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Analytical Study of Overlooked Bronze Age and Iron Age Goldwork from Northwest Portugal

Maria Filomena Guerra and Isabel Tissot

Keywords

Bronze Age, Iron Age, Northwest Portugal, gold, alloys, jewellery, bracelet, neck ornament

Abstract

To gain further insight into the use of native gold and intentional gold alloys during the Bronze Age in Northwest Portugal, jewellery in the collections of three Portuguese museums was studied using an in-house built portable X-ray fluorescence spectrometer. Among the analysed objects are the Late Bronze Age bracelet from Monte Airoso (Viseu) and the Iron Age tubular bracelet from Regoufe (Aveiro). To shed more light on the manufacture of gold bracelets, have also been analysed all the components (bracelets, wires and bands) of the hoard of Arnozela (Braga), so far incompletely studied. The results obtained show that from the Middle Bronze Age onwards gold containing 8-11 wt. % silver is used, to which gradually increasing amounts of copper seem to have been added over time. These results support a heterogeneous chronology for the components of the hoard of Arnozela and show that the bracelet from Monte Airoso is made from an intentional gold alloy used in the Portuguese area during the Late Bronze Age. On the contrary, the bracelet from Regoufe is made from native gold without addition of copper. This could indicate that at least during a certain period the goldsmiths used both native gold as found and intentional alloys.

Two other objects, discussed on the light of the data obtained for the bracelets, complete the study. One of them is one of the two Portuguese Early Bronze Age gold sheet ornaments with cut parallel bands (so-called gargantilha de tiras), found in Braga. Without obvious signs of use-wear like other ornaments of the same type, and made, as expected, with gold hammered into sheet, this ornament is made from an alloy that seems to correspond to a later period using gold from another origin. Finally, the composition of some of the components of the Late Bronze Age-Iron Age string from Malhada (Vila Real), a set of gold decorated plaques made from punched gold sheet by using different and asymmetrical punches, supports the addition of small amounts of copper to gold to produce intentional alloys.

Introduction

In opposition to tin and copper, the first more present in the Northwest (NW) and the second in the south of Tagus river (Monteagudo, 1953; Cardoso, 2014-15), gold is quite widespread throughout the Iberian Peninsula. In the Bronze Age (BA), the local gold resources of each region may therefore have been large enough to cover the local demand, even when it has started to grow. It is also conceivable that in rich gold mining regions, a local gold metallurgy gradually developed. 'Visible' native gold occurs in primary deposits (quartz veins) and alluvial deposits. Gold can be recovered by simple panning and washing in alluvial deposits, which are considered the earliest sources of gold in the Iberian Peninsula (details in e.g. Guerra and Tissot, 2021). Native gold is, however, an alloy containing variable amounts of silver and sometimes small amounts of copper (Boyle, 1987; Butt, Hough and Verrall, 2020; Chapman, et al., 2021) that give to the collected nuggets, dust and flakes variable optical properties, i.e. variable colour and reflectivity (Guerra, 2021), as well as different mechanical properties (Grimwade, 2009). The perhaps early desire to control all these properties and the absence of the necessary alloys in native deposits has certainly increased

the search for metallurgical processes related to the production of intentional gold alloys. The intentional gold alloys may have been used along with native gold, but may have quickly replaced it. The exploitation of different sources of gold along the Atlantic façade and the introduction of intentional gold alloys may thus explain, at least partially, the quite varied compositions observed for BA bracelets from Portuguese finds (Tissot and Guerra, 2017). This ring-shaped ornament, well represented in Iberian jewellery, became in the Late Bronze Age (LBA) the jewel of choice between rivers Tagus and Douro (Silva, 1994; Vilaça, 2004).

Indeed, among the BA objects found in the Iberian Peninsula, sometimes in hoards, are numerous gold bracelets. The diversity of their types and the quality of their manufacture are well illustrated by those contained in the hoard of Villena, Alicante, Spain (Soler García, 1969; Schüle, 1976; Perea, 1991; Almagro-Gorbea, 1995), while the diversity of their sizes is well represented in hoard 1 of Caldas de Reis, Galicia, Spain (Domato Castro and Comendador Rey, 1998; Valle Pérez and Peña Santos, 2017). Those contained in the latter are plain rings of rounded section, weighing up to 870 g, which have diameters ranging between 7.9 and 22.6 cm. They have been suggested to be ingots (Maluquer de Motes, 1970; Ruiz-Gálvez, 1995) or to be part of ritual depositions (Perea, 1994; Harding, 2000). However, unlike bracelets, ingots typically have only as-cast surfaces, because finishing is an unnecessary process that is both time and material consuming for an item produced to be reshaped.

In order to shed new light on the use of native gold and intentional gold alloys along the Atlantic façade during the BA, we have concentrated our studies on one of the richest regions in gold of the Iberian Peninsula, the NW (Noronha and Ramos, 1993; Spiering, et al., 2000; MacKenzie, Craw and Mortensen, 2019). The area includes several Portuguese and Spanish districts, but we have focused our studies on objects from NW Portugal, several of them being bracelets. Indeed, although Hartmann (1982) analysed quite a large number of Iberian objects, the elemental composition of several significant pieces from North Portugal and NW Spain remained unknown until recently (e.g. Martín-Torres and Ladra, 2018; García-Vuelta, Montero Ruiz and Villa Valdés, 2020; Guerra and Tissot, 2021; Armada and García-Vuelta, 2021) and the composition of many others remains to be studied.

We investigated four hitherto overlooked gold objects and an incompletely analysed hoard from NW Portugal. The objects are shown in Figures 1A and 1B, and the map of Figure 2 shows the sites where they were found. Two are bracelets. One of them is the LBA plain gold bracelet from Monte Airoso (Penedono, Viseu) in the collection of the National Museum of Archaeology in Lisbon. It was found in an area of influence that includes one of the major NW LBA sites in Portugal, the Castro da Senhora da Guia de Baiões (São Pedro do Sul, Viseu). The bracelet from Monte Airoso is one of the four LBA gold objects excavated in that area. The other bracelet studied here, in the collection of the Museum of Sacred Art of Arouca (Aveiro), is from Regoufe (Covêlo de Paivó, Arouca, Aveiro). This supposedly Iron Age (IA) tubular bracelet (Silva, 1986) is decorated with a motif comparable to that of a unique bracelet found among the twenty gold ring-shaped items that constitute the BA hoard of Arnozela (Fafe, Braga), which is in the collection of the National Museum of Archaeology. Although Hartmann (1982) analysed the main rings of this hoard, the composition of the small wires and thin bands twisted around some of them have not been analysed. For this reason, we analysed and reassessed the whole group for comparison with the bracelet from Regoufe and the other studied pieces.

To complete the study, we investigated two other gold items. One of them, in the collection of the National Museum of Archaeology, is the string from Malhada (Campeã, Vila Real), which is believed to have come from a burial. The string is one of the rare LBA-IA gold objects found in the district of Vila Real. The ornament consists of fifty-six small rectangular punched gold sheet plaques originally strung to be worn around the neck. We discuss here the results obtained for six of the plaques. Finally, in this study we have included one of the two gold sheet ornaments with cut parallel bands represented in Portuguese collections. This type of ornament is said representative of the Early Bronze Age (EBA) gold production of the Atlantic façade. The ornament comes from an unspecified context from the district of Braga and is in the collection of the Gold Museum in Travassos (Póvoa de Lanhoso, Braga).

The objects could not be moved for analysis. Therefore, they were examined in each museum by using a stereomicroscope equipped with a camera and an in-house built portable X-ray fluorescence (XRF)

spectrometer. Only the three major elements of the gold alloys were accurately determined, but further insight into the gold alloys employed in NW Portugal is obtained in the light of the elemental composition of other published gold objects from the same and neighbouring regions.

The analysed objects

The sheet ornament with cut parallel bands from Braga

Gold sheet collars with cut parallel bands, frequently named *gargantilhas de tiras*, which have been attributed to the EBA (e.g. Monteagudo, 1953; Pingel, 1992; Blas Cortina, 1994; Cardoso, 2014-15), are typical ornaments of the Atlantic façade, more precisely of NW Iberian Peninsula and French Atlantic façade. Some of them are associated with funerary contexts (Bóveda Fernández, 1996; 1998). They consist of a rectangular gold sheet, with sometimes so-called fastening holes and often decorated with dot-punched motifs, which has been cut in the front in several parallel bands and rolled into a cylindrical shape (Monteagudo, 1953; Eluère, 1977, 1982; Hernando Gonzalo, 1989; Armbruster, 1999).

Until now, to our knowledge, only nine complete sheet collars have survived. The distribution of the finds can be seen in Figure 3. Three of them were found in western France, one in Saint-Laurs (Deux-Sèvres) and two in Rondssec (Plouharnel, Morbihan) (Eluère, 1977; 1982). All the four Spanish collars were found in Galicia, three in A Coruña, at Monte dos Mouros (Toques) and Cícere (Santa Comba), and one in Pontevedra, at Castro de Goiás, included in the hoard of Agolada (López Cuevillas, 1926; Monteagudo, 1953; Hernando Gonzalo, 1989; Balseiro García, 1992; 2000; Blas Cortina, 1994; Pérez-Outeiriño, 1995; Comendador Rey, 1998). Six fragments contained in hoard 1 of Caldas de Reis, Galicia, could also have been part of one of these collars (e.g. Pingel, 1992; Hernando Gonzalo, 1989; Armbruster, et al., 2004). Only two ornaments of this type were found in Portugal. A third one was found at São Bento de Balugães (Barcelos, Braga), but as for the collar from Saint-Père-en-Retz (Loire-Atlantique, France), only the drawings made during the discovery have survived (Parenteau, 1878, Pl.61; Veiga, 1891, Pl.4).

One of the two surviving Portuguese gold sheet ornaments with cut parallel bands is the collar from Vale dos Moinhos (Santarém), decorated with dot-punched triangular and linear motifs (Armbruster and Parreira, 1983), which was analysed by Hartmann (1982). The other, shown in Figure 1A (no.1), is a smaller piece found in the district of Braga, in an undetermined context. It is exhibited unrolled at the Gold Museum. Such as many pieces in the collection of the museum, the ornament was purchased from a dealer by Francisco de Carvalho e Sousa, a goldsmith born in Travassos. Throughout fifty years of activity, he has collected both pieces and documentation that are today on display in the Gold Museum. The majority of them are related to the manufacture of filigree jewellery, which has been since the IA a major production activity in this Portuguese region (Sousa, 2013). It is recorded in the museum's archives that the dealer indicated that the object was discovered along with other undefined pieces at the base of a boulder in the district of Braga, near the St. Vincent chapel of Sezures or about 11 km away at Gondizalves, during the excavations for a new highway between Braga and Porto (Sousa, 2004).

The piece from Braga is c. 17.6 cm long and c. 3.6 cm high, and thus too short to be worn around the neck. For example, the sheet collar from Vale dos Moinhos and the two from Monte dos Mouros are, respectively, 34, 31 and 40 cm long (Armbruster and Parreira, 1993; Balseiro García, 2018), and the collar from Saint-Père-en-Retz was 46 cm long (Parenteau, 1878). However, over time, different functions have been assigned to these objects (Monteagudo, 1953; Bóveda Fernández, 1998; Ladra, Silva and Sousa, 2003 and cited references). For example, the piece excavated at Mata'l Casare (Asturias, Spain) measures c. 2 cm in diameter and was considered a fin gerring (Blas Cortina, 1994; Armbruster, 2021).

The sheet ornament from Braga, suggested as a hair ornament (Ladra, Silva and Sousa, 2003), could also have been another type of personal ornament, a bracelet or a garment decoration, for example. The most striking particularity of this object is its shape. Unlike the other gold sheet ornaments with cut parallel bands from the Atlantic façade, which have straight edges, the ornament from Braga has one of its edges wave-shaped. Indeed, only the collar found with two bracelets (Figure 4A) in 1920 near Castro de Goiás, during the

excavation work for the construction of a new route between Agolada and Lalín, in the district of Pontevedra in Spain (López Cuevillas, 1926; Monteagudo, 1954), has wave-shaped edges (Figure 4B). The decorations of the sheet ornaments from Braga and Agolada are similar. Both ornaments contain parallel bands cut into two sections, which are separated by a thin uncut strip with dot-punched lines, and their terminals are decorated with embossed parallel lines. In the ornament from Braga those lines, made from the back, are thicker. The same decoration can be seen in one of the two gold sheet collars from Rondsossec (Morbihan, France).

Although the gold sheet ornament from Braga has been flattened (perhaps by the discoverers), it is possible to appreciate the absence of signs of use-wear and tool marks remaining from cutting and decorating the gold sheet. As mentioned, the eleven parallel bands are cut into two sections, separated by a thin uncut strip in between, which is decorated at each side with a dot-punched line made from the front and running from almost top to bottom. On the back, as expected for this type of object, incised guidelines are visible on the gold sheet, drawn to facilitate the cutting process, as shown in Figure 5.

The bracelet from Monte Airoso

The plain gold penannular bracelet from Monte Airoso (Penedono, Viseu) is in the collection of the National Museum of Archaeology (Armbruster and Parreira, 1993). This bracelet (Figure 1A, no.2), found near a modern gold mine (Kalb, 1998), is one of the four gold objects dated to the LBA found in one of the most significant BA areas of North Portugal (Silva 1994; Senna-Martinez, 2005).

This unfinished cast penannular ornament (diameter 7.2 cm) has a solid thick body of square section and slightly expanded edges. In some areas, the surface shows an as-cast structure (Figure 6). On its inner part is visible a fracture and, on its surface, the visible overlapping creases are possibly due to bending and shaping by hammering the gold bar (Figures 6B and 6C). The other three gold objects from this area are the two torcs and one bracelet (Kalb, 1990-92) from the important settlement of Castro da Senhora da Guia de Baiões (São Pedro do Sul, Viseu). Indeed, a "hoard" or "foundry deposit" (Figueiredo, et al., 2010), which included weapons, ornaments, ritual objects, tools and a casting mould, was found inside one of the structures of this settlement (Silva, Tavares da Silva and Lopes, 1984).

The bracelet from Regoufe and the hoard from Arnozela

One gold tubular bracelet found near Regoufe (Covêlo de Paivó, Arouca) in the district of Aveiro (Cardozo, 1967), in the collection of the Museum of Sacred Art of Arouca, has a decoration comparable to certain LBA bracelets from NW Iberia. It was found in 1946 by a woman who was picking roots of heather on the slope of Mount Prova, between Regoufe and Silveirinha, near a Castro culture settlement situated between rivers Vouga and Douro (Simões Júnior, 1962; Silva, 1986; Silva and Pinto, 2012). The bracelet is 3 cm high, 6.75-6.95 cm in diameter, and it weighs 171 g (Simões Júnior, 1962).

This tubular bracelet (Figure 1A, no.3) has been dated to the IA (Silva, 1986). Made by lost wax casting, it is decorated along each border with a pair of parallel lines that run the entire length identically to a third line in the centre of the bracelet's body (Figure 7A). The body is decorated with series of inclined and almost parallel small lines of approximately the same length, deeply incised in the wax, suggesting a herringbone pattern. The overlap of the tool marks indicates (Figure 7B) that the hatched lines were made after the parallel lines that run the entire length.

The bracelet from Regoufe finds an equivalent in the unique bracelet with a chased decoration contained in the hoard of Arnozela (Braga), in the collection of the National Museum of Archaeology, which is one of the most significant groups of ring-shaped gold objects ever found in Portugal. The hoard, which consists of different sorts of ring-shaped ornaments, some with gold wires and bands twisted around their bodies, has been dated to both the LBA (Armbruster and Parreira, 1993) and the MBA (Pingel, 1992; Correia, 2013). However, the hoard may instead contain chronologically heterogeneous items (Armbruster, 2010). The main components of the hoard were analysed by Hartmann (1982). The composition of the small wires and thin bands having been neglected, we analysed the whole group here for a comparison with the bracelet from Regoufe and the other studied pieces.

The hoard of Arnozela is a group of twenty ring-shaped objects, weighing 533 g, found in 1903 by mere chance in an imprecise context, near a boulder, in a site where no trace of an ancient settlement was visible. Severo (1905-8) published a picture of the hoard showing its original arrangement (Figure 1B) and claimed that the hoard of Arnozela resembles the Spanish hoard of Hornachos, although the latter contains gold spirals and a rounded smooth ring (Galán, 2016). He provided a description of the different types of ring-shaped ornaments, which he describes as roughly finished. The majority of them have C- and D-shaped thin bodies and one of them, a solid penannular bracelet with a thin rounded body, severely deformed and broken into two pieces, served as “collector” of the nineteen others. Some bracelets are in pairs, assembled with a gold wire or tightened with a thin gold band (see Figure 1B). Details of bracelets with gold bands and gold wires wound around their body are shown in Figure 8.

Only three of the bracelets from Arnozela are decorated. Two of them are ribbed and very similar to other bracelets found in Portugal, in particular those contained in the Treasure of Beira Alta, dated to the MBA (Correia, 2013). The third decorated bracelet is very different from the others and its decoration is reported by Armbruster (2010) as made by chasing. The motif is comparable to the decoration of the bracelet from Regoufe. Indeed, it consists of two pairs of straight lines that run the full length of the bracelet and a band of hatched lines at each edge, made in opposite sense, suggesting a herringbone pattern. A detail was highlighted in Figure 1B. The decoration of the edges also recalls the motif of other known BA bracelets, like one from A Urdiñeira (Ourense, Galicia) in Spain (Balseiro García, 1994, pp.210-21) and two ribbed pieces from Minice (Mělník) in Czech Republic (Moucha, 2005, pp.130, Pl.174, No.15-17).

The articulated string from Malhada

Six of the fifty-six gold plaques, from an unknown site in Malhada (Campeã, Vila Real), were selected for analytical study. They are among the rare gold objects dated to the 1st millennium BC found in the district of Vila Real (Ladra, 2009). According to the finder, they were found in 1954 with several ceramic pieces. The gold plaques are considered to be components of an articulated string, which, along with the ceramic pieces, should have been part of the funerary goods from one grave (Silva, 1986; Pérez-Outeiriño, 1989). Silva (1986, drawing in Pl.93) suggests that it would have had the shape of a circular crown. However, these plaques, probably originally more numerous, could also have been used as garment decorations (Correia, Parreira and Silva, 2013).

The front and back of one of the identical gold plaques is shown in Figure 1A (no.4). They are made by folding and rolling rectangular gold sheets to obtain a slightly trapezoidal shape, necessary to avoid superposition when wearing a string in the form of a circular crown, corroborating Silva's suggestion (Silva, 1986). As shown in Figure 9, the plaques can be strung by simply passing a wire into each tube formed by rolling the gold sheet, as already suggested by Martins (2008).

Each plaque is decorated with the same geometric motif, obtained by using one single punch, which consists of two almost symmetrical patterns. In the punch, each pattern consists of a pair of two concentric circles framed on three sides by a series of dots and on the fourth side by a line. Between each pair of two concentric circles are two triangles, each consisting of three dots. At each edge of the shorter borders, a set of two parallel lines completes the punch. However, to achieve a balanced motif maintaining the radius of the concentric circles, the area used by each nearly symmetrical part of the punch is not the same. In the smaller area, which should be the top of the plaque when strung, the pair of two concentric circles is framed by 27 dots while on the larger area it is framed by 28 dots. On the back of the plaques, the irregularity of the cutting process of the shorter sides can be appreciated. As shown in Figure 10, the gold sheet was first folded backwards to obtain regular borders and then punched to create the main motif. A second punch (more precisely, several similar punches, the use of two of them is shown in Figure 11) was used to create a ribbed motif on the larger sides of the gold sheet. When these sides are rolled forwards, two stringing tubes with ribbed surfaces are obtained for holding a wire or thread.

Data collection

The objects studied in this work are in the collections of three museums. They cannot be sampled or moved for analysis. Therefore, the composition of their alloys was obtained on site, in each of the museums, by using an in-house built portable XRF spectrometer in planar configuration (Figure 12) developed at LIBPhys-UNL (Guerra, et al., 2014). The spectrometer consists of an Amptek Eclipse II X-ray tube system with an Ag-anode set to 30 kV and 20 μ A, an Amptek MC8000A Pocket MCA multichannel analyser, and an Amptek XR-100CR Si-PIN detector with a 25 μ m Be window and with energy resolution of 190 eV at 5.9 keV. The positioning of the area of analysis at the surface of the object was defined by the alignment of two lasers. In order to analyse smaller areas, the spot beam size was reduced to 4 mm at the focus point by using a Ta collimator.

Spectra processing was performed using WinAxil software (Vekemans, et al., 1994). Only the major elements of the gold alloys have been processed and then normalised to 100 wt. %, because the sensitivity and precision of the equipment are limited by the intensity of the output beam from the Ag-anode X-ray tube and the characteristics of the X-ray detector.

The presence of elements like Pt, Pd and Sn in gold alloys is frequently used to relate a gold object with the mining of alluvial deposits (Guerra, 2008; 2020). Tin results from the presence of cassiterite in those deposits (Dube, 2006) and Pt and Pd are, with Rh, the most soluble platinum group elements (PGE) in gold (Harris and Cabri, 1991; Jansen, Hauptmann and Klein, 2016). Trace amounts of these elements are, however, difficult to analyse by portable energy dispersive (ED) X-ray spectrometry, because in the case of this technique, portable equipment provides much poorer detection limits in the case of gold alloys than those attained when using fixed equipment (c. 150-350 ppm, Calliari, Dabalà and Magrini, 2000). Indeed, the X-ray lines of the major elements of the gold alloys (Au, Ag, Cu) cause a continual background in a large energy region, higher in areas containing elements with an atomic number close to those of the major elements due to overlapping of the X-ray lines (e.g. Pt with Au; Pd and Sn with Ag). This is observed even when ion beams, primary targets and specific filters are used (Guerra, et al., 2005; 2008). XRF detection limits can only be effectively improved by using either incident synchrotron radiation (Guerra, et al., 2008; Radtke, et al., 2013) or wavelength dispersive spectrometry (WDS, Radtke, et al., 2016).

The accuracy and precision of the quantitative results obtained in this work have been validated in each museum by analysis of the same reference material, consisting of a set of gold-silver-copper alloys containing comparable amounts of copper and silver with those contained in the jewellery (Tissot, et al., 2013). The data obtained for two consecutive analyses of four standards are summarised in Table 1. The detection limit for copper is 0.1 wt. %.

The very thin layers of corrosion products that can develop on the surface of gold jewellery (Tissot, et al., 2019) do not contribute to the compositions obtained by XRF. Indeed, the depth of analysis by XRF of gold alloys containing until 2 wt. % Cu and 15 wt. % Ag is of c. 13 μ m for the L-lines of Au, c. 30 μ m for the K-lines of Ag and c. 8 μ m for the K-lines of Cu (Troalen, Tate and Guerra, 2014).

The compositions of the bracelets from Monte Airoso and Regoufe, the sheet ornament from Braga and the six analysed components of the string from Malhada are reported in Table 2. The values represent the average concentrations with standard deviation obtained for two to five repeated analyses of the objects. Table 3 summarises the average concentrations with standard deviation obtained for two repeated analyses of all the components of the hoard from Arnozela.

The gold alloys: results and discussion

The ornament from Braga

The supposedly more ancient object analysed in this work is the gold sheet ornament with cut parallel bands from Braga. The amounts of silver and copper obtained for this ornament are compared to those published for comparable objects in Figure 13. These are the sheet collars with cut parallel bands from

Quinta do Vale dos Moinhos and Rondossec, analysed by Hartmann (1982), the two sheet collars from Monte dos Mouros, analysed by Guerra and Tissot (2021), and the fragment contained in hoard 1 of Caldas de Reis, analysed by Hartmann (1982). The results published by Rovira Llorens, Montero Ruiz and Consuegra Rodríguez (1997) for the ring from Mata'l Casare was also taken into account, as well as the information given on the collar from Saint-Père-de-Retz by Parenteau (1878, p.74): "...en or martelé allié à de l'argent dans la proportion d'un 8ème..." (in hammered gold alloyed with silver in the proportion of one-eighth). As the amount of copper is not provided, this collar is represented in the diagram by a line. Neither the collar from Agolada, the only known gold sheet ornament with cut parallel bands similar to the ornament from Braga, nor the collar from Cícere, identical to the missing piece from São Bento de Balugães, have been examined for the composition of their alloys.

It is obvious from the diagram that, like the small ring from Mata'l Casare, the ornament from Braga contains lower amounts of silver and slightly higher copper contents than the other plotted objects, which contain expected amounts of silver and copper. Indeed, analysis of the earliest Iberian goldwork indicates that the objects contain on average up to 20 wt. % Ag and, with a few exceptions, up to 0.2 wt. % Cu (e.g. Rovira Hortalà, et al., 2014; Murillo-Barroso, et al., 2015; Guerra and Tissot, 2016; Valério, et al., 2017), quantities that are expected for native gold (e.g. Pérez-García, Sánchez-Palencia and Torres-Ruiz, 2000; Chapman, Leake and Style, 2002; Moles, Chapman and Warner, 2013; Butt, Hough and Verrall, 2020). The EBA and MBA objects found in Galicia are also shown to contain up to c. 0.2 wt. % Cu and 5-25 wt. % Ag (Guerra and Tissot, 2021). Only the two supposedly MBA bracelets from Lamela (Silleda, Pontevedra) (García Alén, 1968; Montero Ruiz and Rovira Llorens, 1991) found in the same site as a copper ingot (Montero Ruiz, et al., 2010-2011) and which could be a more recent production, are shown to contain higher amounts.

Therefore, the ornament from Braga is made either with native gold from another region or with local native gold but with unintentional introduction of copper during the metallurgical processes. Otherwise, if copper was intentionally added to the native gold, the ornament could date from a more recent period than the EBA. In Figure 14, this ornament is plotted with EBA gold objects from North Portugal and three others from neighbouring Spanish provinces. Among them are those from the three burials containing gold grave goods included by Brandherm (2007) in the group of Vilavella-Atios. One of the objects is the diadem from tomb 240 excavated in Vilavella (Puentes de García Rodríguez, A Coruña) (Monteagudo, 1953). The others are the two ribbed tubular pieces from the cist of Atios (Porriño, Pontevedra), which were found with two silver spirals (Harrison, 1974), and the sheet diadem from the cist of Quinta da Água Branca (Vila Nova de Cerveira, Viana do Castelo), which was found with two gold rings and two gold spirals (Fortes, 1905-08). We have also added to the diagram the gold archer's armband (Taylor, 1994; Fokkens, Achterkamp and Kuilpers, 2008) from an undetermined context in Vila Nova de Cerveira (Viana do Castelo), which has been dated to the EBA (Fitzpatrick, 2011). It was found at the same time as a gold spiral, which is believed to form a group with the archer's armband (Spindler, 1993; Correia, Parreira and Silva, 2013). All these objects have been already analysed by Hartmann (1982). Finally, were added to the diagram six gold beads from the site of Buraco da Pala (Miraneda, Bragança) analysed by Cavalheiro and Sanches (1995), which were found a little further from the Atlantic façade in a context dated by 14C (Sanches, 1997). A line represents the copper contents below the detection limits.

Unlike the ornament from Braga, the EBA objects (Figure 14) contain less than 0.2 wt. % Cu and the objects that contain more than 20 wt. % Ag are all from Galicia. Only the beads from Buraco da Pala and the ring from Mata'l Casare contain identical lower amounts of silver. Interestingly, these are the only objects that come from sites a little further from the Atlantic façade. Therefore, the different composition observed for the ornament from Braga could be caused not only by chronological but also by regional differences. Two gold disks from a Bell Beaker's context in Asturias appear to also contain, on average, 7-8 wt. % Ag and less than 0.1 wt. % Cu. The investigations of Fernández Moreno, et al. (2018) provided, however, copper contents below the detection limits and very variable amounts of silver for the repeated analyses. Interestingly, the tubular LBA gold bracelet found in Cueva del Silo (Spain), situated even further east, in the province of Burgos, contains c. 3 wt. % Ag (Pérez-Romero, et al., 2018) like the ornament from Braga.

Based on the data presently available, we are unable to provide further discussion on the results obtained for the ornament from Braga.

The bracelets from Regoufe and Arnozela

The results obtained by XRF for the bracelet from Regoufe and for all the components of the hoard from Arnozela were plotted in the diagram of Figure 15. As the hoard from Arnozela contains bracelets of different types and as typology may be related to chronology, we used the types as described by Armbruster and Parreira (1993) to plot the objects. Data published by Hartmann (1982) for samples analysed by Optical Emission Spectroscopy (OES) for some of the items contained in this group were added to the diagram to show the good agreement of the data obtained using both techniques. Indeed, only the composition provided by Hartmann for the unfinished open ring with overlapping pierced ends (Table 2, Au 68: 19 wt. % Ag and 1.8 wt. % Cu) is different. The composition obtained by XRF shows the presence of 10.6 wt. % Ag and 0.1 wt. % Cu, amounts regularly found for the items from Arnozela. As the amounts of silver provided by Hartmann are sometimes approximate values (Taylor, 1980; Hartmann, 1982) contrary to those of copper, the composition of that bracelet appears to be incorrect. We observed the same type of error for the bracelet from Cantonha, Portugal (Guerra and Tissot, 2021).

No separate groups regarding the ring-shaped pieces and the thin wires (black square) and bands (black star) coiled around their bodies appear in Figure 15. Nevertheless, and although all bands contain equivalent amounts of copper and silver, the wires are made from different alloys. In addition, the components of some of the groups formed by tightening two ring-shaped items differ in their composition (see wires and bands in Table 2). One example is two bracelets (Table 2, Au 70 and Au 71) tied together with a wire, all three made from different alloys. The ribbed bracelets, the ring-shaped pieces of convex-concave section and the bracelet used as collector of the others are made from gold containing c. 10 wt. % Ag and 0.1-0.6 wt. % Cu. Could this correspond to a metallurgical change resulting in the presence of small quantities of copper in the same gold alloys? Indeed, none of the earliest objects contains more than 0.3 wt. % Cu and the same small increase of copper has been observed for goldwork from Galicia dated to the same period (Guerra and Tissot, 2021). The other components of the hoard contain quite variable silver contents, but some contain less than 0.3 wt. % Cu and none contains more than 0.6 wt. % Cu. Indeed, the silver contents observed for all the pieces from Arnozela are contained within the range defined by the earliest objects plotted in Figure 15, but very few contain more than 12 wt. % Ag and more than 0.6 wt. % Cu. Among these are some of the twisted bands and the only thick wire with two thin wires twisted around the body (Au 63, shown in Figure 11), which contains c. 1 wt. % Cu. The objects containing less than c. 0.2 wt. % Cu should have been made with native gold used as found. A group of eight bracelets dated to the MBA (Correia, 2013), which are typologically similar to those from Arnozela, were analysed by Hartmann (1982). These bracelets, which constitute the so-called Treasure of Beira Alta (Pingel, 1992; Armbruster and Parreira, 1993), were added to Figure 15. They contain less than 0.3 wt. % Cu and 9-11 wt. % Ag, within the range of compositions observed for the earliest gold objects, like the majority of the objects from Arnozela. The highest copper amount was observed for the only bracelet decorated with a chased motif (2.3 wt. % Cu, Au 66 in Table 3), indicating the use of an intentional alloy. It could be a later production, made by adding copper to locally available native gold.

The bracelet from Regoufe, which has been dated to the IA, contains 8.3 wt. % Ag and 0.2 wt. % Cu (Table 1, MSAAD.19) and is included in Figure 15 in the group defined by the MBA bracelets from Beira Alta and the majority of the objects from Arnozela. In order to check if this is due to a possible regional difference, we added to the diagram three LBA bracelets analysed by Hartmann (1982), found like the bracelet from Regoufe in the district of Aveiro. From the original sixteen bracelets found with a "pyramidal piece" in Baralhas (Macieira de Cambra, Aveiro) in 1896 by a shoemaker when excavating the foundations of his house (Vasconcelos, 1896), remain two whole pieces and one fragment. The fragmented piece is unfinished, like the bracelet from Monte Airoso.

We can observe in Figure 15 that, as the majority of the plotted objects, the bracelets from Baralhas contain c.

10 wt. % Ag (one of them contains 10-15 wt. % Ag). They contain however higher copper contents similarly to the bracelet from Arnozela decorated by chasing ("-" in the diagram). These results seem to support the production of intentional alloys by addition of copper to local native gold. The simultaneous use of native gold and intentional alloys in the LBA-IA remains a possibility to explain the results obtained for the bracelet from Regoufe.

The bracelet from Monte Airoso and the string from Malhada

The penannular bracelet from Monte Airoso and the string from Malhada are dated the first to the LBA and the second to the LBA-IA.

As mentioned above, the bracelet from Monte Airoso and the gold bracelet and two gold torcs from Castro da Senhora da Guia de Baiões are the only four LBA gold objects found in an important northern Portuguese area of influence at that time. As the objects from Castro da Senhora da Guia de Baiões were analysed by Hartmann (1982), we plotted all four objects with the six plaques from Malhada analysed in this work in Figure 16. We added to the diagram the data obtained here by XRF for the objects from Arnozela and for the bracelet from Regoufe, as well as data published by Hartmann (1982) for the bracelets from Beira Alta and Baralhas, already considered in Figure 15. We can observe that the objects split into two groups according to the amounts of copper. One of these groups, characterised by amounts of copper on average under 0.2 wt. %, contains the MBA bracelets from Beira Alta (in yellow inside the square named MBA), the bracelet from Regoufe and many of the objects from Arnozela. This group can be considered to correspond to the use of local native gold and the pieces from Arnozela contained in the group could have been made during or before the MBA. The other group, which is characterised by the same amounts of silver and higher amounts of copper, possibly corresponds to the use of intentional alloys made by simply adding copper to the same native gold. In this group are contained the LBA-IA plaques from Malhada, the LBA bracelets from Baralhas and Monte Airoso, one of the LBA pieces from Castro da Senhora da Guia, and some of the objects from Arnozela (inside rectangle named LBA). The other objects from Castro da Senhora da Guia contain different amounts of silver, which may correspond either to the use of different gold or to incorrect values. The plaques from Malhada and the objects from Arnozela contain, however, less copper than the bracelets from Baralhas and Monte Airoso and the torc from Castro da Senhora da Guia de Baiões, splitting the group into two subgroups according to the amounts of copper. The objects from Arnozela contained in this group could have been made during the LBA. Finally, it is noticeable that all the pieces from Castro da Senhora da Guia, the bracelets from Baralhas and the only chased bracelet from Arnozela contain the highest amounts of copper observed.

A third group emerges in Figure 16 when are added to the diagram the earliest gold objects from North Portugal mentioned above (in green, inside rectangle named EBA). The EBA gold needle found in an undetermined site at Areia, in the district of Aveiro (Cruz, 2005; Cardoso and Sousa, 2014), which was analysed by Hartmann (1982), was also considered in the diagram. This third group shows the use of native gold containing as expected up to 0.2 wt. % Cu, but a little higher silver content, which could correspond to exploitation of other deposits. Interestingly, a few objects contained in the hoard of Arnozela have the same range of silver contents as the EBA objects, which tends to support the chronological heterogeneity of the hoard.

Conclusion

Despite the little attention that has been given until now to the objects studied in this work, they proved to be full of information.

We were able to show that, in addition to a dot-punched decoration, the gold sheet ornament from Braga contains on the back the guidelines for the cutting process of the bands, which have been identified in other ornaments of this type (Eluère, 1982; Armbruster, 1999; Armbruster, et al., 2004; Guerra and Tissot, 2021). Such as in the case of the two gold sheet collars from Monte dos Mouros (Guerra and Tissot, 2021), we could

not find any signs of use-wear. Made from a quite thin gold sheet (c. 0.5 mm), the ornament was certainly not easy to wear and therefore simply may have had a funerary function. However, contrary to what was expected, its alloy does not match the composition observed for EBA objects from North Portugal analysed so far. Indeed, if the low amount of silver observed could be due to the use of another native gold (perhaps from a region situated farther from the Atlantic façade or from a small particular alluvial deposit), the amount of copper observed (0.6 wt. %) is not high, but higher than expected for the earliest productions. EBA objects from North Portugal analysed so far have shown to contain on average less than 0.2 wt. % Cu and the MBA bracelets from Beira Alta have also shown to contain less than 0.3 wt. % Cu. Higher amounts of copper were here observed for objects dated from the LBA onwards.

It is possible that the amount of copper contained in the sheet ornament from Braga was unintentionally added during the metallurgical processes. It is also possible that this ornament is not an EBA production, but perhaps an ornament of the same type produced later, although still during the BA. This would explain the small size and the wave-shaped edge of the gold sheet and the bands cut into two sections, decorations that have only been observed in this and the collar discovered at Agolada (Galicia), distancing the later from the Vilavella-Atios group (Bóveda Fernández, 1996; Brandherm, 2007). Future analysis of the gold sheet collar from Agolada may shed new light on the ornament from Braga.

The bracelet from Regoufe is made by lost wax casting, a technology that has been considered to be employed from the MBA onwards (Perea, 1995; Armbruster, 2013). If this shaping technique was thus in use in the IA, the bracelet is made from native gold used as found. The amounts of copper and silver observed correspond to those contained in the MBA bracelets considered in this work. Indeed, although both the EBA and MBA objects contain less than 0.2 wt. % Cu, the EBA objects contain more than 11 wt. % Ag, whilst the MBA objects contain on average 8-11 wt. % Ag. Therefore, the results obtained for the bracelet from Regoufe indicate two possibilities. The bracelet may have been made before the IA, perhaps in the LBA (although the MBA remains a possibility) when intentional gold alloys were just starting to be produced justifying the use of a native alloy. Otherwise, it could have been made in the beginning of the IA, because the native alloys may still have been in use in the production of gold objects along with the new intentional alloys. It is also possible to consider the reuse in the IA of BA obsolete and broken objects, which have been melted down to make new ones.

Contrary to the bracelet from Regoufe, both the bracelet from Monte Airoso and the string from Malhada, the first dated to the LBA and the second to the LBA-IA, are made from alloys containing higher amounts of copper than the EBA and MBA objects, but containing amounts of silver within the range defined by the MBA objects. The bracelet from Monte Airoso contains, such as the LBA bracelets and torcs from Castro da Senhora da Guia and Baralhas, more than 1 wt. % Cu. These objects form a chemical group that also contains the only bracelet from Arnozela with a chased motif, made therefore from an alloy used in the LBA. The gold alloys that were used to make the components of the string from Malhada contain, however, 0.6-0.7 wt. % Cu, values only observed in a few items from Arnozela.

From the results obtained for all the components of the hoard from Arnozela emerges a chemical pattern that seems to correspond to a chronologically heterogeneous group of objects. The large majority of the objects from Arnozela contain amounts of silver within those observed for the MBA and LBA objects, but the copper contents observed are variable. If, as mentioned, the only bracelet of the group with a chased decoration is compositionally consistent with the LBA objects from Monte Airoso, Castro da Senhora da Guia and Baralhas, the majority of the other objects are contained in the group formed by the MBA bracelets from Beira Alta. Several others, containing 0.3-0.6 wt. % Cu, form a group with the plaques from Malhada. The items from Arnozela containing the amounts of silver observed for the MBA pieces, could have been made with local native gold used from the MBA onwards, to which copper have been added in amounts that could have increased over time. The few items in this hoard, of very diverse typology, containing more than 11 wt. % Ag could have been made with the same gold exploited in the EBA, perhaps by reuse, or with gold coming from another region or deposit.

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Figures and Tables

Figure 1. The objects studied in this work: A): 1) The front and back of the gold sheet ornament with cut parallel bands found in the district of Braga exhibited at the Gold Museum; 2) The penannular bracelet from Monte Airoso (Penedono, Viseu); 3) The bracelet from Regoufe (Covêlo de Paivó, Arouca); 4) Front and back of one of the identical fifty-six gold plaques contained in the string from Malhada (Campeã, Vila Real). Photos: M. Tissot. B): The treasure from Arnozela (Fafe, Braga) as found in 1903 (from Severo, 1905-8, Pl.4, public domain), with a detail of the decorated tubular bracelet. Photo: M.F. Guerra.

Figure 2. Map of Portugal showing the sites where the studied objects have been found: 1) Braga (Gondizalves and Sezures); 2) Arnozela (Fafe, Braga); 3) Malhada (Campeã, Vila Real); 4) Monte Airoso (Penedono, Viseu); 5) Regoufe (Covêlo de Paivó, Arouca). Drawing: I. Tissot.

Figure 3. Map of the Atlantic façade showing the distribution of finds containing gold sheet ornaments with cut parallel bands. Complete ornaments today in collections: 1) Rondosseac (Plouharnel, Morbihan); 2) Saint-Laurs (Deux-Sèvres); 3) Mata'l Casare (Asturias); 4) Monte dos Mouros (Toques); 5) Cícere (Santa Comba); 6) Castro de Goiás (Lalín, Pontevedra); 7) Braga (Gondizalves and Sezures); 8) Vale dos Moinhos (Santarém). Ornaments known from the drawings: A) Saint-Père-en-Retz (Loire-Atlantique); São Bento de Balugães (Barcelos, Braga). Fragments supposedly from ornaments: C) Caldas de Reis (As Silgadas, Pontevedra). Drawing: I. Tissot.

Figure 4. A): The treasure of Agolada (Pontevedra), which consists of two bracelets and one gold sheet collar with cut parallel bands (López Cuevillas, 1926, Pl.19, public domain). B): The half of the symmetric collar, unrolled. Drawing: I. Tissot (Based on López Cuevillas, 1926; Monteagudo, 1953; Bóveda Fernández, 1998).

Figure 5. Detail of the back of the gold sheet ornament from Braga showing the guidelines for cutting the bands. Photo: M. Tissot.

Figure 6. Details of the penannular bracelet from Monte Airoso (Penedono, Viseu) showing: A): The as-cast surface structure; B): The fracture in the inner part; C): The overlapping creases. Photos: M. Tissot

Figure 7. Details of the decoration of the bracelet from Regoufe (Covêlo de Paivó, Arouca) showing: Parallel lines (A) that run the entire length and the series of inclined lines that were made (B) after the three parallel lines that run the full length. Photos: M. Tissot.

Figure 8. Details of objects contained in the treasure from Arnozela (Fafe, Braga): A): The open wire with small pieces of wire wound around the body; B): Wound wire around one of the bracelets; C): The two bands tightening two thin flat rings; D): Wound band around one of the bracelets. Photos: M.F. Guerra.

Figure 9. Reconstruction of a possible alignment of the plaques from Malhada (Campeã, Vila Real) to obtain a circular crown-shaped neck ornament. Photos: M. Tissot.

Figure 10. Reverse of the shorter sides of three plaques from Malhada (Campeã, Vila Real) showing the irregular cut and folding of the gold sheet. Photos: M. Tissot.

Figure 11. Two of the ribbed stringing tubes of the plaques from Malhada (Campeã, Vila Real) showing the use of different punches. Photos: M. Tissot.

Figure 12. Portable XRF spectrometer during analysis of the bracelet from Regoufe in the Museum of Sacred Art of Arouca. Photo: M. Tissot.

Figure 13. The Cu and Ag contents obtained by XRF for the ornament with cut parallel bands from Braga compared to published EBA objects of the same type (Rovira Llorens, Montero Ruiz and Consuegra Rodríguez, 1977; Hartmann, 1982; Guerra and Tissot, 2021). The lines represent the amount of Cu in the collar from Saint-Père-de-Retz, not mentioned

by Parenteau (1878), and the range given by Hartmann (1982) for the amounts of Ag contained in the fragment from Caldas de Reis. PT – Portugal, SP – Spain, FR – France.

Figure 14. The Cu and Ag contents obtained by XRF for the ornament with cut parallel bands from Braga compared to the composition of published EBA objects from North Portugal and Galicia (analysed by Hartmann, 1982; Cavalheiro and Sanches, 1995). The line represents the amounts of Cu under the detection limits for the beads from Buraco da Pala. PT – Portugal, SP – Spain.

Figure 15. The Cu and Ag contents obtained by XRF for all the components of the hoard from Arnozela and for the bracelet from Regoufe. Have been added for comparison the results published by Hartmann (1982) for the ring-shaped objects from Arnozela and for the bracelets from Beira Alta and Baralhas. The rectangle contains the MBA bracelets from Beira Alta and the line indicates the limit of 0.2 wt. % Cu observed for the EBA objects.

Figure 16. The Cu and Ag contents obtained by XRF for the bracelets from Monte Airoso and Regoufe, the six plaques from Malhada, and all the objects from Arnozela. The data for the bracelets from Beira Alta and Baralhas are from Hartmann (1982). For comparison, the data for the objects from Castro da Senhora da Guia de Baiões as well as the EBA objects from North Portugal have been added. The dashed rectangles show the areas containing EBA, MBA and LBA objects.

Table 1. Composition obtained by two consecutive XRF analyses of four gold standards. Std – certified value, M – measured value.

Table 2. Average composition with standard deviation obtained by XRF for the gold bracelets and ornaments studied in this work, normalised to 100 wt. %, with the exception of the hoard from Arnozela. GMT – Gold Museum in Travassos; NMA – National Museum of Archaeology; MSA – Museum of Sacred Art of Arouca.

Table 3. Average composition with standard deviation obtained by XRF for the gold bracelets, wires and bands contained in the hoard from Arnozela, normalised to 100 wt. %. Types and cross-sections as described by Armbruster and Parreira (1993). NMA – National Museum of Archaeology.

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