New insight into Bronze Age goldwork from northern Portugal. Bracelets, spirals and torcs from the region of Vila Real

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Abstract

This work is the first analytical study carried out on a representative group of early gold objects found in the region of Vila Real, situated in one of the richest gold-bearing areas in the northwest of the Iberian Peninsula. The objects were examined by optical microscopy and mobile XRF in four museums. Among them are the unique Early Bronze Age spiral ring from Vila Real (found at Fraga da Pitorca), one of the three chains of gold spirals from North Portugal and one of the few Villena-Estremoz type bracelets found in the Northwest (both from Alto da Pedisqueira). They constitute the Treasure of Chaves. Two of the three Bronze Age bracelets from Vila Real (found at Torgueda and Vinhós) have also been analysed together with typical productions of the region of Chaves, the Iron Age torcs from Rendufe, Codeçais and Paradela do Rio. Two similar torcs found in Lugo were analysed for comparison. When the earliest objects from Vila Real are compared to those found in neighbouring regions (Bragança, Minho and Galicia), they suggest regional exploitation of local gold resources. In Vila Real, native gold seems to be used as found until at least the end of the Bronze Age, but search and implementation of metallurgical practices related to several stages of production of intentional gold alloys seem to begin in the Early Iron Age. Some objects are made with gold containing a little more copper but amounts of silver within the same range as earlier objects, suggesting the addition of copper to local native gold, whereas others, particularly the torcs, contain increasing amounts of silver and copper. When copper and increasing amounts of silver are added to a gold containing 10 wt% Ag and 0.2 wt% Cu (native alloy observed at Vila Real) the theoretical debased alloys cover the chemical range defined by objects dated from the Iron Age to the early Roman period found north of Douro river, suggesting the use of well-established metallurgical practices.

Keywords: Vila Real, North Portugal, Bronze Age, Iron Age, gold work, gold alloys

1. Introduction

The main gold sources in nature are hydrothermal quartz veins and placer deposits. In these deposits, gold occurs in native state, as a metal or an alloy, often associated with silver and sometimes with copper (Boyle 1987, Moles et al. 2013, Chapman et al. 2021). Alluvial deposits, which form along rivers, allow native gold to be recovered in the form of nuggets, dust and flakes by simple panning and washing (Boyle 1987, Butt et al. 2020). It is thus possible to suggest that the first gold objects produced by man were made from alluvial gold, used as found.

Native gold often contains different amounts of silver and copper, variable from one alluvial deposit to another, sometimes even within the same deposit (*e.g.* in the Fresnedoso placer in the Moraleja Basin of the West Iberian Massif were found three textural and compositional groups, Alves et al. 2020). Since the optical properties of the gold alloys vary with the amounts of alloying elements, the visual aspect of native gold depends on the deposit being mined. When native gold contains high amounts of silver, the nuggets are perceived as greenish yellow or whitish with high reflectance, whilst when native gold

contains low amounts of silver and sometimes some copper, nuggets are perceived as yellow or yellowish with lower reflectance. It is not only the colour and reflectance of native gold that changes with increasing amounts of the alloying elements, the melting point also changes as well as other properties of the gold alloys, which are relevant in jewellery making like hardness and strength (Rapson 1990, Wise 1964, Grimwade 2009, Fischer-Bühner 2010).

It may be supposed that the native gold qualities accessible in local alluvial deposits to the earliest metallurgists were sufficient to satisfy their needs, but some dissatisfaction certainly set in quickly. The desire for gold alloys with particular properties as well as the difficult access to native gold with a wide range of compositions have certainly led to a rapid development of a gold metallurgy involving increasingly complex processing steps (Guerra 2021a). Although refining native gold to obtain purer gold is considered a quite early metallurgical practice (Bayley 1992, Rapson 1992, Craddock et al. 1999), the addition of copper and silver to native gold must be among the very first steps in a process, conceivably gradual, of production of intentional gold alloys.

To approach the early stages of the production of intentional gold alloys along the Iberian Atlantic façade, it is necessary to obtain information on the composition of gold objects dated to the Bronze Age (BA), preferably from those that are contextualised. The BA is one of the key moments in the metallurgical evolution of the Iberian Peninsula, when processes such as lost wax casting and hard soldering were implemented, leading to the emergence of a gold production that is both technically and typologically characteristic of the Atlantic façade (Maluquer de Motes 1970; Armbruster and Perea 1994, 2009; Perea 1995; Hernández Pérez and Soler Díaz 2005; Armbruster and Comendador Rey 2015). The occurrence of extensive gold deposits in the Iberian Peninsula made gold easily accessible to early metallurgists. In the richest gold bearing regions, such as the Northwest (NW), which includes the NW of Spain and the north of Portugal, one can imagine that, on the one hand, the metallurgy of gold developed locally in each region and, on the other hand, the earliest objects were produced with local native gold used as found.

Figure 1 - Map showing the location of the region and town of Vila Real and of the towns of Braga (A) and Bragança (B) in Portugal and Lugo (C) in Spain. On the right, the map of the Vila Real region where are indicated the findspots: 1) Fraga da Pitorca (Santo António de Monforte), 2) Paradela do Rio (Montalegre), 3) Codeçais (Santo Estêvão de Faiões, Chaves); 4) Alto da Pedisqueira (Chaves), 5) Rendufe (Valpaços), 6) Torgueda e Vinhós (Peso da Régua).

On the basis of those two hypotheses and using the data obtained by analysis of gold objects found in Galicia (a region of NW Spain that includes the provinces of A Coruña, Lugo, Ourense and Pontevedra), we were recently able to make suggestions regarding the production of intentional gold alloys (Guerra and Tissot 2021a). In Galicia, native gold seems to be used as found during a quite long period, because the amounts of copper contained in many objects remained below 0.2 wt % and the amounts of silver remained within the range defined by the Early Bronze Age (EBA) objects from local finds so far analysed. Indeed, the analytical data obtained indicate that at the end of the Late Bronze Age (LBA), possibly also at the beginning of the Iron Age (IA), native gold was used as found simultaneously with native gold containing a slightly higher copper content. The small increase in the copper content in some of the analysed objects can result from unintentional introduction of this metal during gold processing. However, the similar amounts of copper observed for those containing less than 1 wt % suggest a change in the metallurgical process, challenging to explain, but possibly towards the production of intentional alloys. This first change is followed by addition of higher amounts of copper to the native gold and a subsequent metallurgical step, which is the addition of both copper and silver to gold. The use of recycling (Guerra 2021b) is another possibility.

Although the native gold used during the earliest period in North Portugal does not seem to contain the same amounts of silver as the native gold used during the same period in Galicia, the few objects from North Portugal that were to date analysed seem to suggest an increase with time of the copper content (Guerra and Tissot 2021b). To gather further information on the production process of intentional gold alloys, and to assess the initial use of native gold containing increasing amounts of copper, it is necessary not only to analyse a larger number of objects found in the Portuguese territory, but also to take into account the geographical origin of the finds.

We found it interesting to concentrate our research on the Portuguese regions closest to Galicia, north of Douro river, allowing the use of the data gathered for this region for comparison purposes. In those regions, where run several rivers (Minho, Lima, Cávado, Ave, Tâmega, etc.), alluvial gold was easily accessible. Among these regions is Vila Real (Figure 1). Although a relatively small number of gold objects were among the earliest finds from this region, very few have been submitted to analytical examination. Indeed, Severo (1905-1908a, p. 283) provided in the beginning of the 20th century the composition of the solid penannular bracelet from Telões (Vila Pouca de Aguiar), but among the many gold objects found in the Iberian Peninsula analysed by Hartmann (1982) only four and two fragments were found in the region of Vila Real. One tubular bracelet, one torc and two fragments of torcs are contained in the Treasure of Lebução (Valpaços), dated to the Late IA (Ladra 2009, Martins 2010), and the two other objects are the bracelets from Torgueda and Vinhós (Peso da Régua), dated to the LBA (Armbruster and Parreira 1993). More recently, one gold torc dated to the IA, presumably from Chaves, was analysed by Alves et al. (2002), and six of the more than fifty plates that constitute the LBA-IA string from Malhada (Campeã) were analysed by Guerra and Tissot (2021b).

We selected for science-based study some of the most representative objects of the early work of gold in the region of Vila Real (Figure 1). The objects, which can be seen in Figure 2 and whose dimensions and weight are provided in Table 1, are in the collections of three Portuguese museums, the Museum of the Flaviense Region in Chaves, the Vila Velha Museum in Vila Real, and the National Museum of Archaeology in Lisbon. Three of the selected objects, although dated to different periods and not found together, are often referred to as the Treasure of Chaves (Figure 2b). They are the LBA bracelet and the Middle Bronze Age (MBA) chain of spirals found in Alto da Pedisqueira (Chaves) and the EBA spiral ring found in Fraga da Pitorca (Santo António de Monforte, Chaves). Three other objects selected for study are IA gold torcs with hollow double reel shaped terminals with flat circular ends decorated with a floral motif, typical of the Castro Culture production in the NW of the Iberian Peninsula (Figure 2c). They were found in Rendufe (Santa Maria de Émeres, Valpaços), Codeçais (Santo Estêvão de Faiões, Chaves) and Paradela do Rio (Montalegre). Two fragments of torcs of the same type in the collection of the Provincial Museum of Lugo, one also decorated with a floral motif and the other undecorated (Figure 2d), found in undetermined contexts in Lugo (Galicia), were also analysed for comparison. Additionally, the two mentioned bracelets from Torgueda (Vila Real) and Vinhós (Peso da Régua) (Figure 2a) were analysed for a reassessment of the composition of their alloys.

Figure 2 - The objects studied in this work. a) The bracelets from Vinhós (on the left) and Torgueda (on the right) (drawings from Botelho 1904, 1907). b) The Treasure of Chaves. From left to right: the bracelet and the chain from Alto da Pedisqueira and the spiral ring from Fraga da Pitorca. c) The torcs from Vila Real found at, from left to right, Paradela do Rio, Codeçais and Rendufe. d) The two torc fragments from Lugo, one decorated (on the left) and the other undecorated (on the right).

As the objects could not be moved for analysis, they were examined in each of the three museums using a stereomicroscope and *in-house* built portable X-ray fluorescence (XRF) spectrometers. A brief description of the finds and of the manufacturing details of the objects is given below followed by a discussion on the results obtained for the composition of their alloys. In order to shed new light on the early work of gold in the region of Vila Real, the results obtained in this work are compared with the composition of the few other objects from this region so far analysed as well as from the neighbouring region of Galicia. Finally, by comparing the composition of the objects found in Vila Real and Galicia with the composition of objects found in sites situated in the neighbouring northern Portuguese regions, we tried to assess whether a chemical “pattern” emerges for the gold alloys used in Vila Real.

Table 1 – Information on the analysed objects, including museum accession number, findspot, dimension and weight. MFR – Museum of the Flaviense Region, NMA – National Museum of Archaeology, VVM - Vila Velha Museum of Vila Real and PML – Provincial Museum of Lugo.

Object	Acc. no. (museum)	Findspot	Diameter (cm)	Weight (g)
Treasure of Chaves				
Bracelet	944/753 (MFR)	Alto da Pedisqueira, Chaves	6.5	121
Chain of spirals	944/754 (MFR)	Alto da Pedisqueira, Chaves	2 to 5	213
Spiral ring	nr/753 (MFR)	Fraga da Pitorca, Santo António de Monforte, Chaves	2.3	21
Bracelets				
Torgueda	Au 50 (NMA)	Torgueda, Vila Real	7	35
Vinhós	Au 37 (NMA)	Vinhós, Peso da Régua	6.7	17
Torcs				
Rendufe	2003.9.6901 (VVM)	Santa Maria de Émeres, Valpaços	18 (max.) 15 (min.)	250
Codeçais	Au 1139 (NMA)	Santos Estêvão de Faiões, Chaves	15.8 (max.) 14.5 (min.)	212
Paradela do Rio	Au 566 (NMA)	Montalegre	15.4 (max.) 13.8 (min.)	183
Fragments of torcs (Galicia)				
Decorated	1974/2/20 (PML)	Region of Lugo	9 (length)	48
Undecorated	1974/2/39 (PML)	Region of Lugo	10 (length)	56

2. Methods and instrumentation

The gold objects studied in this work could not be moved for analysis. Therefore, they were examined *in-situ* under a SMZ1 Nikon stereomicroscope equipped with a 5 MP Dino-Eye Eyepiece Camera and using a Dino-Lite USB microscope with 1.3 MP resolution. Their composition was obtained by moving to each of the three museums two *in-house* built portable XRF spectrometers, which have already been used for the analysis of several groups of gold jewellery (*e.g.* Adrimi et al. 2009 and Guerra and Tissot 2021a). One of them was developed at LIBPhys-UNL (Guerra et al. 2014) and the other at the Laboratory of C2RMF (de Viguerie et al. 2009). The first consists of an Amptek Eclipse II X-ray tube system with an Ag anode set to 30 kV and 20 μA and an Amptek XR-100CR Si-PIN detector with energy resolution of 190 eV at 5.9 keV. The second consists of a Moxtek Bullet X-ray tube system with an Ag anode operating at 35 kV and 95 μA and a Ketek AXAS-V Peltier cooled SD detector with energy resolution of 140 eV at 5.9 keV. Both systems have an Amptek MC8000A Pocket MCA multichannel analyser, two lasers pointers to define by intersection the area of analysis, and collimators that reduce the spot beam sizes at the focus point to approximately 1 mm^2 in the case of the C2RMF equipment and to 4 mm^2 in the case of the LIBPhys-UNL equipment.

Spectra processing was performed using WinAxil software (Vekemans et al. 1994). Unlike fixed XRF equipment that make it possible to determine the elements present in gold with detection limits attaining a few hundreds of ppm (*e.g.* Calliari et al. 2000), mobile equipment offers poorer detection limits. With our equipment, it is not possible to accurately determine the presence of trace elements in the gold alloys (Guerra et al. 2008, Guerra 2008, 2018). The sensitivity and precision being limited by the energy distribution of the X-ray tube output beams, in our case only the major elements of the gold alloys have been processed and then normalised to 100 wt %. The concentration of copper was obtained with a detection limit of 0.05-0.1 wt %.

The accuracy and precision of the quantitative results have been validated in each museum by analysis of reference material, consisting here of a set of gold-silver-copper alloys containing amounts of copper and silver comparable with those contained in the objects analysed. The same set of standards was used for the analysis of BA gold objects in other museums, as discussed in Guerra and Tissot (2021a, 2021b).

In order to avoid the effect of surface geometry, we have chosen to analyse only the flattest parts of the objects. Therefore, certain components of the torcs have not been considered in this study. Data obtained for the analysed objects are summarised in Table 2 and discussed below.

3. Description of the analysed objects

3.1 The Treasure of Chaves

Three of the studied objects – one gold penannular bracelet, one chain of gold spirals, and one gold spiral ring – are in the collection of the Museum of the Flaviense Region in Chaves. They constitute the so-called Treasure of Chaves (Correia 2013), shown in Figure 2b. The gold bracelet and the chain of gold spirals were found in Alto da Pedisqueira, in the centre of Chaves. Cardozo (1944, 1959) reports that the two objects were found in 1944, separately, by three soldiers from a squadron stationed in the Fort of S. Francisco, built in Alto da Pedisqueira, next to the outer side of the west wall, when carrying out some earth removal work. Questions have been raised about this find, because the Fort was built where a Franciscan Convent already existed, both requiring earth removal during construction.

The bracelet is a typical Villena-Estremoz production (V/E), dated to the LBA (Cardozo 1959, Almagro Basch 1969). Made by lost wax casting, this type of bracelets has been extensively described by several authors (*e.g.* Almagro-Gorbea 1974, Armbruster 1995, Perea 2005), including the use of rotary tools for their decoration (Armbruster and Perea 1994, Perea 1995). The marks left by these tools in the wax are visible on the surface of the bracelet from Chaves, as shown in Figure 3a. Showing several hoe marks made during discovery, this penannular bracelet is decorated with ribs alternating with rows of spikes, in the shape of an elongated truncated pyramid, forming a motif that is repeated and contained between the edge ribs. The motif consists of two rows of spikes between plain ribs incised with parallel line segments inclined in opposite senses to suggest a herringbone pattern. No signs of use-wear were observed. The edge ribs are also incised with parallel lines in opposite senses to suggest again a herringbone pattern. Lines from the work of the wax are visible between the rows of spikes, along the grooves (Figure 3b), and the tool marks on the edges (Figure 3c) indicate that the bracelet was cut after being cast. This corroborates the suggestion of Perea (1995) that these bracelets are originally tubular, made using wax cylinders, and then cut in section to obtain a penannular piece.

Figure 3 - Details of the bracelet contained in the Treasure of Chaves, found in Alto da Pedisqueira, showing: a) marks of the use of a rotary tool; b) tool marks inside the grooves from the work of the wax; and c) cutting marks on one of the edges.

The second object found in Alto da Pedisqueira is a chain of fifteen gold ring-shaped components made from gold wire of rounded shaped section, obtained by hammering according to Armbruster (2000). The wires, coiled two to five times into a spiral, like a skein, are of different thickness and length and some of them have thinner ends (Figure 4a,b). The sixteenth component is a gold strip (Figure 4c). None of the components show signs of use-wear. If some spirals are assembled by twisting gold wires around their bodies (Figure 4b) and two of them with the gold strip, the spirals are mainly intertwined.

Figure 4 - Details of the chain contained in the Treasure of Chaves, found in Alto da Pedisqueira, showing: a) the thinner end of one of the wires; b) wires of different thicknesses; and c) the gold strip.

Cardozo (1950), describes the chain from Chaves as wires “ready to use” by goldsmiths, and dates the find to the IA. However, isolated finds are difficult to date, and chains can be LBA and possibly MBA productions (Jorge 1986). Two other chains of gold skeins or spirals were found in northern Portugal, in Braga, a neighbour region west of Vila Real. One of them, made of six spirals and dated to the MBA, was found at Sequeade buried inside a ceramic container with a flat lid (Soeiro 1982, Jorge 1984). The

other, consisting of five spirals of regular dimensions, was found in Góios when digging a hole in the ground away from any ancient or modern habitat (Cardozo 1950). It is noticeable that such as in the case of the chain from Chaves, gold strips tighten the spirals found in Sequeade (Soeiro 1982). We can also mention the case of the ring-shaped components contained in the Treasure of Arnozela found in 1903 also in the region of Braga (Severo 1905-8b). Some of them, among which three that look like "offcuts" of spirals or skeins, have tightening gold strips around their bodies.

The third component of the Treasure of Chaves is the EBA gold spiral ring found in 1990 in Fraga da Pitorca (Santo António de Monforte, Chaves) with a narrow-butted flat copper axe and fragments of ceramics and of a millstone (RPDM 2015). Inhabitants of the immediate village of Curral das Vacas, who were using a metal detector, found the objects in a natural shelter of the granite formation of Fraga da Pitorca. The gold ring was obtained by hammering a cast ingot into a rectangular cross-section bar with tapering ends coiled to form a spiral. Only the outer side of the spiral has been entirely finished, although in some small areas the as-cast surface is still visible (Figure 5). No signs of use-wear were observed at the surface of the ring.

Figure 5 - Details of the spiral ring from contained in the Treasure of Chaves, found in Fraga da Pitorca, showing the outer side finishing and the inner side as-cast surface.

3.2 The gold bracelets

Although the discovery of several solid gold bracelets in the region of Vila Real has been reported, to our knowledge only three items remain. One of them was found in 1905 in Telões (Vila Pouca de Aguiar) by a mason when digging foundations of a wall between Vila Chã and Soutelinho (Severo 1905-8a). Severo (1905-8c) indicates that this solid penannular bracelet with octagonal section and thinner ends is made from an alloy containing 18.7 wt% Ag and "slight traces of copper". He also indicates that the bracelet is similar to the two bracelets and one fragment of a bracelet, remaining of the sixteen bracelets found in 1896 by a shoemaker in Baralhas (Macieira de Cambra, Aveiro, central Portugal) during the excavation of the foundations of his house (Vasconcelos, 1896). Dated to the LBA, the bracelets from Baralhas are made by hammering a gold bar (Armbruster and Parreira 1993).

Shepherds found the other two known bracelets from Vila Real. One was found near Moções in Torgueda (Botelho 1904) and the other near Vinhós (Botelho 1906) with three other bracelets that have been melted down (Cardozo 1967). They are thin tubular pieces of different types. The bracelet from Vinhós is thin with C-shaped section and the bracelet from Torgueda is a little thicker with rectangular section. The mentioned Treasure of Arnozela found in the region of Braga also contains bracelets of the same types. Suggested as probably being LBA productions, the bracelets from Torgueda and Vinhós are both made by hammering a cast annular ingot (Armbruster and Parreira 1993). According to Hartmann (1982) they contain very low amounts of copper (0.1 wt%) but very different amounts of silver (Torgueda 11 wt% Ag; Vinhós 5 wt% Ag). In this work they were reassessed for their elemental composition.

3.3 The torcs

Three IA gold torcs found in the region of Vila Real (Figure 2c) and two fragment of torcs found in Galicia (Figure 2d), all typical of the Castro Culture production (Silva 1986, Perez Outeiriño 1989, Balseiro 1994, Molina 1996, González-Ruibal 2004, Ladra 2009, Garcia-Vuelta 2016), were selected for study. Made using the same technology, they all have cast diamond cross-sectioned bodies (or neck-rings) and hollow double reel shaped terminals with flat circular ends (Cardozo 1942, Balseiro 1994, Molina 1996, Armbruster and Perea 2000, Perea 2003, García-Vuelta 2007, Ladra 2009, Martins 2010). Silva (1986, p. 236) includes these torcs in a single typological group (subgroup 2 of group D), the one containing Castro Culture torcs typical of the area of Chaves (named "flaviense"). With the exception of one of the fragments from Galicia (undecorated), they all have the flat circular end of the terminals decorated with a geometrical floral motif (rosette) enclosed in a circle. In the three torcs found in Vila Real, more precisely at Rendufe (Santa Maria de Émeres, Valpaços), Codeçais (Santo Estêvão de Faiões, Chaves) and Paradela do Rio (Montalegre), the motif was enhanced with granulation. Additionally, the

torcs from Rendufe and Paradela do Rio have decorated bodies, but only the torc from Rendufe has the back of the terminals decorated.

The torc from Rendufe, which was found by a peasant in a rill while working the land (Silva 1986, Silva 2013), is in the collection of the Vila Velha Museum in Vila Real, while the torcs from Codeçais and Paradela do Rio are in the collection of the National Museum of Archaeology in Lisbon. They were found in identical contexts. The torc from Codeçais was found in 1941 by a farmworker, who was digging a vineyard for a tavern owner (Cardozo 1942). The torc from Paradela do Rio appeared in 1958 with other two torcs of different types on the steep slope of a mountainous site, on the shovel of one of the diggers, during the opening of a road between Paradela do Rio and Outeiro (Cardozo 1959).

The similarity of the decoration of the flat circular ends of the terminals of these three torcs is shown in Figure 6a, b and c. The decoration consists of a six-petal rosette (the torc from Paradela do Rio is asymmetrically decorated as one terminal shows a seven-petal rosette and the other a six-petal rosette, both are shown in the Figure 6c) enclosed in a circle of petals or half-petals. The points of intersection are outlined with small concentric circles or with granules. One unprovenanced gold torc in the collection of the British Museum (Acc. no. 1960,0503.1) that is said from Ourense (Galicia), probably found in the north of Portugal or in NW Spain (Almagro-Gorbea 1962) and typologically related to those produced in the area of Chaves (Silva 1986, Molina 1996), also has a diamond cross-sectioned body and double reel-shaped shaped terminals. The flat circular ends of the terminals are decorated with a rosette enclosed in a simple circle enhanced with granulation at the points of intersection (Figure 6D), like the torcs from Paradela do Rio and Codeçais.

Figure 6 - The decoration of the flat circular ends of the torcs from: a) Rendufe, b) Codeçais, c) Paradela do Rio (the two different terminals are shown), d) said from Ourense, British Museum (Acc. no.1960,0503.1; drawing based on Botelho 1904, 1906), and e) Lugo.

The motifs that decorate the flat circular ends of the terminals of all the torcs were obtained by punching consecutive dots in the wax, as shown in Figure 7a for the torc from Rendufe and one of the fragments from Lugo, where misalignments and later modifications with correction of the decorative motifs can also be seen. The incised guidelines drawn before punching to facilitate the process are visible in Figure 7b in the case of the body of the torc from Paradela do Rio.

The two fragments of torcs of the same type found in Galicia, in 1859 or 1869, in an undetermined site in Lugo, are in the collection of the Provincial Museum of Lugo (Balseiro 1994). Molina (1996, no. 35.3 and 35.6) includes them in the group VI of Balseiro (1994) with the torcs from Paradela do Rio, Codeçais and Rendufe. One of them is undecorated, but the other has the flat circular end of the terminal decorated like the other studied torcs, as shown in Figure 6e.

Figure 7 - a) Details of the decoration of the flat circular ends of the torcs from Rendufe on the left and Lugo on the right. b) Incised guidelines in the body of the torc from Paradela do Rio.

4. The gold alloys

Table 2 summarises the data obtained by XRF for the composition of the studied objects. In the case of the bracelets from Torgueda, Vinhós and Chaves and the spiral ring from Fraga da Pitorca, the results provided in the table correspond to the average of two or three repeated analyses of each of the objects. For the chain from Chaves are provided the results obtained for each of the single analysed region of the sixteen chain components. Due to the spot beam size of the XRF equipment, in order to avoid the effect of surface geometry is only reported for each torc in Table 2 the average of two repeated analyses of the body and of the flat circular ends of the terminals.

Table 2 – Average elemental composition with standard deviation obtained by XRF, normalised to 100 wt % (one single analysis of each component of the chain of spirals). In brackets are given the amounts of copper rounded to the nearest hundredth when under 0.2 wt%. MFR – Museum of the Flaviense Region, NMA – National Museum of Archaeology, VVM - Vila Velha Museum of Vila Real and PML – Provincial Museum of Lugo.

4.1 The Treasure of Chaves and the bracelets from Torgueda and Vinhós

It is noticeable the low amounts of copper contained in the three objects that constitute the Treasure of Chaves, although dated from different periods.

The EBA spiral ring is made from native gold used as found. It contains less than 0.2 wt % Cu like the earliest gold objects so far analysed found in (mainly the south of) the Iberian Peninsula (*e.g.* Perea 1991, Montero and Rovira 1991, Blasco and Ríos 2010, Murillo-Barroso et al. 2015, Valério et al. 2017). When the amount of copper is rounded to the nearest hundredth (given in brackets in Table 2) we observe that the spiral ring contains 0.17 wt%. This amount of copper corresponds to the average value of 0.17 wt % Cu obtained by Moles et al. (2013) for the composition of 500 placer gold grains from the Mourne Mountains in Northern Ireland. Low amounts of copper are commonly observed in native gold. For example, the analysis of *c.* 250 gold grains from several placer deposits in Cameroun has shown the presence of 0.00-0.34 wt% Cu (Nguimatsia et al. 2019, Nono et al. 2021) and the analysis of *c.* 150 gold grains from lode and placer deposits in the United States revealed the presence of 0.003-0.3 wt% Cu (Antweiler and Sutton 1970).

The spiral ring also contains low amounts of silver, less than 8 wt%. Interestingly, this value corresponds to the amounts published by Cavalheiro and Sanches (1995) for the six gold beads excavated in the EBA site of Buraco da Pala (Mirandela). This site is situated in Bragança, a neighbour region east of Vila Real. The two regions form together the natural region of Trás-os-Montes ("beyond the mountains"), bounded to the west by the Tâmega river and on the south by the Douro river. No further analysis of EBA gold objects from Trás-os-Montes is available for comparison.

The components of the chain of spirals contain *c.* 6 to 13 wt % Ag and less than 0.2 wt % Cu; only the gold strip contains 0.3 wt % Cu. These results indicate that the chain components are made from native gold used as found. The variety of the amounts of silver could be due to the use of geographically or chronologically different gold, *i.e.* gold from deposits exploited in different places or at different times.

Figure 8 - The Cu and Ag contents obtained by XRF for the objects contained in the Treasure of Chaves and for the bracelets from Torgueda and Vinhós compared to data published for EBA objects from Portuguese regions situated north of Douro river mentioned in the text. The EBA objects from Galicia have also been added for comparison. The dashed lines represent the copper and silver ranges provided for certain objects. The rectangles contain the EBA objects by region.

The objects from Chaves and the six gold beads from Buraco da Pala were plotted in Figure 8 with the few analysed EBA objects from Portuguese regions situated north of Douro river. The majority was found in Vila Nova de Cerveira, in Viana do Castelo, a region that constitute with the region of Braga the natural region of Minho, situated west of Trás-os-Montes and south of Galicia. These objects are two spiral rings, a gold diadem and a pair of gold rings found inside the Bell Beaker cist of Quinta da Água Branca with a tanged dagger (Fortes, 1905-08a), and one spiral ring believed to form a group with the only archer´s wrist-guard in gold found in Portugal (Fitzpatrick 2011, Correia et al. 2013). They were analysed by Hartmann (1982). The beads from Buraco da Pala (Cu under 1 wt%, Cavalheiro and Sanches 1995) are represented in Figure 8 by a dashed vertical line.

Unlike the EBA objects from Trás-os-Montes, which contain 7-8 wt% Ag, the EBA objects from other regions contain more than 11 wt % Ag. We added to Figure 8 the EBA objects from Galicia analysed to date. Those analysed by Hartmann (1982) are the diadem from Vilavella (Puentes de García Rodríguez, A Coruña) (Monteagudo 1953), the two ribbed tubular pieces from the cist of Atios (Porriño, Pontevedra) (Harrison 1974, Brandherm 2007), and one of the fragments contained in hoard 1 of Caldas de Reis (Pontevedra) suggested to be from an EBA sheet collar (Pingel 1992). The other objects are the two sheet collars with cut parallel bands from Monte dos Mouros (Toques, A Coruña) (Balseiro 1994, 2018) analysed by Guerra and Tissot (2021a). With the exception of one of the collars from Monte dos Mouros and the one from Villavella (arrows in Figure 8), they contain higher amounts of silver than the Portuguese objects. Therefore, the EBA objects from Trás-os-Montes, Minho and Galicia seem to be generally made from gold containing different amounts of silver, suggesting the exploitation of local deposits.

We have also considered in Figure 8 the two bracelets from Vila Real, found in Torgueda and Vinhós, which were reassessed in this work for the composition of their alloys. Containing both *c.* 0.1 wt % Cu,

they are made from native gold used as found. The amount of silver observed by XRF for the bracelet from Vinhós (7.9 wt%) is higher than the value of 5 wt% provided by Hartmann (1982), but interestingly close to the amounts of silver contained in the EBA objects from Trás-os-Montes. The amounts of silver observed for both bracelets are contained within the range defined by the chain of spirals contained in the Treasure of Chaves.

Figure 9 - The Cu and Ag contents obtained by XRF for the V/E type bracelet from Chaves compared to V/E type bracelets and their IA evolutions from North Portugal (NP) and from Galicia (G). EBA objects from those two areas were added for comparison. The square contains the V/E type bracelets and the arrow indicates the amount of Cu outside the considered range.

The Treasure of Chaves comprises a LBA bracelet of V/E type, which also falls compositionally within the range of silver values defined by the chain of spirals, as shown in Figure 8, and which contains less than 0.3 wt% Cu. We have plotted in Figure 9 with the EBA objects from North Portugal and Galicia mentioned above the bracelets from NW Iberia recorded by Perea (2005) and Pérez-Romero et al. (2018) as V/E type bracelets. As the bracelet from Toén (Spain) has not been analysed, in addition to the bracelet from Chaves, were plotted in the diagram the bracelet from Ourense (Galicia) analysed by Guerra and Tissot (2021a) and the bracelet from Monte da Saia (Barcelos, Braga) analysed by Hartmann (1971). They all contain identical amounts of silver and less than 0.3 wt% Cu. They are probably made from native gold used as found.

Although considered by Perea (2005) as IA evolutions of V/E productions, the bracelets from A Urdiñeira and Toques (Galicia), the bracelet from Cantonha (Guimarães, Braga), and the bracelet from Lebução (Valpaços, Vila Real) were added to Figure 9. The first three have been analysed by Guerra and Tissot (2021a), who analysed the three components of the bracelet from Cantonha, which consists of a V/E type bracelet between a pair of Sagrajas/Berzocana type bracelets (Cardozo 1937). The fourth, analysed by Hartmann (1971), is one of the components of the Treasure of Lebução, found in 1899 in Valpaços by a farm worker when digging a vineyard (Severo 1905-8b). The IA evolutions contain all more silver than the V/E type bracelets. With the exception of the bracelet from Toques that contains 0.2 wt% Cu, they also contain more copper. The richly decorated bracelet from Lebução is considered more recent (Pingel 1991), which explains the use of a very different gold alloy. One pair of tubular bracelets from North Portugal analysed by Hartmann (1971, fig. 2), considered by Silva (1986, 1996) as an IA evolution of V/E type productions, also contain higher amounts of both silver and copper (20 wt% Ag and 4-4.3 wt% Cu).

Unlike in the case of the EBA objects, for the V/E type bracelets and their IA evolutions no compositional difference is observed for the Portuguese and Galician productions.

4.2 The gold torcs

4.2.1 The Portuguese torcs

To provide further insight on gold alloys used in the region of Vila Real, we provide below the composition of the three IA torcs from Codeçais, Paradela do Rio and Rendufe, all of which have diamond cross-sectioned bodies and hollow double reel shaped terminals with flat circular ends decorated with a floral motif. We plotted in Figure 10 the torcs with all the other objects from Vila Real so far analysed. In addition to those already mentioned, were considered in the diagram the six gold plates from the LBA-early IA necklace found at Malhada (Silva 1986) analysed by Guerra and Tissot (2021b), and the gold torc presumably from Chaves analysed by Alves et al. (2002). The other components of the Treasure of Lebução, analysed by Hartmann (1971), have also been considered. In addition to fragments of torcs, this treasure contains a complete gold torc with hollow double reel shaped terminals with decorated flat circular ends (Cardozo 1942, Blanco Freijeiro 1958). The elaborate motif contains a rosette enclosed in a circle. Only the bracelet from Telões, analysed by Severo (1905-8c) using an unspecified technique, has not been considered.

Figure 10 - The Cu and Ag contents contained in the gold objects from the region of Vila Real and in the EBA beads (Buraco da Pala) and LIA torc (Vilas Boas) from the neighbouring region of Bragança.

The objects split into two groups, one characterised by low amounts of copper and silver and another by higher amounts of these two elements. The latter contains objects made using intentional gold alloys containing more than 20 wt% Ag and higher amounts of copper: all the torcs, with the exception of the one from Rendufe, and the bracelet from Lebução. Dated to the IA, the torcs have been made in periods when metallurgical practices related to the production of intentional gold alloys were probably well established. The elaborate LIA torc found in Vilas Boas (Hartmann 1982, Ladra 2011), near Vila Flor in the neighbouring region of Bragança, added to the diagram plots with the other represented torcs.

The torc from Rendufe, containing lower amounts of silver (13-14 wt%) and slightly less copper (less than 1 wt%) than the other torcs, plots in the second group with the other objects from Vila Real represented in the diagram. The three analysed parts of this torc, among which the two different flat ends of the terminals, are made from different alloys. Unlike the cast body, the flat ends contain some more copper and silver than the single component of the chain of spirals containing more than 11.3 wt% Ag. A similar amount of copper (*c.* 0.7 wt% Cu) was observed for the six plates from the LBA-IA string from Malhada, which contain, however, less silver (*c.* 10 wt%, like the bracelet from Torgueda and the V/E type bracelet from Chaves). Several components of the chain from Chaves also contain equivalent amounts of silver.

The chemical pattern that emerges from Figure 10 seems to suggest the use in Vila Real during the earliest periods of gold containing less than *c.* 10 wt% Ag and the use of gold with higher amounts of silver, but not exceeding 13 wt%, in the following periods, including in the beginning of the IA. With the exception of the bracelet from Vinhós, all LBA or later productions contain 10 wt% Ag or more. The amounts of copper only increase in objects containing more than 10 wt% Ag. This can be related to the exploitation of different gold deposits and perhaps to unintentional and then intentional changes in the metallurgical processes, possibly towards the use of intentional alloys.

On the basis of this chemical pattern, the bracelet from Vinhós could be an earlier production than the bracelet from Torgueda and the chain of spirals from Chaves could be a group of chronologically heterogeneous components. If some of the components of the chain match compositionally the EBA objects, one has identical composition to the cast body of the IA torc from Rendufe and several others contain the same amounts of silver as the LBA-IA string from Malhada and the LBA bracelet from Chaves.

4.2.2 The torcs from Lugo

In order to shed more light on the gold alloys employed in the NW of the Iberian Peninsula, we analysed in this work two fragments of the same type of torc, found in Lugo, Galicia. However, they are made from alloys that do not entirely correspond to those used in the production of the torcs from Vila Real and the one from Vilas Boas (Bragança), as shown in Figure 11, where the two fragments were plotted with torcs found in the same region (published by several authors: Hartmann 1982; García-Vuelta and Montero-Ruíz 2007; Martinón-Torres and Ladra 2018). Four are unprovenanced and the others were found in Castro de Viladonga (Castro de Rey), Castro da Recadieira (Mondoñedo), Ribadeo, A Madorra (Cospeito), and Burela (Cervo). The EBA objects from Galicia and Portuguese regions situated north of Douro river, plotted in Figure 8, were also added to the diagram.

The two fragments of torc contain amounts of silver observed for EBA objects from Galicia. They could have been made by adding a small amount of copper to native gold. Several other torcs from Lugo are in the same situation, including all those from A Madorra. Like the body of the torc from Rendufe, one of the five torcs from A Madorra (Acc. no. 1972/64-4, National Archaeological Museum in Madrid) contains the same low amounts of silver and copper. It consists of a diamond cross-sectioned body and hollow double reel-shaped terminals (García-Vuelta and Montero-Ruiz 2007, pl. 1-4). Its body is decorated by punching consecutive dots in the wax, a technique used to decorate the torcs from Vila Real.

The other torcs from Lugo contain much higher amounts of silver and copper, such as the torcs and the bracelet comprised in the LIA Treasure of Lebução, found in Portugal and plotted in Figure 10.

Figure 11 - The Cu and Ag contents contained in the torc fragments from Lugo compared to torcs from Lugo published by several authors (see text). The two lines indicate the range of silver contents defined by the EBA objects found in Galicia, added to the diagram. The EBA objects from Portuguese regions situated north of Douro river have also been added and are contained in the square.

4.2.3 Torcs and late treasures: looking for debasement

As in periods of difficult access to gold (Molina, 1996) it is expected the use of alloys containing lower amounts of gold (debasement), we reconsider in Figure 12 the composition of the objects from Vila Real plotted in Figure 10 with two treasures found north of Douro river, in the neighbouring region of Porto, situated southwest of Vila Real.

One of them is the Treasure of Gondeiro, analysed by Hartmann (1971), which was found at Salvador do Monte in Amarante (Cardozo 1930). Amarante is situated between Trás-os-Montes and Minho, at *c.* 40 km from Vila Real. The treasure consists of two coiled cast decorated diamond cross-sectioned gold torcs with expanded square-shaped ends, and an undecorated small spiral and a spiral fragment both of square section with expanded square-shaped ends. Silva (1996) suggests the same period of manufacturing for these objects and the string of Malhada. The other group of objects considered is the Treasure of Estela, found a little further, in Póvoa de Varzim, and recently analysed by Armada and García-Vuelta (2021). More recent, dated to the Roman period (Silva 1996), this treasure contains jewellery and ingots (Fortes 1905-8b), and includes a gold hollow double reel-shaped terminal with decorated flat end (Hartmann 1971, fig 1). Only the jewellery was considered in the diagram.

We observe in Figure 12 that, with the exception of the torc from Rendufe, all the torcs from Vila Real, the Treasure of Gondeiro and the Treasure of Estela are contained in a group characterised by high amounts of both silver and copper. This group also includes the torc from Bragança. All the other objects plot closer to the Portuguese EBA objects from north of Douro river. Therefore, the composition of the cast body of the torc from Rendufe suggests the use of native gold used as found whereas the terminals seem to be made from gold alloys obtained by adding small amounts of copper to a “northern” native gold. The same pattern is observed for the six so far analysed plaques from Malhada.

Although the use of recycling cannot be ignored and would explain (at least partially) the amounts of silver and copper contained in the torcs from Vila Real and Bragança as well as in the jewellery from Gondeiro and Estela, the alloys used to make these objects seem to be obtained by continuously adding more silver and copper to a "northern" gold. Indeed, these alloys contain high amount of silver, reaching more than 40 wt% or even exceeding it as in the case of a fragment of torc from Lebução, which contains 50 wt% of this element.

Figure 12 - The Cu and Ag contents contained in the torcs from Vila Real compared to the objects from Vila Real analysed in this work as well as the plates from Malhada analysed by Guerra and Tissot (2021b), the EBA objects from North Portugal (in Figure 8), the Treasure of Gondeiro and the torc from Bragança analysed by Hartmann (1971), and the jewellery contained in the Treasure of Estela analysed by Armada and García-Vuelta (2021). The hypothetical alloys estimated by adding silver and copper to a native gold containing 10 wt% Ag and 0.2 wt% Cu were added to the diagram.

Although a "Vila Real native gold" is difficult to define, it is possible to make a rough estimation of the intentional gold alloys that can be obtained by adding increasing quantities of silver and copper to a native gold, which was probably locally available when the metallurgical practices related to the production of intentional gold alloys started to be implemented. This means native gold used before or during early IA. The string from Malhada, dated to the LBA-early IA by Silva (1986), appears to be made from a gold alloy obtained by addition (perhaps unintentional) of a small amount of copper to native gold used as found. This gold contains *c.* 10 wt% Ag, a value close to the average silver content (9.3 wt% Ag) of the Treasure of Chaves. Indeed, silver is commonly present in native gold in amounts within the range 5 to 15 wt% (Boyle 1979, 1987).

On the basis of a native gold containing 10 wt% Ag and 0.2 wt% Cu (amount commonly observed for native gold), we estimated the alloys that are obtained by adding 1 part of silver (100 wt%) to decreasing parts of native gold (8 to 1) and increasing copper contents (1.5 to 12 wt%) that compensate for the

colour of the gold alloy. We considered native gold instead of pure gold, because native gold would need to be refined, which is an expensive and unnecessary operation for a metal intended to be mixed with other metals. This estimation does not consider the possible recycling operations and thus the contribution of the amounts of copper and silver present in recycled gold and silver items.

The values obtained have been added to Figure 12 showing that the chemical range defined by the jewellery is entirely covered by the hypothetical alloys and that debasement of locally available gold is a possibility to be considered.

5. Conclusion

Although found in one of the richest gold-bearing areas of the Iberian Peninsula, very few of the early gold jewellery item from the region of Vila Real, in North Portugal, had to date been studied using science-based techniques. In this work, several of the most representative objects of this region have been analysed. Among them, those that constitute the Treasure of Chaves. One of the three items comprised in this treasure is the spiral ring from Fraga da Pitorga, obtained by hammering and coiling a gold bar. This ring is the first EBA object from Vila Real to be analysed. The second object, found at Alto da Pedisqueira in Chaves, is one of the three chains of gold spiral components found in North Portugal. It consists of ring-shaped components made from coiled rounded wire, intertwined or assembled by twisting gold wires and a gold strip around their bodies. The third component of the treasure is a V/E type bracelet. Cast in a tubular wax mould decorated using rotary tools, and cut after casting, the bracelet, also found at Alto da Pedisqueira, is one of the scarce northern examples of this type of jewel. While all three components of the Treasure of Chaves have shown to be made using the expected manufacture technologies, it should be noted that none of them show signs of wear.

To gather further information on the work of gold in Vila Real, were analysed two solid LBA bracelets, one found at Torgueda and the other at Vinhós, and three torcs, all of the same type, typical productions of the area of Chaves ("flaviense" region). With diamond cross-sectioned bodies and hollow double reel-shaped terminals with decorated flat ends, they were found in Rendufe, Codeçais and Paradela do Rio. Made using the same technique, the decoration of their bodies and of the flat ends of their terminals was obtained by punching consecutive dots in the wax following the incised guidelines of the motif. Two fragments of the same type of torc found in Lugo, Galicia, were also analysed for comparison.

The composition of the analysed objects has provided significant clues to the gold alloys employed over time in the region of Vila Real. The EBA spiral ring from Fraga da Pitorga contains the same amount of silver as the six EBA beads from Buraco da Pala (published by Cavalheiro and Sanches 1995). The 7-8 wt% Ag observed for these EBA objects from the natural region of Trás-os-Montes is lower than the amounts of silver contained in EBA objects found in neighbouring regions. The chemical splitting suggests that during the earliest periods was exploited in the northern natural regions of Trás-os-Montes, Minho and Galicia local native gold, which was used as found.

The majority of the objects produced in Vila Real in the following periods and until early IA contain higher amounts of silver than the EBA objects from Trás-os-Montes. However, these amounts do not exceed 13 wt%. On the basis of these results, the bracelet from Vinhós could be an earlier production than the bracelet from Torgueda. Higher amounts of silver are observed only for the gold torcs and the bracelet from Lebução, all IA productions. Regarding the copper, the amounts of this element only start to increase in objects produced from the LBA-early IA onwards, suggesting the use until this period of unprocessed native gold.

The components of the chain of spirals from Chaves, containing variable amounts of silver and very low copper contents, seem heterogeneous. Some spirals have the same composition as the EBA objects, whereas another is compositionally identical to the body of the IA torc from Rendufe and several others contain the amount of silver observed for the V/E type bracelet from Chaves, for the LBA bracelet from Torgueda and for the six plaques contained in the LBA-IA string from Malhada. Considering that all the components were made from local gold, we can suggest that this chain either is a group of chronologically heterogeneous components or contains components made by recycling gold from previous periods. The Treasure of Arnozela (Armbruster 2010), found in the neighbouring region of Braga (Severo 1905-8b) and comprising gold ring-shaped items, wires and bands is in the same situation. The analysis by Guerra and Tissot (2021b) of all the components of this treasure has shown the

variability of the alloys and allowed to suggest the LBA-early IA as *terminus post quem* of the deposition.

With the exception of the torc from Rendufe, all the torcs from Vila Real contain much higher amounts of silver and copper than the other objects found in this region analysed to date. The body and the terminals of the torc from Rendufe are made from slightly different alloys. The body contains the same amount of silver as one of the components of the chain of spirals and 0.3 wt% Cu, suggesting the use of unprocessed native gold. Like the six components of the LBA-IA string from Malhada, the terminals of the torc contain a little more copper, but under 1 wt%, suggesting the use of alloys obtained by unintentional or intentional addition of copper to local native gold. The composition of the two fragments of torc from Lugo, as well as the composition of other torcs from the same region, also suggest the use of alloys obtained by adding a little copper to local native gold. Interestingly, one of the torcs from A Madorra (Galicia), the one with the same composition of the torc from Rendufe, is decorated by punching consecutive dots in the wax as all the "flaviense" torcs studied in this work.

The small amount of copper observed for the torc from Rendufe (and for the string from Malhada) may correspond to either an unintentional or an intentional change in the metallurgical process, conceivably towards the use of intentional alloys. All the other torcs considered in this work are made using intentional gold alloys. Dated to different phases of the IA, they were certainly made when the metallurgical practices related to the production of intentional gold alloys were well established.

Based on a possible local native gold containing 10 wt% Ag and 0.2 wt% Cu, we have estimated the composition of the debased alloys that can be obtained by adding copper and increasing amounts of silver to this gold. When plotted with the torcs and jewellery from North Portugal dated from the IA to the early Roman period, the hypothetical compositions span the chemical range defined by the gold objects, suggesting debasement as a possible metallurgical practice.

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