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RESEARCH

Metallurgical technology and resources mobility in the El Argar culture: An archaeometallurgical study at Laderas del Castillo (Callosa de Segura, Alicante)

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Abstract
Metallurgy has been defined as a pivotal activity in understanding of the development of El Argar society. Nonetheless, comprehensive studies of extractive metallurgical processes based on archaeometallurgical analyses remain lacking. This article examines the production remains found at the El Argar site of Laderas del Castillo, documented from 2150 to 1950 cal BC, including samples of slag, crucibles, copper prills and artefacts. Laderas del Castillo emerges as a key site for understanding the technology and organization of metallurgical production in the El Argar world. There are few sites with archaeometric analysis of metallurgical remains, and the present case allows for an almost complete view of the entire metallurgical *chaîne opératoire* in this period. Microstructural analyses of smelting remains by SEM-EDS reveal a technological tradition that mirrors the previous Copper Age one and the direct exploitation of complex arsenical copper ores with occasional copper sulphides. Despite the existence of closer mineralizations, lead isotope analyses show the exploitation of various copper resources and sources far from the settlement (Linares, the Interior of the Baetic Cordilleras and Almagrera, about 300, 200 and 140 km respectively). These active exchange networks in Laderas del Castillo reflect the same pattern found in the rest of the El Argar territory, which is based on the intensive exploitation of mineralizations in the interior of the Baetics and the Southeast of the peninsula. Some of these sources have also been observed in Copper Age metallurgy, suggesting that the mobility and exchange networks operating in the southeast during the El Argar period were rooted in earlier archaeological phases, indicating a wide and complex exchange network in the region.

Keywords Metallurgical slag · Arsenical copper · SEM-EDS · ICP-MS · LIA · Copper ores · Prehistoric mining · Bronze age metallurgy · Southeast iberia

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Introduction

In prehistoric archaeology, if the well-known El Argar group (2250 – 1550 cal BC) from southeastern Iberia has stood out in any way, it has been due to the importance and variety of metallic objects, most of them discovered in funerary contexts (Lull 1983; Siret and Siret 1890). Over 1,000 tombs at the El Argar site in Almería (Fig. 1) have been pivotal in emphasizing the crucial role that copper, silver, and gold metallurgy played in the El Argar economy (Ibarz et al. 2023). These activities have also been linked to significant environmental impacts, including deforestation and the eventual decline of the El Argar society itself (Lull 1983).

The 1990s marked a turning point in understanding the El Argar metallurgical production. The research undertaken indicated a significant intensification in metallurgical production processes during the development of the El Argar society with respect to the Copper Age period (3100 – 2250 cal BC), focusing primarily on the production of ornaments and weapons (Montero 1993; 1994). Early lead isotope analyses of sites like Gatas, El Argar, La Bastida de Totana and Fuente Álamo (Stos-Gale 2001; Stos-Gale et al. 1999) began to trace the mineral sources, although initial geochemical characterization of many of the existing mineralizations in the Iberian Southeast were incomplete, challenging their correct attribution.

This situation improved with the launch of various research projects. Significant progress was made with the extensive composition analyses series by the Archaeometallurgy of the Iberian Peninsula Project (Rovira and Montero Ruiz 2018) and the Peñalosa Project (Contreras Cortés 2000), which revealed extensive mining activities in Sierra Morena during the El Argar period. Settlements and forts were established to exploit open pit mines, and lead isotope analyses confirmed that copper ore was transported for smelting at sites like Peñalosa (Arboledas Martínez et al. 2008; Moreno Onorato 2000). Peñalosa became a metallurgical settlement due to its proximity to the mines and the abundant remains of metallurgical production documented at the site. From there, the metal would be distributed, including already manufactured objects, to other areas both inside and beyond the El Argar territory. Recent studies estimate that 20% of analyzed metal in El Argar areas originated from Sierra Morena, a similar proportion to that of other mineralisations in the El Argar territory (the interior of the Betic mountain ranges or the mineralisations of the coastal areas of Almería and Murcia) and even from outside the El Argar territory itself. The arrival of metal from other mining areas such as Los Pedroches, the SW or the NE of the Iberian peninsula has also been identified (Murillo-Barroso et al. 2015, 2025). These studies reveal the variability and diversity of resources in exploitation, their wide distribution and circulation, and the extensive exchange networks that were operating during the El Argar period.

In recent decades, archaeometallurgical studies have broadened, incorporating various lines of research. A series of all types of objects is already available; a large battery of compositional analyses has been conducted as well as a better isotopic characterization of the mineralizations and an outstanding range of lead isotope analysis. However, comprehensive studies of metallurgical processes in various

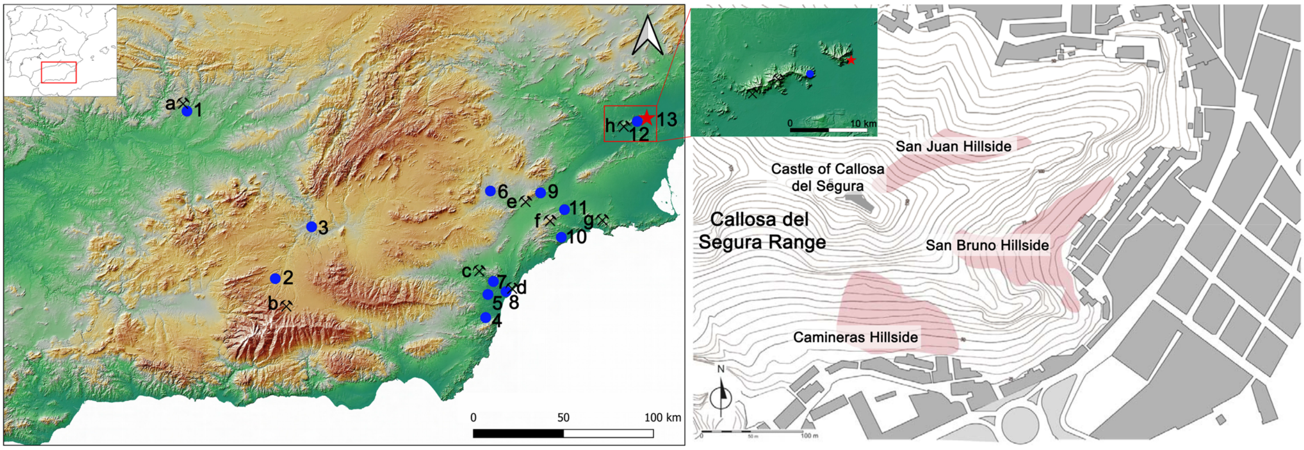

Fig. 1 Left: Location of Laderas del Castillo (red star) and the archaeological sites and mines mentioned in the text. Settlements: 1. Peñalosa, 2. Cuesta del Negro, 3. Terrera del Reloj, 4. Gatas, 5. El Argar, 6. Agua Amarga, 7. Fuente Álamo, 8. Almizaraque, 9. La Bastida, 10. Parazuelos, 11. La Ceñuela, 12. San Antón, 13. Laderas del Castillo. Mines: (a) José Martín Palacios (Linares district), (b) Santa Constanza (Baetic Cordillera- Granada), (c) Cerro Minado, (d) Sierra Almagrera, (e) Sierra Tercia, (f) Sierra Almenara, (g) Cartagena/Mazarrón, (h) Santomera. Right: Location of Laderas del Castillo on the Callosa de Segura Range. The site is extended over the three hillsides. Recent excavations were conducted on the San Bruno hillside

settlements, especially those distant from major copper sources, remain lacking. The traditional greater interest in finished objects, what could be sold to Museums all over the world, and the indifference to the production remains means that many of the crucibles and slags recovered in ancient excavations are lost today and little attention was paid to metallurgical remains from further excavations due to the weaker tradition of analytical studies on slags and crucibles in Iberia, leaving gaps in our understanding of smelting and melting processes in the El Argar society, for which we only have the preliminary study of some slag from Peñalosa and the analysis of a crucible from El Argar (Mongiatti and Montero-Ruiz 2019; Moreno Onorato et al. 2010; 2017).

In this sense, this article aims to fill some of these gaps through an archaeometallurgical analysis of an important set of evidence from the El Argar site of Laderas del Castillo. The findings contribute to resolving existing questions and pose new ones for future research to address.

The site of Laderas Del Castillo

The El Argar settlement of Laderas del Castillo is located southwest of the city of Callosa de Segura, on one of the southern promontories of the Callosa mountain range (Fig. 1). Initial excavations, carried out in 1908, uncovered a wide variety of objects that the excavator associated with the grave goods of various types of poorly preserved tombs, including small "mounds", large ceramic urns and flagstone cists, denying the status of settlement to the site (Furgús 1937). Further work by Josep Colominas between 1924 and 1925 also revealed numerous graves, though these excavations left minimal documentary record (Colominas 1936).

Systematic excavations at the site did not resume until 2012, as part of a research project on the El Argar communities in southern Alicante, supported by the Provincial Council of Alicante through the Museum of Archaeology (MARQ). From 2013 to 2021, annual excavation campaigns were conducted on the eastern slope, San Bruno hillside (Fig. 2). The excavated area showed dense occupation, starting at the end of the Copper Age and continuing throughout the entire El Argar sequence, from 2200 – 1550 cal BC, despite erosion on the steep slope has resulted in the loss of records from the most recent phases of the Bronze Age. These excavations revealed the typical El Argar settlement pattern: built at height, with rectangular houses on terraced areas and tombs located inside the houses. The settlement was reorganised through time and three different phases could be identified (see López-Padilla et al. 2024 for further information on the site).

Phase I (2300 – 2050 cal BC)

This phase began, in the late Copper Age period, with the construction of a large terracing wall and a sub-rectangular, almost square structure (CE-B2) with thick walls, possibly serving as an advanced bastion or buttress tower. An elongated building with an oval floor plan and clay walls on a stone plinth (CE-C1) was built atop this. The exterior featured an equidistant series of holes for wooden posts, with five preserved (Fig. 2). Similar buildings (CE-G, CE-IH and CE-J) were found to the south, all based on the geological substrate. A carbonized cereal seed dating (Beta 419057: 3920 ± 30 BP) suggests the possible existence of a previous Late Copper Age settlement which would be dismantled during El Argar constructions, explaining the sporadic

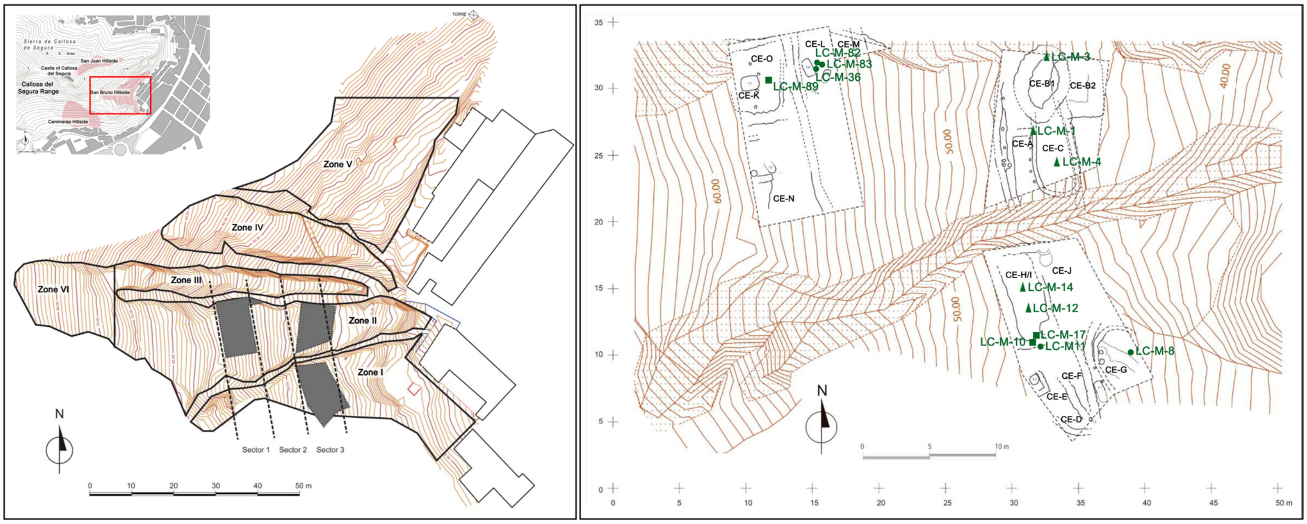

Fig. 2 Left: Sectors excavated at the San Bruno hillside. Right: Structures recovered and location of metallurgical debris

bell-beaker sherds found among terrace fills. In sector 1, Phase I is evidenced in two buildings (CE-M and CE-O) whose walls and floors rest on the rock. In sector 3, at the end of Phase I (2150 – 2050 cal BC), a second tower (CE-B1) with a pseudo-circular plan and an associated house were built on collapsed structures (CE-C2).

Phase II (2050 – 1800 cal BC)

This phase appears to start with extensive settlement remodeling, as well as new terraces walls and buildings. Building CE-A, with masonry walls and numerous internal post wedges, was constructed on CE-B1 and CE-C2. It featured a long continuous bench attached to the western wall. In sector 1, the CE-KN building (approximately 1950 – 1800 cal BC) shows a succession of pavements indicating continuous housing units over a long period of time, with floors reaching considerable lengths, and different areas of activity separated inside by means of clay benches and pavement projections.

Phase III (1800 – 1750 cal BC onwards)

Phase III would start around 1800 – 1750 cal BC, though no stratigraphic references or other evidence define its end due to site erosion. The latest contexts documented so far are tombs 1 and 5, at lower levels than the habitat contexts, dated by radiocarbon (Beta 649694: 3420 ± 30 BP and Beta 447232: 3400 ± 30 BP) to around 1700 – 1650 cal BC. It is currently considered probable that Laderas del Castillo was abandoned around 1575/1550 cal BC, consistent with other El Argar settlements.

The archaeometallurgical record of Laderas del Castillo

The archaeometallurgical record of Laderas del Castillo is extensive and diverse. Excavations conducted by Furgús documented metal ornaments, tools and weapons currently preserved at the MARQ in Alicante. These include a pair of gold rings, several silver spirals, three axes, two halberds and two daggers, all of which were found in graves. Additional items, such a saw, four arrowheads, a "spearhead", a large "hook", a "sword fragment", and an undetermined number of awls, were found in less precise contexts. The discovery of "a crucible of cooked earth" and "fragments of others", as well as a small amount of slag (Furgús 2007: 362) confirms metallurgical production activities at the settlement, although these items have been preserved. Colominas reported the discovery of a halberd, two knives or daggers and two awls, one with a bone handle (Colominas 1936: 39, Fig. 66). These items are now housed at the Museu d' Archeology of Catalonia (MAC) in Barcelona.

Over the last few decades, the site has continued to yield finds, resulting in a third collection of metal objects, hosted at the Municipal Archaeological Museum of Callosa de Segura. This collection includes three halberds, two knives and fragments of three others, three axes, three arrowheads, an awl and a sandstone casting mould, which may have been used to make halberds or ribbed knives.

Recent excavations have documented various metal objects and evidence of metallurgical production in domestic contexts, typically on the floors of dwellings or associated with landfills (Fig. 2). Metallurgical production remains are documented from Phase I, which include smelting remains (LC-M-8), a smelting crucible fragment (LC-M-89) and metal remains (LC-M-82 and LC-M -83). These production activities continued in Phase II, as evidenced by ceramic fragments with slag adherences (LC-M-17 and LC-M-10), slag fragments (LC-M-18) and metal remains (LC-M-11 and LC-M-36). The latest evidence of metallurgical production dates to Phase II, around 1950 cal BC, with the discovery of a smelting prill (LC-M-30) (Table 1). No evidence of metallurgical activity has been found in Phase III, although due to erosive processes this phase is the worst preserved in the excavated area. A complete overview of the archaeological contexts of the metallurgical record studied here can be found in López-Padilla et al. (2024).

Prior to our study, elemental composition analyses were carried out on several metallic objects from Laderas del Castillo (Junghans et al. 1960, 1968, 1974; Simón 1998). The Stuttgart team analysed four halberds and two daggers from the Museu Arqueológic de Catalunya (MAC), with inventory numbers 994 to 997, 1005 and 1006 in their analysis series (Junghans 1968). L. Simón García analysed a total of 23 pieces from Laderas del Castillo and nine of dubious origin between Laderas and San Antón de Orihuela, currently deposited at the MARQ, using SEM-EDS on a extracted sample (Simón 1998: 212–214). Several of the objects analysed by Simón (1998) using SEM-EDS (items with a suffix -S in Table 5) have been re-analysed by us using ICP-MS in order to get better accuracy, however, results obtained by Simón are also reported in Table 5 for comparison. Although the results are similar, the dissimilarities in the quantification of trace elements are due to the application of different analysis techniques and sample preparation.

Materials and methods

A set of 25 objects and metallurgical remains were selected for archaeometallurgical analysis (Table 1). Six objects found at the site before our excavations are included in this set, being three axes (LC-M-52, LC-M-53

Table 1 Materials analysed. Objects ID refers to excavation numbers (those starting with LC) or to Museum's ID (those starting with CS or SVI). Phases: I = 2300–2050 BC, II = 2050–1800 BC, III = 1800–1550 BC. (S) = analyses conducted by Simón (1998)

Analysis ID	Object ID	Type	Phase	SEM-EDS	ICP-MS	LIA	PIXE	pXRF
LC-M-3	LC'13 UE 31,000 INV 213 M 2	Awl	Surface			X	X	X
LC-M-6	LC'14 UE 31,500 INV 210	Awl	Surface			X		X
LC-M-8	LC'14 UE 31,519 INV 86 M 6	Smelting prill/slag	I	X		X		
LC-M-89	LC'20 UE 11,072 INV 124	Slagged pottery	I	X		X		
LC-M-82	LC'18 UE 11,026 INV 32	Metal fragments	I	X		X		
LC-M-83	LC'18 UE 11,026 INV 33	Metal fragment	I	X		X		X
LC-M-12	LC'15 UE 31,543 INV 35 M 7	Chisel	I		X	X		X
LC-M-1	LC'13 UE 31,029 INV 2	Arrowhead	I			X	X	X
LC-M-14	LC'15 UE 31,543 INV 36 M 8	Arrowhead	I			X		X
LC-M-4	LC'14 UE 31,048 INV 12 M 4	Awl	I		X			X
LC-M-10	LC'15 UE 31,529 INV 120 ZONA 1B	Slagged pottery	II	X		X		
LC-M-17	LC'15 UE 31,531 INV 89 ZONA 1B	Slagged pottery	II	X		X		
LC-M-18	LC'16 UE 11,001 INV 86	Slag frag.	II	X		X		
LC-M-36	LC'17 UE 11,026 INV 12	Smelting prill/slag	II	X		X		X
LC-M-11	LC'15 UE 31,529 INV 120 ZONA 1B	Metal fragments	II			X		
LC-M-30	LC'17 UE 11,017 INV 163	Smelting prill/slag	II			X		X
LC-M-63	SVI-174	Halberd	II		X	X		
LC-M-64	SVI-172	Halberd	II	X (S)	X	X		
LC-M-25	LC'17 UE 11,015 INV 202	Awl	II			X		X
LC-M-27	LC'17 UE 11,016 INV 55 M 10	Awl or hook	II			X		X
LC-M-29	LC'17 UE 11,016 INV 57 M 11	Awl	II			X		X
LC-M-66	SVI-175	Halberd	II	X (S)	X			
LC-M-52	CS 1540	Axe	III	X (S)	X	X		
LC-M-54	CS 1577	Axe	III	X (S)	X	X		
LC-M-50	CS 1546	Axe	III			X		X

and LC-M-54) and three halberds (LC-M-63, LC-M-64 and LC-M-66) (Fig. 3). SEM-EDS has been chosen for microstructural analyses of metallurgical remains, and other techniques with higher accuracy (ICP-MS, pXRF or PIXE) have been favored for elemental composition analysis of artefacts. Following a cost-effective strategy, and the evaluation of metal preservation, pXRF has been conducted in a larger set of artefacts (n. 13) while ICP-MS was further conducted on a selection of objects (n. 7). In three cases, sample extraction or patina removal was not possible due to conservation constraints and pXRF has been only qualitatively conducted on surface patina to identify if these objects had some As or Sn. PIXE was available thanks to the collaboration of one of us with the PIXE-Accelerador Grand Louvre Centre de Recherche et Restauration des Musées de France (C2RMF), and two samples were also analysed by PIXE in order to compare the accuracy of the techniques used. Lead isotope has been conducted for provenance analysis. Analytical conditions of each technique are as follow.

Microstructural analysis by Scanning Electron Microscopy was conducted on a set of 8 samples (6 smelting/melting debris and 2 metal fragments) (Table 1) which were mounted in epoxy resin and polished to 0.25 µm. Samples were examined by scanning electron microscopy with energy-dispersive spectrometry (SEM-EDS) using a high resolution Zeiss SUPRA40VP microscope of variable pressure at the Scientific Instrumentation Centre (CIC) of the University of Granada and a Hitachi S-3400n (Type II) one at the Institute of History-CSIC in Madrid. In both cases analyses were performed using an accelerating voltage of 20 kV, a spot size of 5.3, and a process time of 5, corresponding to a dead time of c. 30%; acquisition time was 60 s. Data was processed using EDAX Sprit (v.2.1) software by Bruker and INCA software by Oxford, outputting data as elements for metal phases, and adding oxygen by stoichiometry in ceramics and slag phases. Unless otherwise noted, results are normalised to 100% to facilitate comparisons, but analytical totals are reported to evaluate the precision of the equipments. Bulk analyses compositions are the average of between 3 and 5 area analyses (500 × 400 µm) trying to avoid large inclusions and corroded areas. In order to evaluate composition variability,

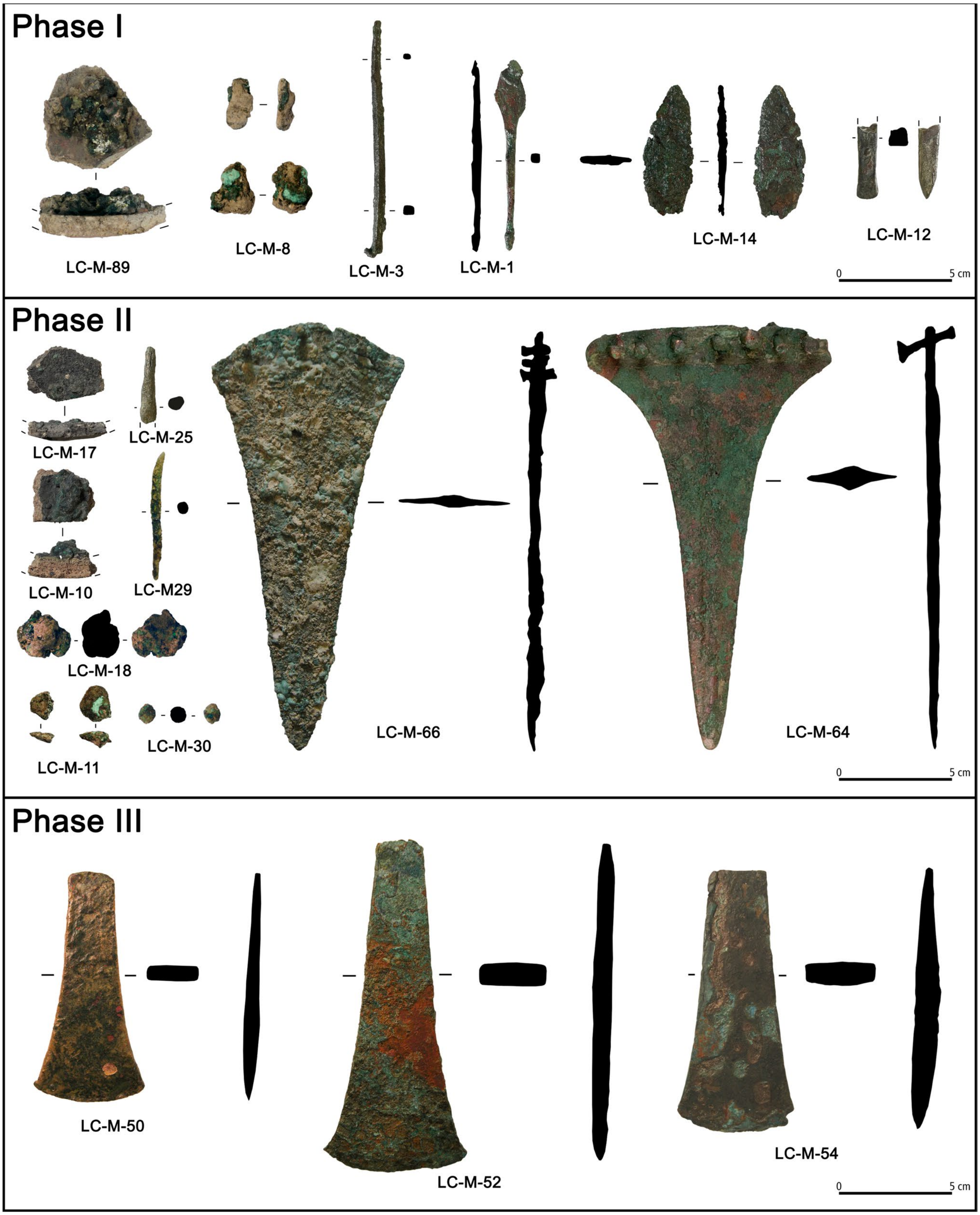

Fig. 3 Selection of materials analysed in this work

a selection of between 4 and 11 prills per sample were analysed under the same analytical conditions and variable magnification depending on the prills' size.

A set of 6 samples of smelting/melting debris, and 15 metal objects or fragments (Table 1) were selected for further lead isotope analysis (LIA), which was conducted at the Geochronology and Geochemistry Service (SGIker) of the University of the Basque Country (Spain) using a multi-collector inductively-coupled plasma - mass spectrometer (MC-ICP-MS) following the methodology described in Rodríguez et al. (2020). The reliability and reproducibility of the method were verified by regular measurements of the certified reference material NBS981 interspersed between the measurements of the samples, and in the same conditions. The average uncertainties (2SE) for the NBS981 during this study are 0.0004 for the 206Pb/204Pb ratio, 0.0005 for the 207Pb/204Pb, 0.0014 for the 208Pb/204Pb, 0.0001 for the 208Pb/206Pb and 0.00003 for the 207Pb/206Pb (NBS981 values used are those proposed by Baker et al. 2004). Uncertainties of measurements are given in Table 6, and are smaller than the symbols used in all graphs.

Seven samples from metallic objects were also removed for further trace element composition by ICP-MS at the SGIker-Geochronology and Isotope Geochemistry Facility of the University of the Basque Country UPV/EHU (Spain) (Table 1). The samples were dissolved with either aqua regia (for Sn; Hf, Sb and Au determinations) or concentrated HNO3 (the rest of elements) in closed Savillex PFA vessels set on a hot plate at 120 °C during 24 h. Dilution after dissolution is done gravimetrically using an electronic balance with precision of 0.1 mg in order to prevent errors induced by volumetric dilutions. Quantitative determination of analytes of interest was done by means of a Thermo XSeries 2 inductively coupled plasma mass spectrometer (ICP-MS) equipped with collision cell (CCT), an interphace specific for elevated total dissolved solids (Xt cones) and shielded torch. A concentric nebulizer and quartz expansion Peltier-cooled chamber were employed. Rh and In solution, used as internal standard, and multielemental solutions for the initial tuning and calibration of the mass spectrometer, and for quality control (QC) of the results were prepared from 10 ppm high-purity standard solutions for ICP, stabilized in diluted HNO3. Internal standard was added online with an automatic addition kit in order to prevent random errors. Further details on the instrumental method are given in García de Madinabeitia et al. (2008). The recoveries in % for the QC solutions are given in the Table of results. Error estimation for each element is established using the error propagation equation of Miller and Miller (2004). Uncertainty of the results corresponds to a 95% confidence level.

All lead isotope and elemental analysis samples were extracted using 0.5 mm high speed steel (HSS) drill bits. In some cases tungsten carbide tipped drill bits have been used. In general, the objects from the oldest phases presented a strong state of corrosion, especially the thinner ones, and the sampled areas for elemental analysis were cleaned as much as possible with the use of a binocular magnifying glass. However, in some cases the corrosion of the objects affected the core, preventing the analysis of healthy metal (for example LC-M-27). In these cases, trace element analyses were not carried out and their elemental composition was quantified after removing the patina in a small area by X-Ray Fluorescence using a portable spectrometer Niton XL3T-950 GOLD from the Antonio Arribas Palau Laboratory of Archaeometry of the Department of Prehistory and Archaeology of the University of Granada. The analytical mode 'General Metals' optimised for the analysis of archaeological metals and a collimate beam of 3 mm were used with an acquisition time of 60s. The arsenical copper standard 32XCAS3 was used to monitor the reliability of the analyses (Table 2). Data was only qualitatively considered to evaluate the presence of tin bronze or arsenical copper alloy in the cases in which the objects were heavily corroded and the patina was not removed.

Two samples (LC-M1 y LC-M3) were also analysed by Particle-induced X-ray emission at the PIXE-Accelerador Grand Louvre Centre de Recherche et Restauration des Musées de France (C2RMF) (Table 1). For PIXE analyses, samples from the metallic core were extracted and pressed to form compact pellets. Two filters were used during the analysis: a cobalt one of 25 µm for the HE1 line and an aluminum one of 200 µm for the HE3 and HE4 spectral lines. Calibration of the results was done using the certified materials CTIF-4, CTIF-5, CTIF-6 and DRN (see Escanilla 2016 for further methodological questions).

Table 2 Arsenical Copper standard 32XCAS3 used to control the reliability of the pXRF analyses performed

SAMPLE	Cu	As	Sn	Pb
32xCAS3	96.71	2.92	0.34	0.02
32xCAS3	96.70	2.92	0.34	0.01
32xCAS3	96.70	2.92	0.34	0.01
32xCAS3	96.70	2.92	0.34	0.02
32xCAS3	96.69	2.91	0.35	0.02
32xCAS3	96.69	2.92	0.35	0.01
32xCAS3	96.68	2.93	0.35	0.02
Average	**96.70**	**2.92**	**0.34**	**0.01**
Standard	**96.70**	**2.92**	**0.36**	
Abs. Deviation	**0.00**	**0.00**	**0.02**	**-0.01**

Results

Microstructural analyses of production remains

Of the eight samples selected for microstructural analysis, two samples turned out to be severely corroded metal remains (LC-M-82 and LC-M-83), two turned out to be copper prills resulting from a smelting process with some slag still adhered to them (LC-M-8 and LC-M-36), and the remaining four samples, loose slag remains (LC-M-18) or slag layers adhered to ceramic fragments (LC-M-10, LC-M-17 and LC -M-89).

Sample LC-M-83 is a completely corroded metal remain in which no metallic core is preserved and only 0.4% As is detected in the pXRF analysis. Sample LC-M-82 does preserve a metallic core of a rectangular section surrounded by a thick layer of secondary corrosion products (Fig. 4). This metal, with average levels of 3.8% As in area analysis in the SEM, and 4.48% As in the pXRF analysis, presents a granular microstructure with arsenic segregates at the grain boundaries, where the δ phase appears, reaching > 20% As (Fig. 4). Inclusions rich in Ag and Sb were also documented, reflecting the polymetallic composition of the ores used. Remarkably, these two samples were considered slag samples on a first macroscopic examination and could be considered as scrap metal possibly to be re-melted. LC-M-83 still had a charcoal piece adhered on its surface, and LC-M-82 was found with other small slag fragments.

Samples LC-M-36 and LC-M-8 are copper prills resulting from a smelting process with slag still adhered in some areas. Smaller copper prills and charcoal remains were identified in these slaggy areas (Fig. 5D), some of them reaching up to 14% As. Presence of chalcocite, sometimes occurring as rounded prills (Fig. 5B and C), suggests the use of sulphides in the smelting process.

Samples LC-M-10, LC-M-17, LC-M-18 and LC-M-89 are clear evidence of the smelting activity in the site. Slags have a bulk composition rich in SiO_2, Al_2O_3 and FeO with significantly high values of CaO ranging from 11.6 to 20.8% CaO. This can be the result of the high reactivity between the slag and the ceramic, which also present high lime contents (up to 25.3%) (Table 3). However, the ratio Si/Al is higher in the slag (up to 2.7%) than in the ceramic compositions (all of them at 1.3%) and therefore some additional contribution of silica in the charge may have come from quartz or other silicates from the ore gangue. FeO content in bulk compositions is comparatively lower than CaO (between 5 and 8%). The exception would be sample LC-M-10 with higher FeO (c. 22%) than CaO (c. 12%) values (Table 3). This high FeO values compared to the bulk ceramic composition (c. 8%) is probably reflecting the higher contribution of a ferrous ore gangue in sample LC-M-10. These bulk compositions would reflect the selection of two different ores: one of quartz and dolomitic or calcium rich gangues in samples LC-M-18 or LC-M-89, and a ferrous one in LC-M-10. Some levels of sulfur (up to 2%wt) are documented in bulk analyses of all slag samples, which together with the occurrence of chalcocite in sample LC-M-36 could reflect the presence of sulphidic ore in the charge.

High alkali contents (up to 8.1% and 8.5% in slag bulk and matrix compositions respectively) together with the high presence of lime and some potash in matrix compositions, may suggest the use of bone as fuel together with charcoal. However, bulk and matrix compositions of crucibles also show high lime content (≈ 20%) and thus part of the lime content in slag may have also come from the interaction with the ceramic paste.

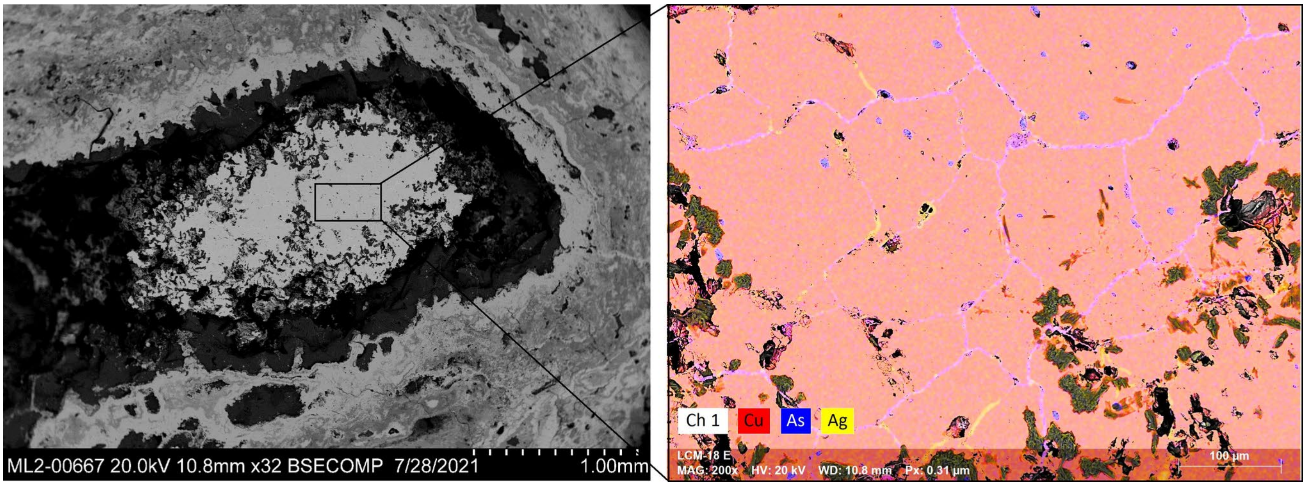

Fig. 4 SEM-BSE image of sample LC-M-82 where As and Ag segregates in grain boundaries can be observed (right)

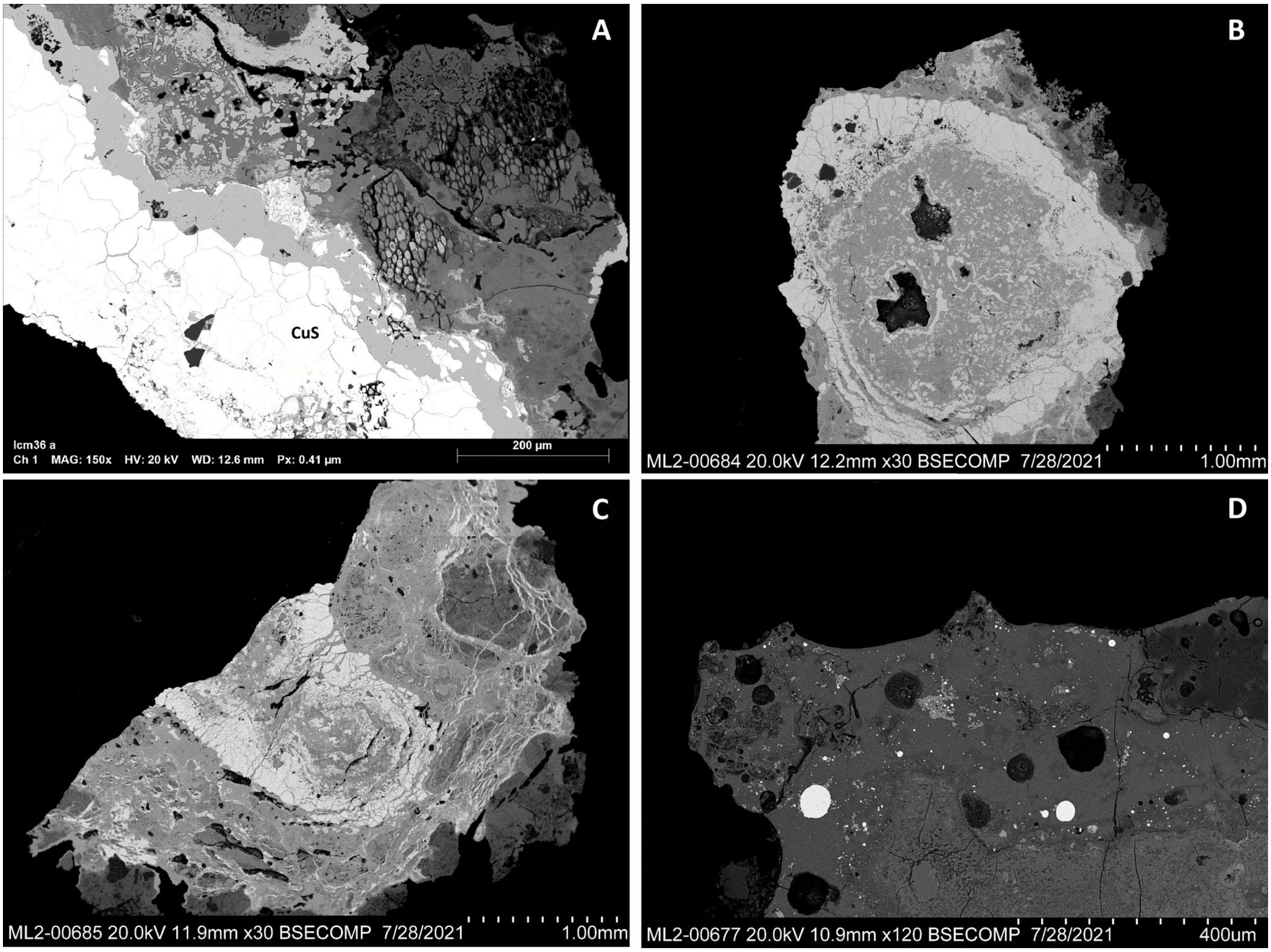

Fig. 5 SEM-BSE imagen of samples LC-M-36 and LC-M-8. A: A large chalcocite layer in LC-M-36a with slag and charcoal remains, B: rounded prill with slag remains still adhered to it (LC-M-36b), C: Prill with some slag and corrosion products (LC-M-36c), D: Sample LC-M-8: some copper prills trapped in slaggy areas adhered to a bigger prill (not in the image)

The bulk compositions of the ceramic crucibles are quite similar to that of their matrices, obtained by SEM-EDS area scans that avoided large inclusions. They show exceptionally high concentrations of alkali and alkaline earth oxides (averages of 26.8% and 30.3% respectively in bulk and matrix compositions) due to their high lime levels. These compositions highlight by their high lime content when compared with other prehistoric Iberian crucibles (Fig. 6), but resembles common and domestic pottery in that area (e.g. del Curbelo et al. 2024), and is consistent with the local high-lime clay resources (IGME 1987). These elemental compositions significantly differ from proper refractory ceramics documented from Medieval times which are characterized by high alumina and low alkali and alkali earth levels (Martinón-Torres and Rehren 2009). The particularly high alkali and alkali earth levels documented in the Laderas del Castillo crucibles would have diminished their thermal stability (Martinón-Torres and Rehren 2014), what indicates that specific clays were not selected for the manufacture of these metallurgical ceramics, their compositions reflecting local clay catchment. Moreover, clays reflect a thin granulometry, although the coarse grain size and abundance of quartz temper documented would have contributed to improve their performance (Fig. 7). This lack of refractoriness is the reason for the significant interaction between the slag and the molten ceramic in its inner surface documented in some of the sherds (Fig. 7). Yet, the physic-chemical properties of the crucibles used were enough to smelt copper, even if they were exposed to the limit of their thermal capability. In this, they are comparable to most of the prehistoric crucibles studied in Iberia, Europe and the Near East (Bayley and Rehren 2007; Murillo-Barroso et al. 2017).

Slag layers, some of them reaching up to > 5 mm thick (Fig. 7), are indicative of smelting activity. They show a greenish surface colour, denoting their copper-rich composition, and small charcoal remains and mineral relics

Table 3 ‘Bulk’ and matrix slag and ceramic compositions obtained by SEM-EDS. Average of up to 6 analyses per sample are reported. Areas analysed were selected trying to include all representative features and avoiding big inclusions, voids and corroded zones. Data are normalised in wt% and analytical totals are provided. Oxygen has been added by stoichiometry except for S and C. for which data obtained as elements is reported. Tr = traces (below 0.5%)

		Sample	Na2O	MgO	Al2O3	SiO2	P2O5	S	Cl	K2O	CaO	TiO2	FeO	CuO	As2O3	Total
Slag	Bulk	LC-M-10	< LOD	1.5	6.4	35.2	< LOD	tr	2.2	1.8	11.6	0.6	21.9	12.7	4.0	106.8
		LC-M-17	0.2	1.0	5.2	17.1	tr	tr	0.6	2.4	4.7	tr	28.2	45.2	8.2	102.3
		LC-M-18	3.0	2.5	10.6	28.8	< LOD	0.8	0.8	1.6	20.8	0.6	8.7	15.4	3.8	102.8
		LC-M-89	3.2	3.0	23.9	36.6	0.3	1.4	0.5	3.2	16.1	0.0	5.5	5.2	1.1	101.3
	matrix	LC-M-10	1.9	0.7	9.1	39.5	1.4	< LOD	< LOD	1.4	22.7	0.9	13.8	2.4	6.1	102.8
		LC-M-17	3.0	2.8	10.1	42.0	tr	< LOD	< LOD	5.4	14.3	tr	10.1	4.3	7.4	105.7
		LC-M-18	4.7	< LOD	12.8	33.7	1.9	< LOD	< LOD	2.7	20.4	1.3	15.2	2.5	4.2	103.5
		LC-M-89	4.9	< LOD	12.2	38.3	< LOD	< LOD	< LOD	2.5	11.0	1.3	8.7	13.3	7.6	96.2
Ceramic	Bulk	LC-M-10	2.4	3.3	28.1	37.0	tr	tr	tr	2.0	17.5	1.4	8.0	< LOD	< LOD	98.1
		LC-M-17	2.3	2.7	19.7	43.0	< LOD	1.1	tr	3.1	19.5	< LOD	6.5	0.5	< LOD	102.9
		LC-M-18	< LOD	3.9	28.2	35.9	0.9	tr	tr	2.9	25.3	tr	2.7	< LOD	< LOD	78.4
		LC-M-89	3.3	3.2	24.9	38.1	tr	1.1	tr	3.3	16.8	0.5	5.7	< LOD	< LOD	92.5
	matrix	LC-M-10	1.1	7.5	25.5	32.1	< LOD	< LOD	< LOD	1.1	21.6	< LOD	11.1	< LOD	< LOD	104.3
		LC-M-17	2.3	3.5	17.9	40.2	< LOD	2.1	< LOD	2.8	25.4	< LOD	5.5	< LOD	< LOD	101.95
		LC-M-18	1.8	3.8	23.2	37.7	< LOD	< LOD	< LOD	1.8	25.3	< LOD	6.3	< LOD	< LOD	98.7
		LC-M-89	< LOD	2.0	25.1	41.1	< LOD	< LOD	< LOD	0.4	20.8	< LOD	10.6	< LOD	< LOD	99.3

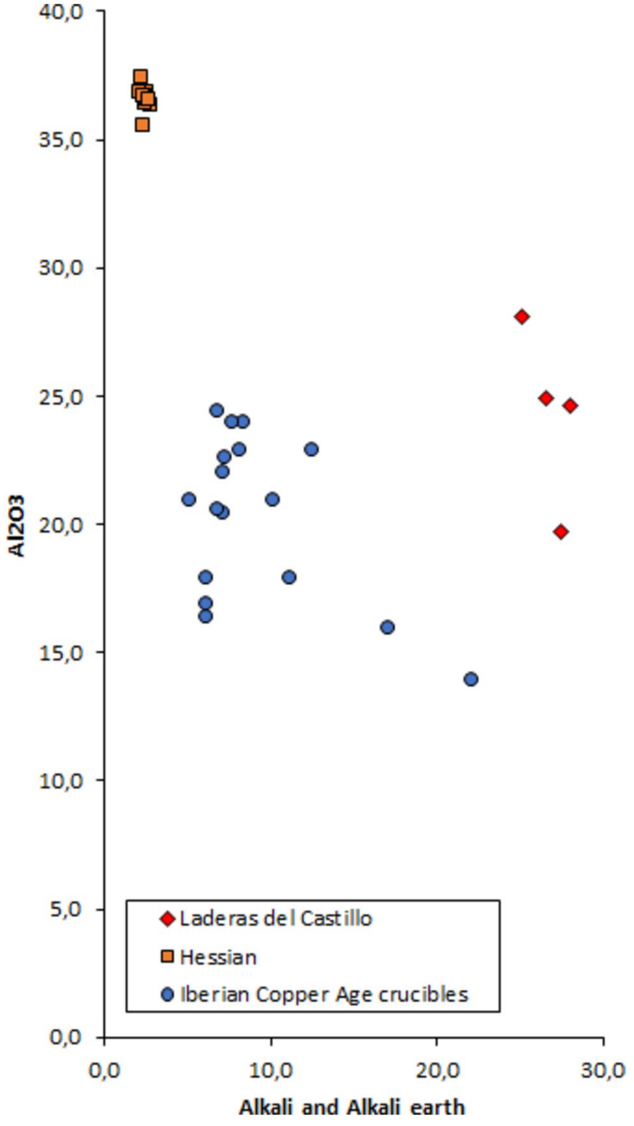

Fig. 6 Proportion of Al2O3 vs. alkali and alkali-earth materials in the Laderas del Castillo ceramic crucibles compared to other Iberian Copper Age crucibles and proper refractories Hessian ones

are also identifiable. Mineralogical composition of slag is mostly composed by magnetite (Fe_3O_4) and delafossite ($Cu^{1+}Fe^{3+}O_2$) as well as by a high presence of CuO and copper chloride possibly as a consequence of post-depositional weathering (Fig. 8). The high presence of CaO in the system also contributed to the formation of calcium-rich crystals as tabular and rhombohedral hedenbergite ($Ca^2Si_2O_6$) from the pyroxene group. This co-existence of metal oxides with silicates is consistent with a variable pO2 atmosphere with oxidizing conditions in which magnetite or delafossite could grow but reaching reducing enough gas atmospheres for silicates to develop and for metallic copper to precipitate and to retain high levels of arsenic (up to 25% As) (Table 4).

Chalcocite inclusions are frequently documented, mostly as layers on top of the crucible and filling ceramic cracks, what could be the consequence of post-depositional processes in an environment rich in sulfur, as the one of this site, with natural presence of gypsum. The fact that some S is also detected in ceramic bulk compositions and in the patina of metallic objects (see below), may be indicative of this post-depositional enrichment in sulfur. Nonetheless, the presence of a certain amount of sulphides in the charge, probably as impurities in the predominantly oxidic ores, or as a co-occurring sulphdic-oxidic ores cannot be completely discarded, as some levels of S are also detected in some of the metallic prills trapped in the slag matrix of LC_M_10 (up to 1.6% S) and some round chalcocite prills have also been documented (Fig. 5) However, presence of some sulfur in the system would have not contributed to improve the reducing conditions but on the contrary; it would have favored the oxidation due to sulfur's affinity for oxygen. It could have been the case of LC_M_89 where large masses of CuO are documented together with delafossite and magnetite. In this case, the CuO layer in close contact with the ceramic still contains some traces of S (up to 0.3%) while S is not detected in the more surface CuO layer (Fig. 9). Nonetheless, some gypsum entrapped in cracks is visible, and sulfur is not detected in copper prills of LC_M_89, therefore the presence of sulfur as a consequence of post-depositional contamination cannot be discarded (Fig. 9).

Heterogeneity in the copper ores used was probably not too high, as both mineralogy of the slag layers and copper prills compositions only present a limited amount of elements, that is S, Fe, Cu, As, and in the case of the slag Ca. Copper prills trapped in the slag matrix present variable arsenic contents reaching up to 25.1% As and one prill near to a speiss composition with 12.3% Fe, 52.9% Cu and 25.1% As (Table 4). This high variability on prills compositions is characteristic of primary arsenical copper smelting as evidenced both archaeological and experimentally (e.g. Hanning et al. 2010; Lackinger et al. 2024; Murillo-Barroso et al. 2017). No other impurities such as Ag, Sb, Co, Ni or Pb have been detected.

Elemental compositional analysis

Of the 18 objects analysed, all except the axe LC-M-50 are made of arsenical copper without the presence of tin. Tin does appear as an impurity in the samples analysed by ICP-MS, but in the two objects analysed by PIXE and most of the objects analysed by pXRF is below the detection limit (Table 5 and Supplementary Information 1). The axe LC-M-50 is made of tin-bronze with 5.72% Sn and a minor arsenic content (0.84% As). This object stands out as the only one with certain levels of silver (0.52% Ag).

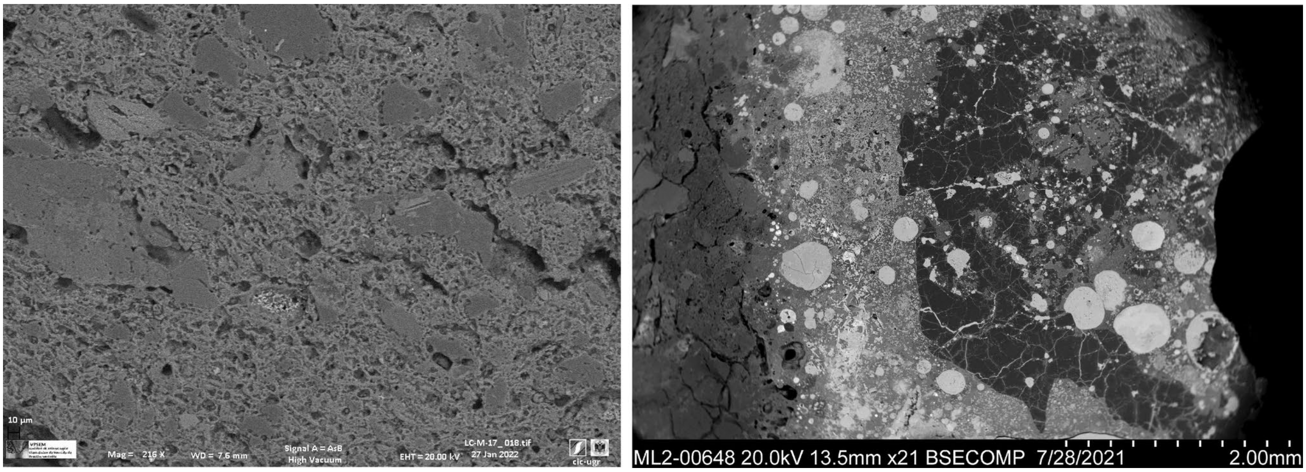

Fig. 7 Left: general view of the ceramic paste of crucible of LC-M-17. Right: slag layer of c. 5 mm showing high interaction with the ceramic paste of the crucible

Excluding the analyses carried out on patina, the average arsenic content of the set of arsenical coppers is 2.7%, with values ranging between 1.15% and 5.34%. The highest arsenic values occur in the awl LC-M-4, with 5.00% As, and in two of the three halberds analysed: LC-M-63 with 4.79% As and LC-M-66 with a 5.34% As, the highest value in the analysis series. The lowest arsenic values are found in one arrowhead (LC-M-1) and one awl (LC-M-6), with arsenic percentages below 1.5%.

Based on the ICP-MS results, the sum of metallic impurities typically ranges from 0.1 to 0.3% of the total metallic mass, with Fe being the dominant impurity, followed by Zn, Bi and to a lesser extent Ni. Co and Sb are present but their distribution is more irregular (Fig. 10). In the two axes analysed with this technique (LC-M-52 and LC-M-54), the higher Sn values stand out compared to the rest of the objects, especially in LC-M-52, with 0.16% Sn. Besides tin, LC-M-54 also presents the highest lead values in the series (0.04% Pb). As for Co and Sb, they are prominent in two of the halberds, but are otherwise anecdotal elements in the rest of the series analysed.

The higher detection limits of PIXE, SEM or pXRF compared to ICP-MS mean that, with the exception of iron, additional impurities observed in the ICP-MS series are not detected. Noteworthy is the presence of 0.09% Hg detected by PIXE in LC_M_3, as well as traces of S in some pieces (elements not analysed in the ICP-MS series), providing information on the use of ores with sulphidic minerals.

Lead isotope analysis

Lead isotope compositions of 4 slag samples, 3 smelting prills with some slag still adhered to them and 16 artefacts were also measured. Their proportions range from 18.267 to 18.815 in the ^{206}Pb/^{204}Pb ratio, 15.635–15.968 for ^{207}Pb/^{204}Pb and 38.360–39.328 for ^{208}Pb/^{204}Pb (Table 6). Regarding the type of artefacts, slag samples are grouped in three groups: attending the ^{206}Pb/^{204}Pb ratio, one sample presents values of 18.75, another one of 18.32, being these two the extreme values for slag samples and having another two values of 18.40–18.44 for ^{206}Pb/^{204}Pb (Fig. 11). This linear distribution might reflect the exploitation of three (or at least two) different ore sources at the site (see discussion below). Interestingly, two smelting prills and some of the artefacts plot in between these slag samples with a similar tendency (Fig. 11).

Discussion

Laderas del Castillo emerges as a key site for understanding the technology and organization of metallurgical production in the El Argar world, since there are few sites with archaeometric analysis of metallurgical remains. The site's in-situ metallurgical activity and the use of complex ores with high levels of As and potentially S, documented throughout its occupation period, are significant since local copper resources are scarce and appear to have been systematically ignored during recent prehistory.

Technological development at Laderas del Castillo, as evidenced by the microstructures of slags, mirrors the previous Copper Age metallurgical tradition (3200 – 2250 cal BC), which relied on open-air crucible metallurgy with minimal atmospheric control. This is indicated by the presence of crystallizations such as magnetite and delafossite alongside pyroxenes or metallic prills with high As contents, resulting in viscous and poorly liquefied slags. These are a result of direct ore smelting without flux addition, limiting

Table 4 SEM-EDS composition of copper prills trapped in slag matrices. Results in %wt elements, normalized to 100% with analytical totals indicated. Note high degree of oxidation in some of the prills

ID	O	Cl	S	Fe	Cu	As	Total
LC-M-8					92.7	7.3	98.5
					90.9	8.1	98.9
	7.8				92.2		97.3
	20.7			2.3	67.8	8.4	96.8
	21.0			2.7	68.4	6.7	96.1
	21.6			3.7	63.7	9.2	103.6
LC-M-10	1.3	1.0	0.6	2.6	79.1	15.4	80.6
	6.2	1.9	1.6	12.3	52.9	25.1	90.4
	1.3	1.1	0.6		95.6		81.0
	21.3	1.0		1.5	72.9		88.2
	20.4			2.3	74.2	3.0	92.6
	20.4			2.0	74.7	2.8	102.0
	21.0			2.0	70.2	5.9	101.0
LC-M-17	1.5			1.8	96.7		108.6
	1.3			4.4	92.8	1.5	102.4
	2.2			4.7	93.0		106.6
	10.3	0.2			89.4		103.9
	10.7	0.4		0.3	88.5		106.8
	13.0			1.4	82.4	3.0	108.6
	1.1			1.8	96.9	0.1	106.7
LC-M-18	3.1			1.0	80.1	10.0	94.2
	21.3			5.5	64.3	7.2	92.7
	32.9				55.6	7.1	107.0
	20.5			0.4	72.8	5.9	97.6
	21.5		0.1	1.1	55.3	21.5	96.4
	20.8			0.6	65.1	13.4	97.9
	20.9			1.4	64.3	12.6	95.1
	21.4			1.5	61.9	13.8	98.1
	20.8			0.7	67.1	10.9	96.3
	20.2			0.8	78.7		93.8
	20.9			1.0	65.5	12.2	96.2
LC-M-89	20.5			1.6	74.0	3.4	93.8
	20.6			1.8	70.9	3.0	96.0
	20.3			1.7	76.0	1.9	90.2
	10.1			0.5	89.4		97.4

metal/slag separation and causing significant metal loss in the slag (Müller et al. 2004; Murillo-Barroso et al. 2017; Rovira 2002).

The use of complex ores containing sulfur (of the *fahlore* type) is uncommon in the prehistoric metallurgy of the Iberian Peninsula, though some occasional samples indicate that oxide and sulfide minerals were directly smelted together. Examples include Almizaraque (Müller et al. 2004), Agua Amarga (Escanilla et al. 2016), Parazuelos (Escanilla 2016), La Ceñuela (Escanilla 2016; Rovira 2002), Carboneros (Escanilla 2016; San Nicolás del Toro 1998) and the El Argar site of Peñalosa (Moreno Onorato et al. 2010). However, such slags are a minority in these sites, suggesting sulfur presence in the system was likely accidental due to sulfides, oxides and carbonates coexisting in the same ore. In fact, chalcopyrite or tennantite is documented alongside oxides and carbonates in some of these sites (Escanilla 2016). These minerals were not previously roasted, and the presence of sulfur in the charge would have increased system oxidation, something observed in sample LC_M_89. In Laderas del Castillo samples, matte formation in slag is not documented, implying sulfur likely entered the charge as copper sulfide or as *fahlore* (such as covellina or tennantite) rather than as chalcopyrite.

Metallurgical production at Laderas del Castillo is concentrated from 2150 – 1800 cal BC, with no evidence of production from 1800 onwards, although this is the least known phase of the site. Two samples of production

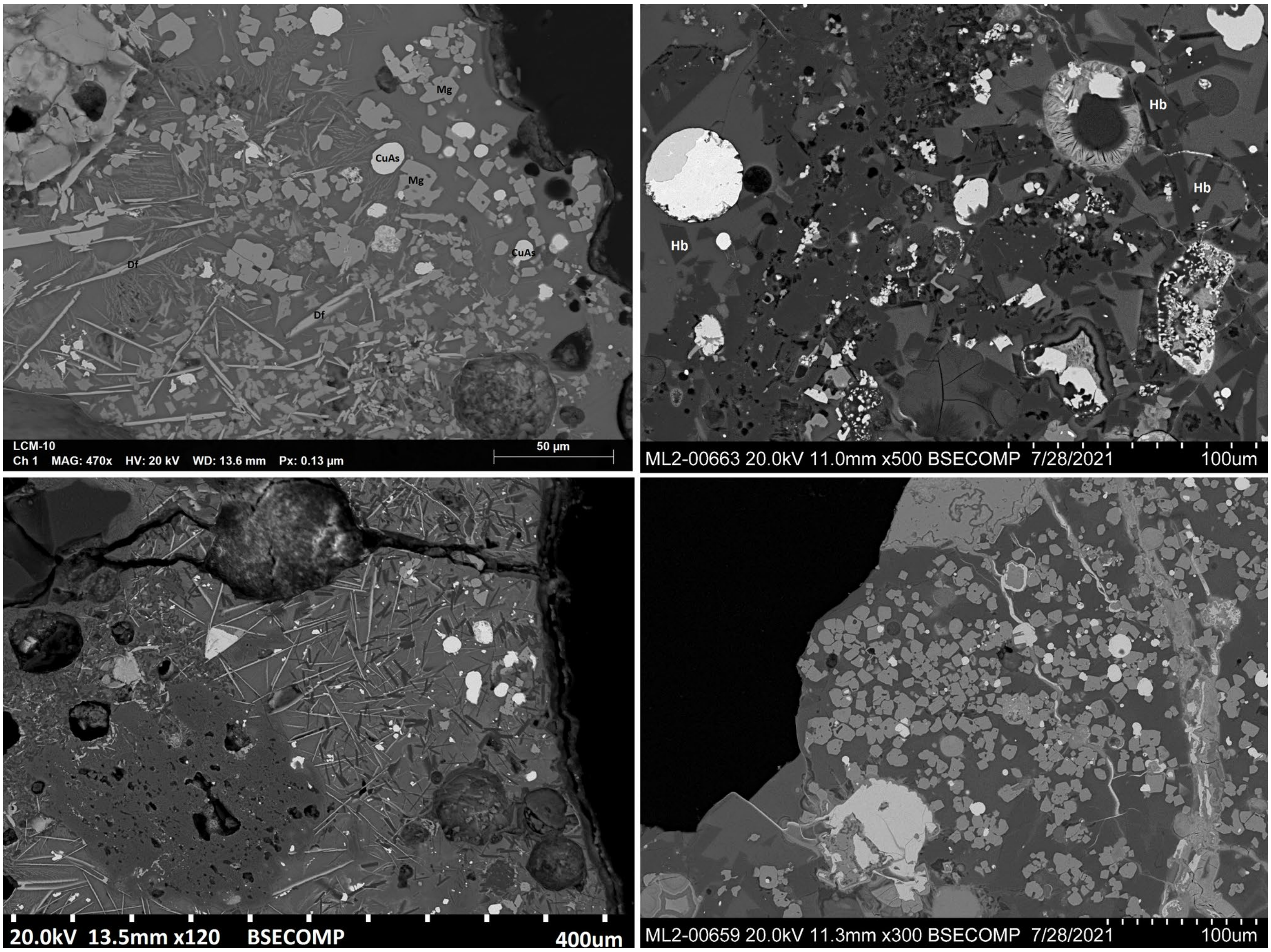

Fig. 8 SEM-BSE images of slag samples LC-M-10 (top left), LC-M-18 (top right), LC-M-17 (bottom left) and LC-M-18 (bottom right). Mg: Magnetite, Df: Delafosite, Hb: Hedenbergite, CuAs: Arsenical Copper

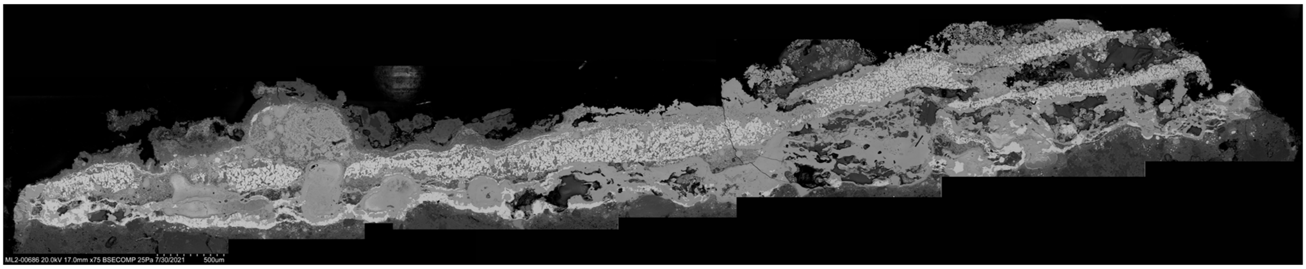

Fig. 9 SEM-BSE layer on LC-M-89 next to a large Cu prill. Large masses of CuO are documented together with delafossite and magnetite. Note that the CuO layer in close contact with the ceramic still contains some traces of S (up to 0.3%) while S is not detected in the more surface CuO layer

debris (one slag and one copper prill with some slag still adhered) and five objects were selected for further LIA analysis from phase I (2150 – 2050 cal BC); five production samples (three slag fragments and two smelting prills) and seven artefacts from phase II (2050 – 1800 cal BC); and only three axes and no production debris from phase III (1800 – 1550 cal BC). These axes, lacking further archaeological context, were found in tombs excavated in ancient works. We attribute them to phase III due to their typology and funerary provenance, as axes typically appear in tombs from 1800 BC onwards (Lull et al. 2018). Some differences can be inferred from the different phases,

Table 5 Elemental composition analysis of archaeological objects. Results presented in %wt normalized to 100%. Complete table of results can be downloaded from supplementary information 1. < LOD: below the limit of detection. Analyses conducted on patina are not normalised and Si, Al or P are the remaining elements up to 100%. Artefacts LC-M-64, LC-M-66, LC-M-52 and LC-M-54 were analysed by SEM in 1998 and have been reanalysed by ICP-MS in order to get better accuracy

ID	Technique	Type	Fase	Cu	As	Ag	Zn	Co	Ni	Sn	Sb	Pb	Bi	Fe	Au	S	Reference
LC-M-3	PIXE	Awl	Surface	98.00	1.56	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.028	< LOD	< LOD	This paper
LC-M-3	pXRF	Awl	Surface	96.68	1.69	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.03	< LOD	< LOD	0.8	This paper
LC-M-6	pXRF	Awl	Surface	97.21	1.15	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.04	< LOD	0.52	This paper
LC-M-1	PIXE	Arrowhead	I	98.56	1.43	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.006	< LOD	< LOD	This paper
LC-M-1	pXRF	Arrowhead	I	98.83	0.37	< LOD	0.18	< LOD	< LOD	0.04	< LOD	< LOD	0.04	0.48	< LOD	< LOD	This paper
LC-M-12	ICP-MS	Chisel	I	96.70	3.26	< LOD	0.024	0.002	0.009	0.000	0.003	0.001	0.003	0.11	< LOD	< LOD	This paper
LC-M-12	pXRF	Chisel	I	97.13	2.68	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.05	This paper
LC-M-83	pXRF (patina)	Metal Frag.	I	85.87	0.42	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.29	< LOD	0.24	This paper
LC-M-4	pXRF	Awl	I	94.84	5.00	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.02	0.11	< LOD	< LOD	This paper
LC-M-14	pXRF	Arrowhead	I	94.08	2.92	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.02	< LOD	< LOD	0.05	This paper
LC-M-36	pXRF	Metal Frag.	I	97.78	1.74	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.48	< LOD	< LOD	This paper
LC-M-30	pXRF	Copper Prill	I	97.92	1.36	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.04	0.55	< LOD	< LOD	This paper
LC-M-63	ICP-MS	Halberd	II	95.14	4.79	< LOD	0.046	0.001	0.001	0.002	0.001	0.002	0.012	0.045	nd	< LOD	This paper
LC-M-64	ICP-MS	Halberd	II	97.94	1.95	< LOD	0.054	0.001	0.012	0.001	0.023	0.007	0.018	0.017	0.001	< LOD	This paper
LC-M-64-S	*SEM-EDS*	*Halberd*	*II*	*95.93*	*2.77*	< LOD	*1.09*	< LOD	*< LOD*	*0.12*	*< LOD*	*0.05*	< LOD	< LOD	< LOD	< LOD	*Simón*,1998
LC-M-64-S	*SEM-EDS*	*Rivet*	*II*	*96.60*	*2.29*	< LOD	*1.15*	< LOD	*< LOD*	*0.26*	*0.12*	*< LOD*	< LOD	< LOD	< LOD	< LOD	*Simón*,1998
LC-M-66	ICP-MS	Halberd	II	94.56	5.34	< LOD	0.023	0.026	0.026	0.000	0.001	0.002	0.024	0.089	< LOD	< LOD	This paper
LC-M-66-S	*SEM-EDS*	*Halberd*	*II*	*93.53*	*5.87*	< LOD	*1.23*	< LOD	*< LOD*	*0.04*	*< LOD*	*< LOD*	< LOD	< LOD	< LOD	< LOD	*Simón*,1998
LC-M-25	pXRF	Awl	II	97.56	1.51	< LOD	0.19	< LOD	< LOD	< LOD	< LOD	< LOD	0.03	0.66	< LOD	< LOD	This paper
LC-M-27	pXRF (patina)	Awl or hook	II	85.28	0.67	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.20	< LOD	3.32	This paper
LC-M-29	pXRF (patina)	Awl	II	85.04	1.61	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.07	< LOD	3.04	This paper
LC-M-52	ICP-MS	Axe	III	97.10	2.66	< LOD	0.023	0.001	0.035	0.162	0.007	0.002	0.019	0.050	0.001	< LOD	This paper
LC-M-52-S	*SEM-EDS*	*Axe*	*III*	*97.1*	*2.2*	< LOD	*0.2*	< LOD	*0.3*	*< LOD*	*< LOD*	*0.04*	< LOD	< LOD	< LOD	< LOD	*Simón*,1998
LC-M-54	ICP-MS	Axe	III	96.64	3.21	< LOD	0.021	0.009	0.015	0.018	0.006	0.044	0.031	0.038	< LOD	< LOD	This paper
LC-M-54-S	*SEM-EDS*	*Axe*	*III*	*95.9*	*3.7*	< LOD	*0.5*	< LOD	*0.04*	*< LOD*	*< LOD*	*0.1*	< LOD	< LOD	< LOD	< LOD	*Simón*,1998
LC-M-50	pXRF	Axe	III	92.57	0.84	0.52	< LOD	< LOD	< LOD	5.72	0.10	0.06	0.01	0.09	< LOD	< LOD	This paper

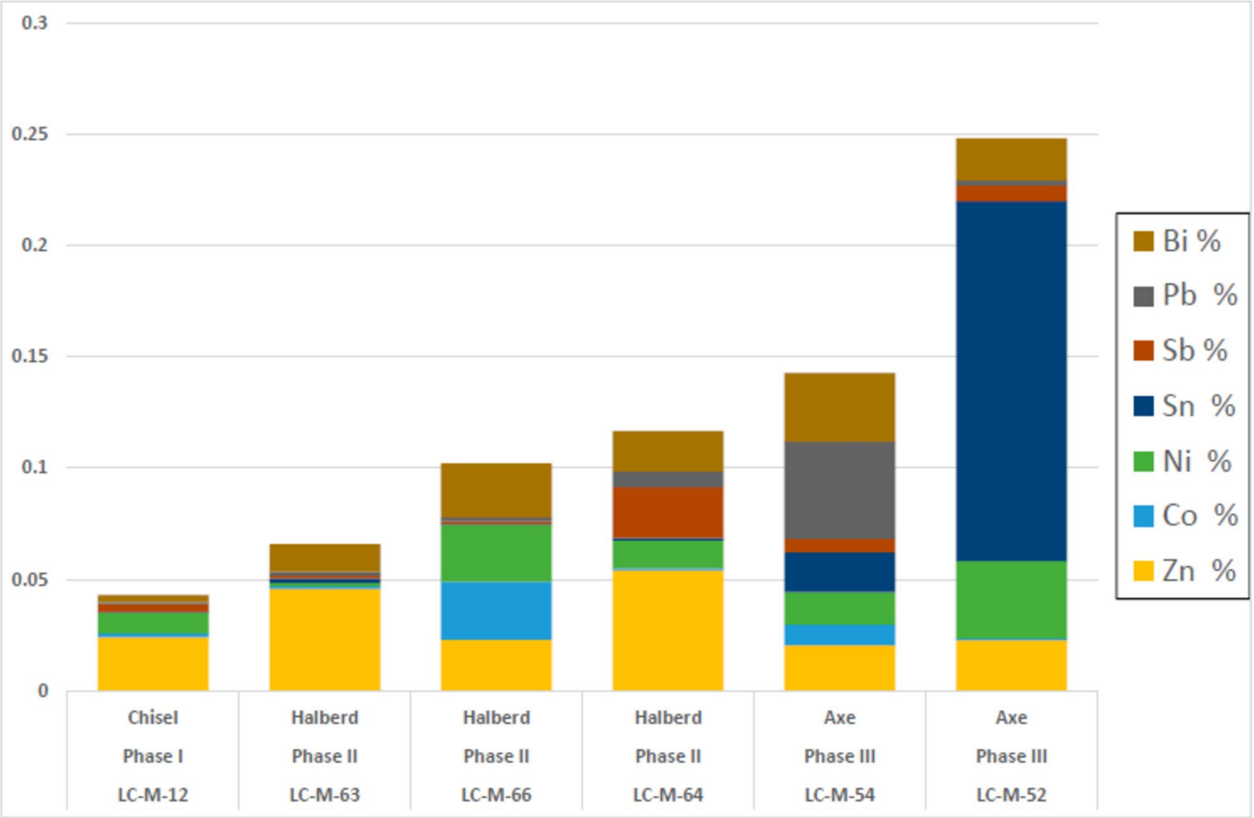

Fig. 10 Main impurities analysed by ICP-MS of metal objects from Laderas del Castillo

though resources exploited seem to persist from 2150 to 1550 cal BC.

In phase I, two different sources appear to be exploited at the site (the slag sample LC-M-89 with 18.32 and the smelting prill LC-M-8 with 18.55 values for ^{206}Pb/^{204}Pb) (Fig. 12). Interestingly, some objects match or plot close to these production samples, suggesting they were produced from the same resources, while two other metallic samples which could be considered as scrap metal possibly to be re-melted (LC-M-82 and LC-M-83, see Sect. 5.1) reflect a different origin with c. 18.8 for ^{206}Pb/^{204}Pb.

During phase II, the availability of copper resources seems to expand. One caption area (including one or several mining districts) is reflected by slag samples LC-M-10 and LC-M-17, and smelting prills LC-M-30 and Lc-M-36, with Pb values from 18.37 to 18.45 for ^{206}Pb/^{204}Pb. The second caption area is represented by the slag sample LC-M-18 with 18.75 for ^{206}Pb/^{204}Pb, similar to the two scrap metal samples from phase I (Fig. 12). This indicates that these resources were known and already exploited in phase I by other communities, possibly reaching Laderas del Castillo through exchange or brought by their owners, either as scrap metal for re-melting or as artefacts. Later on, these resources were directly exploited by Laderas del Castillo metallurgists during phase II, though not on a priority basis. Conversely, resources represented by the smelting prill LC-M-8 of phase I were no longer exploited at the site in subsequent phases, though they continued to be used elsewhere, as three artefacts from phase II could match these resources. Similarly, resources represented by LC-M-89 in phase I were not exploited at the site in phase II but were still used elsewhere. Interestingly, the two halberds (LC-M-63 and LC-M-64) plot very close to this sample.

The three axes ascribed to phase III, which lack documented production debris, as well as one awl recovered on surface, match the tendency reflected in phases I and II. This pattern suggests an intermittent resource exploitation system, with several mining districts being exploited simultaneously by multiple sites.

To identify potential exploitation areas, production samples (slags and prills) were compared to different mining districts (Fig. 13). Current geological information on the region's copper deposits suggests that nearby mining districts such as Santomera, Sierra Almenara or Sierra Tercia, located up to 60 km away (Fig. 1), could be discarded as potential sources due to their low levels of As and S (Brandherm et al. 2022; Escanilla 2016) and the higher radiogenic values of Santomera mine (Brandherm

Table 6 Lead isotope compositions by MC-ICP-MS

ID	Type	Phase	$^{206}Pb/^{204}Pb$	Error (2SE)	$^{207}Pb/^{204}Pb$	Error (2SE)	$^{208}Pb/^{204}Pb$	Error (2SE)	$^{207}Pb/^{206}Pb$	Error (2SE)	$^{208}Pb/^{206}Pb$	Error (2SE)
LC-M-6	Awl	Surface	18.5692	0.0012	15.6842	0.0011	38.8430	0.0027	0.84463	0.00006	2.09180	0.00002
LC-M-8	Smelting remain	I	18.5486	0.0009	15.6739	0.0008	38.6349	0.0022	0.845017	0.000015	2.082898	0.00005
LC-M-89	Slagged pottery	I	18.3236	0.0010	15.6481	0.0010	38.4935	0.0030	0.853984	0.000022	2.100761	0.00008
LC-M-83	Metal frag.	I	18.7973	0.0011	15.6870	0.0010	38.8423	0.0027	0.834538	0.000016	2.066379	0.00005
LC-M-82	Metal frag.	I	18.8159	0.0011	15.6867	0.0010	38.8339	0.0027	0.833693	0.000018	2.063887	0.00006
LC-M-12	Chisel	I	18.4813	0.0007	15.6622	0.0007	38.6645	0.0018	0.84747	0.00001	2.09209	0.00004
LC-M-1	Arrowhead	I	18.5537	0.0007	15.6632	0.0007	38.6558	0.0019	0.844210	0.000011	2.083457	0.00004
LC-M-3	Awl	I	18.4445	0.0006	15.6698	0.0008	38.6539	0.0024	0.849566	0.00008	2.095688	0.00002
LC-M-14	Arrowhead	I	18.2671	0.0007	15.6366	0.0007	38.4242	0.0019	0.855998	0.000016	2.103468	0.00005
LC-M-17	Slagged pottery	II	18.4480	0.0008	15.6571	0.0009	38.5885	0.0025	0.848717	0.000018	2.091744	0.00007
LC-M-10	Slagged pottery	II	18.4056	0.0009	15.6624	0.0009	38.5579	0.0027	0.850958	0.000019	2.094898	0.00007
LC-M-18	Slag frag.	II	18.7516	0.0008	15.6836	0.0007	38.9898	0.0020	0.836387	0.000013	2.079278	0.00004
LC-M-36	Smelting remain	II	18.3799	0.0008	15.6335	0.0009	38.3603	0.0026	0.850578	0.000020	2.087081	0.00008
LC-M-11	Metal frag.	II	18.2998	0.0010	15.6479	0.0010	38.4667	0.0026	0.855084	0.000015	2.102028	0.00005
LC-M-30	Smelting prill	II	18.3714	0.0010	15.6602	0.0012	38.4891	0.0034	0.852427	0.000027	2.095058	0.00010
LC-M-63	Halberd	II	18.3347	0.0013	15.6559	0.0012	38.4861	0.0030	0.85389	0.00002	2.09909	0.00006
LC-M-64	Halberd	II	18.3051	0.0007	15.6553	0.0007	38.4784	0.0022	0.85525	0.00001	2.10207	0.00006
LC-M-25	Awl	II	18.5224	0.0037	15.6801	0.0033	38.6243	0.0083	0.846546	0.000037	2.085273	0.00011
LC-M-27	Anzuelo	II	18.5934	0.0013	15.6671	0.0012	38.6138	0.0033	0.842617	0.000023	2.076750	0.00008
LC-M-29	Awl	II	18.6123	0.0010	15.6751	0.0011	38.8264	0.0030	0.842190	0.000019	2.086066	0.00007
LC-M-52	Axe	III	18.6195	0.0008	15.6862	0.0007	38.5426	0.0021	0.84246	0.00001	2.07001	0.00005
LC-M-54	Axe	III	18.2796	0.0011	15.6401	0.0009	38.4432	0.0025	0.85560	0.00002	2.10306	0.00007
LC-M-50	Bronze Axe	III	18.4873	0.0015	15.6855	0.0014	38.7052	0.0036	0.84845	0.00002	2.09362	0.00007

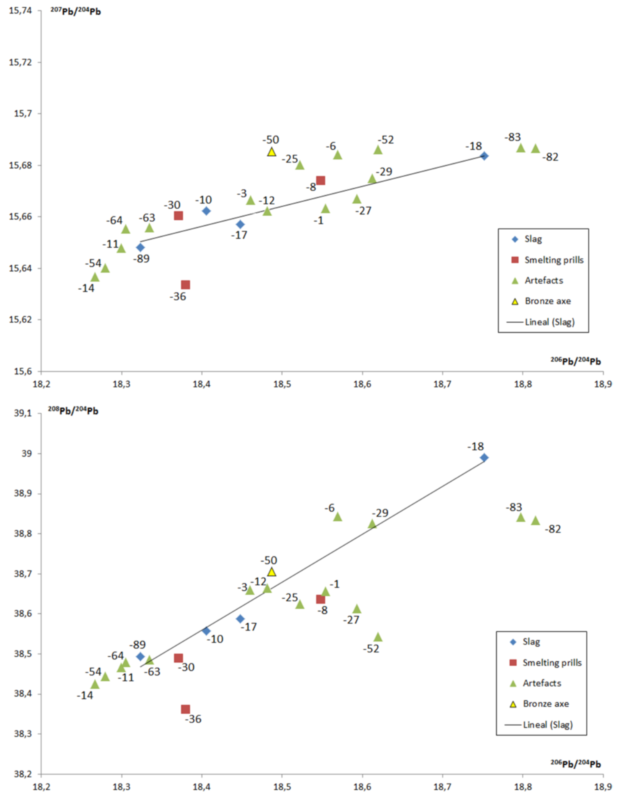

Fig. 11 Lead isotope ratios of metallurgical remains (slag and smelting prills) and CuAs and Bronze objects found at Laderas del Castillo. In order to lighten up the information in the graphs, 'LC-M' has been removed from all samples IDs, and only their final numbers are represented

et al. 2022), at barely 5 km from the site. However, isotopic characterization of other deposits from Sierra Almenara or Sierra Tercia is worthwhile.

Some smelting samples show good concordance with other deposits exploited by the El Argar communities. The smelting slag LC-M-18, with a $^{206}Pb/^{204}Pb$ value of 18.75, could relate to the copper ores of Cartagena/Mazarrón or Sierra Almagrera in the southeast (Fig. 13). The Cartagena/Mazarron mining district, 50 km away from Laderas del Castillo, primarily contains galena, with copper occurring occasionally, though some surface copper deposits and prehistoric mining works are documented (Antolinos Marín and Soler Huertas 2008; Domergue 1990), notably Filón Consuelo, which has prehistoric works and copper mineralization with high arsenic content (Escanilla and Delgado 2015; Escanilla 2016). Sierra Almagrera, 140 km away, is a well-known copper mining district. Copper oxides and carbonates are well documented there, as are tennantite and tetrahedrite, consistent with the composition of copper prills of LC-M-18, with high As levels (up to 25%) and some CuS presence. The closest match, according to Euclidean distances, is a sample from Sierra Almagrera (0.0068). This, together with the better concordance with its isotopic field in some graphs (Fig. 13B and D) leads us to consider Almagrera as the likely source for slag LC-M-18.

At the other end of the graphs, slag sample LC-M-89 could relate to the Linares mining district, though it plots just at the limit of this isotopic field. Exploitation of this district is well-documented during the El Argar chronology, possibly starting during the Copper Age (Arboledas Martínez et al. 2015; Hunt Ortiz et al. 2011; Moreno Onorato et al. 2010). Early mining works at the Jose Palacio mine in the Linares mining district are dated to 2205 – 2030 2σ cal BC (Arboledas Martínez et al. 2015), consistent with the chronology of LC-M-89. The arrival of copper ores from Linares to Laderas del Castillo as early as 2200/2100 BC suggests that the connectivity of the El Argar territory is rooted in the earlier Copper Age background.

The smelting prill LC-M-36, with high S levels plots close to a sample from Los Pedroches mining district, outside the El Argar territory, in northern Córdoba province. These mining districts have ample evidence of prehistoric mining works, and provenance from Los Pedroches has been proposed for some El Argar objects (Murillo-Barroso et al. 2015, 2025).

For the remaining production samples (LC-M-8, LC-M-10, LC-M-30 and LC-M-17), two options are possible, though not necessarily mutually exclusive: some (mostly the crucible LC-M-10 and the smelting prill LC-M-30) could be associated with copper deposits of the interior Baetic Cordillera in Granada province. Some of these mines have evidence of prehistoric works and are related to metal artefacts of nearby El Argar sites such as Terrera del Reloj or Cuesta del Negro (Murillo-Barroso et al. 2025). Alternatively, they could result from co-smelting of copper ores from Sierra Almagrera in Almería and Linares in Jaén.

Examining the dispersion of metal objects, the situation appears somewhat similar. However, in this case, the isotopic correspondence with mining districts far from the site may result from the exchange of objects rather than the transport of ore to the site, as suggested by the smelting slags mentioned above. A provenance on the Linares mining district can be proposed for the arrowhead LC-M-14 (dated in phase I, 2150 – 2050 cal BC) and the axe LC-M-54 (phase III, 1750 – 1550 cal BC) (Fig. 14). This suggests early exploitation of these resources (also evidenced by the slag LC-M-89) and its continuity. In both cases, their closest matches according to Euclidean distances are samples from the Linares mining district (0.0039 and 0.0087 respectively).

At the other end of the graph, the two corroded metals LC-M-82 and LC-M-83 (both dated to phase I) could be related to the copper mining districts of the southeast, possibly Cerro Minado, although they plot at its limit in one graph (Fig. 14). Their closest matches according to Euclidean distances are one sample from Málaga (0.0163

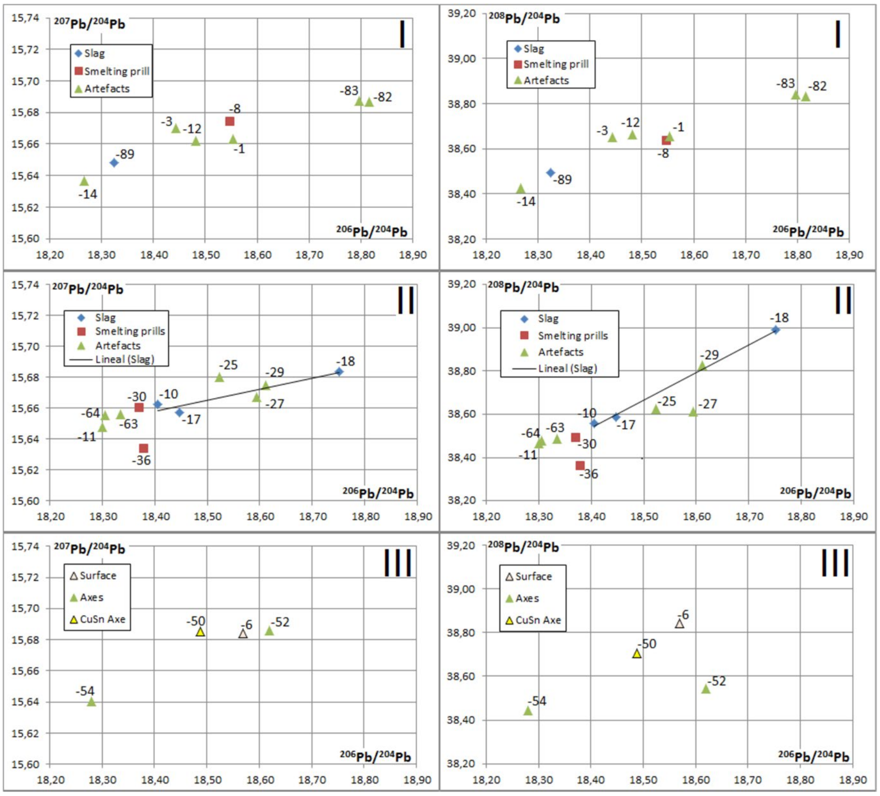

Fig. 12 Lead isotope ratios of metallurgical remains and metal objects found at Laderas del Castillo in phases I, II and III so the chronological evolution of caption areas can be inferred. In order to lighten up the information in the graphs, 'LC-M' has been removed from all samples IDs, and only their final numbers are represented

and 0.0141, respectively) and Mallorca (0.0247 and 0.0318, respectively), although this seems less probable due to the low archaeological evidence in these areas (Llull et al. 2021; Rodríguez Vinceiro et al. 2018). For LC-M-82, the third closest sample is from Cerro Minado at a Euclidean distance of 0.0438. For LC-M-83, there are four other closest samples from Cabo de Gata (with Euclidean distances between 0.0354 and 0.0435) although the main ores at Cabo de Gata are mostly lead, silver and gold minerals. For this sample, the ore from Cerro Minado is at a Euclidean distance of 0.0565.

The four awls (two dated to phase II and two with an indeterminate chronology) and the bronze axe (phase III) could be related to the mining districts of the Interior of the Betic Cordillera in Granada province (Fig. 14). The use of these resources is also documented in the El Argar sites of Cuesta del Negro and Terrera del Reloj, up to 40 km away from these mines (Fig. 1) (Murillo-Barroso et al. 2025). These objects might have been exchanged with these sites, or brought by the inhabitants of Laderas del Castillo. Awls are usually associated with women of the upper class, for whom higher mobility is also proposed (Villalba-Mouco et al. 2022). Therefore, it is reasonable to consider that women traveled carrying their own belongings. Unfortunately, these awls were recovered in domestic contexts and thus were not directly associated with any individual remains, preventing us from testing this hypothesis through anthropological and mobility analyses.

The tendency line of the four awls differs significantly from that of the rest of artefacts, which could indicate the use of different resources. Excluding them, the tendency lines of the arsenical copper artefacts and the production remains match quite well, suggesting the co-smelting of different ores and possibe artefacts production at the site (Fig. 14). Due to the productive limits of crucible smelting metallurgy, massive objects such as axes would likely require the pouring of several crucibles, resulting in a more likely metal mixing if several mines were exploited simultaneously. This could be the case, if manufactured in the site, for the samples LC-M-1, LC-M-12, LC-M-63 and LC-M-64 which plot in the mixing line of the resources processed at the site (Linares and Almagrera).

Regarding the results of the elemental composition, the almost total absence of bronzes stands out, only represented by the axe LC-M-50. This is because the majority of samples are from ancient phases, prior to 1800 cal BC, when objects with tin began to appear sporadically in the El Argar record (Montero Ruiz and Murillo-Barroso 2022). Nor did the analyses carried out by L. Simón García (1998: 210–211) detected the presence of tin-bronzes in the Furgús collection from Laderas del Castillo. Bronze objects are equally scarce in San Antón, where only a

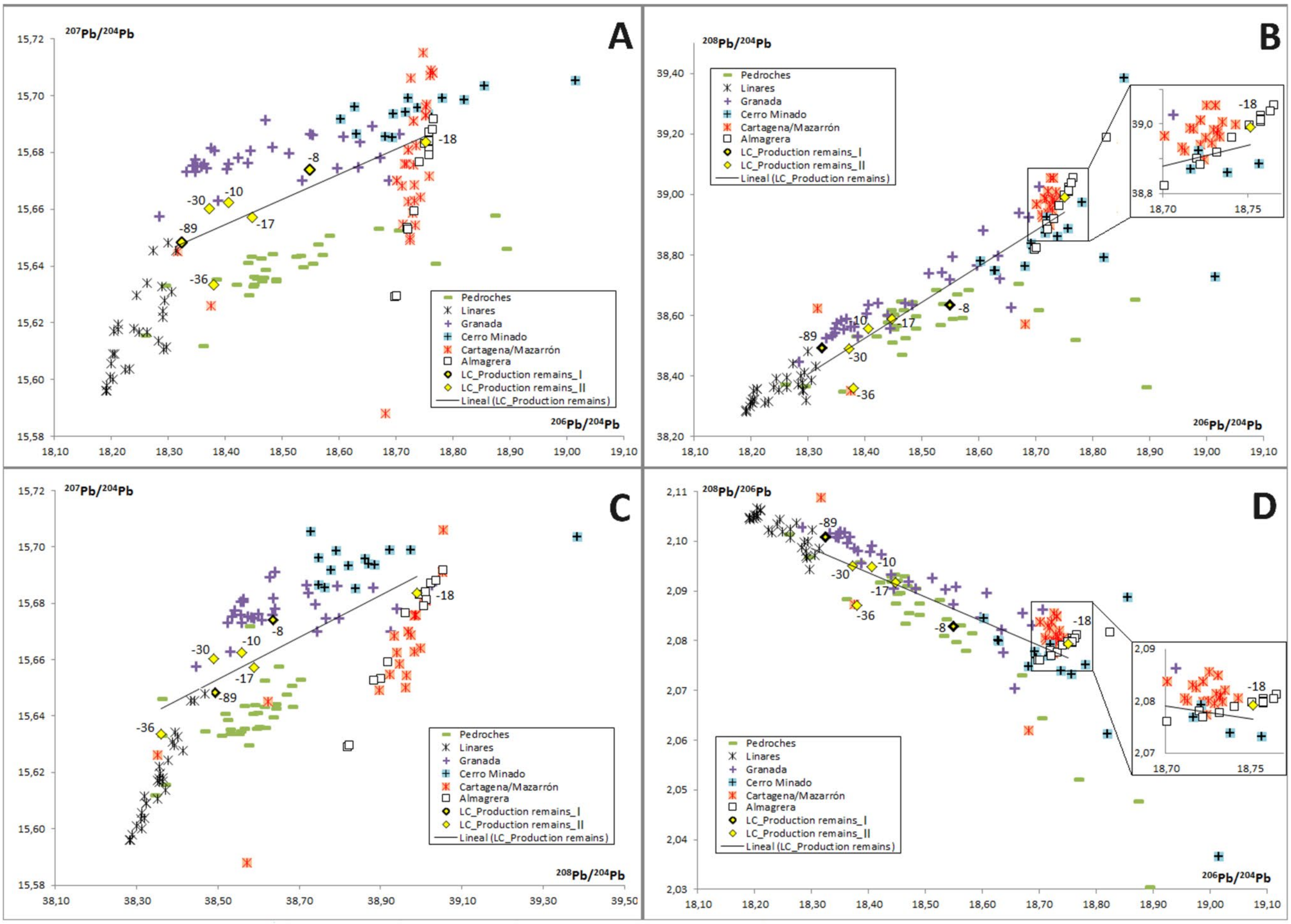

Fig. 13 Lead isotope ratios of metallurgical remains found at Laderas del Castillo compare the isotopic fields of copper mineralisations of Los Pedroches (Córdoba), Linares (Jaén), the Interior Betic Belt (Granada), Cerro Minado and Almagrera (Almería). LIA data from geological deposits from: Arribas and Tosdal 1994; Baron et al. 2017, Klein et al. 2009; Murillo-Barroso et al. 2019, 2020, 2025; Santos Zaldegui et al. 2004, Stos-Gale et al. 1995. In order to lighten up the information in the graphs, 'LC-M' has been removed from all samples IDs, and only their final numbers are represented

dagger and an axe have a tin concentration of 5.6% and 3.11% Sn, respectively (Brandherm 2022: 136; Simón García 1998: 210).

As for arsenic, it generally occurs in the range between 2 and 5%, with an average of 3%. These are the average ranges of El Argar objects and maintain the trend of a higher concentration of arsenic in bladed artifacts such as halberds (Lull et al. 2017: 377; Harrison et al. 1981: 161). This arsenic content matches the average of the analyses carried out by L. Simón García in Laderas del Castillo (1998: 212–214), which stands at 3.2% and rises to almost 4% if only bladed artifacts are considered. For the analysed axes, the arsenic average is close to 2.5%.

Recent analyses of the nearby settlement of San Antón (Brandherm et al. 2022) show a similar trend to those of Laderas del Castillo in terms of lead isotopes (Fig. 14). The arsenic average increases above 4% when discarding the two objects that have tin in their composition. This high average could be due to the fact that, except for an axe, the rest of the analyses correspond to dagger and halberd blades.

The average of the set of impurities between 0.1 and 0.3% is also close to the usual values in the El Argar metallurgy, which are generally very pure arsenical coppers, mostly identified in Group 2 of Pollard et al. (2018: 86 and 100). Only iron is systematically detected, though it very rarely exceeds 0.5%. In the analyses of Laderas del Castillo, iron occurs between 0.01% and 0.1%, indicating technological processes under poor reducing conditions, where practically no slag was generated (Craddock and Merks 1987; Pernicka 1999).

The tin impurities of the two axes LC-M-52 and LC-M-54 stand out, which with 1373 ppm and 171 ppm respectively, exceed the rest of the analysed objects by an order of magnitude. Furthermore, in LC-M-54 lead also exceeds the rest of the objects by an order of magnitude, going from an average of 20 ppm to 415 ppm Pb. This

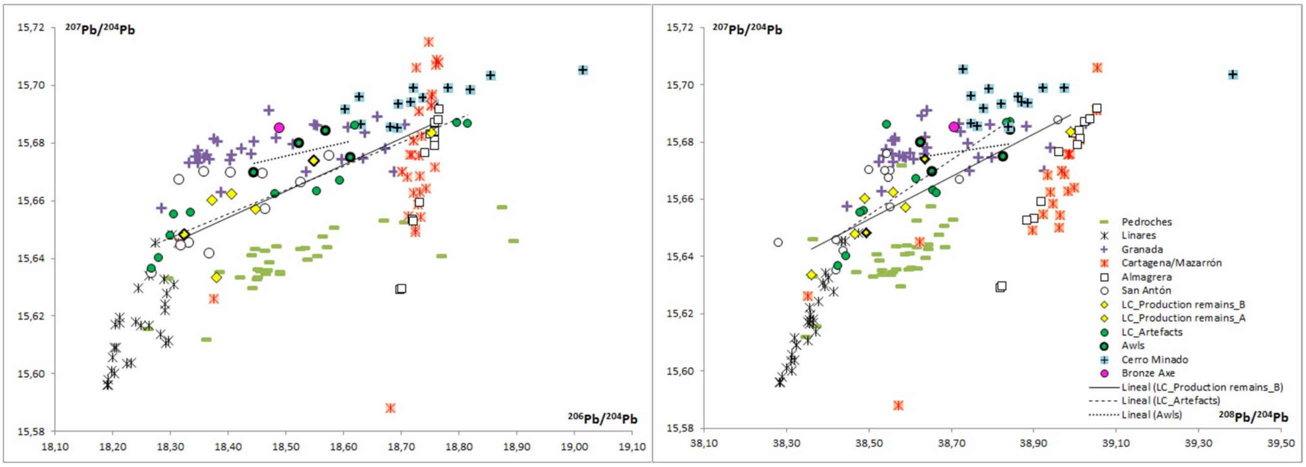

Fig. 14 Lead isotope ratios of metallurgical remains and metal objects found at Laderas del Castillo compare the isotopic fields of copper mineralisations of Los Pedroches (Córdoba), Linares (Jaén), the Interior Betic Belt (Granada), Cerro Minado and Almagrera (Almería). LIA data from geological deposits from: Arribas and Tosdal 1994; Baron et al. 2017, Klein et al. 2009; Murillo-Barroso et al. 2019, 2020, 2025; Santos Zaldegui et al. 2004, Stos-Gale et al. 1995. Note the different trend of awls in comparison to the rest of the objects. For better discrimination of isotopic fields, 208Pb/204Pb vs. 207Pb/204Pb (as in Fig. 13C) instead of 206Pb/204Pb vs. 208Pb/204Pb (Fig. 13B) has been chosen

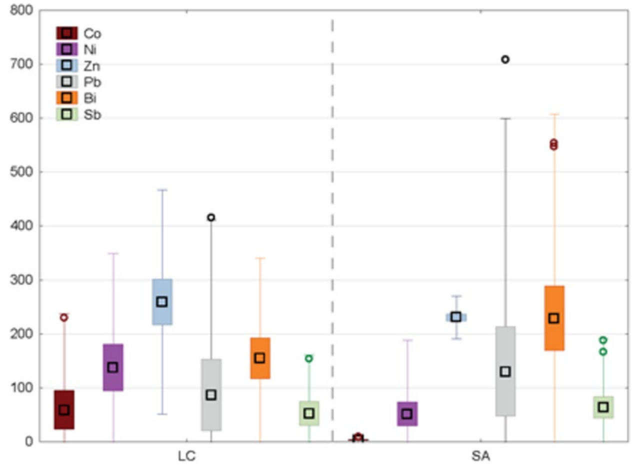

Fig. 15 Comparison of main impurities (ppm) analysed by ICP-MS on artefacts from Laderas del Castillo (this paper) and the nerby site of San Antón de Orihuela (Brandherm et al. 2022; Table 2)

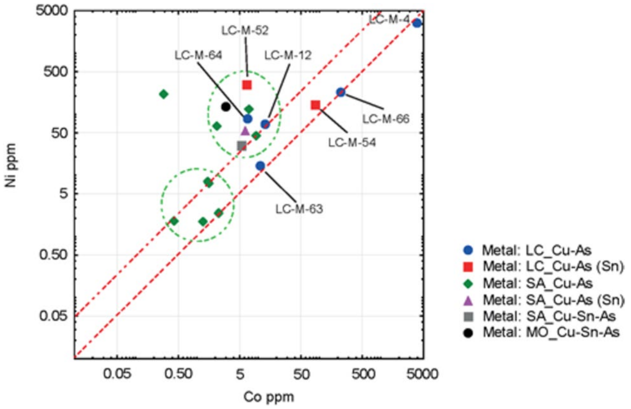

Fig. 16 Co-Ni Ratio on ICP-MS results from Laderas del Castillo (this work) and San Antón de Orihuela (Brandherm et al.: Table 2)

reinforces the relationship that lead isotopes establish between LC-M-54 and Linares, where copper-lead minerals are abundant.

The two axes from phase III break the pattern of impurities that we see in the rest of the objects, in which Zn, Bi and Ni are configured as the main impurities after iron.

Comparing the trace elements of Laderas del Castillo with those of San Antón (Brandherm 2022), we see certain common patterns (Fig. 15), specifically in the relationship between Zn, Pb, Bi and Sb, which suggests, just like the isotopes, a common origin of the base metal (Fig. 14). Co and Ni, in turn, present differentiated patterns, with extremely low values in the San Antón objects in Co and lower than in Laderas in Ni.

The Co-Ni relationship is quite stable during the smelting process (Pernicka 1999), so it has been represented in Fig. 16 to observe possible trends of different metals used in both sites and if these varied with the presence of tin as an impurity or in alloy. The graph shows a set of objects on the R 1 line with a Co/Ni ratio between 0.5 and 1%, and another on the R 0.2 line with several artifacts with a Co/Ni ratio close to 0.2. Finally, a third group of objects stands out for having a Co/Ni ratio of less than 0.15 and where Co does not exceed 10 ppm. Two of the halberds (LC-M-63 and 66), one of the axes (LC-M-54), and the LC-M-4 awl align on line R2 without forming a group, matching the objects with a higher percentage of As from the Laderas del Castillo series, indicating the known relationship of arsenic with minerals rich in cobalt and nickel, such as in Cerro Minado and hydrothermal mineralizations in the southeast.

Conclusions

Laderas del Castillo was a large Argaric settlement, spanning nearly 2 hectares and occupied for over 600 years. Its foundation, around 2200/2150 cal BC, along with other settlements from the final reaches of the Segura and Vinalopó rivers, demonstrates that this part of the intrabetic basin fully participated in the formation of what we recognize as the El Argar society (Hernández Pérez et al. 2021). The archaeometallurgical study carried out from a set of evidence that includes smelting vessels and slags, crucibles, copper nodules and metal artefacts, provides a comprehensive view of the metallurgical production processes carried out at the settlement throughout its various phases of occupation, despite the copper sources being located at a considerable distance within the El Argar social sphere.

The relevance of this study is significant because, until now, although remains of metallurgical production have been documented at other El Argar sites (Arribas et al. 1989; Contreras Cortés 2000; Lull et al. 2010; Martínez Rodríguez et al. 1996; Montero Ruiz 1994; Mongiatti and Montero-Ruiz 2019; Moreno Onorato and Contreras Cortés 2010; Simón García 1998; Siret and Siret 1890), extractive metallurgical remains have only been analysed in Peñalosa (Moreno Onorato et al. 2010, 2017). At Laderas del Castillo, it has been possible to document and analyse, for the first time, nearly the entire metallurgical *chaîne opératoire* in an El Argar settlement outside the metallurgical core of the Rumblar valley. Unlike the Vega Baja of the Segura river, mining played an essential role in the economy of the Rumblar valley populations (Arboledas Martínez et al. 2014).

Microstructural analyses of production remains indicate continuity in the Copper Age metallurgical tradition, with crucible technology, open-air structures that allowed for little atmospheric control and low performance. Smelting metallurgical activity begins to be documented in the first phase of occupation (2300 – 2050 BC) and continues, with more evidence, in phase II (2050 – 1800 BC). No metallurgical evidence is documented in the final phase of the site (1750 – 1550 BC), but this phase is also the least known of the site due to erosive processes.

Among the results presented, the evidence of co-smelting of oxidic and sulfidic minerals without prior roasting stands out. The study of extractive remains using SEM-EDS suggests the use of carbonates with inclusions of gray copper sulfides and/or chalcopyrite, rather than an express selection of sulfides.

Isotopic and elemental composition analyses show the exploitation of various copper resources and sources far from the settlement (Linares and Almagrera, about 300 and 140 km respectively) from the earliest periods documented on the site (2300 – 2050 BC). Despite the existence of closer mineralizations in the Orihuela mountain range, such as Cerro de la Mina, or the Cartagena/Mazarrón mining district that presents copper arsenates, the active exchange networks in Laderas del Castillo repeat the same pattern as found in the rest of the El Argar territory (e.g. Murillo-Barroso et al. 2015, 2025). This pattern is based on the intensive exploitation of the mineralizations of the interior of the Baetics and the Southeast of the peninsula, along with the use of resources that partially repeat the patterns observed in the Copper Age metallurgy of the east, where the arrival of copper ores from Cerro Minado has already been documented in the site of Sancho Llop (Pascual Beneyto et al. 2021). This suggests that the mobility and exchange networks operating in the southeast during the El Argar period were rooted in earlier archaeological phases. This mobility or exchanges is also inferred from the isotopic signatures and elemental composition patterns of the objects, which show distant origins such as the interior of the El Argar area (Linares or Granada) or the central core of the Vera basin (Cerro Minado and Almagrera), reflecting the interconnection that these Argaric groups must have had.

These data, some of which resemble those available for the nearby settlement of San Antón, allow us to infer that the exchange networks for copper ore, metal scrap for remelting, and artefacts made from different sources were stable throughout the historical dynamics of those peasant populations, using both land transport and, probably, the sea route through strict cabotage. In short, from the above it can be inferred that.

1. In its social space and throughout its historical development, El Argar society maintained different exchange routes of copper in different stages of production to cover the social needs acquired by the bulk of the population, whether linked to productive activities or those of a sumptuous, coercive, or social reproduction nature.
2. There does not seem to have been a centralised system of control over the production, exchange and distribution of copper in the phases represented at Laderas del Castillo, although metallurgy and metal objects played an essential role in El Argar society, both as work instruments and as objects of gender identity, but especially, as demarcating social position, as has been extensively documented from the metallic objects within the tombs (Lull 1983; Lull et al. 2010).
3. In El Argar, as an initial class society (Jover 1999; Arteaga 2000), the dominant groups of the different regions would control and manage the processes of

production, exchange and distribution of metal among the population. Although peasant groups tried to be governed by the principles of self-supply and self-sufficiency (Jover et al. 2020), the creation of new social needs, such as those linked to metals, gave the dominant groups a privileged position by taking over the surplus value implicit in exchange processes. Hence the importance that metals acquired and that has not yet been fully recognized in the research of European societies of recent Prehistory. It is within this context that the metallurgical record of Laderas del Castillo —reflecting the intensification of metallurgical production documented in El Argar society and the appearance of new metallic objects such as weapons and ornaments— should be understood.

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Author contributions MMB and NE contributted equally to the development of this study. All authors contributed to the study conception and design. Archaeological excavations were directed by JALP and JJM. Reaearch funding was obtained by MMB, JALP and JJM. Material preparation, data collection and analysis were performed by MMB, NE and AL. Analytical results were discussed by all authors. The first draft of the manuscript was written by MMB and NE, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Data availability Data is provided within the manuscript or supplementary information files.

Declarations

Competing interests The authors declare no competing interests.

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