France and Tunisia, which present comparable isotope patterns with low Euclidean distances to these samples (*d* between 0.002 and 0.050). The nearest Euclidean neighbours for sample K685 ($d = 0.007$) are ore sources from Greece, whilst Pb isotope ratios of sample K282 are indicative from Morocco ($d = 0.019$).

In an attempt to carry out an overall discussion, Table 7 summarizes all available data, including the typological

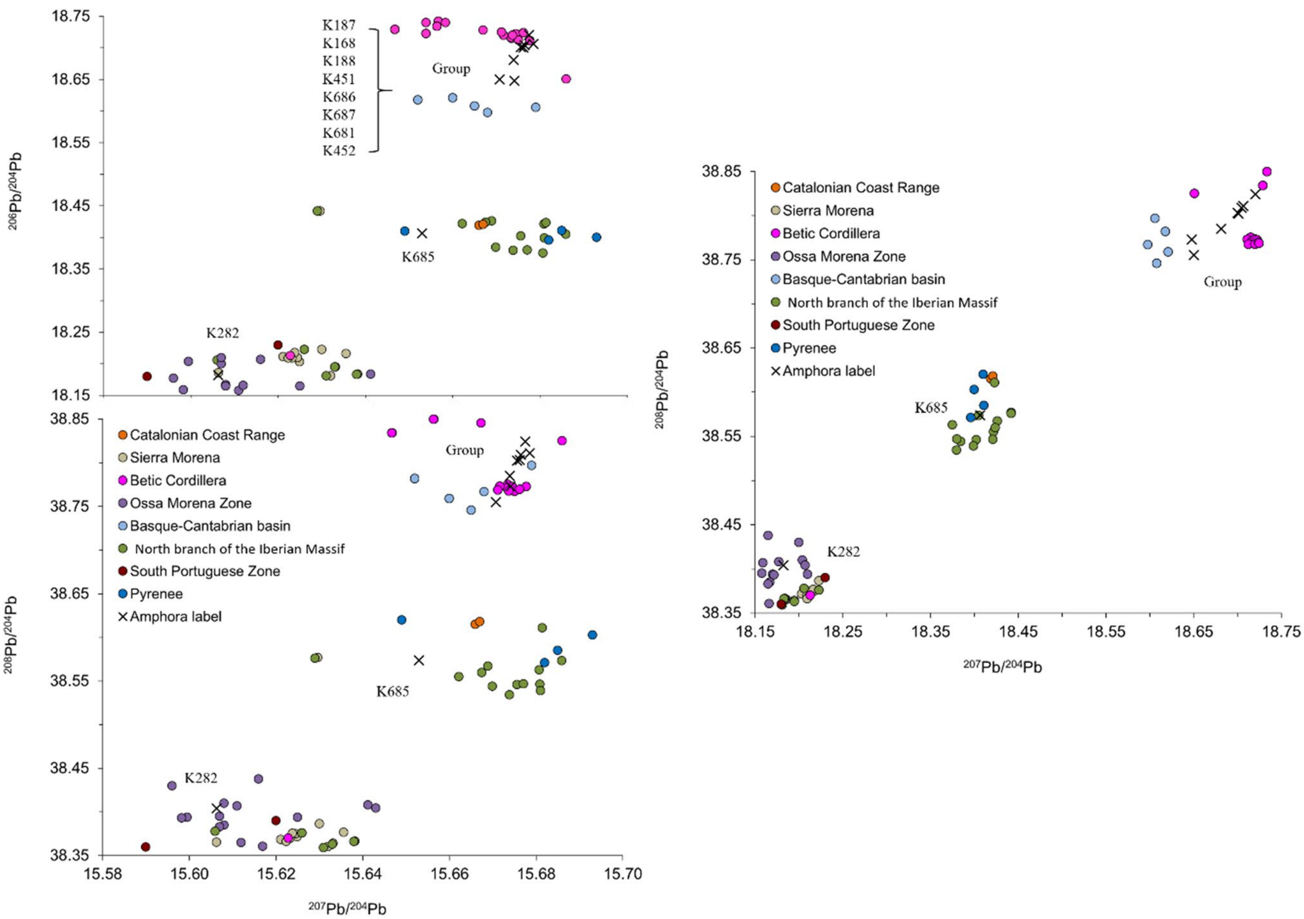

Fig. 7 Diagrams with $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$ for the Roman lead labels recovered in the Arade River compared with Euclidean neighbours from the Iberian Peninsula ($d < 0.05$)

classification of samples, in addition to Pb isotope ratios and minor/trace elements contents of ten lead labels from the Arade River (Table 7).

Most samples of the main Group suggest that lead originated from the litharge reduction (Ag ≤ 100 mg kg^{-1}), whilst sample K452 could also be the result from the smelting of a silver-poor galena (100 mg kg^{-1} < Ag < 400 mg kg^{-1}). The probable provenance of these lead samples indicates several sources in the Mediterranean region, although being difficult to attribute a specific source due to the high similarity of the Pb isotopic pattern of those regions. However, it is important to note that marine motifs in plaques from the Arade River are very similar to the ones displayed by lead labels from a Roman shipwreck discovered near Annaba, Algeria (Quevedo and Fernández-Diaz 2020). Moreover, some authors consider that salted fish products were an important foodstuff exported from African regions controlled by the Romans during the Late Empire (Lequément 1975; Slim et al. 2007). The importance of these resources to the Roman African economy is demonstrated by numerous mosaics showing fishing-related sceneries, a local reality of the Roman African coastline (Slim et al. 2007). Therefore, the presence of Tunisia among the nearest Euclidean neighbours ($d < 0.02$) of most samples from the main Group (note that Tunisia is always present among Euclidean neighbours with $d < 0.05$, see additional supporting information S1) strongly suggests this region as the source of lead used in those labels, specially from the Nappe and Domes Zones.

The label K685 with a rosette suggests a lead produced by the smelting of silver-poor galena or a litharge reduction also with probable provenance from Mediterranean regions, such as France and Greece. The small plaque K282 with a perforation and incised inscription indicates a lead produced by litharge reduction. This different typology agrees isotopically with an argentiferous galena from Azuaga mine ($d = 0.005$, see additional supporting information S1) in Ossa Morena Zone (Iberian Peninsula), which is characterised by massive sulphides enriched in galena (Tornos and Chiaradia 2004; Santos Zalduegui et al. 2007; Milot et al. 2021). Therefore, this label probably used in textile products seems to be an exception with lead provenance from the Iberian Peninsula.

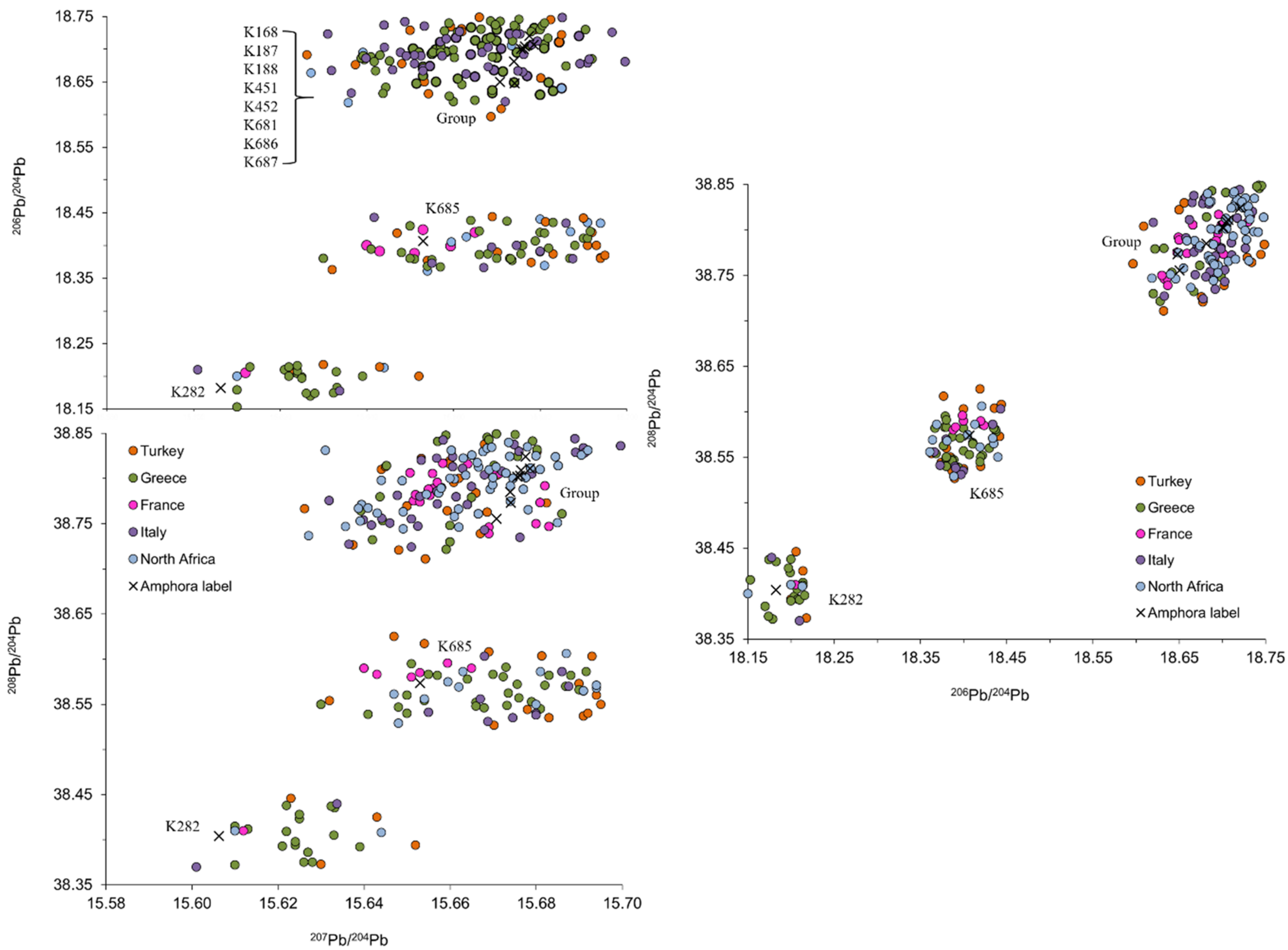

Fig. 8 Diagrams with $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ versus $^{206}Pb/^{204}Pb$ for the Roman lead labels recovered in the Arade River compared with Euclidean neighbours of lead ores sources from Italy, Greece, France, North Africa and Turkey ($d < 0.05$)

Conclusion

The study concerning lead plaques and similar artefacts recovered in the Arade River estuary allowed a new insight on trade routes and cultural exchanges during the Late Antiquity in the southwestern Iberian Peninsula. The combination of archaeological and analytical data was invaluable to overcome the overlap of Pb isotope signatures from distinct areas in the Mediterranean region.

The lead artifact collection is composed of 37 samples in which a chemical pattern was divided into two main groups mainly due to Sn and As contents. In the first set, 18 Roman labels are included, 16 being related to amphorae used as containers for fish-processed products and two small labels associated to textile products. The typology and motifs depicted in relief on most labels present high resemblances with Roman North African labels. Also, the incised Roman numerals in two artefacts are testimonies of the Roman world, surely ascribed to the Roman Empire period. The remaining set is composed of 17 undecorated plaques of unknown functionality, with the exception of two fish-net weights and a possible ingot. A chronology cannot be ascribed to this set, since the artifact typology is recurrent from Antiquity until the present day.

High-Sn contents (Sn > 100 mg kg^{-1}) in lead samples have been usually associated, in the archaeological literature, to the use of recycled lead having remnants of tin solder. However, lead raw material with high-Sn content may also have its origin in argentiferous galena from some mining deposits or in the use of complex lead-silver ores containing tin, as it happens with jarosites of the Iberian Pyrite Belt. Thus, further studies on the distribution of trace elements in lead from the different manufacturing processes would give a more accurate answer.

The distribution of Pb isotope ratios of Roman lead labels indicates distinct probable sources of raw materials. Assuming that the raw material does not result from mixing of different sources, Pb isotopic signatures of

Table 7 Iconographic features, probable raw material, and nearest Euclidean neighbours ($d < 0.02$) of lead labels from Portimão Museum (Portugal) recovered in dredging programs in the Arade River from the Iberian Peninsula and other Mediterranean regions mentioned in text (number of samples)

Inventory	Iconographic features	Raw material	Euclidean neighbours
K168	Fish	Litharge	Greece (2) Italy (1)
K187	Ear of corn or palm	Litharge	Tunisia (5) Greece (3) Italy (3) Turkey (2)
K188	Trident	Litharge	Greece (4) Tunisia (1) Turkey (1)
K451	Ear of corn or palm	Litharge	Greece (3) Italy (1) Tunisia (1) Turkey (1)
K452	Ear of corn or palm	Litharge/silver-poor galena	Italy (2)
K681	Trident	Litharge	Italy (3) France (1) Turkey (1)
K686	Ear of corn or palm	Litharge	Greece (4) Tunisia (1)
K687	Trident	Litharge	Greece (3) Tunisia (1) Italy (1) Turkey (1)
K685	Rosette	Litharge/silver-poor galena	France (3) Greece (1)
K282	Roman numerals	Litharge	Ossa Morena Zone (3) Morocco (1)

eight amphora labels match isotopic fields of lead ores from Greece, Italy, Turkey, France, and Tunisia. The latter seems to be a more probable source of lead considering the iconographic and archaeological attribution made by comparison with known examples, i.e. all these labels seem to be associated with North African amphorae that carried fish products. Furthermore, the label with a perforation and incised Roman numerals points to the Ossa Morena Zone as a likely provenance, suggesting a trade of textile products within the Iberian Peninsula, whilst a lead label with an unusual inscription for North Africa showed a signature compatible with deposits from Greece and France.

Finally, the present work demonstrates that Pb isotope research backed up by trace element patterns can give important answers about ancient trade networks despite the known drawbacks of possible recycling of raw materials. In this particular case, the short period of use of this labelling technique would reduce the mixing of Pb from different sources, relating most of the studied labels with ores from Africa Proconsularis.

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Author contribution Susana Sousa Gomes contributed to the conceptualization, methodology, validation, formal analysis, investigation and first draft of the manuscript; Vera Teixeira de Freitas contributed to the archaeological work, investigation, artefacts drawing and resources; Carlos Fabião contributed to the archaeological investigation and resources; Pedro Valério contributed to investigation, formal analysis and design; António M. Monge Soares contributed to investigation and conceptualization and Maria Fátima Araújo contributed to conceptualization, validation, formal analysis, investigation and resources. All authors reviewed and approved the final manuscript.

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Data availability All relevant data are within the manuscript.

Declarations

Ethics approval Not applicable.

Competing interests No potential conflict of interest was reported by the authors.

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