archaeometry

Archaeometry **58**, 4 (2016) 593–609 doi: 10.1111/arcm.12197

A COMPOSITIONAL AND MICROSTRUCTURAL STUDY OF EIGHTH-CENTURY BC BRONZES FROM MOITA DA LADRA (TAGUS ESTUARY): HOW DID THE SPREAD OF THE PHOENICIAN METALLURGY TAKE PLACE IN WESTERN IBERIA?*

P. VALÉRIO,[1]† A. M. M. SOARES,[1] M. MONTEIRO,[2] A. PEREIRA,[2] M. F. ARAÚJO[1] and R. J. C. SILVA[3]

[1]*Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10 (km 139,7), 2695-066 Bobadela LRS, Portugal*
[2]*EMERITA—Empresa Portuguesa de Arqueologia, Apartado 32—BEC Oeiras, 2781-901 Oeiras, Portugal*
[3]*CENIMAT/I3N, Departamento de Ciência dos Materiais, Faculdade de Ciências e Tecnologia, FCT, Universidade Nova de Lisboa, 2829-516 Monte de Caparica, Portugal*

Metals from a votive deposit at Moita da Ladra (Tagus Estuary) dating to the eighth century BC *were studied by micro-EDXRF, optical microscopy and Vickers testing to investigate the adoption of Phoenician innovations by indigenous communities. Artefacts are made of bronze alloys with suitable tin contents (11.6 ± 2.3 wt%) and very low iron impurities (<0.05 wt%), and were often manufactured using the long post-casting sequence. Comparisons with indigenous and Phoenician metallurgies from western Iberia revealed a conservative technology suggesting that the spread of Phoenician innovations was very slow. In this region, the adoption of a diversified copper-based metallurgy and reduction furnaces only seems to occur during the Post-Orientalizing Period, c. sixth to fourth centuries* BC.

KEYWORDS: WESTERN IBERIA, EARLY IRON AGE, PHOENICIAN, METALLURGICAL INNOVATIONS, BRONZE ALLOYS, EDXRF, OPTICAL MICROSCOPY

THE ORIENTALIZING PERIOD IN PORTUGAL

The beginning of the Phoenician expansion to the far west of the Mediterranean can be ascribed to the ninth century BC (Arruda 1999–2000; Aubet 2008; González de Canales *et al*. 2004, 2008). The high proportion of Phoenician pottery among the ceramic recovered at Plaza de las Monjas (Huelva), together with a few shards with origins in Greece and the Central Mediterranean (González de Canales *et al*. 2004), argues in favour of a regular presence of Phoenician traders at Huelva from the start of that century. The foundation of the first Phoenician colonies in Iberia, not only on the southern coast but also on the western Atlantic coast, took place at the end of the ninth century and their *floruit* can be dated to the following century.

The Phoenician presence in Iberia seems to be strongly connected with activities related to ore and metallurgical exploitation and production (Ruiz Mata 2001; Arruda 2005; Gauss 2013; Schiavon *et al*. 2013). In these fields, several technological innovations can be ascribed to their

---

*Received 26 March 2014; accepted 2 February 2015
†Corresponding author: email pvalerio@ctn.ist.utl.pt

influence, namely the smelting of iron ores, the art of blacksmithing, the cupellation of silver, lost-wax (*cire perdue*) casting, the use of copper-based ternary alloys and true smelting furnaces; that is, a developed furnace technology with slagging instead of using ceramic crucibles on simple hearths, as was usual in the Iberian Peninsula until the arrival of this Eastern Mediterranean people. Besides these metallurgical innovations, other important technological advances can be added to the Phoenician presence, such as the potter's wheel or new architectural features and solutions. Also, the introduction of the script in Iberia must be imputed to the Phoenicians. All these technological innovations trigger the cultural change; in other words, it is with the arrival and the settlement of Phoenicians that the Iron Age, the so-called Orientalizing Period, begins in the Iberian Peninsula.

How the contacts between the indigenous populations and the Phoenician traders and colonialists took place is still a matter of debate. In the Portuguese territory, the Phoenician presence has been recorded on the southern coast (e.g., at Castro Marim, Tavira and Rocha Branca, all in Algarve), in the Sado Valley (at Alcácer do Sal, Abúl and Setúbal), in the Lisbon Peninsula and in the Lower Tagus Valley (at Lisbon, Quinta do Almaraz and Santarém), and in the Lower Mondego Valley (at Santa Olaia and Conimbriga) (Arruda 1999–2000, 2005; Maia 2003) (Fig. 1 (a)). The Phoenician presence is strong and well attested by archaeological excavations at these settlements located on the seaboard, in the estuary or in the lower river valley of the main rivers of southern and central Portugal. Santa Olaia and Abúl were founded *ex nihilo*, while at Quinta do Almaraz, Lisbon, Santarém, Alcácer do Sal and Tavira, the Phoenician implantation occurred in pre-existing indigenous settlements. Nevertheless, little is known about how its influence spread inland from these places, due to the lack of archaeological excavations and chronological determinations, namely absolute dates, concerning indigenous settlements where some ceramic or metallic Orientalizing artefacts have been found. It is, however, known that from the end of the sixth century to the beginning of the fourth century BC, during the so-called Post-Orientalizing Period (Jiménez Ávila 2008; Jiménez Ávila and Ortega Blanco 2008), technological innovations brought by Phoenicians were in use in the southwestern Iberian inland. At Alentejo, for instance, a potter's wheel and the Orientalizing metallurgy (i.e., with copper, binary and leaded bronzes) were recorded at Cabeço Redondo, a site dated to the fifth century BC with a monumental building—an aristocratic residence—located in the middle of the country (Soares 2012; Soares and Soares in press). In the same region, bronze and silver artefacts also ascribed to Orientalizing metallurgy were recorded at the Palhais necropolis, dated to the sixth to fifth centuries BC (Valério *et al.* 2013a). Nevertheless, at the settlement of Castro dos Ratinhos, located not far from the previous sites, the eighth to seventh century BC bronze metallurgy does not show any trace of Orientalizing influence (Valério *et al.* 2010), despite some pottery imports and architectural features with an Orientalizing affiliation uncovered during recent archaeological excavations (Berrocal-Rangel and Silva 2010).

The innovations imputed to the Phoenician presence are described in the literature—for instance, the ninth to eighth century BC (Barros and Soares 2004) metallic artefacts from Quinta do Almaraz display typical Orientalizing technological features (Valério *et al.* 2012)—but little is known about the introduction of those innovations in coeval settlements where the true presence of Phoenician settlers is not recorded. However, the recent discovery and excavation of a votive deposit at Moita da Ladra, Vila Franca de Xira (Monteiro and Pereira 2013), located in the upper Tagus Estuary, led to the discovery of an interesting and important material culture of the people who lived nearby and opened up the opportunity to gain an insight into the influence of the first Phoenician colonies in the neighbouring coeval indigenous settlements.

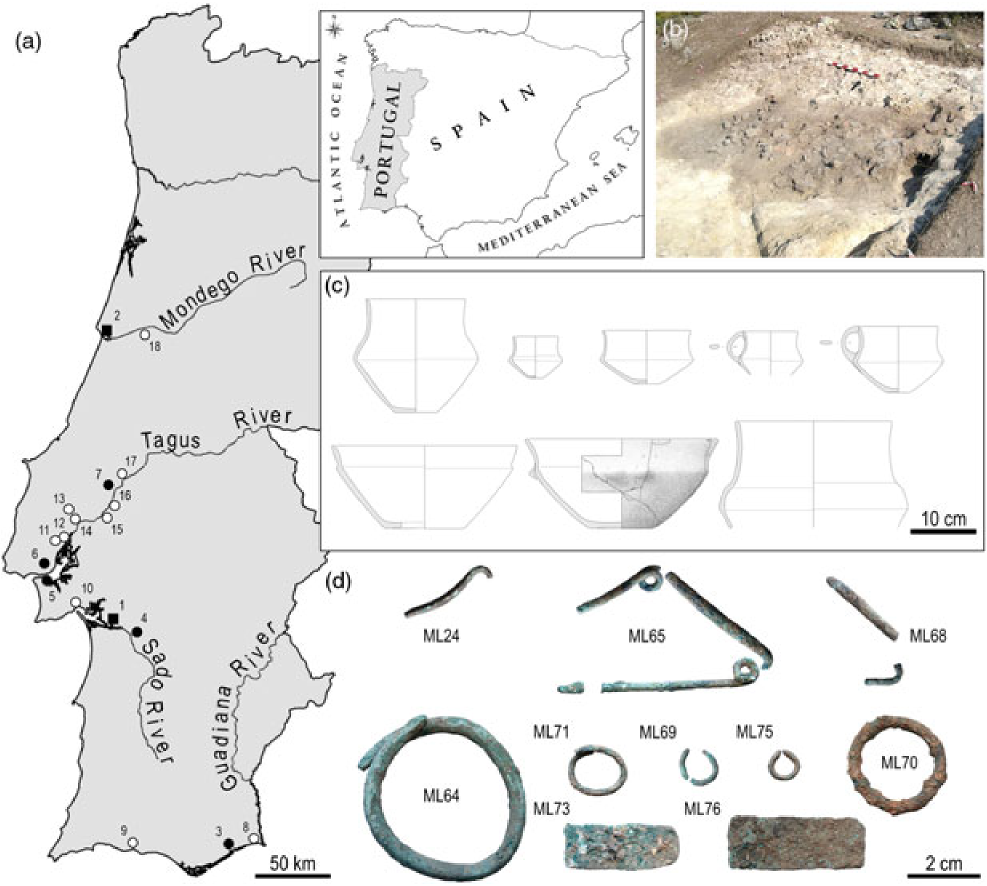

Figure 1 *(a) A map of the Portuguese territory with Early Iron Age sites marked. ■, Phoenician settlements founded ex nihilo (1, Abúl; 2, Santa Olaia); ●, indigenous settlements with a Phoenician presence (3, Tavira; 4, Alcácer do Sal; 5, Quinta do Almaraz; 6, Lisbon; 7, Santarém); ○, indigenous settlements with Orientalizing influences (8, Castro Marim; 9, Rocha Branca; 10, Setúbal; 11, Moita da Ladra; 12, Santa Sofia; 13, Castro do Amaral; 14, Quinta da Marquesa II; 15, Alto do Castelo; 16, Porto do Sabugueiro; 17, Chões de Alpompé; 18, Conimbriga). (b) The larger votive deposit at Moita da Ladra. (c) Some of the ceramic vessels. (d) Copper-based metallic artefacts recovered during the archaeological excavations.*

THE VOTIVE DEPOSIT AT MOITA DA LADRA (TAGUS ESTUARY)

A Chalcolithic walled settlement (Moita da Ladra 1) was identified and studied in the course of archaeological surveys and excavations that took place during 2003 at Moita da Ladra, an archaeological site located on the right bank of the Tagus River (Cardoso and Caninas 2010). A few remains of Early Neolithic occupation were also identified in a small area of the settlement, while Late Bronze Age (LBA) pottery was recorded in some superficial layers.

Additionally, two or three small pits were identified and excavated close to the north of this settlement, in which LBA pottery, including whole vessels, and burnt animal bones were collected. These contexts were interpreted as ritual deposits but without a funerary character (Cardoso 2013). A larger coeval votive deposit located nearby was excavated later, probably being associated with those small pits, since the pottery shapes, pastes and surface features (burnished surfaces) show great similarities. Additionally, many animal bones, most of them burnt, were recorded in all deposits. A preliminary study of this larger deposit was published recently (Monteiro and Pereira 2013).

The larger votive deposit fills a natural depression (4 m long by 3 m wide and 0.7 m deep) dug into the limestone substratum (Fig. 1 (b)). Pottery was the most abundant type of artefact recovered from this context. Although the vessels were found in a very fragmentary state, probably broken *in situ*, about 50 whole containers of various dimensions and forms were found arranged in a random way in the filling of the pit (Fig. 1 (c)).

Food remains, namely animal bones and marine shells, many of them burnt, and pieces of charcoal were also abundant. The food remains together with the fact that the most part of the ceramic ware can be totally reconstructed suggest that all these materials may be remnants of rituals of commensality. Besides pottery and food remains, some metals and a muscovite bead were also recovered during the archaeological excavation. The metals are all copper-based alloys with two exceptions, which are shapeless iron fragments. These iron remains may indicate contacts with Phoenician trade or, alternatively, may be evidence of a pre-colonial trade, since iron artefacts have been recorded in inland contexts with chronologies prior to the ninth century BC (Vilaça 2006). The copper-based metals comprise two small prills and several artefacts, some of them in a very fragmented state. The artefacts include fibulae (possibly *ad occhio* types), several rings (overlapping ends, open and closed types), two rectangular sheets and a few fragmented rods of unknown functionality (Fig. 1 (d)).

THE CHRONOLOGY OF THE VOTIVE DEPOSIT

The dimensions of the deposit and the morphologies of the pottery and the metallic artefacts point to the formation of this context in a very short period of time. Since several organic remains (mammal bones, charcoal and marine shells) were recovered during the archaeological excavations, it was possible to test this hypothesis and to obtain an absolute chronology for the deposit using the method of radiocarbon dating. The radiocarbon dates determined for this site are presented in Table 1 and Figure 2, together with radiocarbon dates determined for other Orientalizing sites located in the Lower Tagus Valley, namely Quinta do Almaraz, Santa Sofia and Santarém. As mentioned before, Quinta do Almaraz and Santarém were occupied or had strong trading ties with the Phoenicians (Arruda 1999–2000, 2005), while Santa Sofia was a settlement where some Orientalizing imports such as wheel-thrown pottery were recovered during the archaeological excavations (Pimenta and Mendes 2010–11; Pimenta *et al.* 2013).

Additional radiocarbon dates were determined for the same sampled stratigraphic units of Quinta do Almaraz, but using marine shell samples (Barros and Soares 2004). The results are statistically no different from those obtained with associated terrestrial samples, and for the sake of a better visualization of the data they are not presented here. Also, a radiocarbon date using a charcoal sample associated with the marine shell sample Sac-2865, both from Moita da Ladra, was obtained with the value 2720 ± 30 BP (Beta-356040). Nevertheless, this value must be considered as an outlier and, consequently, it is not presented in Table 1. If we apply a chi-squared test ($\chi^2_{:0.05} = T$) as the statistical criterion for the definition of contemporary samples collected in the votive deposit, we obtain the following values: $T = 4.83$; $\chi^2_{:0.05} = 7.81$ (terrestrial samples) and $T = 5.43$; $\chi^2_{:0.05} = 9.49$ (marine samples) with pooled means of 2580 ± 22 BP (804–768 cal BC) and 3034 ± 18 BP (815–729 cal BC), respectively. Thus, the hypothesis that the deposit was formed in a short time is not contradicted by the radiocarbon data. Its formation would have

Table 1 *Radiocarbon dates for Early Iron Age sites in the Tagus Valley*

Laboratory reference	*Sample*	^{14}*C age* (BP)	*Calendar date (cal* BC*)**	
			1σ	*2σ*
Quinta do Almaraz				
Sac-1655	Bones	2780 ± 70	1003–843	1110–809
Sac-1656	Bones	2710 ± 45	896–821	971–960, 936–800
ICEN-926	Bones	2660 ± 50	892–876, 847–795	915–776
Sac-1636	Charcoal	2630 ± 120	925–734, 689–662, 648–546	1027–409
ICEN-927	Bones	2570 ± 60	809–747, 685–666, 641–587, 581–556	839–507, 501–490
Sac-1362	Bones	2510 ± 50	782–733, 690–661, 649–545	798–477, 444–431
Santarém				
Beta-131488	Charcoal	2650 ± 70	900–785	979–740, 687–664, 646–549
ICEN-532	Charcoal	2640 ± 50	888–881, 844–786	917–756, 679–671, 604–598
ICEN-525	Charcoal	2470 ± 70	760–677, 674–512	776–410
Moita da Ladra				
Sac-2864	Bones	2640 ± 35	825–796	894–869, 850–778
Sac-2855	*Solen marginatus*	3090 ± 50	870–771	943–731
Sac-2854	*Venerupis decussata*	3080 ± 35	841–768	895–746
Sac-2850	Bones	2550 ± 60	801–745, 686–665, 644–551	820–482, 440–434
Sac-2866	*Chlamis varia* + *Nodipecten corallinoides*	3020 ± 35	795–732	843–647, 642–640, 632–630
Sac-2865	*Chlamis varia* + *Flexopecten flexuosus*	3000 ± 50	797–690, 682–664	830–542
Sac-2852	Bones	2540 ± 40	795–749, 684–667, 640–588, 578–566	802–727, 719–704, 695–541
Sac-2851	Bones	2540 ± 50	796–747, 685–666, 642–586, 585–555	807–513
Sac-2853	*Venerupis decussata*	2980 ± 40	779–663	796–553
Santa Sofia				
Sac-2296	*Venerupis decussata*	2950 ± 35	755–620	774–535
Sac-2295	*Venerupis decussata*	2860 ± 45	650–455	714–398

*Calendar dates were calculated using the calibration curves IntCal13 (terrestrial samples) and Marine13 (shell samples) (Reimer *et al.* 2013) and the calibration program CALIB 7.0 (Stuiver and Reimer 1993). A *ΔR* value of +95 ± 15 ^{14}C yr was used for marine samples (Soares and Dias 2006).

occurred during the first half of the eighth century BC, when the Phoenician presence in the Tagus Estuary and the Lower Valley was already well established.

The comparison of the four groups of dates obtained for the Orientalizing sites in the Tagus region show the contemporaneity of the respective four human occupations, at least at some time interval during the eighth century BC. The Phoenician occupation seems to occur first at Santarém and later at Quinta do Almaraz, considering the material culture recovered at those two sites (Arruda 1999–2000, 2005). Moita da Ladra and Santa Sofia, with only indigenous occupations, and with radiocarbon-dated archaeological contexts much probably later than the previously mentioned Phoenician occupations, received some goods from their foreign neighbours, probably the iron artefacts recorded at Moita da Ladra and certainly the wheel-thrown pottery at Santa Sofia.

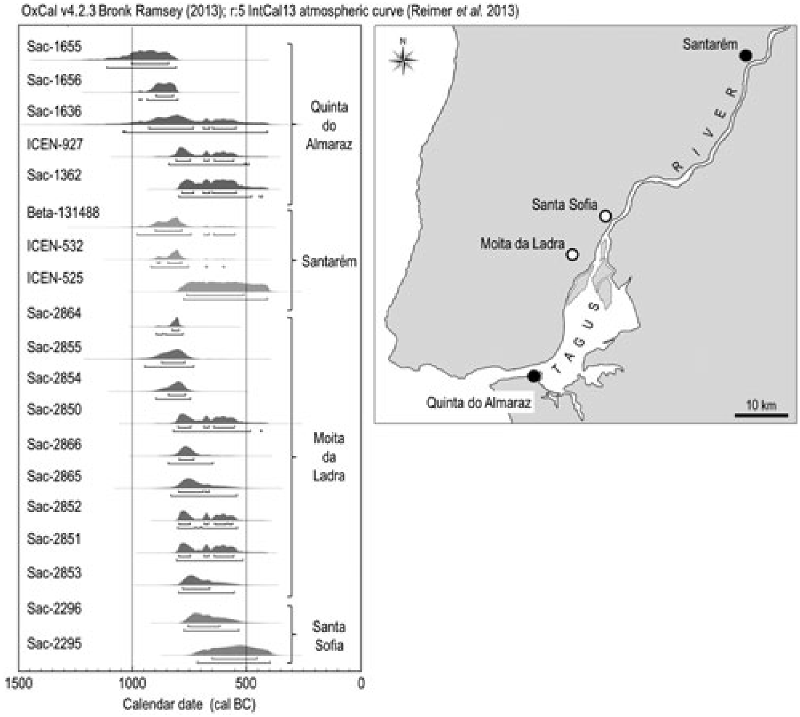

Figure 2 *Radiocarbon dates and locations of Phoenician settlements (●, Quinta do Almaraz and Santarém) and indigenous settlements (○, Moita da Ladra and Santa Sofia) in the Tagus Valley (for details of samples and calendar dates, see Table 1).*

As mentioned before, several copper-based artefacts were recovered at Moita da Ladra, but for this type of material only an elemental and microstructural study can assess the presence of technological improvements superposed on the indigenous metallurgical tradition. Therefore, in this work the technological characteristics of those copper-based artefacts will be used to evaluate how tight the Orientalizing contacts were and if any metallurgical innovation was included in them; that is, the metallurgical innovation will be used as a proxy to infer the presence or absence of cultural change induced by the Phoenician presence in this region of the western Iberian Peninsula.

EXPERIMENTAL

The metallic collection (Table 2) was first analysed by energy-dispersive X-ray fluorescence spectrometry (EDXRF), without removing the superficial corrosion layer to promptly identify the metal or alloy. Analyses were performed on a Kevex 771 spectrometer equipped with a Rh X-ray tube, secondary targets and a liquid nitrogen cooled Si(Li) detector with a resolution of 165 eV at 5.9 keV. The experimental conditions involved the Gd secondary target, 57 kV of tube voltage, 1.0 mA of current intensity and 300 s of live time. A detailed description of the analytical procedure has been published previously (Valério *et al*. 2007).

Table 2 *Metallic collection and analyses performed on artefacts from Moita da Ladra*

Type	*Description*	*Reference*	*EDXRF*	*Micro-EDXRF*	*OM*	*Vickers*
Prill	Rounded shape (0.7 cm length)	ML67	×	×	×	–
Prill	Irregular shape (1.0 cm length)	ML99	×	–	–	–
Fibula	Arched bow (ellipsoidal section)	ML24	×	×	×	×
Fibula	Arched bow (ellipsoidal section)	ML65	×	×	×	×
Fibula(?)	– (ellipsoidal section)	ML68	×	–	–	–
Ring	Overlapping ends	ML64	×	×	×	×
Ring	Overlapping ends	ML71	×	×	×	×
Ring	Open	ML69	×	×	×	×
Ring	Open	ML75	×	×	×	–
Ring	Closed	ML70	×	×	×	–
Rod	Circular section	ML63	×	×	×	×
Rod	Circular section	ML72	×	×	×	×
Rod	Ellipsoidal section	ML74	×	×	×	×
Rod	Ellipsoidal section	ML77	×	×	×	×
Rod	Ellipsoidal section	ML78	×	×	×	×
Sheet	Rectangular shape	ML73	×	×	×	×
Sheet	Rectangular shape	ML76	×	×	×	×
Fragment	–	ML66	×	–	–	–

Preparation for subsequent microanalyses involved the cutting of small sections (~2–3 mm) with a jeweller's saw. Samples were mounted in epoxy resin and polished with increasingly fine silicon carbide papers (1000, 2500 and 4000 grit size) and diamond pastes (3 μm and 1 μm). Alternatively, a small area of about ~3–5 mm diameter on the surface of the closed ring (ML70) was polished with diamond pastes (15 μm to 1 μm). The prill ML99, fibula ML68 and fragment ML66 were heavily corroded and had no traces of bulk metal for microanalysis.

The metallic surfaces were analysed with an ArtTAX Pro micro-EDXRF spectrometer equipped with a Mo X-ray tube, a focusing polycapillary lens and a silicon-drift detector with a resolution of 160 eV at 5.9 keV. Samples were analysed at three independent spots, using 40 kV of tube voltage, 0.5 mA of current intensity and 100 s of live time. Quantifications utilized fundamental parameters and experimental calibration factors calculated using the British Chemical Standards Phosphor Bronze 551 reference material. The quantification limits of significant elements are as follows: 0.5 wt% Ag, 0.5 wt% Sn, 0.5 wt% Sb, 0.2 wt% Zn, 0.10 wt% Ni, 0.10 wt% Pb, 0.10 wt% As and 0.05 wt% Fe. The accuracy is better than 2% for alloying elements such as Cu and Sn, and better than 10% for Sb, Ni, Pb, As and Fe. Additional experimental details have been published previously (Valério *et al.* 2007).

Optical microscopy observations were made using a Leica DMI 5000 M reflected light microscope, with magnifications from 50× to 1000×. A multifocus imaging mode allows accurate observation of non-mounted samples such as the closed ring ML70 (Figueiredo *et al.* 2013b). The metallic surfaces were observed after etching with an aqueous ferric chloride solution for the enhancement of microstructural features.

Vickers microhardness testing involved a Zwick-Roell Indentec with an indentation of 0.2 kg for 10 s. At least three indentations were made on each mounted artefact to obtain a relative standard deviation lower than 10%. Some of the microstructures exhibit significant intergranular corrosion, which prevented suitable testing.

RESULTS

Alloy composition

Non-invasive EDXRF analyses of the collection of metals from the votive deposit of Moita da Ladra have identified the exclusive presence of bronze alloys. The micro-EDXRF quantification of prepared samples characterized these bronze alloys with average tin contents around 11.6 ± 2.3 wt% (Table 3). The sheet ML73 is the only low-tin bronze presenting 5.0 wt% Sn and 1.19 wt% Sb. A low-tin bronze can be the result of the loss of tin by preferential oxidation during melting, so it can be explained by the use of bronze scrap. For instance, a 10 wt% tin bronze will be reduced to about 3 wt% of tin after a few recycling cycles (Sarabia 1992). However, the antimony content is also strongly and similarly affected by metallurgical operations (Tylecote *et al.* 1977); thus the higher amount of antimony in this alloy (ML73) makes the recycling option improbable. Moreover, the unusually high antimony content of this artefact suggests the use of different metal sources (ores?), which can imply importation from another region. The remaining collection presents a regular amount of tin (~12 wt%) and no significant differences were found in the alloys used for fibulas, rings or rods. Nevertheless, the two rings (ML75 and ML71) present somewhat different minor element patterns, namely higher amounts of Pb (1.37 wt%) and Ni (0.34 wt%), respectively. Nickel is a good indicator of source, as the Cu/Ni ratio will be the same for both ore and metal (Tylecote *et al.* 1977), suggesting a different source for the ring ML71.

The very low iron content (<0.05 wt%) of the analysed bronze alloys is also very significant. The importance of a low amount of iron in ancient metal was first acknowledged by comparison of LBA and Phoenician bronzes from Iberian sites, and was considered to be an indication of smelting with a poorly reducing atmosphere (Craddock and Meeks 1987). In Iberia, this circumstance is related with the use, well into the Iron Age, of smelting crucibles instead of true furnaces (Rovira and Montero Ruíz 2013).

Table 3 *The elemental composition of artefacts from Moita da Ladra (values in wt%)*

Type	*Reference*	*Cu*	*Sn*	*Pb*	*As*	*Sb*	*Ni*	*Fe*
Prill	ML67	89.3 ± 0.4	10.5 ± 0.5	0.19 ± 0.01	<0.10	n.d.*	n.d.	<0.05
Fibula	ML24	89.6 ± 0.1	10.1 ± 0.1	0.19 ± 0.05	<0.10	n.d.	n.d.	<0.05
Fibula	ML65	86.1 ± 0.1	13.8 ± 0.1	n.d.	<0.10	n.d.	n.d.	<0.05
Ring	ML64	89.7 ± 0.2	10.1 ± 0.2	0.14 ± 0.02	<0.10	n.d.	n.d.	<0.05
Ring	ML69	88.7 ± 0.1	11.0 ± 0.1	n.d.	0.21 ± 0.01	n.d.	n.d.	<0.05
Ring	ML70	86.0 ± 0.5	13.7 ± 0.4	0.28 ± 0.04	<0.10	n.d.	n.d.	<0.05
Ring	ML71	87.6 ± 0.5	12.0 ± 0.5	n.d.	n.d.	n.d.	0.34 ± 0.01	<0.05
Ring	ML75	85.7 ± 0.3	12.9 ± 0.4	1.37 ± 0.12	n.d.	n.d.	n.d.	<0.05
Rod	ML63	87.1 ± 0.1	12.8 ± 0.1	n.d.	<0.10	n.d.	n.d.	<0.05
Rod	ML72	88.0 ± 0.4	11.4 ± 0.4	0.14 ± 0.01	0.42 ± 0.01	n.d.	n.d.	<0.05
Rod	ML74	87.0 ± 0.3	13.0 ± 0.4	n.d.	n.d.	n.d.	n.d.	<0.05
Rod	ML77	86.4 ± 0.2	13.4 ± 0.2	0.14 ± 0.03	n.d.	n.d.	n.d.	<0.05
Rod	ML78	87.4 ± 0.3	12.5 ± 0.2	n.d.	n.d.	n.d.	n.d.	<0.05
Sheet	ML73	93.6 ± 0.4	5.0 ± 0.4	n.d.	<0.10	1.19 ± 0.08	n.d.	<0.05
Sheet	ML76	89.7 ± 0.6	10.1 ± 0.5	n.d.	0.11 ± 0.01	n.d.	n.d.	<0.05

* n.d., Not detected.

Microstructure

Optical microscopy observations of the small bronze prill ML67 identified a dendritic microstructure with small Cu_2S inclusions (Fig. 3). Dendrites are underlined by coring and by corrosion along interdendritic boundaries. This fine as-cast structure indicates a relatively fast cooling rate that is consistent with a metal droplet that fell during melting or pouring. The small prill ML67 together with the other one (ML99; completely corroded) appear to be the only direct evidence of prehistoric metallurgical operations somewhere in the vicinity of Moita da Ladra, which most probably involved the casting of bronze artefacts.

The microstructural characterization of bronze artefacts identified mostly α-phase matrices with abundant Cu_2S inclusions (Table 4). Vestiges of the $\alpha+\delta$ eutectoid are also present in a few cases, such as the rings ML70 and ML71 or the rods ML74 and ML78. The uncontrolled cooling rate of the molten metal in common prehistoric casting operations initiates the development of the δ phase at low tin contents; that is, alloys with ~4–5 wt% Sn may have this tin-rich phase despite being very distant from the ~14 wt% solubility limit of tin

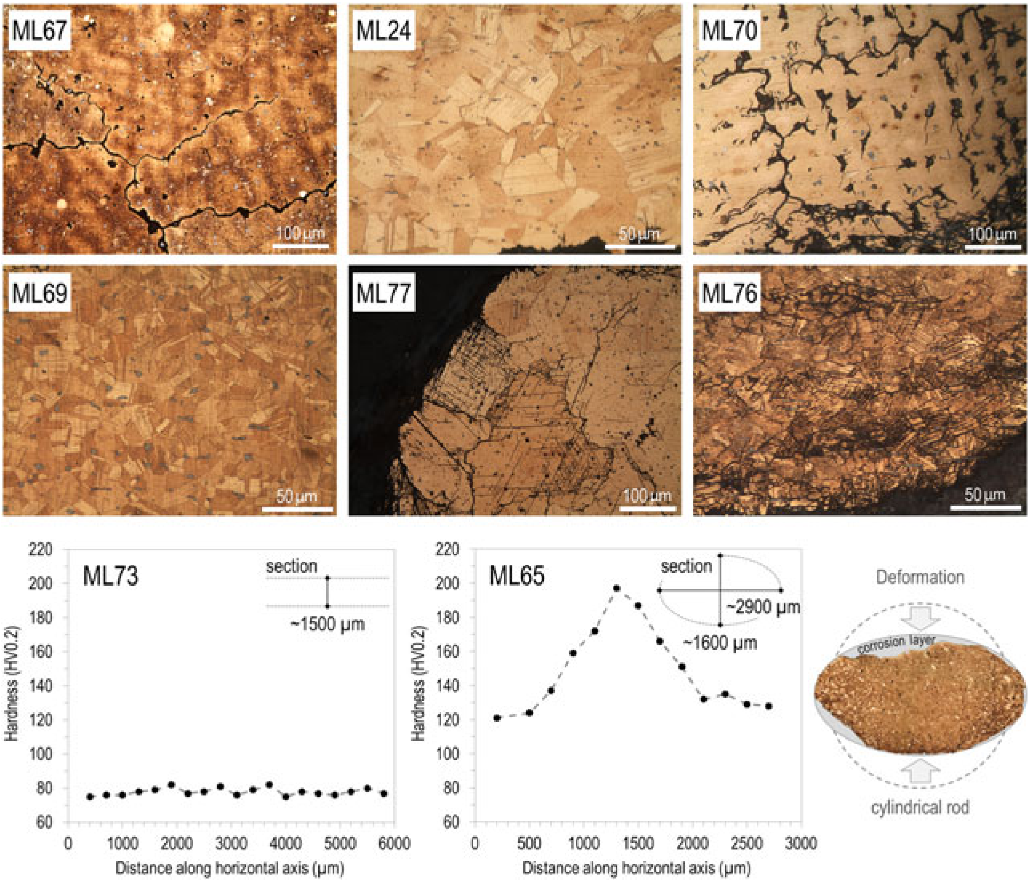

Figure 3 *The microstructures of prill ML67, fibula ML24, rings ML70 and ML69, rod ML77 and sheet ML76 (OM-BF images; etched) and hardness profiles of sheet ML73 and fibula ML65, together with the post-casting deformation proposed for this fibula (see explanation in text).*

Table 4 *Microstructural features, post-casting work and the hardness of artefacts from Moita da Ladra (wt% Sn given by micro-EDXRF)*

Type	*Reference*	*Sn*	*Phases*	*Inclusions*	*Post-casting work*	*HV0.2*
Fibula	ML24	10.1	α	Cu_2S	(F + A) + FF†	117 ± 10
Fibula	ML65	13.8	α	Cu_2S	(F + A) + FF	149 ± 25
Ring	ML64	10.1	α	Cu_2S	(F + A) + FF↓	109 ± 3
Ring	ML69	11.0	α	Cu_2S	(F + A) + FF	126 ± 6
Ring	ML70	13.7	α, α + δ	Cu_2S	F↓	–
Ring	ML71	12.0	α, α + δ	Cu_2S	F↓	126 ± 4
Ring	ML75	12.9	α	Cu_2S	(F + A) + FF↓	–
Rod	ML63	12.8	α	Cu_2S	(F + A) + FF	187 ± 8
Rod	ML72	11.4	α	Cu_2S	(F + A) + FF↓	119 ± 6
Rod	ML74	13.0	α, α + δ	Cu_2S	(F + A)↓ + FF	138 ± 4
Rod	ML77	13.4	α	Cu_2S	(F + A)↓ + FF	127 ± 7
Rod	ML78	12.5	α, α + δ	Cu_2S	(F + A) + FF↓	121 ± 4
Sheet	ML73	5.0	α	Cu_2S	(F + A) + FF↓	78 ± 2
Sheet	ML76	10.1	α	Cu_2S*	(F + A) + FF	180 ± 5

*Highly elongated.

†F, Forging; A, annealing; FF, final forging; ↓, low amount.

in copper (Hanson and Pell-Walpole 1951). The presence of the eutectoid on some artefacts illustrates less intense post-casting works that were insufficient to completely homogenize the alloy.

The manufacturing features of the two fibulas (ML24 and ML65) include deformed equiaxial grains with annealing twins and slip bands (Fig. 3). Post-casting processing of these highly valued ornaments consists of the long sequence comprising one or more cycles of hammering and annealing plus a final hammering operation. The rings (ML64, ML69, ML70, ML71 and ML75) show distinct microstructures ranging from dendritic (ML70 and ML71) to equiaxial with annealing twins and slip bands (Fig. 3). The dendritic microstructures show slip bands from mechanical work, while the post-casting work of the remaining rings involved hammering plus annealing cycles prior to a final hammering. The variable density of the slip bands indicates the differing intensity of final hammering on different artefacts. The variable post-casting sequences of these rings are not related with their typologies (overlapping ends, open or closed shapes). Instead, these are probably associated with the purpose of each artefact (e.g., finger ring, earring, connecting element etc.), which is impossible to establish due to the context of their discovery.

Rods ML63, ML72 and ML78 present deformed equiaxial microstructures with annealing twins and deformation bands. However, there are two examples (ML74 and ML77) displaying coarse microstructures with few annealing twins but many slip bands (Fig. 3). In these cases, the cycle of hammering plus annealing was less significant, but the final deformation was considerable. It is possible that these two fragmented rods constitute unfinished artefacts, thus being other possible evidence of metallurgical operations in the vicinity of Moita da Ladra. The microstructures of the rectangular sheets ML73 and ML76 (thicknesses of 1.5 mm and 1.9 mm, respectively) provide evidence of the long post-casting sequence (Fig. 3). The thinner sheet was much more worked, as evidenced by heavily elongated Cu_2S inclusions. The post-casting processing has involved efficient hammering and annealing cycles (evidenced by a smaller grain size) and a strong final hammering (a high density of slip bands, which are underlined by corrosion).

Hardness

Vickers microhardness testing on mounted samples established the hardness of the metallic artefacts from Moita da Ladra (Table 4). Despite having been work-hardened, these bronzes show quite different hardnesses (78–187 HV0.2), mostly due to the variable manufacturing conditions (i.e., tin content, intensity of final deformation, number of processing cycles and annealing parameters, such as temperature and time of operation). No significant differences were observed between typologies and, in fact, similar artefacts have distinct hardnesses. For instance, the stronger post-casting processing applied to the sheet ML76 resulted in a much harder alloy (180 HV0.2) than the low-tin bronze sheet ML73 (78 HV0.2). The hardness of the majority of these bronzes is rather constant along each section (standard deviation lower than 5%, see, for instance, the longitudinal hardness profile of sheet ML73, in Fig. 3), but the fibula ML65 shows particularly variable values (149 ± 25 HV0.2: standard deviation ~17%) that are explained by a microstructure that is also variable. The central section presents a smaller grain size, thus being harder than the lateral areas, as can be observed in the longitudinal hardness profile (Fig. 3). This feature can be explained by a higher deformation applied on the central section of a cylindrical rod to obtain the final ellipsoidal shape of this fibula.

DISCUSSION

The Phoenician trade and colonies, involving goods, people and knowledge, initiated the so-called Orientalizing metallurgy, shared by many Mediterranean regions, such as Egypt, Greece, Cyprus, Crete, Sicily and Sardinia. Overall, these regions present a diversified copper-based metallurgy comprising copper, bronze and leaded bronze artefacts. For example, the metals of mainland Greece and offshore islands during the Early Iron Age (EIA, *c*.1100–900/850 BC) and the Archaic Period (*c*.900/850–500 BC) were mostly composed of bronze with randomly variable tin content, while unalloyed coppers and leaded bronzes became increasingly more common and were often chosen for minor items and castings, respectively (Kayafa 2006).

The Orientalizing metallurgical tradition was already present in the Tagus Estuary at the time of formation of the Moita da Ladra votive deposit (eighth century BC). This is proven by metals from Orientalizing contexts of Quinta do Almaraz comprising mostly binary bronzes, but also a few unalloyed copper artefacts (a fibula, a cauldron handle, two rivets and small fragments) and leaded bronzes (a fish-hook and a pair of tweezers) (Araújo *et al*. 2004; Valério *et al*. 2012). However, the present study has revealed that the indigenous people inhabiting the immediate vicinity of Moita da Ladra during this period only seem to have used binary bronze artefacts. Moreover, the tin content of these alloys (11.6 ± 2.3 wt% Sn) is substantially different from the lower tin content of Phoenician bronzes from Quinta do Almaraz; that is, 5.4 ± 2.0 wt% Sn (Fig. 4). This indigenous bronze metallurgy with ‘good’ tin contents—that is, 10.1 ± 2.5 wt% Sn—has already been identified in the inland region of the southern Portuguese territory, namely at the eighth to seventh century BC settlement of Castro dos Ratinhos (Valério *et al*. 2010).

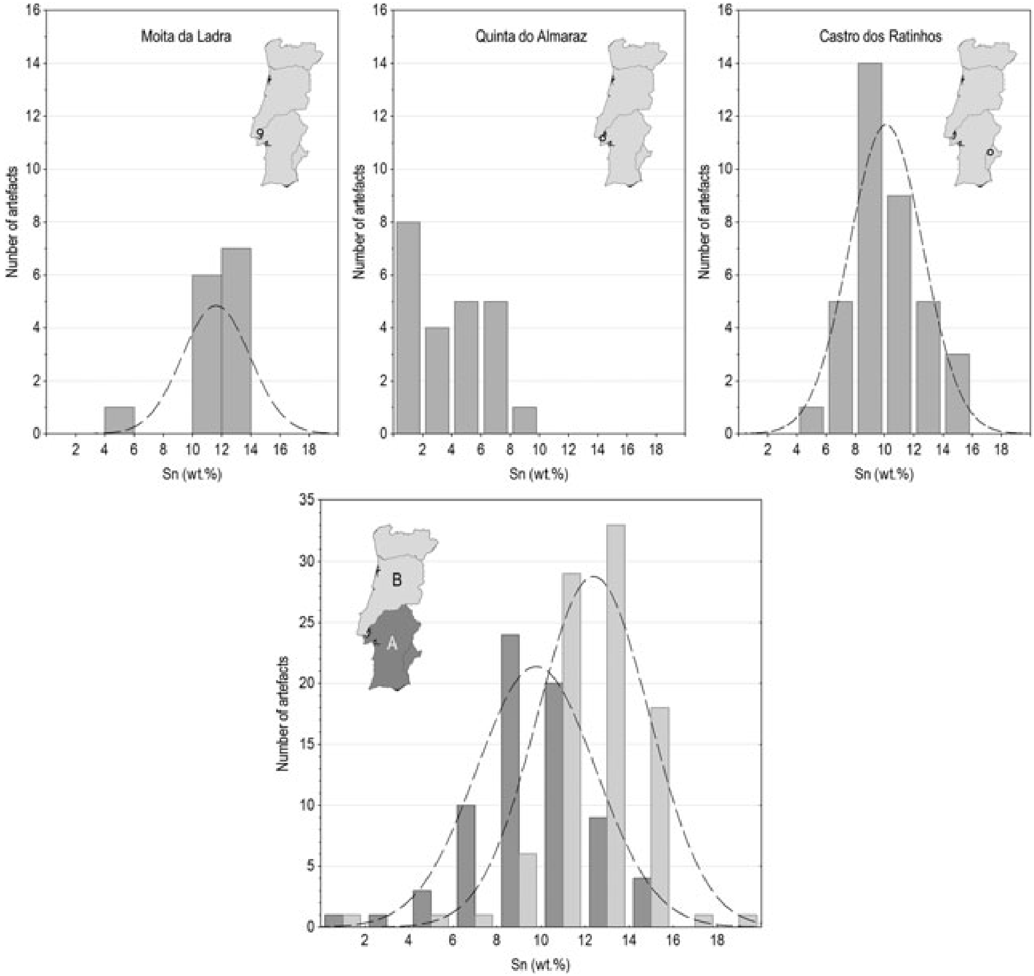

Figure 4 *Histograms of the tin contents of artefacts from Moita da Ladra, Quinta do Almaraz, Castro dos Ratinhos and the southern region (A, 72 artefacts from Castro dos Ratinhos [Valério et al. 2010], Santa Margarida, Salsa 3, Quinta do Marcelo, Baleizão, Évoramonte and Cerro da Mangancha [Valério et al. 2015], Entre Águas 5 [Valério et al. 2013c] and Outeiro do Circo [Valério et al. 2013b]) and central/northern region of the Portuguese territory (B, 91 artefacts from Moita da Ladra [this work], Baiões/Santa Luzia [Figueiredo et al. 2010], Freixanda [Gutiérrez Neira et al. 2011], Casais da Pedreira, Moinho do Raposo and Vila Cova de Perrinho [Bottaini et al. 2012] and Medronhal [Figueiredo et al. 2013a]). The dashed lines are the normal distributions of the tin content; see the averages and standard deviations in the text.*

The steady tin content of the indigenous bronzes from Castro dos Ratinhos (~10 wt%) and Moita da Ladra (~12 wt%) suggests suitable access to the sources of tin. Moreover, the alloys from Castro dos Ratinhos seem to be somewhat poorer in tin, but the reduced number of samples in these two collections hinders a proper interpretation. Therefore, considering the technological continuity of the indigenous metallurgy in the Portuguese territory during the LBA (*c*.1200–800 BC) and the EIA (*c*.800–400 BC), a comparison was made between the tin content of indigenous metals from sites in the southern and central/northern regions (Fig. 4). Similarly to the results from Castro dos Ratinhos and Moita da Ladra, the southern group also shows a lower average tin value than the central/northern set (9.7 ± 2.7 wt% Sn, *n* = 72 and 12.3 ± 2.5 wt% Sn, *n* = 91, respectively). Specific artefacts were also compared to see if this distinction could be due to the different incidence of typologies in the two collections. The rings were the only typology with a significant number of samples to allow a meaningful comparison, and the results also indicate the poorer bronze metallurgy in the southern region (10.6 ± 2.9 wt% Sn, *n* = 21 and 13.2 ± 1.4 wt% Sn, *n* = 36 for Castro dos Ratinhos and Moita da Ladra, respectively). Overall, the difference probably reflects the availability of tin, or to put it in a different perspective, it can be related to the distance to the ore sources, which are located in the central/northern region. A more difficult access to the sources of tin imply the usage of poorer bronze alloys and/or a higher utilization of scrap metal, which lowers the tin content of the alloy due to tin losses during melting. Another implication is that other possible tin sources in the neighbouring regions to the east (e.g., in the Caceres region; Rovira 2002) were not exploited extensively despite being closer to this southern region of the Portuguese territory.

Later, after the collapse of the Castro dos Ratinhos settlement, the indigenous communities in the inland southwestern Iberian Peninsula adopted the Orientalizing copper-based metallurgy. Some examples can be found in sites such as Cancho Roano, Medellín (Montero Ruíz *et al.* 2003) and El Palomar (Rovira *et al.* 2005), in the Badajoz region, or El Risco, El Torrejón de Abajo (Gómez Ramos and Rovira Lloréns 2001) and Talavera la Vieja, in the Cáceres region (Montero Ruíz and Rovira Lloréns 2006). However, the chronologies of the metals from those sites were obtained from typological comparisons, broadly belonging to the seventh to fifth centuries BC, which hinders a more precise conclusion about the progress of the Orientalizing technological innovations. As we have seen, an analogous situation happens in inland Portugal, where technological innovations brought by the Phoenicians only arise from the sixth century BC onwards.

The amounts of different types of alloy at each of those sites also depend on the availability of raw materials, artefact typology and manufacture and, of course, the metallurgical knowledge of their inhabitants. For instance, most of the fifth to fourth century BC Orientalizing copper and bronze statuettes from Alcácer do Sal present high amounts of lead, allowing better castings due to the low melting point and low solubility of lead (Schiavon *et al.* 2013). Another example was given by the Orientalizing settlement of Morro de Mezquitilla (Málaga), where most of the minor items are made of unalloyed copper, probably to avoid using the more expensive tin (Giumlia-Mair 1992).

As mentioned before, the iron content in the alloy is also a distinguishing factor between indigenous and Phoenician metallurgical traditions. Orientalizing metal presents higher values of iron—for example, Quinta do Almaraz, 0.43 ± 0.28 wt% (Valério *et al.* 2012); Palhais, 0.68 ± 0.22 wt% (Valério *et al.* 2013a); Cancho Roano, 0.29 ± 0.20 wt% (Montero Ruíz *et al.* 2003); El Palomar, 0.38 ± 0.15 wt% (Rovira *et al.* 2005); Talavera la Vieja, 0.18 ± 0.08 wt% (Montero Ruíz and Rovira Lloréns 2006); and Morro de Mezquitilla, 0.36 ± 0.33 wt% (Giumlia-Mair 1992)—when compared with the coeval indigenous metal, such as that from Castro dos Ratinhos, where the iron content is almost always very low (<0.05 wt%). At the votive deposit of Moita da Ladra, the iron content of the artefacts is also very low, including the metallic set in the indigenous LBA metallurgical tradition. The relatively high-pO_2 working atmosphere for smelting crucibles hinders the reduction of iron impurities plus their incorporation into the metallic nodules formed during smelting. This smelting method has been used since prehistoric times in Iberia and so the prehistoric metal also presents very low amounts of iron—see, for instance, the metallic nodules, prills and artefacts from the tenth to ninth century BC metallurgical workshop at Entre Águas 5 (Valério *et al.* 2013c), or from the Late Bronze Age habitat site of Castro da Senhora da Guia de Baiões (Figueiredo *et al.* 2010).

The usage of casting, forging and annealing to produce bronze artefacts has been well established since prehistoric times, and the majority of artefacts from Moita da Ladra, Castro dos Ratinhos and Quinta do Almaraz comprise the long manufacturing sequence involving hammering plus annealing cycles and a finishing hammering of variable intensity. The average hardness values from the bronzes of Moita da Ladra (131 ± 30 HV0.2, $n = 12$) are very similar to those from the indigenous bronze alloys of Castro dos Ratinhos (144 ± 30 HV0.2, $n = 7$). However, the higher hardness of a few bronzes from Quinta do Almaraz (~200–240 HV0.2) seems to demonstrate improved hammering and annealing operations in Orientalizing metallurgy even though these are low-tin alloys; that is, ~5–6 wt% (Valério *et al.* 2012). Therefore, the *chaîne opératoire* of Phoenician bronze artefacts can be more efficient despite sharing the general characteristics of the indigenous metallurgy.

CONCLUSIONS

The binary bronze artefacts from the eighth century BC votive deposit of Moita da Ladra provide evidence of the persistence of those indigenous communities in the use of technological choices established since prehistoric times. A conservative metallurgy had already been observed in the inland region of the Portuguese territory, namely at the eighth to seventh century BC settlement of Castro dos Ratinhos, following the LBA trend of an almost exclusive use of bronzes with suitable tin contents and suggesting good access to the tin sources located in the central/northern region of the Iberian Peninsula.

Inhabitants in the vicinity of Moita da Ladra surely had contacts with Phoenician traders travelling in the Tagus River Valley, between Quinta do Almaraz and Santarém. The remains of iron artefacts recovered from the votive deposit may indicate such contacts or, alternatively, may be evidence of pre-colonial trade. At Santa Sofia, about 10 km upstream, some coeval or perhaps slightly later archaeological contexts present a few shards of wheel-thrown Orientalizing pottery that clearly attest to those Phoenician contacts.

However, no trace of technological innovations related to metallurgical processes brought into the region by Orientalizing contacts was recorded at these archaeological sites. Moreover, it must be noted that the local demand for metal was probably low, since the archaeological record suggests small-scale domestic metallurgy. For instance, at Moita da Ladra the evidence of a local metallurgical production can be summarized as two small prills, probably resulting from the casting of bronze artefacts.

Given all these data, it can be concluded that the spread of Phoenician technological innovations, namely those related to metallurgy, was a very slow process. The current knowledge shows that in western Iberia it seems to have occurred during the Post-Orientalizing Period (*c.* sixth to fourth centuries BC). Until then, those technological innovations would be only implemented in places where the Phoenicians settled (e.g., Quinta do Almaraz), while in coeval indigenous settlements, such as Moita da Ladra and Santa Sofia, the Orientalizing imports (sheet ML73?) that eventually came to exist are from trade rather than local manufacturing. So it seems that over the course of two centuries, the technological innovations were restricted to colonial sites, being performed by people with an Orientalizing origin, and that it is only at a late stage of acculturation that the spread of Phoenician innovations occurs.

ACKNOWLEDGEMENTS

C2TN/IST authors gratefully acknowledge the FCT support through the UID/Multi/04349/2013 project. The work was funded by FEDER funds through the COMPETE 2020 Programme and National Funds through FCT under the project UID/CTM/50025/2013. The Department of Conservation and Restoration (Faculdade de Ciências e Tecnologia, Universidade Nova de Lisboa) is acknowledged for the use of the micro-EDXRF spectrometer.

REFERENCES

Araújo, M. F., Barros, L., Teixeira, A. C., and Melo, A. A., 2004, EDXRF study of prehistoric artefacts from Quinta do Almaraz (Cacilhas, Portugal), *Nuclear Instruments and Methods in Physics Research B*, **213**, 741–6.

Arruda, A. M., 1999–2000, *Los Fenicios en Portugal*, Cuadernos de Estudios Mediterráneos, Barcelona.

Arruda, A. M., 2005, O 1° milénio a.n.e. no Centro e no Sul de Portugal: leituras possíveis no início de um novo século, *O Arqueólogo Português*, **23**, 9–156.

Aubet, M. E., 2008, Political and economic implications of the new Phoenician chronologies, in *Beyond the homeland: markers in Phoenician chronology* (ed. C. Sagona), 247–59, Peeters, Leuven.

Barros, L., and Soares, A. M. M., 2004, Cronologia absoluta para a ocupação orientalizante da Quinta do Almaraz, no estuário do Tejo (Almada, Portugal), *O Arqueólogo Português*, **22**, 333–52.

Berrocal-Rangel, L., and Silva, A. C., 2010, *O Castro dos Ratinhos: escavações num povoado proto-histórico do Guadiana*, O Arqueólogo Português, Suplemento 6, Museu Nacional de Arqueologia, Lisboa.

Bottaini, C. E., Silva, A. L. M., Covita, D. S., Moutinho, L. M., and Veloso, J. C. F. A., 2012, Energy dispersive X-ray fluorescence analysis of archeological metal artifacts from the Final Bronze Age, *X-Ray Spectrometry*, **41**, 144–9.

Bronk Ramsey, C., 2013, Oxcal 4.2 Manual, Web interface build number: 80. Last Updated: 24/5/2014; https://c14.arch.ox.ac.uk/oxcalhelp/hlp_contents.html

Cardoso, J. L., 2013, Moita da Ladra 2 (Vila Franca de Xira), um sítio ritual do Bronze Final da Região de Lisboa, *Revista da Faculdade de Letras Ciências e Técnicas do Património*, **12**, 49–67.

Cardoso, J. L., and Caninas, J. C., 2010, Moita da Ladra (Vila Franca de Xira), resultados preliminares da escavação integral de um povoado calcolítico muralhado, in *Transformação e mudança no Centro e Sul de Portugal: o 4.° e 3.° milénios a.n.e*, 65–95, Câmara Municipal, Cascais.

Craddock, P. T., and Meeks, N. D., 1987, Iron in ancient copper, *Archaeometry*, **29**, 187–204.

Figueiredo, E., Araújo, M. F., Silva, R. J. C., and Vilaça, R., 2013a, Characterisation of a proto-historic bronze collection by micro-EDXRF, *Nuclear Instruments and Methods in Physics Research B*, **296**, 26–31.

Figueiredo, E., Silva, R. J. C., Araújo, M. F., and Braz Fernandes, F. M., 2013b, Multifocus optical microscopy applied to the study of archaeological metals, *Microscopy and Microanalysis*, **19**(5), 1248–54.

Figueiredo, E., Silva, R. J. C., Senna-Martinez, J. C., Araújo, M. F., Braz Fernandes, F. M., and Inês Vaz, J. L., 2010, Smelting and recycling evidences from the Late Bronze Age habitat site of Baiões (Viseu, Portugal), *Journal of Archaeological Science*, **37**(7), 1623–34.

Gauss, R., 2013, The development of metallurgy on the Iberian Peninsula: technological and social patterns of a long-term innovation process, in *Metal matters: innovative technologies and social change in prehistory and antiquity* (eds. S. Burmeister, S. Hansen, M. Kunst, and N. Müller-Scheessel), 209–29, Marie Leidorf, Rahden Westf.

Giumlia-Mair, A. R., 1992, The composition of copper-based small finds from a west Phoenician settlement site and from Nimrud compared with that of contemporary Mediterranean small finds, *Archaeometry*, **34**, 107–19.

Gómez Ramos, P., and Rovira Lloréns, S., 2001, Aspectos metalúrgicos de ‘El Risco’ y de ‘El Torrejón de Abajo’ (Cáceres), in *El Risco: excavación de urgencia en Sierra de Fuentes (Cáceres). 1991–1993* (eds. J. J. Enríquez Navascués, A. Rodríguez Días, and I. Pavón Sodevilla), 195–213, Editora Regional de Extremadura, Mérida.

González de Canales, F., Serrano, L., and Llompart, J., 2004, *El emporio fenicio precolonial de Huelva, ca. 900–770 a.C*, Biblioteca Nueva, Madrid.

González de Canales, F., Serrano, L., and Llompart, J., 2008, The emporium of Huelva and Phoenician chronology: present and future possibilities, in *Beyond the homeland: markers in Phoenician chronology* (ed. C. Sagona), 567–92, Peeters, Leuven.

Gutiérrez Neira, P. C., Zucchiatti, A., Montero-Ruiz, I., Vilaça, R., Bottaini, C., Gener, M., and Climent-Font, A., 2011, Late Bronze Age hoard studied by PIXE, *Nuclear Instruments and Methods in Physics Research B*, **269**, 3082–6.

Hanson, D., and Pell-Walpole, W. T., 1951, *Chill-cast tin bronzes*, Edward Arnold, London.

Jiménez Ávila, J., 2008, El final del Hierro Antiguo en el Guadiana medio, in *Sidereum Ana I: el Rio Guadiana en Época Post-Orientalizante* (ed. J. Jiménez Ávila), 101–34, Instituto de Arqueología de Mérida, Mérida.

Jiménez Ávila, J., and Ortega Blanco, J., 2008, El poblamiento en llano del Guadiana Medio durante el período post-orientalizante, in *Sidereum Ana I: el Rio Guadiana en Época Post-Orientalizante* (ed. J. Jiménez Ávila), 251–82, Instituto de Arqueología de Mérida, Mérida.

Kayafa, M., 2006, From Late Bronze Age to Early Iron Age copper metallurgy in mainland Greece and offshore Aegean Islands, in *Ancient Greece: from the Mycenaean palaces to the age of Homer* (eds. S. Deger-Jalkotzy and I. S. Lemos), 213–31, Edinburgh University Press, Edinburgh.

Maia, M., 2003, Fenícios em Tavira, in *Tavira: território e poder*, 57–72, Museu Nacional de Arqueologia, Lisboa.

Monteiro, M., and Pereira, A., 2013, Um depósito votivo da Idade do Bronze na Moita da Ladra (Vila Franca de Xira): síntese dos trabalhos realizados e resultados preliminares, *Cira Arqueologia*, **2**, 63–94.

Montero Ruíz, I., and Rovira Lloréns, S., 2006, Comentários sobres las composiciones de los metales del conjunto, in *El conjunto Orientalizante de Talavera la Vieja (Cáceres)* (ed. J. Jiménez Ávila), 109–14, Junta de Extremadura, Cáceres.

Montero Ruíz, I., Gómez Ramos, P., and Rovira, S., 2003, Aspectos de la metalurgia orientalizante en Cancho Roano, in *Cancho Roano IX: los materiales arqueológicos* (ed. S. Pérez), 195–210, Instituto de Historia, Madrid.

Pimenta, J., and Mendes, H., 2010–11, Novos dados sobre a presença fenícia no vale do Tejo: as recentes descobertas na área de Vila Franca de Xira, *Estudos Arqueológicos de Oeiras*, **18**, 591–618.

Pimenta, J., Soares, A. M., and Mendes, H., 2013, Cronologia absoluta para o povoado pré-Romano de Santa Sofia (Vila Franca de Xira), *Cira Arqueologia*, **2**, 181–94.

Reimer, P. J., Bard, E., Bayliss, A., Beck, J. W., Blackwell, P. G., Bronk Ramsey, C., Buck, C. E., Cheng, H., Edwards, R. L., Friedrich, M., Grootes, P. M., Guilderson, T. P., Haflidason, H., Hajdas, I., Hatté, C., Heaton, T. J., Hoffmann, D. L., Hogg, A. G., Hughen, K. A., Kaiser, K. F., Kromer, B., Manning, S. W., Niu, M., Reimer, R. W., Richards, D. A., Scott, E. M., Southon, J. R., Staff, R. A., Turney, C. S. M., and van der Plicht, J., 2013, IntCal13 and Marine13 radiocarbon age calibration curves, 0–50,000 years cal BP, *Radiocarbon*, **55**(4), 1869–87.

Rovira, S., 2002, Metallurgy and society in prehistoric Spain, in *Metals and society* (eds. B. S. Ottaway and E. C. Wager), 5–20, Archaeopress, Oxford.

Rovira, S., and Montero Ruíz, I., 2013, Iberia: technological development of prehistoric metallurgy, in *Metal matters: innovative technologies and social change in prehistory and antiquity* (eds. S. Burmeister, S. Hansen, M. Kunst, and N. Müller-Scheessel), 231–9, Marie Leidorf, Rahden Westf.

Rovira, S., Montero Ruíz, I., Ortega, J., and Jiménez Ávila, J., 2005, Bronce y trabajo del bronce en el poblado orientalizante de 'El Palomar' (Oliva de Mérida, Badajoz), *Anejos de AEspA*, **30**, 1231–40.

Ruiz Mata, D., 2001, Tartessos, in *Protohistoria de la Península Ibérica* (eds. M. Almagro-Gorbea, O. Arteaga, M. Blech, D. Ruiz Mata, and H. Schubart), 1–190, Ariel, Barcelona.

Sarabia, F. J., 1992, Arqueología experimental: la fundición de bronce en la prehistoria reciente, *Revista de Arqueología*, **130**, 12–22.

Schiavon, N., Celauro, A., Manso, M., Brunetti, A., and Susanna, F., 2013, Early Iron Age bronze statuettes in southern Portugal: combining archaeological data with EDXRF and BSEM + EDS to assess provenance and production technology, *Applied Physics A*, **113**(4), 865–75.

Soares, R. M., 2012, O Cabeço Redondo: um edifício da Idade do Ferro pós-Orientalizante na Herdade do Metum (Moura), M.Sc. thesis, Faculdade de Letras da Universidade de Lisboa, Lisboa.

Soares, A. M. M., and Dias, J. M. A., 2006, Coastal upwelling and radiocarbon—evidence for temporal fluctuations in ocean reservoir effect off Portugal during the Holocene, *Radiocarbon*, **48**(1), 45–60.

Soares, R. M., and Soares, A. M. M., in press, O Cabeço Redondo (Moura): um edifício monumental e singular na margem esquerda do Guadiana, in *Sidereum Ana III: el Río Guadiana y Tartessos*, Instituto de Arqueología de Mérida, Mérida.

Stuiver, M., and Reimer, P. J., 1993, Extended ^{14}C data base and revised CALIB 3.0 ^{14}C age calibration, *Radiocarbon*, **35**(1), 215–30.

Tylecote, R. F., Ghaznavi, H. A., and Boydell, P. J., 1977, Partitioning of trace elements between the ores, fluxes, slags and metal during the smelting of copper, *Journal of Archaeological Science*, **4**(4), 305–33.

Valério, P., Araújo, M. F., and Canha, A., 2007, EDXRF and micro-EDXRF studies of Late Bronze Age metallurgical productions from Canedotes (Portugal), *Nuclear Instruments and Methods in Physics Research B*, **263**(2), 477–82.

Valério, P., Silva, R. J. C., Araújo, M. F., Soares, A. M. M., and Barros, L., 2012, A multianalytical approach to study the Phoenician bronze technology in the Iberian Peninsula—a view from Quinta do Almaraz, *Materials Characterization*, **67**, 74–82.

Valério, P., Soares, A. M. M., Araújo, M. F., Silva, R. J. C., and Santos, F. J. C., 2013a, The distinctive grave goods from Palhais (Beja, Portugal): new insights into the metallurgical evolution under Orientalizing influence in the southwestern end of Iberia, *Trabajos de Prehistoria*, **70**(2), 361–71.

Valério, P., Soares, A. M. M., Araújo, M. F., Silva, R. J. C., Porfírio, E., and Serra, M., 2013b, Estudo de metais e vestígios de produção do povoado fortificado do Bronze Final do Outeiro do Circo (Beja), in *Arqueologia em Portugal: 150 anos* (eds. J. M. Arnaud, A. Martins, and C. Neves), 609–15, Associação dos Arqueólogos Portugueses, Lisboa.

Valério, P., Silva, R. J. C., Soares, A. M. M., Araújo, M. F., Braz Fernandes, F. M., Silva, A. C., and Berrocal-Rangel, L., 2010, Technological continuity in Early Iron Age bronze metallurgy at the south-western Iberian Peninsula—a sight from Castro dos Ratinhos, *Journal of Archaeological Science*, **37**(8), 1811–919.

Valério, P., Soares, A. M. M., Silva, R. J. C., Araújo, M. F., Rebelo, P., Neto, N., Santos, R., and Fontes, T., 2013c, Bronze production in southwestern Iberian Peninsula: the LBA metallurgical workshop from Entre Águas 5 (Portugal), *Journal of Archaeological Science*, **40**(1), 439–51.

Valério, P., Silva, R. J. C., Soares, A. M. M., Araújo, M. F., Gonçalves, A. P., and Soares, R. M., 2015, Combining X-ray based methods to study the protohistoric bronze technology in Western Iberia, *Nuclear Instruments and Methods in Physics Research B*, **358**, 117–23.

Vilaça, R., 2006, Artefactos de ferro em contextos do Bronze Final do território português: novos contributos e reavaliação dos dados, *Complutum*, **17**, 81–101.