Bottaini *et al. Heritage Science* (2022) 10:97
https://doi.org/10.1186/s40494-022-00736-z

Heritage Science

RESEARCH ARTICLE

Open Access

Islamic copper-based metal artefacts from the Garb al-Andalus. A multidisciplinary approach on the *Alcáçova* of Mārtulah (Mértola, South of Portugal)

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Abstract

A multidisciplinary approach has been applied to investigate the production technology of a collection of copper-based artefacts found during archaeological excavation campaigns carried out in the Almohad neighbourhood of Mārtulah, the Islamic name of modern Mértola (South of Portugal). In stark contrast to other Islamic materials found in the same site such as common and finely decorated pottery, glass, and bone artefacts, metal objects have received less attention despite the high number of artefacts recovered.

This study focuses on the chemical characterisation of 171 copper-based artefacts dating back to the 12th and the first half of the thirteenth centuries. The artefacts are daily use objects and consist of personal ornaments (earrings, rings, and casket ornaments), tools (spindles, spatulas, and oil lamp sticks) and artefacts with unknown functions. The analytical results by X-ray fluorescence Spectroscopy (XRF) provided information not only about technological issues but infer as well on the socio-economic implications of metal consumption in Islamic Mértola. Results revealed that metals were produced using a variety of Cu-based alloys, namely unalloyed copper, brasses (Cu + Zn), bronzes (Cu + Sn), and red brasses (Cu + Sn + Zn), with a variable concentration of Pb, without any apparent consistency, as a likely result of recurrent recycling and mixing scrap metals practices or use of mineral raw materials available locally.

Keywords: Islamic metallurgy, Mértola, Al-Andalus, EDXRF, Medieval metal technology

Introduction

Islamic culture provided a very important contribution to European history and modern science and technology [1, 2]. In fact, since the seventh century AD, scholars from the Muslim world stand out in virtually every field of knowledge, being at the forefront of scientific advance and technological innovation in a wide range of research fields such as astronomy, medicine, mathematics, cartography, and agriculture [3, 4].

Since the arrival of the Muslim army, led by Tariq ibn Ziyad (711 AD), who firstly crossed the Strait of Gibraltar from the North African coast, and up to the end of the thirteenth century, when the Christian Reconquest was completed in the west of al-Andalus, the Iberian Peninsula was gradually and actively involved in a climate of cultural and scientific development, generally known as Islamic Golden Age [5, 6]. Along this period of prosperity that lasted about five centuries, the Iberian Peninsula also played a central role as one of the major points of

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transmission of Islamic culture and technology to the rest of Europe.

Towns in al-Andalus like Seville, Cordoba or Granada progressively became the centre of social life and political power, also establishing themselves as commercial hubs where manufacturing activities (e.g., pottery, metalwork, tannery) and different forms of art (e.g., textiles, illuminated manuscripts, woodwork, architecture, ceramics, and metalwork) flourished. Within this scenario, Mārtulah, located at the confluence between the Guadiana River and the Oeiras creek, benefited from its strategic location as the last navigable point of the river, functioning as a commercial hub able to link the Atlantic Ocean with Northern Africa, al-Andalus and the Mediterranean Sea [7, 8] (Fig. 1).

The twelfth and thirteenth centuries, i.e., the period when the metal artefacts analysed in this paper were produced and used, were characterised by a great development in trade despite a political instability caused both by internal crises within the Islamic community and by increasing external pressure caused by the advancing Reconquest by the Christian Iberian kingdoms. From the beginning of the 2nd millennium AD, periods of political fragmentation (with the creation of the so-called *Taifas*), alternated with periods of reunification, especially under the Almoravid and Almohad dynasties. The conquest of Mértola by Portuguese king Sancho II in 1238 was a key moment in the southward advance of the Christian forces that, in 1249, with King Afonso III, put an end to the Islamic presence in southern Portugal by conquering the Algarve region.

Fig. 1 Location of Mértola (South of Portugal)

The systematic archaeological excavations carried out along more than 40 years in the urban area of the modern Mértola have shed light on this historical period, allowing to reconstruct its history and the evolution of the town since the Iron Age period and across the centuries. Important archaeological vestiges, including monumental buildings, still stand as evidence of Islamic Mārtulah. One of the most relevant excavated areas is the Almohad neighbourhood, located in the Alcazaba, i.e., the walled fortification, in an area located on the northern slope of the castle that overlooks the town.

The area where the Almohad neighbourhood is located stands on an artificial platform built on a late Roman cryptoporticus, already occupied by buildings richly decorated with mosaics during the Late Antiquity (fifth-eighth centuries). Around the twelfth century, it was used for the edification of a neighbourhood that was abandoned soon after the Christian conquest of Mértola. In the following centuries, the area served for different purposes: it was used as a Christian cemetery until the eighteenth century as a vegetable garden and since the beginning of the twentieth century as a football pitch. The metals analysed in this paper are from the excavated area, where 15 houses from the Islamic period have been dug [9] (Fig. 2).

Even though Islamic copper-based artefacts have been recurrently found at different excavations carried out in the urban area of the present-day Mértola, this paper presents, for the first time, the results of a large-scale analytical program aimed at characterising the chemical composition of a collection composed of 171 artefacts. The analytical strategy adopted in this work was oriented towards the on-site acquisition of information on the composition of copper alloys by means of portable and hand-held X-ray fluorescence spectroscopy (XRF). The aim of the research was to provide an overview of the metal production technology in the site while, at the same time, shedding light on social and economic issues related to the use of metal in Mértola during the last phase of Islamic rule.

Fig. 2 The Almohad neighbourhood. Aerial view (**A**), and detail of an archaeological area of the excavation (**B**)

Fig. 3 A selected group of metals analysed in this paper

Materials and methods

Materials

The collection of objects analysed in this paper represents a selection of the copper-based artefacts recovered to date from the Almohad neighbourhood of Mārtulah. The assemblage includes 171 artefacts that, based on their typology, can be subdivided into three different groups, namely: (a) tools (i.e., spindles, oil lamp sticks, and spatulas); (b) ornaments (i.e., rings, earrings, buckles, casket ornaments); and (c) fragmented objects of undetermined function (Fig. 3). Regardless of their typological characteristics, it is important to stress that all the artefacts analysed here come from domestic contexts and can therefore be considered objects of daily use.

Methods

The XRF equipment used was a Bruker TRACER III-SD handheld spectrometer equipped with a rhodium anode tube and a Silicon Drift Detector with a resolution of 140 eV at Mn Kα FWHM 5.9 keV. The operating conditions were 40 kV and 3 μA current with an Al/Ti filter (304.8 μm aluminium/25.4 μm titanium) and 60 s acquisition time. The spectra were acquired using the Bruker S1PXRF v.3.8.30 software and Bruker ARTAX v.5.3.0.0 software for the first spectra evaluation. For system calibration, analyses of certified copper alloys were used, namely five standards from certified reference materials [10] and three standard reference materials (National Bureau of Standards Standard Reference Material 1107, 1110 and 1113). Quantification was performed using Bruker S1CalProcess v.2.2.33 software to find the concentration of the unknown samples.

To reduce the effects of surface enrichment occurring in multi-layered objects like metals, one corrosion-free mechanically cleaned point for each artefact was analysed. Although we are aware of the fact that objects may be heterogeneous in terms of composition, we must also consider that many of the objects analysed were very small with some of the metal fragments only slightly larger than the dimensions of the XRF window. Moreover, the objects were very fragile, and we were not allowed to remove corrosion in multiple areas of each artefact. Therefore, the approach we adopted was chosen to strike a balance between curatorial concerns and the need to avoid potential skewing in the data set that would arise from exclusive reliance on surface analysis where surface segregation effects may have led to discrepancies between the surface composition and the bulk composition of an object [11].

Results and discussion

Results are summarised in Table 1. A first issue to be solved in the interpretation of data was to address the definition of the alloys. In general, the classification of copper alloy types is problematic and the decision to set a certain threshold is usually based on arbitrary options made by each researcher [12–24]. In order to better sort the collected data, we adopted the alloy nomenclature used in the analysis of a fourteenth century AD collection of metals from a Parisian workshop [21], in turn adapted from Bayley [12]. The following threshold were used as starting point to systematise information: < 2% Zn and < 3%Sn for almost pure copper; bronze is defined as an alloy with Sn higher than Zn. Zn = 3 Sn was considered a red brass (Cu + Sn + Zn). Finally, considering that the maximum metallurgical advantage in the addition of Pb is achieved at about 3% Pb [25], from this value on, an alloy was defined as leaded.

According to Fig. 4, brass is the predominant alloy used at Mértola (*c.* 74.8%), followed by red brass (*c.* 12.9%), almost pure copper (*c.* 9.4%), and bronze (*c.* 2.9%). Only 14 artefacts (*c.* 8.2%) analysed showed an amount of lead higher than 3 wt.%. A detailed analysis for each of the alloys identified at Mértola is offered in the next pages with the aim of discussing the probable causes of this variability.

Brass and leaded brass

Brass is the predominant alloy in use at Mértola. About 74% of the entire assemblage analysed in this work fall into this category, comprising 25 casket ornaments, 32 earrings, 10 oil lamp sticks, two rings, 14 spatulas, 39 spindles, and six undetermined objects. Zinc levels range between *c.* 2.3 and 22.4 wt.%, showing an average of *c.* 9.9 wt.% (Fig. 5A). The concentration of Sn does not exceed 2 wt.% in most artefacts (Fig. 5B). Lead occurs above 3 wt.% in a very small fringe of objects (6 out 128 brasses) (Fig. 5C), while impurities are generally low, not overcoming, as a rule, 0.5 wt.% (Fig. 5D).

Only one (AA-02-29) of the analysed objects had a Zn content higher than 22 wt.%, thus falling into the 22–28 wt.% Zn range, that is the typical interval for brasses produced with the method of cementation [26–28]. With the exceptions of four objects, the rest of the collection contained Zn between 4 wt.% and 20 wt.%, that is the range that Craddock considers as typical for objects produced through the mixing of pristine brass with 22 to 28 wt.% Zn with scrap copper-based alloys with lower Zn content [26].

According to P. Craddock [26], when a brass is remelted, the alloy progressively loses about 10 wt.% of its Zn content, and a 4 to 5 wt.% additional Zn should be added to compensate the melting losses. Considering, for instance, the recycling of an ancient brass produced via the so-called cementation process and containing 28% Zn, Zn content may drop to about 25 wt.% after the first remelting, to 22 wt.% after a further remelting, and so on. This means that the moderate Zn level observed in Mértola's brass objects is most likely the result of multiple remelting of scrap metal composed, in turn, of alloys with varying Zn content.

Another point to highlight in this group is the presence of a small cluster of brass artefacts containing just over 20 wt.% Zn. These may be brasses that have undergone two or three remelting cycles or, as an alternative, may have been produced by cementation throughout the so-called medieval method. This involves the reaction of zinc vapour with liquid rather than solid copper, at higher temperatures and in open vessels. The use of this method at a temperature of about 1200 °C makes it possible to produce a brass with about 20 wt.% Zn [29]. Considering that the maximum zinc uptake in cementation process occurs at about 930 °C [30], the fact that higher temperatures could have been reached may also be an indication of the difficulty encountered by metalworkers who produced the artefacts in use in Mértola in properly controlling the temperature inside the crucibles during the cementation process.

As outlined above, the data also suggest that the addition of lead in brass was not a common practice. The variability of Pb does not seem to have any significant correlation with Zn concentration. For example, the highest amounts of Pb were found both in brasses with low Zn content, i.e., an earring (BC-03-120) with 4.1 wt.% Zn and a spindle (PF-02-130) with 5.83 wt.% Zn, and in higher Zn brasses, i.e., an oil lamp stick (EC-01-14) with 19.69 wt.% Zn, and a casket ornament (AA-02-56) with 14.75 wt.% Zn.

Finally, Fig. 6A also confirms the lack of correlation between Zn variability and the functionality of the artefacts, particularly between ornaments and tools. For example, a Zn content in the range of 10–20 wt.% is known to be responsible for a golden yellow colour in the final alloy, making the latter particularly suitable for ornamental objects. When looking at Mértola's data, though, the 50 brasses artefacts that fall in this range are evenly distributed between tools and ornaments (Fig. 6B), suggesting that the brightness and colour nature of an alloy was probably not considered relevant in producing finished objects with specific forms and functions.

Another point on which the p-XRF data on brasses allow to shed light is about the type of metal ore used for zinc. In the case of brass produced by cementation, we know that this process relies upon the use of zinc oxide mixed with copper metal and that two main types of zinc ore were usually employed, namely carbonate ore (smithsonite) and sulphide ore (sphalerite). The former was the most widely used source of zinc in Europe, while the latter was more common in the eastern Mediterranean, probably originating in northern Anatolia [15]. The use of one mineral rather than the other implied adopting

Table 1 Elemental composition of the artefacts from the Almohad neighbourhood of Mārtulah (wt.%); n.d.: not detected

ID	Functionality	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
Brass										
AA-01-02	Casket ornament	91.90	4.95	1.4	0.75	0.3	0.46	0.07	0.02	0.15
AA-01-04	Casket ornament	90.30	6.6	1.6	0.93	0.07	0.2	0.09	0.01	0.2
AA-01-06	Casket ornament	83.75	10.7	2.3	1.47	0.5	0.25	0.85	0.03	0.15
AA-01-08	Casket ornament	84.40	11.0	3.0	1.05	0.15	0.17	0.1	0.03	0.1
AA-01-13	Casket ornament	86.75	12.4	0.48	0.08	0.19	0.1	n.d	n.d	n.d
AA-01-14	Casket ornament	89.35	8.35	0.9	0.4	0.35	0.4	0.09	0.01	0.15
AA-01-16	Casket ornament	91.00	7.2	0.33	0.17	0.3	0.9	0.06	0.01	0.03
AA-01-17	Casket ornament	89.30	8.75	1.2	0.22	0.02	0.42	0.09	n.d	n.d
AA-01-18	Casket ornament	88.60	7.4	2.1	1.1	0.05	0.32	0.08	0.15	0.2
AA-01-20	Casket ornament	88.70	8.45	0.5	0.9	0.3	1.15	n.d	n.d	n.d
AA-02-25	Casket ornament	86.45	10.4	0.6	0.95	0.32	0.8	0.48	n.d	n.d
AA-02-26	Casket ornament	86.91	10.34	0.91	1.15	0.22	0.15	0.32	n.d	n.d
AA-02-27	Casket ornament	88.12	7.9	1.4	1.55	0.25	0.23	0.26	0.02	0.27
AA-02-28	Casket ornament	93.05	4.62	0.7	0.75	0.29	0.15	0.29	n.d	0.15
AA-02-29	Casket ornament	77.31	22.37	n.d	0.1	0.02	0.13	n.d	0.07	n.d
AA-02-30	Casket ornament	83.92	14.52	n.d	1.1	0.26	0.2	n.d	n.d	n.d
AA-02-32	Casket ornament	83.56	14.74	1.1	0.15	0.23	0.2	n.d	0.02	n.d
AA-02-33	Casket ornament	78.31	20.3	1.1	0.05	0.03	0.12	0.06	0.03	n.d
AA-02-36	Casket ornament	86.15	12.07	1.25	0.3	0.1	0.13	n.d	n.d	n.d
AA-02-37	Casket ornament	86.57	10.2	1.7	1.0	0.1	0.19	0.07	0.02	0.15
AA-02-46	Casket ornament	90.46	6.52	1.37	0.95	0.2	0.24	0.07	0.02	0.17
AA-02-54	Casket ornament	88.80	9.95	n.d	0.15	0.08	0.7	0.11	0.17	0.04
AA-03-62	Casket ornament	81.93	14.11	1.4	1.35	0.11	0.9	0.2	n.d	n.d
AA-03-79	Casket ornament	84.52	10.39	3.3	1.4	0.17	0.2	0 n.d	0.02	n.d
AN-01-36	Finger ring	87.51	10.5	0.4	0.55	0.27	0.7	0.07	n.d	n.d
BC-01-06	Earring	91.04	7.07	0.7	0.4	0.05	0.66	0.08	n.d	n.d
BC-01-07	Earring	93.41	4.75	0.65	0.6	0.06	0.15	0.14	0.01	0.23
BC-01-09	Earring	91.40	5.6	1.95	0.57	0.06	0.15	0.1	n.d	0.17
BC-01-11	Earring	89.79	7.98	1.35	0.21	0.03	0.55	0.09	n.d	n.d
BC-01-16	Earring	88.53	9.41	1.05	0.45	0.1	0.31	0.15	n.d	n.d
BC-01-17	Earring	82.88	14.12	1.35	0.65	0.2	0.8	n.d	n.d	n.d
BC-01-18	Earring	85.47	13.08	0.85	0.15	0.07	0.25	0.13	n.d	n.d
BC-01-19	Earring	88.67	9.23	0.65	0.22	0.06	1.1	0.07	n.d	n.d
BC-01-21	Earring	82.98	14.72	1.2	0.25	0.05	0.8	n.d	n.d	n.d
BC-01-26	Earring	86.96	11.55	0.75	0.1	0.06	0.5	0.08	n.d	n.d
BC-01-30	Earring	92.38	4.98	1.16	0.95	0.12	0.4	n.d	0.01	n.d
BC-01-42	Earring	88.27	8.71	0.9	0.48	0.14	1.5	n.d	n.d	n.d
BC-01-46	Earring	91.79	5.76	0.88	0.62	0.1	0.85	n.d	n.d	n.d
BC-01-55	Earring	88.69	7.73	1.85	0.85	0.2	0.4	n.d	0.02	0.26
BC-01-61	Earring	90.92	5.75	1.35	1.0	0.26	0.35	0.11	0.02	0.24
BC-01-62	Earring	87.33	10.32	1.15	0.21	0.03	0.9	0.06	n.d	n.d
BC-01-66	Earring	90.56	5.61	1.8	0.95	0.25	0.55	0.1	n.d	0.18
BC-01-72	Earring	86.07	11.43	1.3	0.5	0.02	0.6	0.07	0.01	n.d
BC-01-76	Earring	91.58	6.44	0.75	0.6	0.04	0.25	0.15	0.01	0.18
BC-01-89	Earring	84.81	12.14	1.63	0.68	0.06	0.35	0.09	0.01	0.23
BC-03-100	Earring	86.64	8.63	1.3	2.3	0.55	0.3	0.09	0.01	0.18
BC-03-101	Earring	83.48	14.74	1.55	0.05	0.03	0.15	n.d	n.d	n.d
BC-03-105	Earring	88.35	9	1.25	0.6	0.2	0.3	0.09	0.01	0.2

Table 1 (continued)

ID	Functionality	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
BC-03-106	Earring	87.23	9.42	1.7	1.05	0.05	0.3	0.07	0.01	0.17
BC-03-108	Earring	83.68	14.32	0.87	0.38	0.1	0.65	n.d	n.d	n.d
BC-03-109	Earring	88.55	8.19	1.15	1.4	0.25	0.3	0.16	n.d	n.d
BC-03-115	Earring	90.60	7.6	0.48	0.6	0.13	0.33	0.08	0.01	0.17
BC-03-118	Earring	83.57	10.94	2.34	2.25	0.32	0.5	n.d	0.08	n.d
BC-03-97	Earring	91.97	5.1	1.3	0.65	0.19	0.7	0.09	n.d	n.d
BC-03-98	Earring	83.50	15.1	1.17	0.05	0.04	0.08	n.d	0.06	n.d
AR-01-35	Ring	85.22	12.07	1.38	0.8	0.26	0.22	n.d	0.05	n.d
DV-02-44	Undetermined	93.28	4.19	0.9	0.25	0.21	0.74	0.43	n.d	n.d
DV-02-75	Undetermined	89.8	7.65	0.95	0.85	0.09	0.47	0.06	0.01	0.12
DV-02-80	Undetermined	89.87	6.45	1.3	1.93	0.12	0.22	n.d	0.01	0.1
DV-03-113	Undetermined	93.98	2.27	0.8	1.35	0.18	0.6	0.16	0.07	0.59
DV-03-86	Undetermined	88.30	8.9	1.55	0.48	0.03	0.48	0.08	0.02	0.16
DV-03-89	Undetermined	89.46	8.1	0.95	0.47	0.35	0.45	0.07	0.02	0.13
EC-01-01	Oil Lamp stick	85.62	8.73	2.12	2.75	0.25	0.12	0.14	n.d	0.27
EC-01-05	Oil lamp stick	87.86	11.65	n.d	0.3	0.07	0.12	n.d	n.d	n.d
EC-01-06	Oil lamp stick	87.76	8.35	2.35	0.9	0.07	0.32	0.11	0.01	0.13
EC-01-07	Oil lamp stick	85.70	14.05	n.d	0.1	0.07	0.03	0.05	n.d	n.d
EC-01-08	Oil lamp stick	82.46	12.62	2.75	1.25	0.05	0.12	0.4	n.d	0.35
EC-01-09	Oil lamp stick	89.74	6.94	1.65	1.15	0.25	0.21	0.06	n.d	n.d
EC-01-12	Oil lamp stick	85.54	12.43	0.1	1.07	0.2	0.47	0.14	0.05	n.d
EC-01-15	Oil lamp stick	88.13	10.67	n.d	0.55	0.03	0.1	0.21	0.31	n.d
EC-01-16	Oil lamp stick	88.89	8.7	0.6	0.38	0.35	0.78	0.14	0.01	0.15
ES-01-01	Spatula	85.69	13.7	0.01	0.5	0.03	0.07	n.d	n.d	n.d
ES-01-06	Spatula	92.77	5.18	0.3	0.54	0.14	0.9	n.d	0.02	0.15
ES-01-08	Spatula	91.43	4.77	1.38	0.95	0.87	0.31	0.26	0.03	n.d
ES-01-11	Spatula	94.88	2.52	0.49	0.43	0.16	1.3	n.d	0.01	0.21
ES-01-12	Spatula	87.94	7.33	2.02	1.55	0.21	0.58	n.d	0.03	0.34
ES-01-20	Spatula	89.03	8.07	0.85	0.77	0.2	0.71	0.14	0.05	0.18
ES-01-22	Spatula	88.14	7.68	1.55	1.7	0.15	0.44	0.12	n.d	0.22
ES-02-23	Spatula	88.66	9.67	0.5	0.51	0.04	0.62	n.d	n.d	n.d
ES-02-26	Spatula	86.74	9.39	2.3	0.81	0.04	0.32	0.13	n.d	0.27
ES-02-27	Spatula	89.26	8.41	0.74	0.28	0.1	1.2	n.d	0.01	n.d
ES-02-29	Spatula	85.99	10.63	1.4	1.1	0.15	0.4	n.d	0.03	0.3
ES-02-30	Spatula	88.77	7.63	0.83	1.8	0.12	0.4	0.2	0.03	0.22
ES-02-36	Spatula	88.22	8.05	2.5	0.58	0.25	0.26	0.14	n.d	n.d
ES-02-54	Spatula	93.94	4.49	0.24	0.61	0.21	0.3	0.2	0.01	n.d
PF-01-02	Spindle	88.39	7.61	1.41	0.9	0.06	1.35	0.27	0.01	n.d
PF-01-06	Spindle	90.36	7.9	0.38	0.62	0.2	0.14	0.15	0.04	0.21
PF-01-100	Spindle	93.10	3.67	1.55	0.52	0.35	0.23	0.27	0.01	0.3
PF-01-104	Spindle	89.45	8.2	0.61	0.66	0.4	0.4	0.11	0.01	0.16
PF-01-106	Spindle	84.99	12.06	0.85	0.88	0.16	1.05	n.d	0.01	n.d
PF-01-107	Spindle	85.23	14.08	0.03	0.32	0.13	0.14	0.07	n.d	n.d
PF-01-11	Spindle	86.88	11.59	0.43	0.25	0.12	0.72	n.d	0.01	n.d
PF-01-16	Spindle	88.66	9.23	1.03	0.13	0.15	0.8	n.d	n.d	n.d
PF-01-20	Spindle	85.81	11.58	1.42	0.32	0.07	0.8	n.d	n.d	n.d
PF-01-27	Spindle	88.54	9.28	0.4	0.65	0.13	0.86	0.14	n.d	n.d
PF-01-30	Spindle	77.77	17.61	3.68	0.74	0.09	0.11	n.d	n.d	n.d
PF-01-31	Spindle	74.97	20.58	1.1	2.2	0.14	0.52	0.18	0.03	0.28

Table 1 (continued)

ID	Functionality	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
PF-01-32	Spindle	80.91	15.48	1.9	0.4	0.16	1.15	n.d	n.d	n.d
PF-01-33	Spindle	91.05	6.67	0.6	0.2	0.05	1.35	0.08	n.d	n.d
PF-01-36	Spindle	81.16	15.78	1.66	0.2	0.23	0.83	0.13	0.01	n.d
PF-01-37	Spindle	83.62	13.83	1.0	0.4	0.25	0.79	0.11	n.d	n.d
PF-01-38	Spindle	80.97	15.44	0.14	2.9	0.11	0.2	0.24	n.d	n.d
PF-01-47	Spindle	93.09	4.9	0.7	0.79	0.28	0.11	0.13	n.d	n.d
PF-01-54	Spindle	83.75	14.87	0.29	0.17	0.07	0.78	n.d	n.d	0.07
PF-01-65	Spindle	87.95	9.26	1.2	0.32	0.04	1.15	0.07	0.01	n.d
PF-01-77	Spindle	84.43	11.9	1.45	1.7	0.18	0.3	n.d	0.04	n.d
PF-01-81	Spindle	92.73	4.39	1.15	0.85	0.19	0.55	0.14	n.d	n.d
PF-01-85	Spindle	89.42	6.98	0.83	0.88	0.58	1.2	0.1	0.01	n.d
PF-02-114	Spindle	94.92	2.68	0.56	1.5	0.09	0.25	n.d	n.d	n.d
PF-02-122	Spindle	79.28	19.83	0.11	0.42	0.08	0.28	n.d	n.d	n.d
PF-02-128	Spindle	84.22	10.78	2.97	1.15	0.48	0.34	n.d	0.06	n.d
PF-02-129	Spindle	88.29	8.7	1.65	0.84	0.24	0.16	0.12	n.d	n.d
PF-02-131	Spindle	88.57	8.95	1.7	0.41	0.07	0.19	0.08	0.03	n.d
PF-02-156	Spindle	84.17	13.09	1.1	0.37	0.15	0.88	0.24	n.d	n.d
PF-02-161	Spindle	84.89	11.74	2.19	0.75	0.15	0.26	n.d	0.02	n.d
PF-02-179	Spindle	91.47	6.86	0.88	0.43	0.03	0.14	n.d	0.19	n.d
PF-02-182	Spindle	86.72	9.23	1.81	1.72	0.2	0.26	n.d	0.06	n.d
PF-02-183	Spindle	88.63	9.67	0.63	0.22	0.1	0.67	0.08	n.d	n.d
PF-02-185	Spindle	84.86	13.43	1.26	0.15	0.06	0.13	0.05	0.06	n.d
PF-02-223	Spindle	90.63	5.92	1.74	0.95	0.05	0.38	0.11	0.02	0.2
PF-02-231	Spindle	87.98	8.25	2.06	1.03	0.1	0.14	n.d	0.44	n.d
PF-02-232	Spindle	82.17	14.86	1.74	0.73	0.2	0.25	n.d	0.05	n.d
Leaded brass										
BC-01-81	Earring	85.07	9.78	0.12	3.7	0.13	1.05	0.15	n.d	n.d
AA-02-56	Casket ornament	71.20	14.75	1.75	10.9	0.32	0.42	0.2	0.02	0.44
PF-02-130	Spindle	85.54	5.83	1.93	5.74	0.4	0.36	0.2	n.d	n.d
EC-01-14	Oil lamp stick	75.84	19.69	0.07	4.19	0.07	0.12	n.d	0.02	n.d
BC-03-120	Earring	90.50	4.1	0.4	3.75	0.2	0.7	0.35	n.d	n.d
PF-01-08	Spindle	69.34	21.27	2.33	5.9	0.17	0.77	0.15	0.07	n.d
Bronze										
BC-01-70	Earring	94.99	0.08	4.25	0.4	0.06	0.2	n.d	0.02	n.d
DV-02-47	Undetermined	91.32	0.15	5.41	1.16	0.25	0.142	1.56	0.01	n.d
DV-02-64	Undetermined	90.54	0.06	9.09	n.d	0.11	0.14	0.06	n.d	n.d
PF-01-01	Spindle	91.34	1.54	4.63	1.1	0.2	0.61	0.17	n.d	0.41
Leaded bronze										
AR-01-53	Ring	75.13	2.12	9.75	11.05	0.55	0.8	n.d	0.05	0.55
Copper										
AA-02-43	Casket ornament	99.52	0.08	n.d	0.21	0.01	0.15	n.d	0.03	n.d
AA-03-59	Casket ornament	96.45	1.9	0.26	0.65	0.07	0.16	0.15	0.06	0.3
AA-03-66	Casket ornament	97.05	0.13	1.3	0.15	0.29	0.65	0.05	0.08	0.3
BC-03-117	Earring	96.13	1.58	0.87	0.45	0.23	0.63	n.d	0.11	n.d
FIV-01-12	Buckle	98.95	0.09	n.d	0.64	0.05	0.04	0.09	0.01	0.13
AR-01-42	Ring	96.63	0.05	0.63	2.3	0.03	0.25	0.11	n.d	n.d
DV-01-04	Undetermined	98.00	0.09	0.1	0.63	0.13	0.09	0.62	0.04	0.3
DV-01-05	Undetermined	98.92	0.1	n.d	0.03	0.25	0.13	0.57	n.d	n.d
DV-01-19	Undetermined	96.48	0.09	0.49	2.3	0.2	0.18	0.09	0.09	0.08

Table 1 (continued)

ID	Functionality	Cu	Zn	Sn	Pb	Fe	As	Ag	Ni	Sb
DV-01-36	Undetermined	98.55	0.09	n.d	0.33	0.1	0.06	0.66	0.02	0.19
DV-02-53	Undetermined	97.70	0.1	0.95	0.8	0.02	0.14	0.11	0.02	0.16
DV-02-54	Undetermined	96.44	1.77	0.85	0.35	0.17	0.24	n.d	n.d	0.18
DV-02-79	Undetermined	98.07	0.07	0.26	0.8	0.04	0.31	0.25	0.02	0.18
PF-01-78	Spindle	91.94	1.94	2.93	0.79	0.2	0.26	1.94	n.d	n.d
PR-02-13	Nail	98.55	0.25	0.07	0.66	0.23	0.2	n.d	0.04	n.d
Leaded copper										
PR-02-06	Nail	93.62	0.11	0.15	4.85	0.24	0.46	0.16	0.03	0.38
Red brass										
PF-01-44	Spindle	92.88	2.24	1.14	2.85	0.04	0.25	0.21	n.d	0.39
AA-01-21	Casket ornament	88.10	3.7	3.05	4.0	0.5	0.37	0.09	0.02	0.17
AA-02-39	Casket ornament	84.68	7.33	5.3	2.15	0.24	0.07	0.1	0.01	0.12
AA-02-49	Casket ornament	87.92	7.76	3.4	0.34	0.5	0.08	n.d	n.d	n.d
AN-01-35	Finger ring	88.52	7.48	2.64	0.5	0.05	0.55	0.08	0.01	0.17
BC-01-05	Earring	94.32	2.47	1.16	1.4	0.1	0.55	n.d	n.d	n.d
BC-01-12	Earring	89.71	5.45	3.65	0.6	0.02	0.27	0.09	n.d	0.21
BC-01-82	Earring	90.13	4.81	2.41	1.25	0.5	0.9	n.d	n.d	n.d
BC-03-116	Earring	86.52	6.27	4.85	1.15	0.2	1.0	n.d	0.01	n.d
PF-01-03	Spindle	89.18	6.13	2.82	0.78	0.15	0.59	0.1	n.d	0.25
PF-01-04	Spindle	90.93	5.08	2.03	0.49	0.15	0.5	0.6	0.01	0.21
PF-01-05	Spindle	91.77	4.23	2.25	0.86	0.14	0.37	0.37	0.01	n.d
PF-01-55	Spindle	87.74	7.03	3.35	1.1	0.29	0.24	0.25	n.d	n.d
PF-01-99	Spindle	90.06	4.15	3.8	1.25	0.15	0.18	0.12	0.02	0.27
PF-02-178	Spindle	87.33	8.06	2.9	1.18	0.15	0.3	n.d	0.08	n.d
PF-02-230	Spindle	91.89	3.33	1.62	2.1	0.3	0.51	0.24	0.01	n.d
Leaded red brass										
BC-01-86	Earring	68.74	18.08	6.25	6.3	0.09	0.3	0.21	0.03	n.d
ES-01-10	Spatula	89.85	2.1	1.25	6.2	0.08	0.52	n.d	n.d	n.d
DV-01-31	Undetermined	82.25	9.3	4.3	3.6	0.15	0.1	0.09	0.01	0.2
PF-01-14	Spindle	82.98	5.71	4.0	6.6	0.51	0.2	n.d	n.d	n.d
DV-02-63	Undetermined	82.71	4.96	2.12	7.85	0.4	0.64	0.2	0.1	1.02
DV-03-95	Undetermined	86.88	3.06	1.71	6.5	0.58	0.45	0.13	0.15	0.54

different operational chains. Whilst smithsonite was mixed with copper metal and charcoal and heated in a crucible to about 1000 °C, allowing the Zn vapour to diffuse into the Cu, the use of sphalerite to make brass required a more complex pre-treatment process. In fact, ore needed to be roasted at high temperature to drive away the sulphur. As a consequence, smithsonite resulted in zinc with more impurities than sphalerite, namely Fe, Pb, and Mn (unless these elements occured in the copper) [15, 16, 31].

The data discussed here display no trace of Mn and relatively little iron for all the type of alloys, with the majority results lying below 0.2 wt.% Fe (Fig. 7A). Given that the amount of iron in smithsonite brass is expected to range from 0.2 to 0.5 wt.% and more [11, 15], and also considering that no particular trend has been observed in the decrease of iron in higher zinc brasses (Fig. 7B), the possible use of sphalerite to produce brass alloys found in Mértola is not to be discarded.

In addition to technological issues (low iron concentration), two other points seem to be in favour of the use of sphalerite as a zinc ore. Firstly, although more common in eastern Mediterranean, the use of sphalerite is not unprecedent in medieval European metallurgy. A collection of metals from Leopoli-Cencelle (central Italy), contemporary with the objects from Mértola analysed here, were recently analysed and the data, characterised by a moderate Zn amount and the reduced levels of Fe, Sn, and Pb, were considered compatible with the use of sphalerite [20]. At the same time, the concentration of

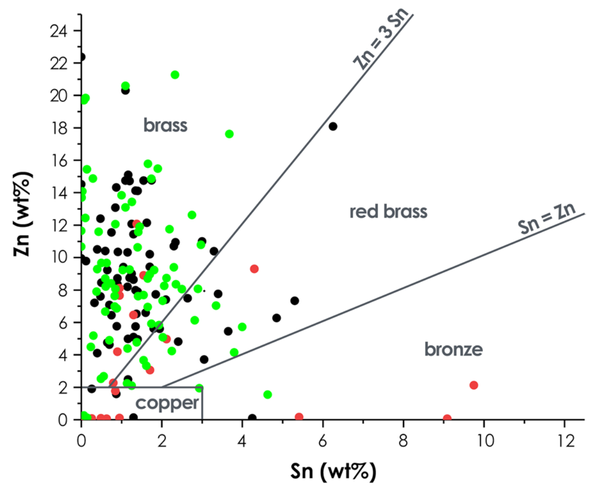

Fig. 4 Zinc and tin contents in all the artefacts analyse from Mértola sorted according to the type of alloy. The lines define the areas of each type of alloy, i.e., brass, copper, red brass, and bronze

iron in Mértola's metal objects is much lower than, for example, iron in an assemblage of copper-based artefacts from a fourteenth century AD Parisian workshop recently analysed, where this element is much higher, pointing out for the use of smithsonite as the zinc ore [21]. Secondly, it is worth remembering that sphalerite is a zinc ore well-known in the Iberian Pyrite Belt, a metallogenic province located in the SW Iberian Peninsula [32–34] and could therefore be easily exploited by local communities as source of zinc. In this case, the technology for brass production using sphalerite may have been passed on to local metalworkers by Islamic craftsmen who moved to al-Andalus.

Bronze (Cu + Sn) and leaded bronze (Cu + Sn + Pb)

Binary bronzes consist of only five objects. Similar to the other alloys discussed so far, also bronze was not used to produce a specific type of artefact. Altogether, one earring, two undetermined objects, one ring and one spindle were made of bronze. The Sn content is variable,

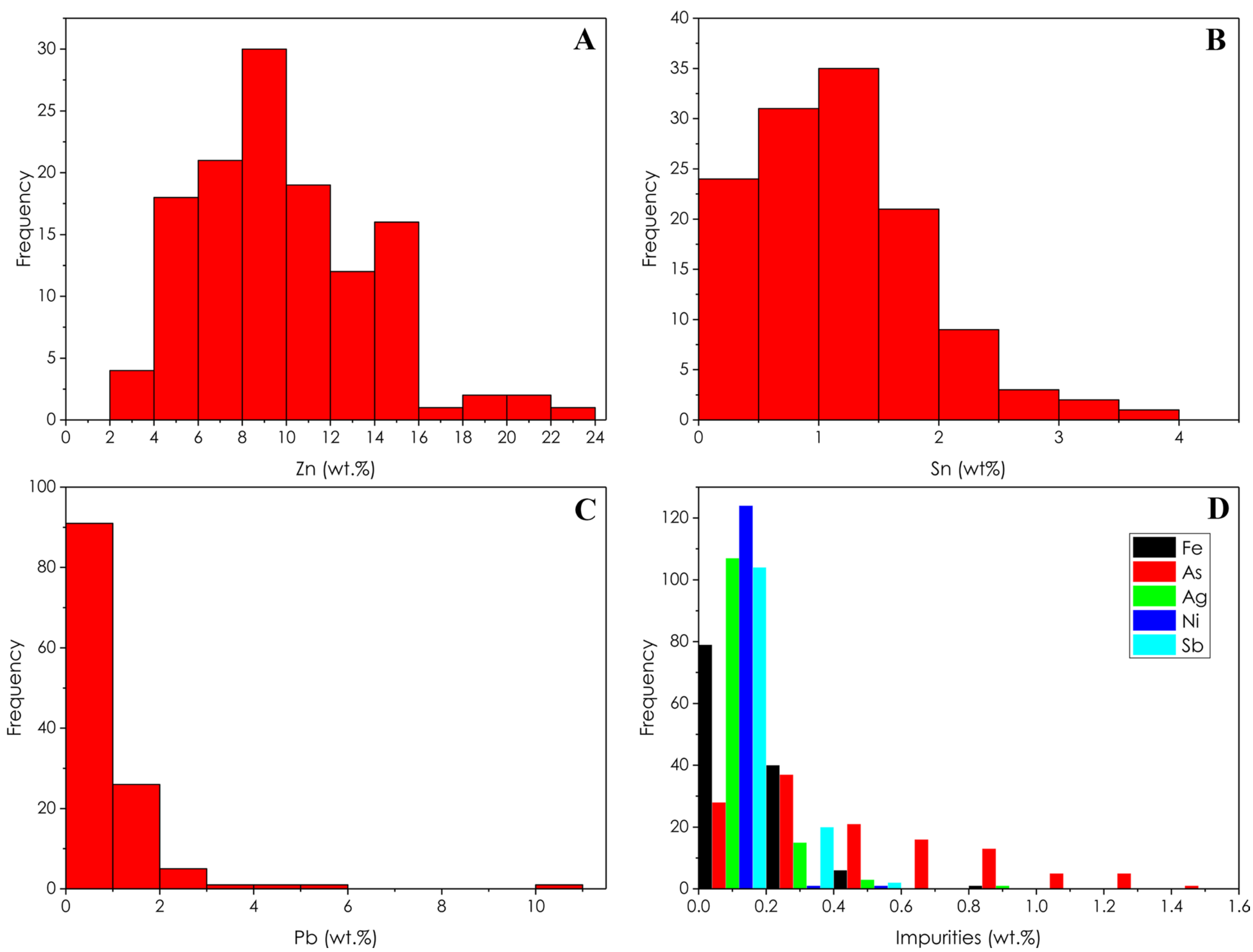

Fig. 5 Zinc (**A**), tin (**B**), lead (**C**), and impurities (**D**) content distribution within brass artefacts

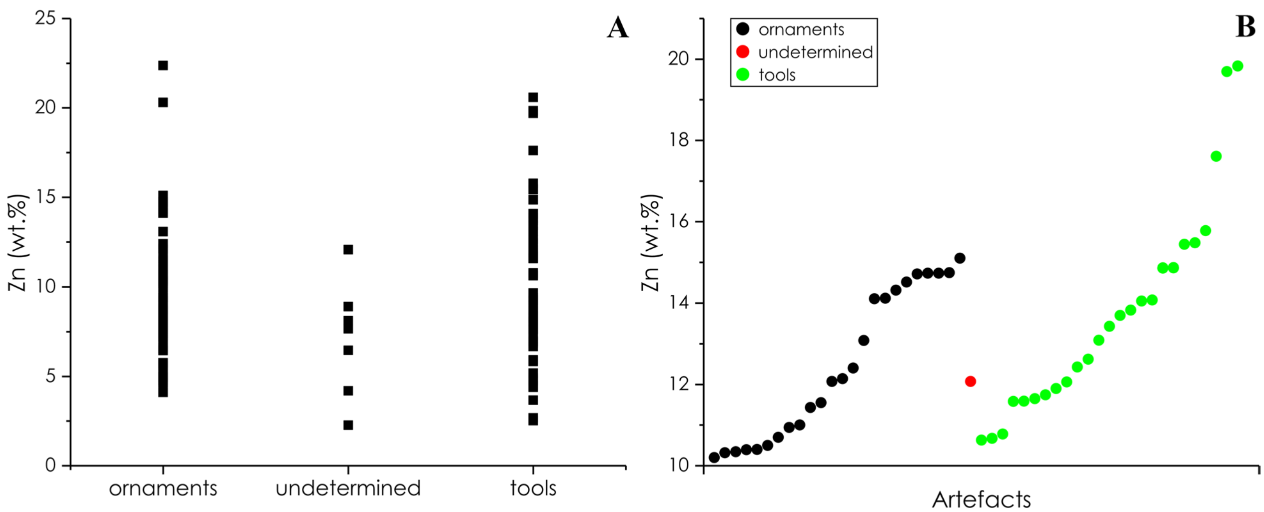

Fig. 6 Zinc variability within the different artefacts categories from Mértola, highlighting the lack of any correlation (**A**). Distribution of ornaments, undetermined objects, and tool in the range 10–20 wt.% Zn (**B**)

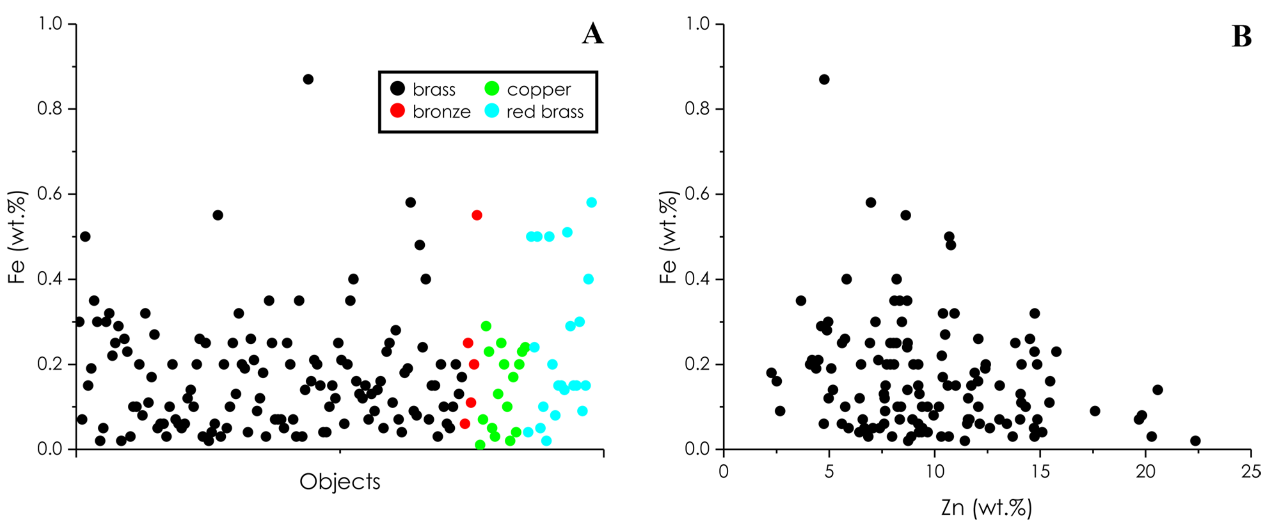

Fig. 7 Iron content within the artefacts analysed from Mértola according to the different type of alloys (**A**). Bivariate scatterplot showing zinc versus iron contents within brass artefacts (**B**)

ranging from 4.25 to 9.75 wt.%. Only one leaded bronze has been detected, i.e., ring AR-01–53 with 11.05 wt.% Pb (Fig. 8A). As for minor elements, they appear to be randomly distributed, reaching a total that is slightly above 4 wt.% (i.e., AR-01-53) (Fig. 8B).

As it is well known, the addition of Sn to Cu lowers the melting temperature of Cu, and improves the mechanical properties of the metal, making the alloy physically more resistant to impacts. In this respect, the mechanical effects that the presence of Sn may have on the finished alloy begin to become evident only at Sn concentrations above 3–4%, with the best results between 10 and 15 wt.% Sn [25, 35].

Considering the Sn content found in the bronzes from the Almohad neighbourhood of Mértola, it is quite evident that the addition of fresh Sn during the melting process was not a technological option for the metalworkers that produced these metals. As such, the reduced content of Sn is a further indication that, at that time, the use of recycled scraps as a raw material, instead of alloying Cu and Sn in suitable proportions, was a well-established practice. The reduced amount

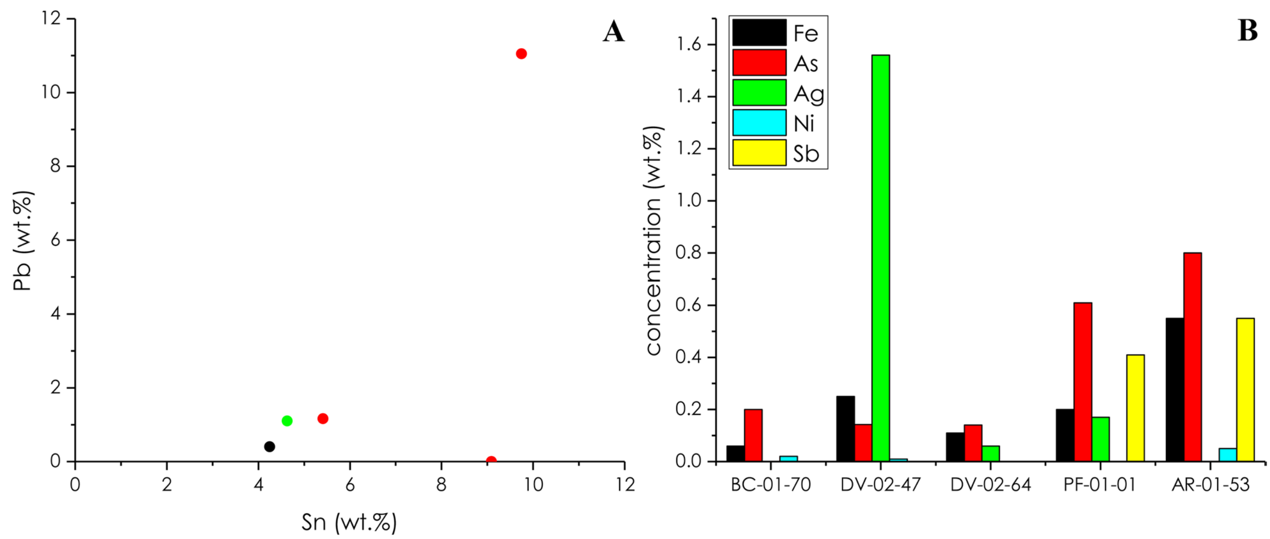

Fig. 8 Bivariate scatterplot showing lead versus tin contents within bronze artefacts (**A**). Distribution of impurities within the five bronze artefacts from Mértola (**B**)

of Sn in the finished objects is a consequence of the decrease in concentration that this element experiences as a consequence of the recycling process. Each time a tin-bronze is remelted, Sn gradually decreases through volatilization, leading to the production of objects with less Sn content than those used as scrap. The higher the number of remelting episodes, therefore, the lower the amount of tin in the final alloy [26].

In any case, when placed in its historical context, the low concentration of tin in the alloys found in Mértola was to be expected. In fact, no tin mines have so far been identified in the South of Portugal with the most likely source of tin at the beginning of the 2nd millennium being located in the Iberian Peninsula northwest, where tin had been exploited since antiquity [36]. However, it is very likely that with the Reconquista underway, these tin mines were no longer accessible to Moors as in the first quarter of the 2nd millennium Iberia northwest was already under the firm control of the Christian kingdoms.

The low concentration of tin in the alloys analysed in this paper could therefore be explained by a shortage of Sn supply due to the interruption of the tin trade to southern Portugal. However, it cannot be underestimated that tin, during the Islamic period, was also used for other craft productions, in particular for pottery glazes [37–43]. Thus, it is also possible that the little available tin, given its scarcity, may have been deliberately restricted to productions of greater social and artistic values such as prestige pottery, rather than for metal objects of daily use.

Copper and leaded copper

Pure coppers are relatively scarce within the assemblage of metals found in Mértola, accounting for about 8.8% of the investigated collection. In these objects, Sn and Zn are variable not exceeding 3.0 wt.% and 2.0 wt.% respectively (Fig. 9A–C). Other impurities occur in variable concentrations, ranging from 0.48 to 8.06 wt.% in total. Of special interest is a spindle (PF-01-78) composed by an alloy rather impure that includes 2.93 wt.% Sn, 1.94 wt.% Zn, and 1.94 wt.% Ag. Figure 9D clearly shows the difference in impurity concentrations, making possible the hypothesis that lower impurities occur in metal produced from fresh copper ore, while higher impurities may indicate metals produced from scrap containing variable and random amounts of elements other than Cu. Lead is general low. The only exception is a nail (PR-02-06) that contains 4.85 wt.% Pb. While the occurrence of Pb in this nail probably results from the intentional mixing of Pb to fresh or scrap copper, the rest of Pb may have originated from its presence in the minerals used by metalworkers.

From a typological perspective, these Cu-artefacts include three casket ornaments, one ring, one earring, one buckle, seven undetermined objects, and two nails. The only two nails analysed in this paper are both made of copper, although with differences in terms of Pb content. Due to the small number of artefacts composed of unalloyed copper, the data does not allow however for any further noteworthy comment to be made.

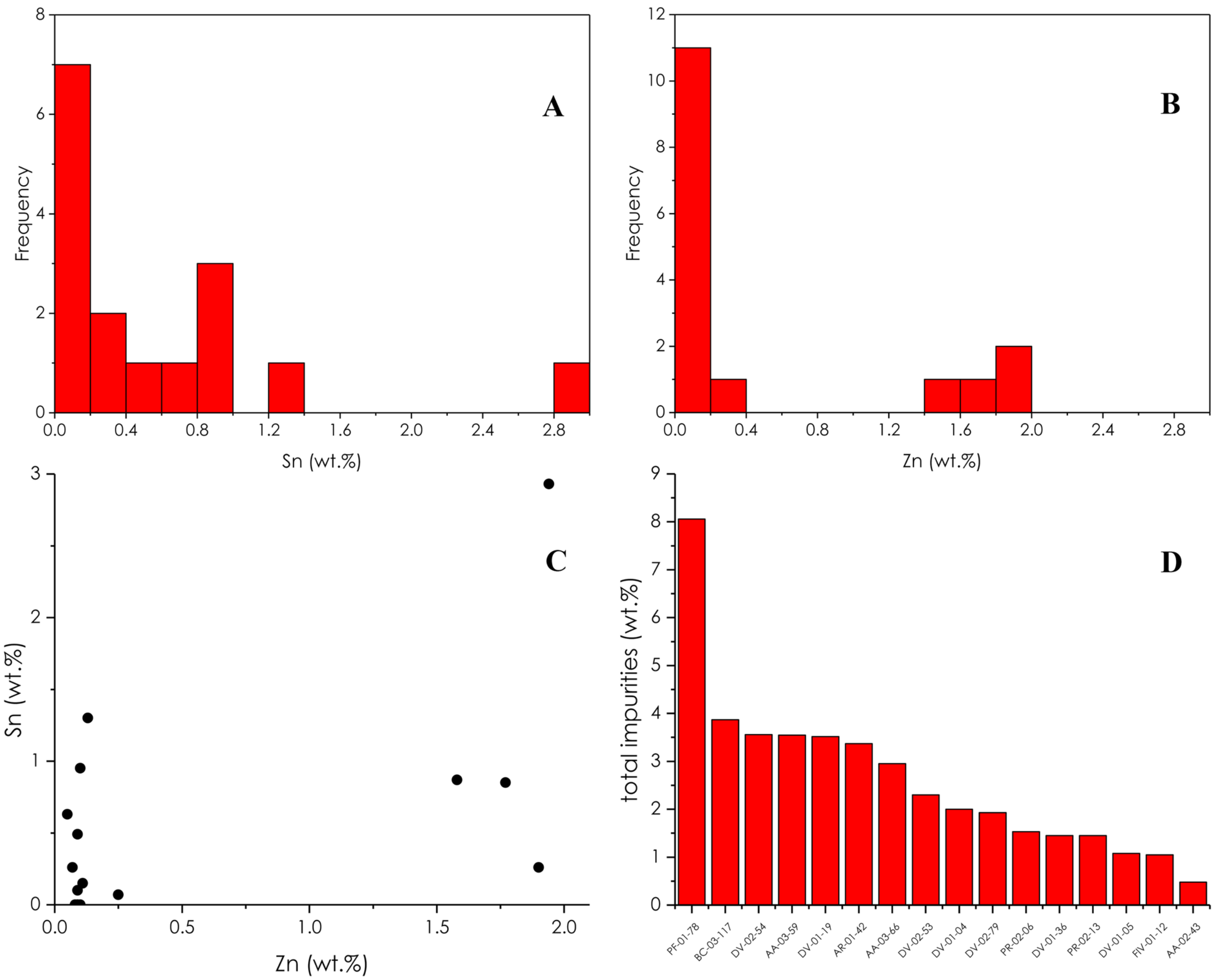

Fig. 9 Histograms showing the concentration of tin (**A**) and zinc (**B**) within copper artefacts. Bivariate scatterplot displaying tin versus zinc content in copper objects (**C**). Histogram showing the total of impurities of the Mértola copper artefacts (**D**)

Red brasses (Zn–Sn–Pb) and leaded red brasses (Zn–Sn–Pb + Pb)

Red brasses represent *c.* 13% of the entire assemblage, and includes spindles, casket ornaments, earrings, and undetermined objects. These objects show very reduced tin concentration, and a variability in composition of both Zn (from 2.1 to 18.08 wt.%), and Pb (from 0.34 to 7.85 wt.%, with six of the 23 red brasses above 3 wt.% Pb) (Fig. 10). Total impurities are reduced, ranging between 0.54 and 2.65 wt.%,

The composition of red brasses alloys appears to be a further argument in favour of a predominantly scrap-based metallurgy in which fresh ores were not added to the melt. In fact, low levels of Zn and Sn, in particular, confirm the hypothesis which regards the use of scrap as raw material to produce new objects as a very common practice of the time.

Impurities

Some of the minor and trace elements contained in metal objects are related to impurities in the ore processed that are unintentionally reduced during the smelting and refining processes and end up being incorporated into the finished artefacts. The elemental concentration of these impurities in the final alloy depends on different factors such as the quantity of impurities in the ore or the smelting technology in use. Attempting to address questions concerning to origin of raw materials through the identification of impurity patterns is a controversial issue in archaeology, although the presence of certain minor elements can give valuable information about the type of ore employed and/or the technology adopted in their production.

Impurities detected in the objects found in Mértola were rather low regardless of the alloy type, ranging from

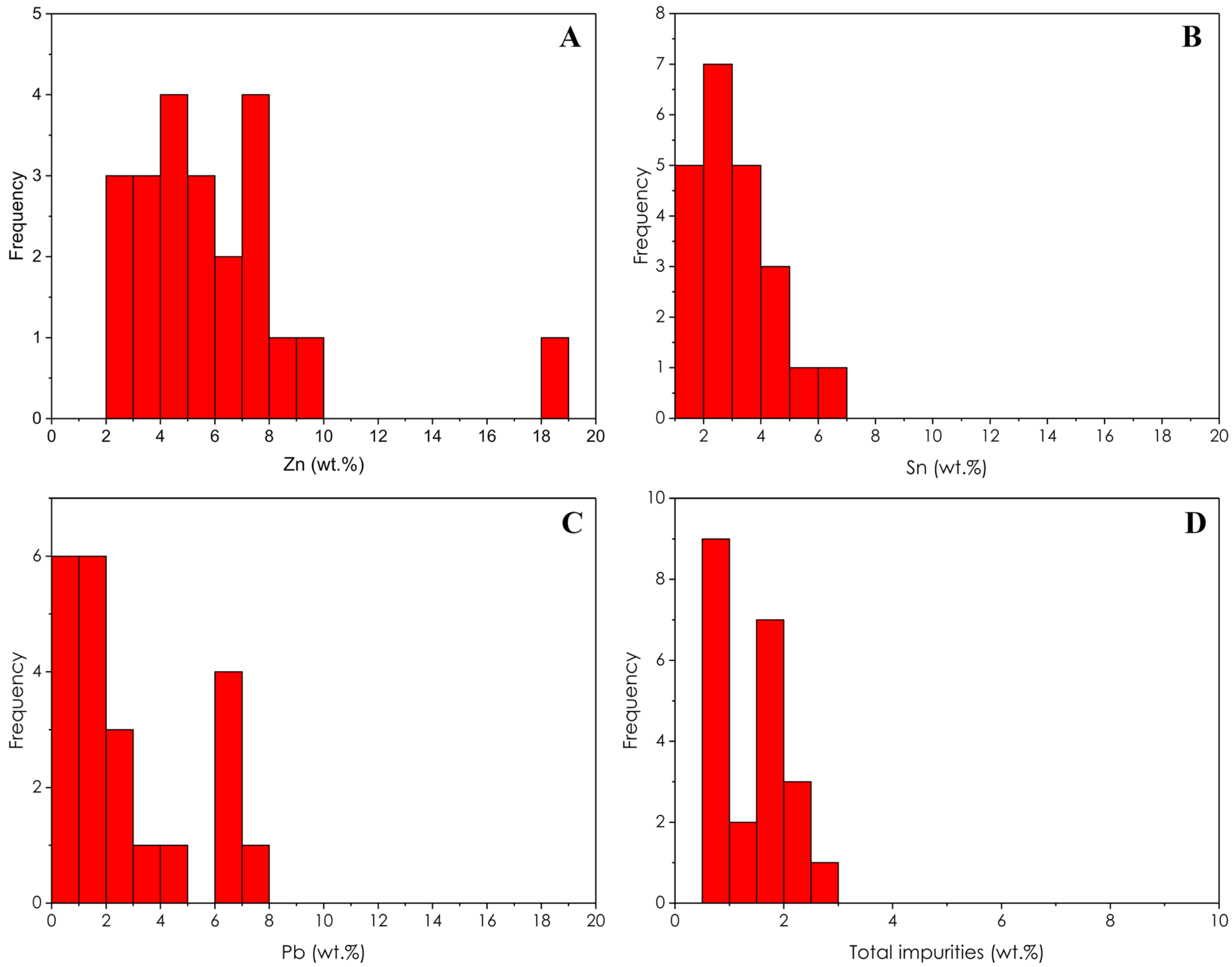

Fig. 10 Histograms displaying the concentration of zinc (**A**), tin (**B**), lead (**C**), and total impurities (**D**) within red brasses

0.21 to 3.53 wt.% (average *c.* 1.56 wt.%) in brasses, and from 0.31 to 3.74 wt.% (average *c.* 1.69 wt.%) in the other alloy types (Fig. 11A). Each individual minor element does not overcome 2 wt.%. Ag, Fe, Sb, and Ni are more concentrated below 0.5 wt.%, while As and Pb display a wider distribution (Fig. 11B–G).

In the last few years, an increasing number of research papers has focused on the analysis of impurities, together with isotopic data, as a means to provide valuable information on how metals were produced and remelted by ancient societies. In fact, this new approach, also known as the Oxford system [44–48], was not merely intended to understand from which mine the metal used for the production of a certain object may have come from, but instead to characterise the changing nature of metal objects in use and in circulation and their social meaning.

This system is based on the presence/absence and variability of some specific trace elements commonly found in ancient metals, like As, Sb, Ag, and Ni. These elements have a different thermodynamic behaviour when dispersed in the liquid metal: while As and Sb tend to decrease, Ag and Ni are less likely to be lost during the (re)melting process.

The trace element patterns of the objects analysed in this paper show that about 30% contains As + Sb + Ag + Ni; about 21% As + Ag; about 14% As + Ni, and As alone; and finally, the impurity patterns consisting of As + Ag + Ni, As + Sb + Ag, As + Sb + Ni, and As + Sb do not exceed 10% in each case. It is important to note that within the analysed object groups (tools, ornament, and others) not all typologies display the same patterns of impurities: for example, tools show eight different patterns (out of 16 copper categories identified by [47]) against six in the case of both ornament and undetermined objects. The order in which the different patterns of impurities appear in succession is different as well (Fig. 12).

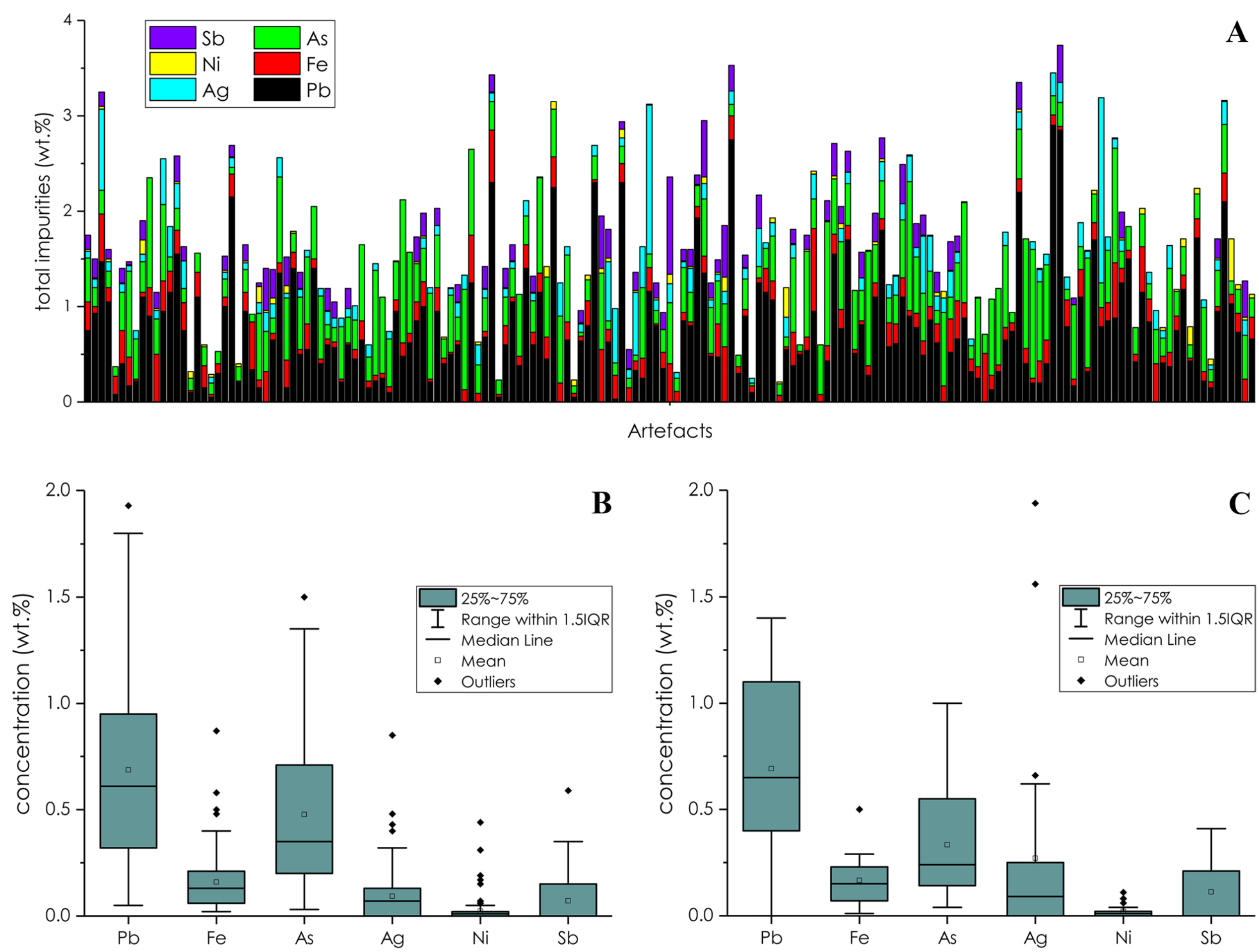

Fig. 11 Stacked columns with the main impurities detected in the 171 artefacts analysed from Mértola, showing differences in the concentration of each element and in the total amount (**A**). Boxplot displaying the variability of impurities in brass artefacts (**B**) and the rest of alloys (**C**)

Finally, no clear patterns can be observed in the distribution of the four impurities mentioned above, even though some differences can be detected between their distribution in brasses and the rest of the alloys. In fact, from the overall distribution we can observe that while As and Ni tends to be higher in brasses, Ag and Sb have larger average amounts in non-brass alloys, as follows: arsenic content is between 0.03 and 1.5 wt.% (0.03–1.5 wt.% in brasses, and 0.04–1.0 wt.% in the rest of the alloys): silver is between 0 and 1.94 wt.% (0–0.85 wt.% in brasses, and 0–1.94 wt.% in the rest of the alloys), nickel is between 0 and 0.44 wt.% (0–0.44 wt.% in brasses, and 0–0.15 wt.% in the rest of the alloys), and antimony is between 0 and 1.02 wt.% (0–0.59 wt.% in brasses, and 0–1.02 wt.% in the rest of the alloys).

Mértola within the Islamic copper-based metalwork from the 12th and 13th centuries

Based on the overall data from Mértola, it is quite clear that the high variability in the concentrations of the main elements, i.e., Zn, Sn, and Pb, found in the objects analysed does not allow clear compositional patterns to be identified. In fact, tools, ornaments, and fragments that do not fall into any of the previous categories seem to be produced seamlessly together with alloys containing variable concentrations of Zn, Sn and Pb (Fig. 13).

Considering the high technological expertise in metal production reached during the Islamic Iberian Peninsula [49–51], it is not at all likely that the metalworkers who produced the objects found in Mértola were unaware of the mechanical properties of the different copper-based alloys to the point of not taking advantage of them.

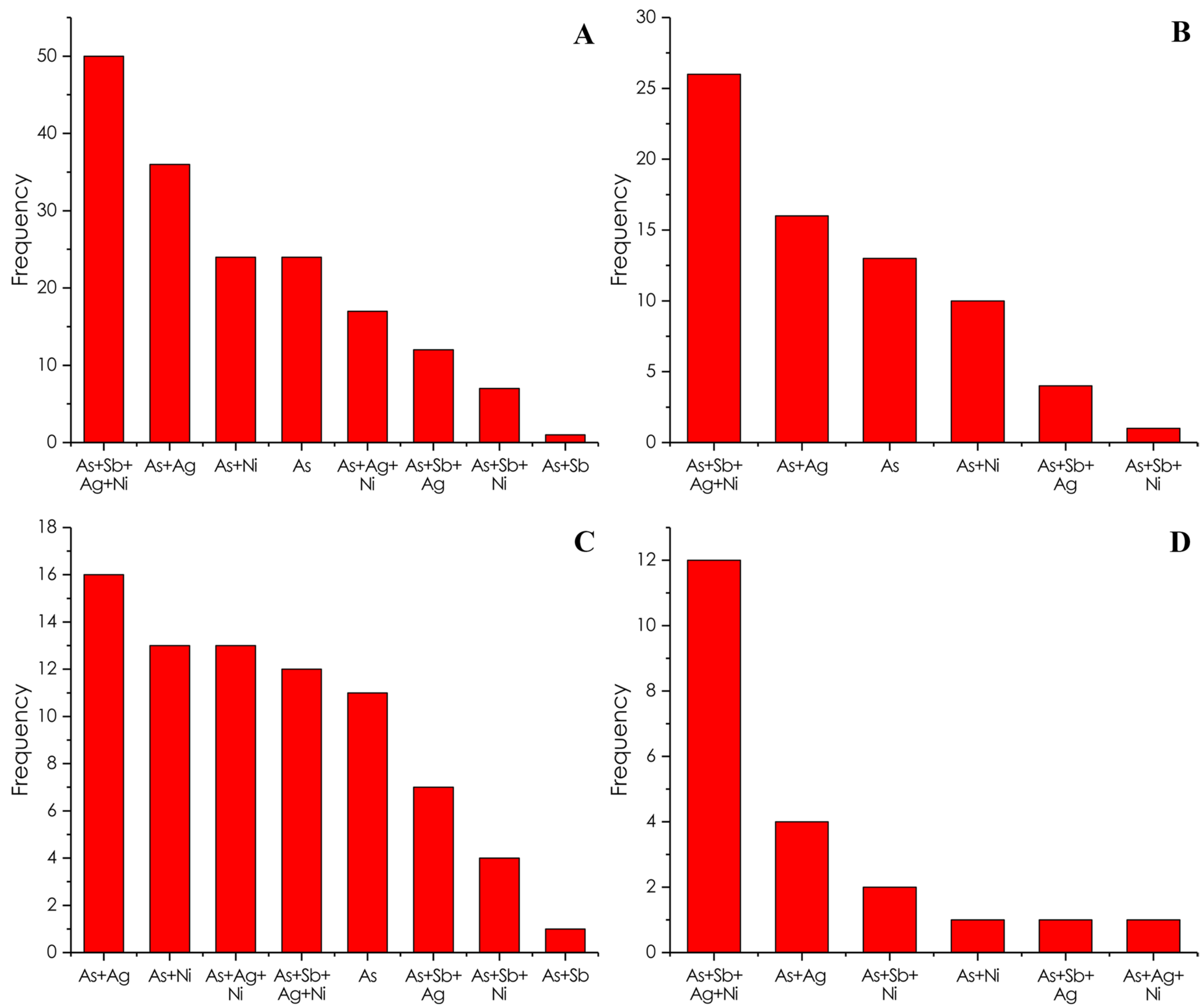

Fig. 12 Distribution of the whole collection (**A**), ornaments (**B**), tools (**C**), undetermined objects (D) within the 16 "copper groups", according to [47]

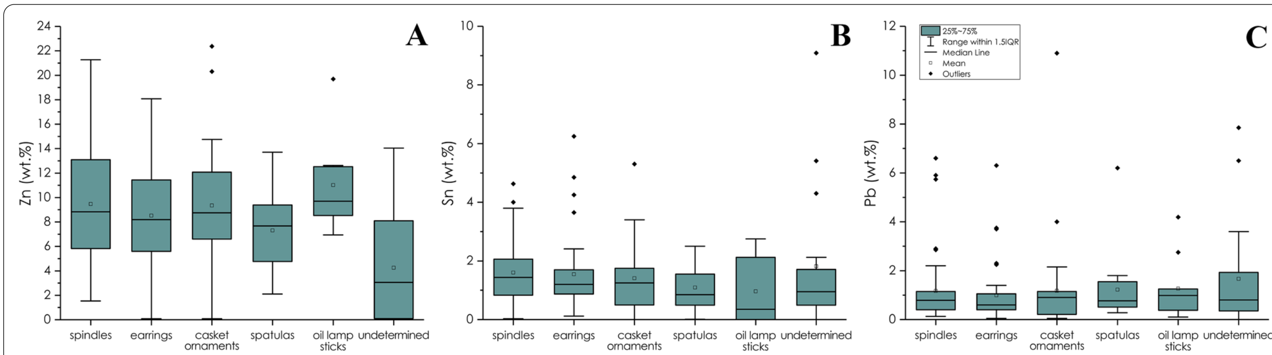

Fig. 13 Boxplot displaying the concentration and variability of zinc (**A**), tin (**B**), and lead (**C**) in the 171 artefacts analysed here from Mértola

However, the data available on metal found at different Islamic site in the Iberian Peninsula, i.e., Madinat al-Zahra (Córdoba, Spain) [52], Qalat Rabah (Calatrava la Vieja, Spain) [53] Denia (Alicante, Spain) [18, 49], a collection of oil lamps from different Portuguese sites [50] and other artefacts from various Spanish sites [54], depict a reasonably consistent picture with the objects analysed in this paper. In general, brass was the predominant alloy, very probably due to the availability of Zn ore regionally, while binary bronzes (Cu + Sn), although still in use, are rather scarce [55]. As outlined above, this low tin content could depend on the fact that its supply could not always be assured due to political tensions and/or economic restrictions, as tin ores were found in territories far beyond the control of the Islamic communities in southern Portugal and the rest of the Iberian Peninsula.

At the same time, the composition of Islamic brasses from al-Andalus, where Islamic tradition might have been expected to exercise some influence in the production of metals, seems to be very close to contemporary European brasses. For example, this is the case with metal collections from NW Europe [56, 57], France [21], and Italy [20], among others. The data from Mértola points out, for the first time, some differences in the iron content, likely due to the use of local available sphalerite. However, on this point, a larger dataset of analysis would be needed to confirm this trend.

Conclusion

This research has shown that a variety of different Cu-based alloys were in use in Mértola during the 12th and the first half of the thirteenth centuries. Tin bronze artefacts are the smallest group, while brass appears to be the preferred alloy to produce objects of daily use. Moreover, bronzes and brasses were further mixed to produce red brass alloys (Cu + Sn + Zn). Occasionally, Pb was randomly added to the different alloys.

The overall data suggest that objects were not produced with well-defined and predetermined composition and the results clearly revealed that no link can be found between the functions or the forms of the artefacts and their composition as similar objects were produced with different alloys, and vice versa, objects with distinct forms and functions were made of alloys with very similar mechanical properties. Metalworker that produced the objects found in the Almohad quarter of Mértola apparently did not possess advanced technical skills or they were not particularly concerned with the final alloy composition of the artefacts, and/or even if they were aware of the advantages linked to the different chemical compositions of the alloys, they chose not to take advantage of this knowledge.

A point to be further investigated in the future is about the use of zinc ore. Iron found at Mértola is lower than other contemporary Islamic sites and this could open up a new scenario to be investigated dealing with the ability of local metalworkers to assimilate a technology brought to al-Andalus by metallurgists arrived in Mértola from the East.

It is important to remind that the data we are dealing with in this paper represent a sort of snapshot and that we had access to objects escaped from recasting for reasons that we do not know. Notwithstanding this ephemerality, however, data from Mértola provide new important information not only on technological issues, but also on socio-political dynamics. In fact, it is very likely that the elemental composition of the metals in Mértola may mirror political barriers and economic constraints of the time that could have deeply influenced the technological options that metalworkers took along the production chain. In this respect, mention has been already made of the great instability experienced in southern al-Andalus during the 12th and the first half of the thirteenth centuries, characterised by periods of strong political fragmentation, i.e., with the formation of the so-called *Taifas*, and periods of reunification, particularly under the Almoravids and Almohads dynasties. Furthermore, since the beginning of the 2nd millennium, an increasing intensification of pressure on Islamic territories by Christian forces lead to the conquering South of Portugal finally achieved in the mid-thirteenth century. This climate of political instability is very likely to have had a negative effect on the metal trade, with al-Andalus communities experiencing ever increasing difficulties in the access to ore mineral resources located in territories they did not control. This is especially true for tin, which unlike zinc ores were not available locally and was also used for other types of production, such as glazed pottery. As a result, metal technology was affected by the widespread political insecurity in al-Andalus at the time, and local craftsmen were forced to adapt their production to circumstances beyond their control and to use as raw material local ores or scrap metals they had easier access to.

Acknowledgements

The authors would like to acknowledge Rute Fortuna from the Campo Arqueológico de Mértola for the archaeological laboratory treatment of the metals. CB also thanks Dr. Lino Mioni (Indiana University Bloomington, US) and Mr. Vasco Rossi (Zocca, Italy). We would like to warmly thank Reviewers for taking the time and effort necessary to review the manuscript. We sincerely appreciate all valuable comments and suggestions, which helped us to improve the quality of the manuscript.

Author contributions

CB: conceptualisation, methodology, XRF analysis, data collection and interpretation, and writing-original draft preparation. RB: data interpretation and writing editing. MB: XRF analysis, data collection and writing editing; JM: writing editing. SGM and LR: archaeological investigation and writing editing. NS: writing editing. All authors read and approved the final manuscript.

Funding
The study was carried out within the project *DE RE METALLICA—DEfining and REdiscovering MEtallurgy and Trade in AL-Andaluz (VIII-XIII century AD): Leaping into Innovative Comprehensive Archaeometric Approaches*. Analyses were carried out with equipment from the HERCULES Laboratory (University of Évora, Portugal) and funded by FCT (UIDB/04449/2020). Funding for open access fee payment was made available by the H2020-MSCA-ITN-EJD ED-ARCHMAT Project funded by the H2020-MSCA-ITN-EJD under GA n. 766311.

Availability of data and materials
The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing interests
The authors declare that they have no competing interests.

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Received: 20 December 2021 Accepted: 9 June 2022
Published online: 22 June 2022

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