The gold of the Carambolo Treasure: New data on its origin by elemental (LA-ICP-MS) and lead isotope (MC-ICP-MS) analysis

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A B S T R A C T

The Carambolo Treasure (Seville, Spain), is a key collection of materials from the 1st Millennium BC Mediterranean. Besides the uniqueness, technical complexity and beauty of this assemblage of gold associated with the mythical name of Tartessos, the treasure has been at the epicentre of debates over the last 50 years regarding the Phoenician presence in the west and the origin of the first great western civilization. However, the absence of a precise archaeological context and systematic analyses aimed at identifying the source of the supply of gold have led to diverse and conflicting interpretations in terms of its functionality (ritual from a Phoenician temple versus ostentation of a palatial royalty), and origin (Atlantic *vs* Eastern Mediterranean).

New chemical (by LA-ICP-MS) and isotopic data (Pb by MC-ICP-MS) are presented in this work, which provide an alternative interpretation. The results suggest that the origins of the gold may not be thousands of kilometres away, in the Atlantic or the Mediterranean, but rather in the same region. We highlight geochemical similarities with the gold of the preceding 3rd Millennium BC civilization, with its main political and economic hub at Valencina de la Concepción, located just 2000 m from the Carambolo itself.

1. Introduction

Gold has fascinated people in most cultures since early times. After the emergence of the first political systems, its principal functions have been the production of goods oriented to the expression-reproduction of power and social inequality and a store of wealth. Such is the case of the most important goldsmithing collection in the Protohistory of the Iberian Peninsula, highlighted at the exhibition *Assyria to Iberia at the Dawn of the Classical Age*, held in the Metropolitan Museum of Art of New York in 2014, now kept in the Archaeological Museum of Seville (Spain): The Carambolo Treasure (Fig. 1).

Its accidental discovery inside a vase, in 1958 (Carriazo, 1958, 1959), was an extremely important event in the world of Spanish archaeology: the twenty-one gold jewels weighing 2.770 kg and showing goldsmithing techniques such as filigree, soldering, granulation, enamelling in blue and probably in white (Navarro and San Martín, 2017) were presented as the first material evidence of the mythical kingdom of Tartessos, recorded in classical sources (e.g. Herodotus, Hecateo, Avianus), whose site numerous researchers, such as A. Schulten (1923, 1924), had attempted to find since the end of the 19th and early 20th centuries (Torres, 2002; Escacena, 2010).

This circumstance, alongside the enormous acceptance of diffusionist ideas (Trigger, 1989) in Spanish archaeology and the scarcity of efficient systematic analyses aimed at precisely identifying the source of the gold, fuelled profuse and controversial debates regarding the functionality and origin of this treasure, although at all times under the same paradigm: the Phoenician presence in the West.

A first interpretation purported that the jewels came from the crown and ornaments from Arganthonios, the mythical long-lived

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Fig. 1. The Carambolo treasure (Copyright: Regional council of culture. Granted by the museum of Seville. Author: J. Morón).

king of Tartessos (Carriazo, 1958, 1959, 1973). Nevertheless, at the turn of the 21st century, the documentation of new gold products in the Carambolo linked to a unique monumental building interpreted as a sanctuary and dated between 2770 ± 50 BP and 2502 ± 40 BP (Fernández and Rodríguez, 2005a,b, 2010), led to an approximation of the chronology of the Treasure (towards the end of the 8th century – early 7th century BC), the possible date of when it was hidden (end of the 7th – early 6th century BC) and this led to articulating alternative functional proposals, such as the ritual dowry of a priest besides an ox and cow for sacrifice in the liturgical commemorations dedicated to the Phoenician gods, Baal and Astarte (Amores and Escacena, 2003; Belén, 2011; De la Bandera et al., 2010; Fernández and Rodríguez, 2010).

In the geochemical field, Kalb and Hartmann (1969) obtained the first elemental results on a sample of the treasure through optical emission spectroscopy and SEM, setting its origin in the East Mediterranean (Gold Type U), due to the presence of a copper alloy technology and alluvial traces of platinum.

The latter element (Pt), along with the presence of copper in the alloy, were determining factors in their traditional interpretation, since they were considered as indicators/tracers of a technological change and a supply of external provenance (Atlantic and/or Eastern Mediterranean), which would determine a chronological threshold (8^{th}-7th century BC) for gold metallurgy on the Iberian Peninsula for which the treasure would be one of its first representatives (Hartmann, 1970, 1982; Kalb and Hartmann, 1969).

This perspective remained almost unaltered since subsequent elemental analyses, despite having increased knowledge of joining techniques in the construction of jewels and other interesting data, have been unable to move forward in evaluating its provenance due to the analytical resolution (detection of major elements: Ag/Au/Cu) of the applied (XRF and PIXE) methodology (Hartmann, 1982; Ontalba et al., 2002; Scrivano et al., 2013; etc.).

The stylistic and typological studies, without entering the debate over the origin of the raw material and after noting the lack of parallels for the jewels in the Mediterranean goldwork of the 1st Millennium BC, would point to an interpretation focused on its origin. The Carambolo Treasure would be the expression of the dawn, at the turn of the 7th century BC, of a new local technological tradition (Tartessian Culture) resulting from the convergence of Atlantic goldsmithing at the end of the Bronze Age with the Egyptian-Mesopotamian tradition of the Eastern Mediterranean; this influx would arrive with the Phoenician colonization, and even at an earlier date (Perea and Armbruster, 1998, 2007; Guerra and Rehren, 2009).

Against this background, we consider that an alternative proposal could be put forward to explain the origin of the Carambolo Treasure, based on a new scientific research programme designed to compare the provenance of the gold and its manufacture based on an analysis of its elemental and isotopic composition, using a combination of the two advanced techniques: laser ablation (by LA-ICP-MS) and lead isotopes (by MC-ICP-MS and LA-MC-ICP-MS). Additionally, we perform a comparative study of the results in space and time, regarding the new records of gold producing recovered in the course of the most recent archaeological interventions in the Carambolo (see above), contemporary and subsequent productions (8^{th}-4th centuries BC) and the gold productions that preceded it almost 2000 years earlier (3rd Millennium BC) in the same geographical area: the Lower Guadalquivir Basin (Fig. 2).

This work presents new chemical and isotopic data which enable an alternative interpretation as to the origin of the Carambolo Treasure. The results suggest a direct link with the gold metallurgy from the 3rd Millennium BC civilization in the Lower Guadalquivir Basin and with its main economic and political hub: Valencina de la Concepción. This settlement spread alongside the Carambolo hill and acted as a gateway for raw materials of regional and transcontinental origin (i.e. copper, gold, ivory, amber, limestone), and a space for their artisanal transformation into products (Nocete, 2001, 2014; Nocete and Nocete, 2015; Nocete et al., 2005, 2008, 2013), including gold metallurgy (Nocete et al., 2014).

2. Materials and method

The Carambolo Treasure comprises twenty-one pieces of jewellery. A necklace -DO5489 - from which hang seven seals; two cylindrical bracelets in a circular section -DO5487 and DO5488; three rectangular shaped plates, among which two formal and decorative sets can be distinguished: eight of them with aligned motifs of semi-spherical bodies with a central umbro alternating with two types of tensioning elements -DO5490-5491-5492-5493-5498-5499-5500-5501- and a further eight with alignments of

Fig. 2. Location of the archaeological sites from the Lower Guadalquivir Basin (Spain) referenced in the text.

capsules sealed with lamina decorated with rosettes, alternating with semi-spherical bodies -DO5494-5495-5496-5497-5502-5503-5504-5505-; and lastly, two necklaces in a rectangular shape with concave sides -DO5485-5486- or in “bull hide” (for a full description, see Perea and Armbruster, 1998; Navarro and San Martín, 2017).

We selected two samples for analysis. A fragment of base sheet (Table 1, No. 1) and a fragment of the semi-sphere (Table 1, No. 2) belonging to one of the plates (DO5505) deposited in the Archaeological Museum of Seville, which was broken at the time of the finding (Fig. 3).

In order to evaluate its relationship with contemporary and subsequent 1st Millennium BC gold productions from its closest geographical area (Lower Guadalquivir Basin), six further samples were selected in the Archaeological Museum of Seville (Table 1, No. 3–8, Figs. 4 and 5). The two gold products documented in the recent excavations of the Carambolo (Table 1, No. 3 and 4, Figs. 4 and 5), two samples from the main series of goldsmithing in the region: Seville (Table 1, No. 5, Figs. 4 and 5) and the Ebora Treasure (Table 1, No. 6, Figs. 4 and 5) in Cadiz, and two additional samples belonging to the main funerary contexts: Cruz del Negro (Table 1, No. 7, Figs. 4 and 5) and El Acebuchal (Table 1, No. 8, Figs. 4 and 5) in Carmona, Seville.

To evaluate its relationship with earlier goldsmithing productions from the 3rd Millennium BC in the same geographical area, seven additional decorated samples belonging to funerary contexts were selected in the Archaeological Museum of Seville (Table 1, No. 9–15, Fig. 6). Four recovered from the main megalithic tombs of Valencina de la Concepción (Seville), located in the surrounding area (Fig. 7) of the Carambolo (Table 1: Los Veinte No. 9, Matarrubilla No. 10–12, Divina Pastora No. 13), a fifth belonging to the tholos tomb of Las Canteras (Table 1, No. 14, Figs. 6 and 8) from Alcalá de Guadaira (Seville) and the last from the hypogeum tomb of Los Algarbes from Tarifa (Table 1, No. 15, Figs. 6 and 8) in Cadiz. Added to these are the results of seven additional undecorated samples, previously analysed, that echo contexts with radiocarbon dating in settlements (Valencina de la Concepción and Cabezo Juré: Table 1, No, 16–21, Fig. 9) and a sample of primary gold (La Sultana mine: Table 1, No, 22) located in the area studied (Fig. 2), in order to contrast potential catchment areas.

All in all, this study was based on the analysis of twenty-two samples of archaeological gold types (Table 1).

Elemental analyses of archaeological gold samples were conducted by laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at the Geochronology and Isotope Geochemistry-SGIker facility of the University of the Basque Country (Spain). Gold samples were cleaned with ultrapure water and set on a 25 mm diameter resin mount with double sided adhesive tape. The analyses involved the ablation of the gold with a UP213 Nd:YAG laser ablation system (NewWave) coupled to a Thermo Fisher Scientific XSeries 2 quadrupole ICP-MS instrument with enhanced sensitivity through a dual pumping system. Spot diameters of ca. 100 μm associated with repetition rates of 10 Hz and laser fluence at the target of ca. 5 J/cm^2 were used for the gold analysis. A pre-ablation was conducted to remove surface contamination, followed by ablation during 60 s. The ablated material was carried into helium and then mixed with argon. The procedure applied was based on that described by Kovacs et al. (2009), i.e., using the so-called 'wet ablation' method which, in this case, involved the use of a Peltier refrigerated spray chamber and a PFA micronebulizer to introduce calibration solutions of known concentration for the quantification of the elements of interest (see Kovacs et al., 2009 for details). Tuning and mass calibration were performed using the NIST SRM 612 reference glass, by inspecting the signal of ^{238}U to obtain ca. 1.400.000 cps/μg g^{-1} and by minimizing the ThO^+/Th^+ ratio to ca. 1.5%.

One of the main problems in the elemental analysis of gold-rich archaeological samples by LA-ICP-MS methods is the absence of certified standards similar in nature to those investigated. To guarantee the reliability of the results obtained in our study, we have applied a variety of approaches. Firstly, after a proper tuning and mass calibration of the instrument, we verified that the amounts of Au and Ag, i.e., the potential main components of the archaeological unknowns, were correctly measured. This was achieved by the repeated analysis of the NIST SRM 481 standard, which consists of six gold-silver alloys with different amounts of the two metals. Using the conditions described above, the analysis of the NIST silver-gold wires satisfactorily reproduced the certified values to less than 1% SE. As for the trace element contents, we used a double strategy. On the one hand, by means of the silicate NIST glasses for which there is abundant information of major and trace element composition. This is a standard procedure in many LA-ICP-MS studies when no standards are available of matrix composition similar to that of the unknowns. In this case, the method employed provided trace element results with less than a 20% deviation from the reported values (Jochum et al. 2011), which is normally regarded as acceptable in this type of analysis. At the same time, we used a secondary standard consisting of a commercially available gold foil (Alfa Aesar 14721-FF) whose composition was carefully established (<10% error for all the elements determined) by solution ICP-MS using the method of standard addition. Once we had verified the validity and repetitiveness of the method, we selected a

Table 1
Gold samples analysed.

Nº	Archaeological site	Lab. No	Museum No,	Context	Description	References
1st Millenium BC						
1	El Carambolo (Camas, Seville)	MIDAS 599	DO5505a	Treasure	Base sheet	Carriazo, 1973; Perea and Armbruster, 1998; Ontalba et al., 2002; Scrivano et al., 2013; Navarro and San Martín, 2017.
2	El Carambolo (Camas, Seville)	MIDAS 600	DO5505b	Treasure	Semi-Sphere	
3	El Carambolo (Camas, Seville)	MIDAS 608	DJ2002/24/1022-165	New findings	Tube with striated decoration (tube-shaped sheet)	Fernández and Rodríguez, 2007; Escacena et al., 2007; Hunt et al., 2010; Perea and Hunt, 2010
4	El Carambolo (Camas, Seville)	MIDAS 609	DJ2002/24/1022-168	New findings	Button/circular fitting with onfalo	
5	Seville	MIDAS 605	DJ2016/02	Treasure	Earring	Fernández, 1997
6	Cortijo Ebora (Cádiz)	MIDAS 604	REP09125	Treasure	Biconical bead with granulated decoration	Carriazo, 1973
7	Cruz del Negro (Carmona, Seville)	MIDAS 613	OD9777	Burial (incineration pit)	Decorated bead	Bonsor, 1889; Amores and Fernández, 2000
8	El Acebuchal (Carmona, Seville)	MIDAS 607	OD0090/OD0100	Burial (Tumulo G)	Hemispherical button	Bonsor, 1889; Fernández, 1997
3rd Millenium BC						
9	Valencina de la Concepción (Seville)	MIDAS 602	REP1996/07-2	Burial (Los Veinte Tholos)	Decorated sheet (geometric motifs in relief)	Fernández, 1997; Fernández and Oliva, 1983, Murillo, 2016
10	Valencina de la Concepción (Seville)	MIDAS 601	REP11723 EP1185	Burial (Matarrubillla Tholos)	Undecorated sheet	Collantes, 1969; Fernández, 1997; Murillo, 2016
11	Valencina de la Concepción (Seville)	MIDAS 603	REP11723 REP11851	Burial (Matarrubillla Tholos)	Undecorated sheet	Collantes, 1969; Fernández, 1997; Murillo, 2016
12	Valencina de la Concepción (Seville)	MIDAS 611	REP11853	Burial (Matarrubillla Tholos)	Decorated sheet (geometric motifs in relief)	Collantes, 1969; Fernández, 1997; Murillo, 2016
13	Valencina de la Concepción (Seville)	MIDAS 612	DJ1996/18-01	Burial (Divina Pastora Tholos T2)	Decorated sheet (geometric motifs in relief)	Arteaga and Cruz-Auñón, 2001; Murillo, 2016
14	Las Canteras (Alcalá de Guadaira, Seville)	MIDAS 614	REP1988/126	Burial (Tholos)	Decorated sheet (geometric and eye-motifs in relief)	Fernández, 1997: Hurtado and Amores, 1984
15	Los Algarbes (Tarifa, Cádiz)	MIDAS 606	OD7028	Burial (Hipogeo 5)	Decorated spherical sheet (geometric motifs in relief)	Possac, 1975; Fernández, 1997; Lorenzo, 1998
16	Valencina de la Concepción (Seville)	MIDAS 487		Residential area	Undecorated sheet	Nocete et al., 2014
17	Valencina de la Concepción (Seville)	MIDAS 505		Production area	Casting spill	Nocete et al., 2014
18	Cabezo Juré (Alosno, Huelva)	MIDAS 378		Fortified area	Undecorated sheet	Nocete et al., 2014
19	Cabezo Juré (Alosno, Huelva)	MIDAS 435		Fortified area	Undecorated sheet	Nocete et al., 2014
20	Cabezo Juré (Alosno, Huelva)	MIDAS 434		Fortified area	Undecorated sheet	Nocete et al., 2014
21	Cabezo Juré (Alosno, Huelva)	MIDAS 506		Fortified area	Undecorated sheet	Nocete et al., 2014
22	La Sultana (Cala, Huelva)	MIDAS 436		Mine	Raw material	Nocete et al., 2014

Fig. 3. The Carambolo Treasure: Samples analysed.

Fig. 4. 1st Millennium BC: Gold samples analysed. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

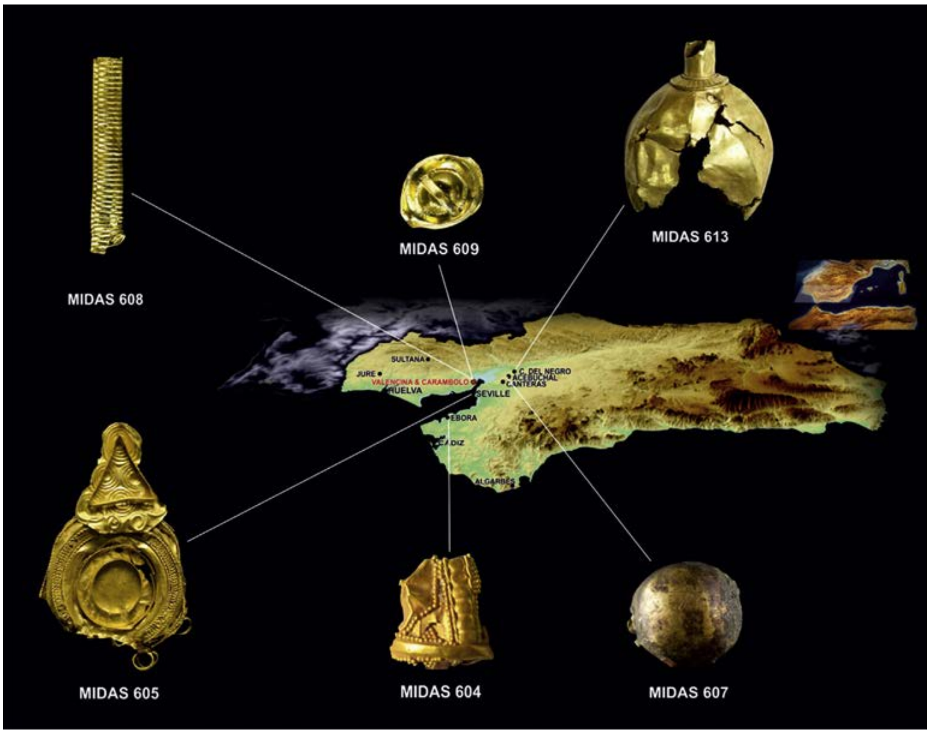

Fig. 5. 1st Millennium BC: Location of gold samples analysed in the Lower Guadalquivir Basin. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

number of elements for their quantification on the basis of previous studies on Au provenance in archaeological remains (e.g. 30; Kovacs et al., 2009; Shlosser et al., 2009; *inter alia*). The calculation of the concentrations was performed assuming that the analysis included all the elements present in the sample. A minimum of 3–6 spots were analysed on each gold sample. Spots using diameters

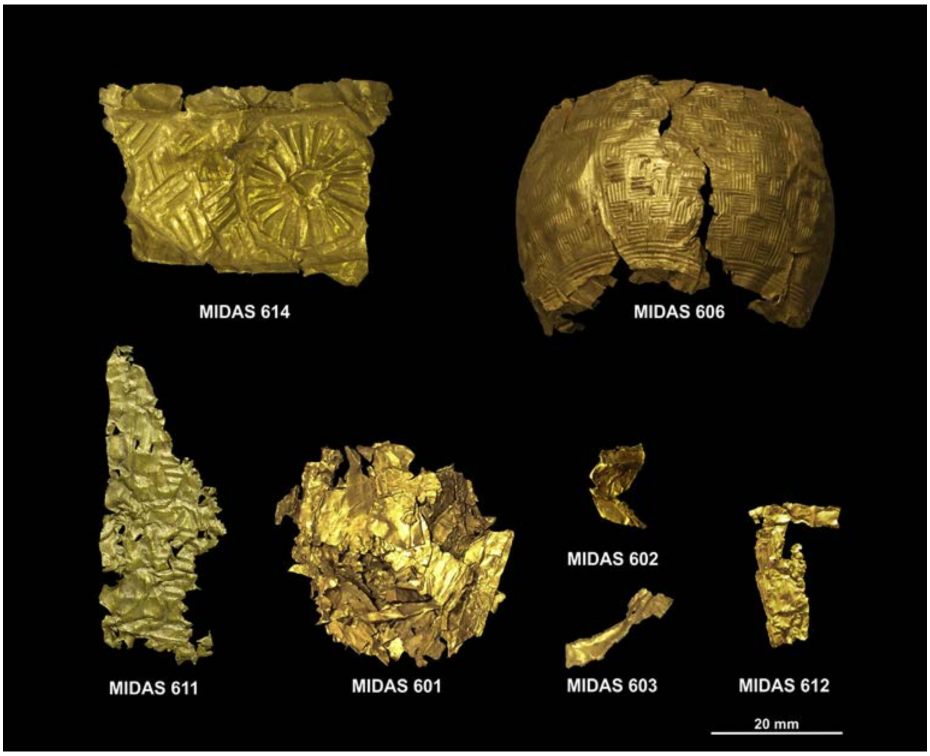

Fig. 6. 3rd Millennium BC: Gold samples analysed. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

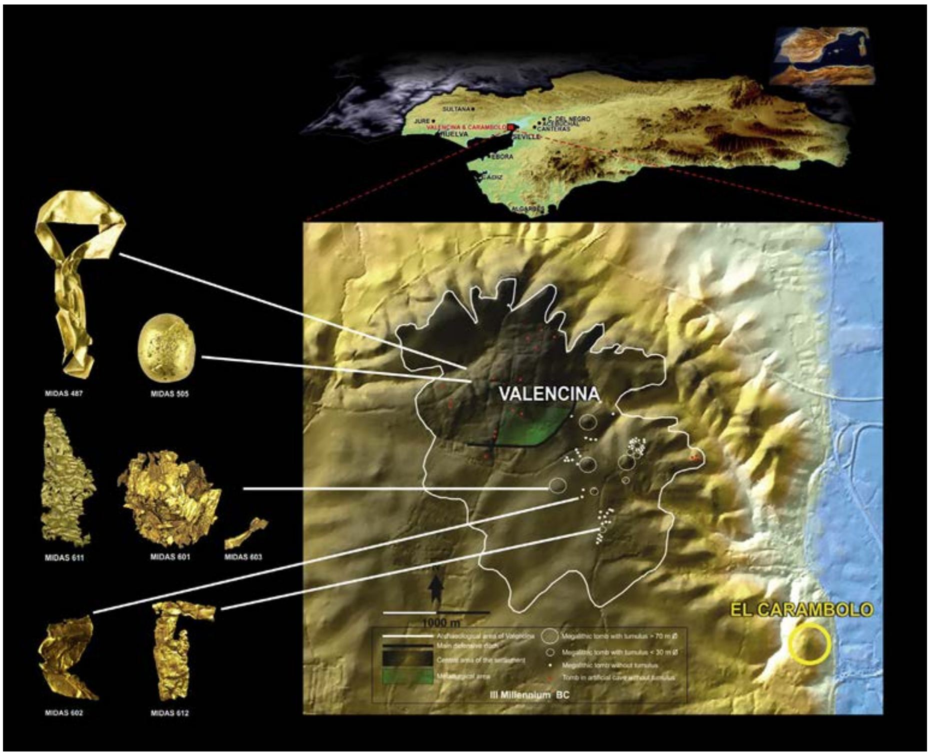

Fig. 7. 3rd Millennium BC: Location of gold samples analysed at Valencina. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

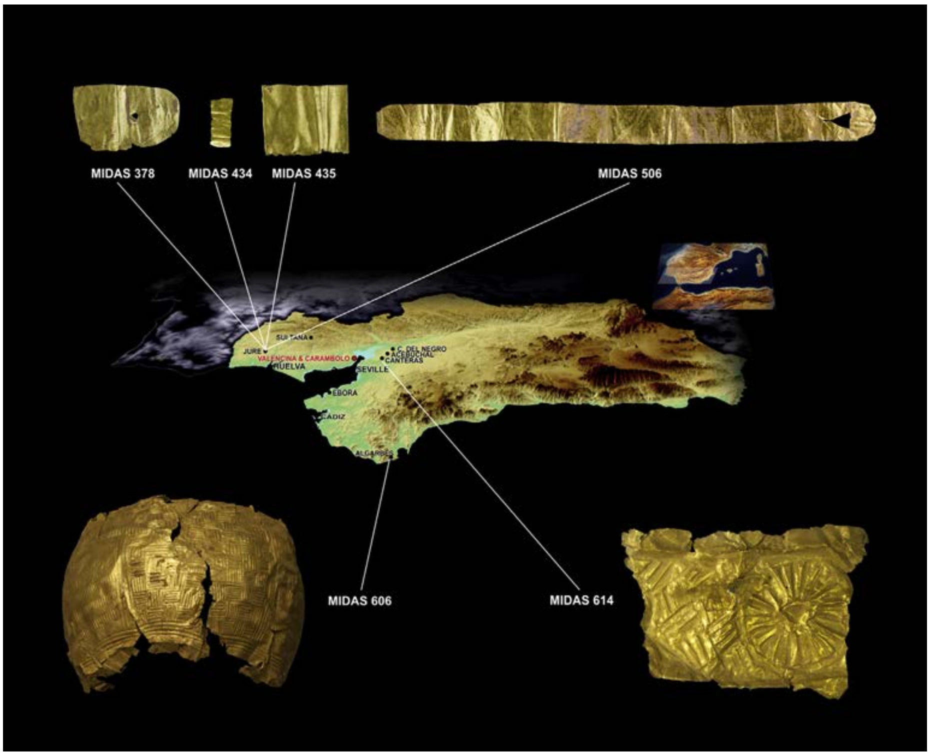

Fig. 8. 3rdMillennium BC: Location of gold samples analysed in the Lower Guadalquivir Basin. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

less than 100 μm yielded significantly increased errors and have not been taken into account.

Lead isotope analyses by MC-ICP-MS were performed at the Geochronology and Geochemistry-SGIker facility of the University of the Basque Country UPV/EHU (Spain). The gold samples were cleaned overnight in hot, nitric acid concentrate, rinsed with ultrapure water and finally dissolved in *aqua regia*. Lead was extracted by conventional ion-exchange chromatography in HBr-HCl media; all reagents employed were triple-distilled at sub-boiling temperature before use. Pb isotope ratios have been obtained using a Neptune MC-ICP-MS instrument with the thallium correction technique (Walder et al., 1993), following procedures similar to Chernyshev et al. (2007). Due to the low Pb contents, samples were introduced into the plasma through an Apex IR inlet system (Elemental Scientific, USA). The accuracy of the method was confirmed by analysis of the NBS 981 lead isotope certified reference material at similar concentrations to the unknown samples. In cases where it was not possible to have samples for conducting analysis by MC-IPC-MS, lead isotope measurements were performed at the Geochronology and Isotope Geochemistry-SGIker facility of the UPV/EHU using a Neptune MC-ICP-MS coupled to a UP-213 laser ablation system (LA-MC-ICP-MS). Data were collected in static mode during 60 s of ablation with a spot size of 100 μm. The isotopes ^{202}Hg, ^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb were monitored to allow for the correction of isobaric interference of Hg on ^{204}Pb. Data reduction was performed with Iolite 3 (Paton et al., 2011) using the SRM610 for correction of the fractionation and SRM612 for quality control. Additional details on the method used that attest to low deviation among spot results in the same sample, good reproducibility and consistency of the data, even for those ratios that include ^{204}Pb values, are provided in García de Madinabeitia et al. (2017).

The results of elemental analysis are given in Table 2 (1st Millennium BC) and 3 (3rd Millennium BC). The results of isotopic analysis are given in Table 4.

3. Discussion of results

3.1. Elemental analysis by LA-ICP-MS

The results of the elemental analysis, by LA-ICP-MS (Tables 1 and 2: MIDAS 599 and 600), of the base sheet and a semi-sphere of one of the plates from the Carambolo Treasure, show a homogeneous manufacture on a very pure gold (Au fineness 961–979 °/$_{00}$) with low silver content (Ag: 1.29–2.39%). The presence of tin (Sn: 34–173 ppm) and platinum (Pt: 23–43 ppm) suggest an origin in placer deposit in fluvial transport (Brostoff et al., 2009; Guerra and Calligaro, 2003; Troalen et al., 2014) and the proportion of copper (Cu: 0.75–1.58%) an alloying metallurgical process (Bendall et al., 2009; Chapman et al., 2006; Ehser et al., 2011a; Hartmann, 1970, 1982). The higher copper in the base sheet of the plate may have been deliberate, to increase hardness and facilitate handling in goldsmithing, or it may indicate that the decorative elements (semi-sphere) were gilded by depletion (Blakelock et al., 2016; La Niece, 1995).

Comparison of these results with those from a series of goldsmithing samples (Table 2), which cover the span of the first half of the 1st Millennium BC (8th-4th century B.C.) in their closest territorial area, Lower Guadalquivir Basin (Figs. 2 and 5), makes it possible to discriminate their singularity in a context of technological diversity and provenances.

The treasure, along with the new findings in the Carambolo and Acebuchal (Table 2: MIDAS 599,600,607,608,609), point to the existence of a series of workshops (8th-7th century BC), involving the supply and production of extremely pure alluvial types of gold (Sn,

Fig. 9. 3rd Millennium BC: Additional gold samples analysed in Nocete et al. (2014). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Pt, Pt > Pd) with low contents of alloyed copper.

This signature stands out from contemporary productions of the same purity with no detectable copper extracted from mines (absence of Cu, Sn,Ti, Pt), such as Cruz del Negro (Table 2: MIDAS 613), and from subsequent productions (6^{th}-4th century B.C.) of less pure types of gold (Au: 85–87%) with a higher copper alloy content (Cu: 2–3%); it is also different from alluvial sources (Ti *vs* Sn, Pd > Pt), such as the Ebora or Seville Treasures (Table 2: MIDAS 604 y 605). Nonetheless, the Carambolo Treasure has some identifying features that could determine differences in the production process (alloy containing copper) and different supply sources. Along with the slight differences in Mg, Cr, As, Pt, Pb and Bi, notable differences are its higher percentage of gold (Au > 96% *vs* Au < 89%), its lower percentage of silver (Ag < 2% *vs* Ag >9%) and the higher values in platinum throughout the series.

Comparison of these results with those from a series of goldsmithing samples dating from the 3rd Millennium BC from the same geographical area (Table 3 and Figs. 2, 7 and 8) and, more particularly, with the decorated productions documented in funerary contexts, identifies chemical similarities and suggests an alternative interpretation: it is possible that the origin of the gold in the Carambolo Treasure may not derive from distant sources in the

Table 2
Results of the elemental analysis. Gold samples from the Carambolo and other 1st Millennium BC sites. Analytical Technique: LA-ICP-MS. Results in μg g^{-1} (ppm); Au and Ag in % wt. Empy boxes: below detection limit.

Context and description	No,	Mg	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Pd	Ag	Cd	Sn	Sb	Te	Pt	Au	Pb	Bi
EL CARAMBOLO (CAMAS, SEVILLE)																						
Treasure	Laboratory:	5.72	3.51	0.16	1.40	16.8	263	0.75	5.75	15239	5.70		3.74	2.31	0.34	143.0	1.57	1.84	43.0	96.12	5.48	0.53
Base sheet	**MIDAS 599**	3.15		0.16	2.27	13.6	220	0.74	5.65	15011	7.75		4.77	2.33	0.40	157.0	2.03	1.68	42.1	96.12	5.59	0.74
	Museum:	2.00		0.07	1.47	20.9	218	0.89	5.65	15820	6.07		3.47	2.39	0.25	173.2	1.90	1.81	42.9	95.98	7.67	0.62
	DO5505 a	3.13		0.09	1.45	18.1	243	1.02	5.56	15369	5.47		3.40	2.31	0.25	172.1	1.82	1.70	43.1	96.10	8.65	0.54
		2.08		0.05	1.24	16.7	221	0.71	5.68	15711	5.37		3.48	2.30	0.24	162.6	1.70	1.61	44.2	96.08	6.34	0.54
Treasure	Laboratory:	3.03		0.04	0.90	20.9	62.4	0.20	2.22	7985	4.36	5.19	3.14	1.32	0.24	36.71	0.89	1.40	23.6	97.86	6.37	0.13
Semi-Sphere	**MIDAS 600**	1.44			0.97	19.2	56.6	0.19	2.25	7996	3.69	5.48	3.13	1.32	0.25	37.16	0.90	1.39	23.5	97.86	6.49	0.13
	Museum:	1.63		0.07	1.30	19.0	37.2	0.17	1.90	7532	4.64	5.63	3.54	1.31	0.27	34.64	0.90	1.40	24.3	97.93	6.21	0.14
	DO5505 b	0.85			1.06	18.5	31.3	0.18	2.16	7729	4.25	5.90	3.28	1.29	0.21	35.07	0.96	1.34	23.2	97.92	6.69	0.15
		0.59			2.41	16.9	28.4	0.17	1.82	7610	4.01	4.63	9.38	1.31	0.23	34.13	0.92	1.32	23.9	97.92	6.39	0.19
New findings	Laboratory:			0.20		27.89	145.0	0.46	0.67	8072	7.81		2.09	10.57	0.27	59.73	11.51	3.99	21.96	88.57	24.74	3.48
Tube with striated decoration	**MIDAS 608**			0.09		34.51	126.2	0.30	1.11	8036	6.97		2.08	10.60	0.26	52.07	12.26	3.75	21.63	88.55	25.56	3.61
(tube-shaped sheet)	Museum:			0.13		22.19	132.4	0.31	0.58	7476	7.15		2.27	10.44	0.26	50.35	10.86	3.76	22.48	88.77	24.28	3.65
	DJ2002/24/			0.17		29.30	187.7	0.38	0.85	7566	9.55		2.07	10.33	0.25	78.84	14.75	3.77	21.52	88.86	26.41	4.15
	1022-165			0.25		42.60	186.4	0.66	1.09	7473	10.17		2.36	10.52	0.25	45.89	10.03	4.08	24.17	88.68	27.85	3.81
New findings	Laboratory:					5.88	126.3	0.18	1.11	12017	2.89		2.32	9.87	0.26	117.80	2.07	3.78	2.44	88.89	14.29	3.69
Button/circular fitting with onfalo	**MIDAS 609**					13.26	98.9	0.17	0.89	12044	2.79		2.03	9.76	0.24	131.63	2.11	3.51	2.41	89.00	15.89	3.83
	Museum:					14.95	101.6	0.16	0.81	11974	3.27		2.00	9.77	0.23	123.60	2.07	3.61	2.34	89.00	15.11	3.87
	DJ2002/24/		2116	0.18	3.12	8.45	149.5	0.21	0.59	12671	11.98		2.44	9.67	0.25	132.38	2.41	3.48	3.08	88.80	12.61	2.44
	1022-168					4.98	97.2	0.16	0.58	11893	3.02		2.12	9.49	0.24	139.58	2.52	3.36	2.28	89.28	16.97	4.75
EL ACEBUCHAL (CARMONA, SEVILLE)																						
Burial Tumulo G	Laboratory:	4.80		0.48		7.81	138.9	0.47	4.78	10601	12.41	1.48	1.01	9.38	0.34	677.80	6.47	4.21	0.72	89.40	64.10	44.63
Hemispherical button	**MIDAS 607**			0.33		7.77	118.4	0.43	4.02	7151	12.01	1.48	0.91	9.19	0.33	556.21	5.93	4.11	0.69	89.96	50.06	43.92
	Museum:			0.31		7.65	128.3	0.65	17.58	27761	12.77	1.36	1.24	9.05	0.35	594.58	5.98	4.07	0.62	88.04	89.13	42.26
	OD0090/100																					
CRUZ DEL NEGRO (CARMONA, SEVILLE)																						
Burial (incineration pit)	Laboratory:				1.68	1.71			0.32					5.17		5.57		2.47		93.43		0.27
Decorated bead	**MIDAS 613**				1.60	2.98			0.38					5.18		5.58		2.39		93.46		0.24
	Museum:				1.61	2.32			0.31					5.28		4.30		2.56		93.35		0.24
	OD9777				1.78	1.21			0.33					5.18		5.19		2.56		93.43		0.26
					1.67	0.90			0.30					5.23		5.65		2.52		93.37		0.26
CORTIJO DE EBORA (CÁDIZ)																						
Treasure	Laboratory:	6.51	26	0.29		4.90	480.4	0.52	5.25	24240	4.37	3.74	11.2	9.82	0.27	136.1	5.91	4.54	2.94	87.68	21.2	4.08
Biconical bead with	**MIDAS 604**	3.53	21	0.33	0.40	4.60	428.2	0.52	5.47	27200	5.94	3.56	12.0	10.2	0.30	141.6	6.18	4.72	3.16	86.99	23.0	4.50
granulated decoration	Museum:	10.2	329	0.26	0.57	8.71	491.1	0.50	5.83	28270	9.74	6.41	10.9	10.5	0.27	163.3	6.97	4.65	3.04	86.55	28.4	5.25
	REP09125	3.66		0.22		7.68	436.8	0.47	5.17	26717	5.50	5.21	11.7	10.4	0.28	160.5	6.59	4.31	2.87	86.90	22.5	4.35
		8.29	170	0.28	0.43	13.9	497.8	0.57	5.71	26753	8.42	7.05	11.2	10.4	0.26	169.8	7.23	4.29	2.95	86.86	24.8	4.50
SEVILLE																						
Treasure	Laboratory:	12.3	807	0.22	1.95	2.29	31.1	0.18		30661	5.78		70.5	11.1	0.26	3.67	1.02	4.55	14.3	85.71	26.9	1.95
Earring	**MIDAS 605**	13.1	878	0.18	1.69	2.19		0.13		29103	3.81		70.9	11.1	0.26	3.24	0.87	4.25	14.5	85.88	27.2	1.91
	Museum:	9.27	548	0.45	2.15	2.77	28.2	0.25		30042	5.54		69.5	11.3	0.29	3.96	1.13	4.70	14.2	85.62	28.9	1.94
	DJ2016/02	28.5	3443	0.23	4.83	2.16	19.2	0.16		28727	5.80		70.5	10.8	0.26	3.87	1.32	4.05	14.9	85.95	25.9	1.74
		30.7	1876	0.13	2.55	3.33	71.2	0.16		23532	6.50		68.6	10.7	0.29	3.54	0.87	4.30	15.0	86.74	27.2	1.24

Table 3
Results of the elemental analysis. Gold samples from the 3rd Millennium BC. Analytical Technique: LA-ICP-MS. Results in $\mu g\ g^{-1}$ (ppm); Au and Ag in % wt. Empy boxes: below detection limit.

Context and description	No,	Mg	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Pd	Ag	Cd	Sn	Sb	Te	Pt	Au	Pb	Bi
CABEZO JURÉ (ALOSNO, HUELVA)																						
Fortified area	Laboratory:	2.2	1.1			2.6	150		1.4	156			1.0	10.0		2381	28	1.8		89.7	15	
Undecorated sheet	**MIDAS 378**	3.3	1.9			3.6	171		1.7	154		1.5	1.0	10.0		2362	27	1.9		89.8	14	
		1.4				2.7	146		3.1	160		5.7		10.1		2547	28	2.0		89.6	14	
Fortified area	Laboratory:	36	17			15	511		1.5	160		3.4	1.1	10.1		2329	25	1.9		89.6	12	
Undecorated sheet	**MIDAS 435**	15	9.1			7.1	443		1.2	149		9.6	1.0	9.9		2454	28	1.8		89.8	16	
		21	31			12	499		1.2	152		2.5	1.2	10.0		2331	25	1.8		89.7	13	
Fortified area	Laboratory:	9.0	2.0			5.8	203		2.6	166		10		11.1		3099	23	2.1		88.5	24	
Undecorated sheet	**MIDAS 434**	9.3	2.5			6.0	208		2.8	167		6.3		11.1		3168	21	2.1		88.5	20	
		12	8.6			8.6	337		3.2	188		9.1		11.1		3896	25	2.1		88.4	20	
Fortified area	Laboratory:	72	2163			67	504	1.2	29	1488	101	11	1.2	4.7	0.6	255	5.7	11	8.1	94.8	16	0.4
Undecorated sheet	**MIDAS 506**	40	1070			61	271	1.2	27	1387	88	12	1.2	4.6	0.6	231	5.3	11	4.2	95.0	16	0.4
		127	2682			78	855	1.3	28	1435	93	25	1.5	4.7	0.6	219	5.1	11	2.3	94.7	15	0.5
		104	3395	4.3		60	196	1.3	28	1309	85	20	1.5	4.6	0.7	199	4.8	12	1.7	94.7	14	0.4
		63	2415	4.0		69	313	1.4	31	1475	91	16	1.5	4.7	0.6	224	5.0	12	3.1	94.8	13	0.4
		77	6506	21		69	285	1.5	32	1578	108	34	1.8	4.7	0.8	235	5.6	12	5.8	94.3	15	0.5
VALENCINA DE LA CONCEPCIÓN (SEVILLE)																						
Residential area	Laboratory:					1.5	749			1698		2.1	29	14.3		1.1	1.1	2.4		85.5		12
Undecorated sheet	**MIDAS 487**					1.2	709			1715		4.1	28	14.6				2.5		85.1		8.9
		2.9				1.5	783			1699		4.2	27	14.4				2.7		85.3		9.1
Production area	Laboratory:					9.7	65		2.7	1312	20			1.8		7.1	12			98.1	6.4	
Casting spill	**MIDAS 505**					7.0	48		1.5	762		1.1		1.8		1.5	2.4			98.2		
			12			8.8	58		2.0	1118	20			1.8		7.2	11			98.1	5.9	
Burial (Matarrubillla Tholos)	Laboratory:	1.51	8.03	0.41		4.47	26.9	0.28	1.68	273	6.79		0.76	1.28	0.35	7.80	2.80	2.06	0.51	98.68	1.23	0.16
Undecorated sheet	**MIDAS 603**	2.14	2.70	0.29		4.29	27.0	0.27	1.52	273	7.14		0.64	1.25	0.37	7.15	2.51	1.98	0.43	98.72	1.10	0.16
	Museum:	2.58	5.43	0.26		4.24	25.0	0.23	1.35	251	6.03		0.55	1.24	0.33	6.25	2.20	1.89	0.29	98.73	0.98	0.16
	REP 11723	1.24		0.24	0.26	4.36	19.5	0.20	1.47	278	8.47		0.48	1.26	0.33	7.34	2.48	1.90	0.36	98.71	1.06	0.15
	REP11851	4.58	5.46	0.20		4.39	30.3	0.20	1.45	247	5.78		0.33	1.25	0.32	6.60	2.17	1.79	0.21	98.72	0.94	0.09
Burial (Matarrubillla Tholos)	Laboratory:	10.1	300	0.10	2.12	3.43	31.1	0.22	0.62	195	16.1		0.76	1.35	0.31	45.57	1.89	1.46	0.27	98.59	4.07	0.11
Undecorated sheet	**MIDAS 601**	7.03	255	0.09	2.33	3.27	21.6	0.22	0.56	189	15.1		0.89	1.31	0.32	42.42	1.69	1.30	0.18	98.63	3.13	0.18
	Museum:	6.46	244	0.10	1.17	3.21	16.2	0.20	0.56	196	15.3		0.45	1.30	0.30	43.75	1.74	1.33	0.15	98.65	3.37	0.08
	REP11723	7.03	226	0.08	1.17	3.36	23.9	0.22	0.62	240	16.1		0.65	1.33	0.29	45.85	1.79	1.40	0.13	98.61	3.67	0.13
	REP11851	5.26	228	0.04	1.79	3.30	7.9	0.21	0.61	194	15.4		0.72	1.34	0.28	44.51	1.76	1.23		98.61	3.41	0.09
Burial (Matarrubillla Tholos)	Laboratory:	22.09	42.52		1.22	1.88		0.09	0.76	92.81				0.63	0.07		0.69	0.49		99.32	1.52	
Decorated sheet (geometric motifs in relief)	**MIDAS 611**		25.33		1.14	1.88		0.09	0.73	96.46				0.62	0.08		0.72	0.44		99.33	1.40	
	Museum:	6.70	33.71		1.20	2.02		0.09	0.74	95.73				0.65	0.08		0.73	0.44		99.30	1.32	
	REP11853	20.18	61.87		1.21	1.78		0.08	0.68	92.11				0.63	0.07		0.70	0.45		99.32	1.31	
					1.05	1.92		0.09	0.72	98.36				0.66	0.07		0.79	0.45		99.30	1.55	
Burial	Laboratory:				2.73	6.51		0.12	0.85	120.84				0.38		0.35	0.30	0.33		99.44	0.33	
(Div. Pastora Tholos 2)	**MIDAS 612**		79.87		2.10	5.58		0.09	0.61	125.47				0.39			0.29	0.32		99.53	0.17	
Decorated sheet (geometric motifs in relief)	Museum:				1.94	5.25		0.09	0.57	117.91				0.39			0.27	0.31		99.53	0.16	
	REP1996/18-01				1.78	5.63		0.10	0.63	117.48				0.39			0.27	0.30		99.51	0.21	
Burial (Los Veinte Tholos)	Laboratory:	6.06	226	0.05	1.80	1.59		0.13	0.29	190	4.50		0.82	0.25	0.55	0.32	11.74	1.12	0.25	99.70	11.1	0.78
Decorated sheet (geometric motifs in relief)	**MIDAS 602**	6.18	24.0	0.05	1.04	1.73		0.14	0.47	165	4.59		0.37	0.25	0.48	0.14	9.02	0.86		99.73	8.23	0.52
	Museum:	6.42	42.2	0.11	1.46	1.72		0.14	0.51	291	8.89		0.82	0.29	0.58	1.10	9.35	1.09	0.56	99.67	9.02	0.63
	REP1996/07-2	5.20	78.4	0.04	0.64	1.24		0.13	0.31	170	2.71		0.26	0.24	0.49		11.46	1.01	0.18	99.73	10.8	0.69
		3.30	9.86		1.96	1.69		0.12	0.36	170	5.65		0.82	0.25	0.50	0.34	11.03	1.05		99.72	10.5	0.69

LAS CANTERAS (ALCALÁ DE GUADAIRA, SEVILLE)																					
Burial (Tholos)	Laboratory:	5.89			1.19	1.05		0.08	0.51	103,69			0.96	0.08		0.25	0.59		99.00	0.51	
Decorated sheet (geometric and eye-motifs in relief)	**MIDAS 614**	5.05			1.15	0.93		0.07	0.53	94.95			0.94	0.06		0.35	0.58		99.03	0.59	
	Museum:				1.17	1.02		0.08	0.51	110.94			0.95			0.27	0.55		99.01	0.33	
	REP1988/126				1.18	0.94			0.48	98.59			0.95			0.46	0.54		99.01	0.43	
			20.29		1.10	0.99		0.08	0.51	98.86			0.97	0.07		0.56	0.73		99.00	0.65	
LOS ALGARBES (TARIFA, CÁDIZ)																					
Burial (Hipogeo 5)	**MIDAS_606**	34.1	2727	0.31	4.59	1.80	208	0.26		15432	8.01	70.5	9.45	0.49	3.11	0.72	3.70	14.8	88.70	15.8	0.61
Decorated spherical sheet (geometric and eye motifs in relief)	**TUMBA 5**	27.0	2336	0.42	4.52	2.31	263	0.37		14016	6.92	70.2	9.43	0.45	3.68	0.89	4.18	14.7	88.90	15.1	0.50
	Algarbes	40.7	3380	0.71	6.85	1.90	110	0.42		17263	9.67	72.7	9.46	0.38	4.20	1.06	3.89	15.3	88.45	19.6	1.11
		25.9	2518	0.22	8.29	1.60		0.18		10972	4.77	73.4	9.13	0.31	3.07	0.66	3.77	15.2	89.51	17.4	0.39
		33.7	2491	0.36	7.81	1.93		0.54		11469	10.49	95.4	9.04	0.40	3.42	0.52	4.51	15.0	89.55	16.7	0.64

Atlantic or Eastern Mediterranean, but rather from a very close place, which was also used from 2000 to 1300 years earlier (Nocete et al., 2014).

In the gold metallurgy of the 3rd Millennium BC (Table 3), not only do we find all the variability of purities in gold and possible provenances (i.e. mine, alluvial with Sn, Ti) from the 1st Millennium BC. We also notice that all of them, including the group of very pure gold types and of alluvial origin as expressed in the Carambolo Treasure, were already present in both funerary (Table 3: Matarrubilla Tomb MIDAS 601, 603) and in gold production contexts (Table 3: Casting Spill MIDAS 505, Figs. 7 and 9) from the main site of the 3rd Millennium BC in the South of the Iberian Peninsula: Valencina de la Concepción. Importantly, also at this site we find remains from the technological process of copper alloy production and the presence of platinum, which was traditionally used to characterize and discriminate gold productions in the 1st Millennium B.C. and to emphasize their overseas origin.

The most significant difference that we find between the gold from the 1st Millennium BC and that from the 3rd Millennium BC is the evident presence of alloyed copper (Fig. 10: A), added in a metallurgical process where fusion reached relatively high temperatures, as shown by the systematic loss of the volatile Zn (boiling point 906 °C) and As (613 °C) (Figs. 10: B and 11: A, B). There are two samples that may reinforce a link between the two: the first, Cruz del Negro (Table 2: MIDAS 613), shows in the 1st Millennium BC the use of unalloyed gold that characterizes the 3rd Millennium BC (Fig. 10: A and 11: A, B, C). The second, and most important for our analysis, Algarbes (Table 3: MIDAS 606), highlights that added copper, with values similar to those for the Carambolo Trasure and the new findings at Carambolo (Table 2: 599, 609; Fig. 10: A, B and Fig. 11: A, B, C), was already present in the Lower Guadalquivir Basin since the close of the 3rd Millennium BC.

The same applies to platinum, since the samples from the 3rd Millennium BC show a range of variability comparable to the values for the 1st Millennium BC; even the highest values in the series, corresponding to the Carambolo Treasure (Table 2 MIDAS 599, 600: Pt 42.1–44.2 ppm), have been recorded in the context of Valencina de la Concepción in the 3rd Millennium BC (Murillo et al., 2015).

This information could be used to push back the possible eastern relationship-influence (technology, raw material) by 2000 to 1300 years, discarding the notion of Phoenician colonization as its origin. Such an early date for the circulation of gold and eastern-western technology might not appear far-fetched, in line with the added copper and the presence of platinum in the 3rd Millennium BC gold productions of Egypt and Mesopotamia (Meeks and Tite, 1980; Troalen et al., 2014), or with the existence of raw material movement networks, on a Mediterranean scale, in the same period, as also supported by the presence of ivory from Asian elephant in Valencina de la Concepción (Nocete et al., 2013). This, however, does not appear to be the case.

On the one hand, the widespread presence of platinum in the gold from the Egyptian-Mesopotamian period is not only apparently later (Meeks and Tite, 1980; Troalen et al., 2014), but also has a different nature. While in the Egyptian-Mesopotamian samples the platinum appears as discrete inclusions (Jensen et al., 2016) rather than in the gold alloy, the homogeneity found in the analytical values for platinum in the different spots of each of the samples of gold from Lower Guadalquivir Basin (Tables 2 and 3), points to a homogenous blend of both metals where the melting point of the gold shows no significant modifications in spite of the presence of small amounts of platinum (Okamoto and Massalski, 1985).

On the other hand, the use of platinum as a fingerprint to determine the provenance of gold (Guerra and Calligaro, 2004; Guerra et al., 2007; Guerra et al., 1999, 2005; Hartmann, 1970, 1982; Jensen et al., 2016) is not significant for the purposes of our analysis.

Table 4
Results of the lead isotope analysis in the gold samples. Analytical Techniques: LA-ICP-MS and LA-MC-ICP-MS.

Archaeological Site	Context and description	Lab No,	$^{206}Pb/^{204}Pb$	2 SD	$^{207}Pb/^{204}Pb$	2 SD	$^{208}Pb/^{204}Pb$	2 SD	$^{208}Pb/^{206}Pb$	2 SD	$^{207}Pb/^{206}Pb$	2 SD
1st Millenium BC												
El Carambolo (Camas, seville)	Treasure: Base sheet	MIDAS 599	18.319	0.001	15.645	0.001	38.429	0.001	2.0977	0.0001	0.8540	0.0001
El Carambolo (Camas, Seville)	Treasure: Semi-Sphere	MIDAS 600	18.309	0.001	15.636	0.001	38.425	0.002	2.0986	0.0001	0.8539	0.0001
El Carambolo (Camas, Seville)	New findings: Tube with striated decoration (tube-shaped sheet)	MIDAS 608*	18.328	0.133	15.606	0.115	38.377	0.279	2.0949	0.0007	0.8518	0.0002
El Carambolo (Camas, Seville)	New findings: Button/circular fitting with onfalo	MIDAS 609*	18.150	0.180	15.580	0.150	38.150	0.360	2.1011	0.0002	0.8581	0.0005
Seville	Treasure: Earring	MIDAS 605	18.449	0.001	15.668	0.001	38.623	0.002	2.0935	0.0001	0.8492	0.0001
Cortijo Ebora (Cádiz)	Treasure: Biconical bead with granulated decoration	MIDAS 604	18.620	0.001	15.684	0.001	38.876	0.002	2.0878	0.0001	0.8423	0.0001
Cruz del Negro (Carmona, Seville)	Burial (incineration pit): Decorated bead	MIDAS 613	18.320	0.001	15.663	0.001	38.500	0.004	2.1015	0.0001	0.8549	0.0001
El Acebuchal (Carmona, Seville)	Burial (Tumulo G): Hemispherical button	MIDAS 607*	18.361	0.042	15.636	0.029	38.384	0.066	2.0892	0.0010	0.8510	0.0003
3rd Millenium BC												
Cabezo Juré (Alosno, Huelva)	Fortified area: Undecorated sheet	MIDAS 378	18.186	0.002	15.623	0.002	38.186	0.004	2.0997	0.0001	0.8591	0.0001
Cabezo Juré (Alosno, Huelva)	Fortified area: Undecorated sheet	MIDAS 435	18.189	0.003	15.628	0.003	38.194	0.007	2.0991	0.0001	0.8592	0.0001
Cabezo Juré (Alosno, Huelva)	Fortified area: Undecorated sheet	MIDAS 434	18.357	0.003	15.653	0.002	38.561	0.006	2.1001	0.0001	0.8526	0.0001
Cabezo Juré (Alosno, Huelva)	Fortified area: Undecorated sheet	MIDAS 506	18.550	0.005	15.681	0.005	38.703	0.014	2.0863	0.0001	0.8453	0.0001
Valencina de la Concepción (Seville)	Residential area: Undecorated sheet	MIDAS 487	18.354	0.016	15.660	0.013	38.506	0.034	2.0979	0.0003	0.8532	0.0001
Valencina de la Concepción (Seville)	Production area: Casting spill	MIDAS 505	18.453	0.019	15.663	0.017	38.585	0.042	2.0909	0.0004	0.8481	0.0001
Valencina de la Concepción (Seville)	Burial (Matarrubilla Tholos): Undecorated sheet	MIDAS 601	18.320	0.001	15.637	0.001	38.315	0.003	2.0913	0.0001	0.8535	0.0001
Valencina de la Concepción (Seville)	Burial (Matarrubillla Tholos): Undecorated sheet	MIDAS 603	18.328	0.001	15.638	0.001	38.433	0.003	2.0969	0.0001	0.8532	0.0001
Valencina de la Concepción (Seville)	Burial (Matarrubillla Tholos): Decorated sheet (geometric motifs in relief)	MIDAS 611	18.199	0.005	15.619	0.005	38.112	0.013	2.094	0.0002	0.8582	0.0001
Valencina de la Concepción (Seville)	Burial (Divina Pastora Tholos 2): Decorated sheet (geometric motifs in relief)	MIDAS 612	18.439	0.028	15.626	0.024	38.478	0.063	2.086	0.0006	0.8474	0.0002
Valencina de la Concepción (Seville)	Burial (Los Veinte Tholos): Decorated sheet (geometric motifs in relief)	MIDAS 602	17.991	0.001	15.605	0.001	38.178	0.002	2.1220	0.0001	0.8673	0.0001
Las Canteras (Alcalá de Guadaira, Seville)	Burial (Tholos): Decorated sheet (geometric and eye-motifs in relief)	MIDAS 614	17.988	0.017	15.613	0.015	37.996	0.038	2.112	0.0005	0.8680	0.0001
Los Algarbes (Tarifa, Cádiz)	Burial (Hipogeo 5): Decorated spherical sheet (geometric motifs in relief)	MIDAS 606	18.439	0.001	15.647	0.001	38.562	0.004	2.0912	0.0001	0.8485	0.0001
La Sultana (Cala, Huelva)	Mine: Raw material	MIDAS 436	18.519	0.001	15.650	0.001	38.501	0.003	2.0789	0.0001	0.8451	0.0001

Apart from the difficulty involved in discriminating between precise provenances of the platinum, even in one of its main source areas (Afghanistan, Iran, Oman, Egypt) (Jensen et al., 2016), we must also add three further factors: i) Its presence in the sources of gold supply from Eastern (Hauptmann and Klein, 2009) and Atlantic Europe (from the Tagus estuary in Portugal to the British Isles) (Ehser et al., 2011b; Standish et al., 2014) does challenge any attempts at provenancing based on this element only; ii) The scarce information available on Iberian placer deposits of gold does not make it possible to rule out closer regional sources of supply; and iii) The possibility that the presence of platinum may derive from the metallurgical process itself (Brostoff et al., 2009; Guerra and Calligaro, 2003), namely, its association with the copper incorporated in the alloys. This latter fact would not be improbable when taking into account the correlation between copper and platinum in the samples analysed and the presence of platinum in numerous copper mineralizations in geographic areas close to the Lower Guadalquivir Basin, such as Aguablanca (Ortega et al., 2004) to the northwest, Serranía de Ronda (Rábano, 2008) to the southeast or Beni-Boussera (Morocco) (Gervilla and Leblanc, 1990) to the south.

All this means that we should take the conclusions on supply/provenance, solely based on trace elements (especially on platinum) with a good degree of caution (for further details on platinum group element inclusions, see Craddock, 2000), leading us to verify this using another methodology with a proven efficiency in

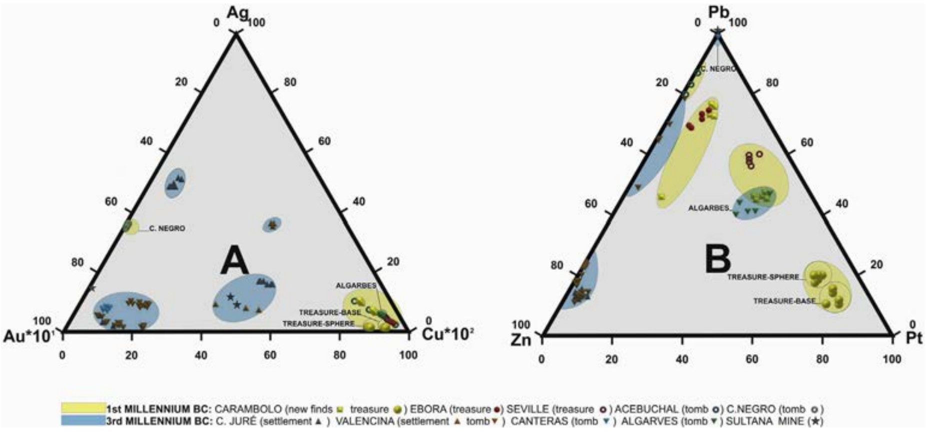

Fig. 10. Ternary diagrams showing the composition of the samples analysed. A: Au/Ag/Cu system; B: Pt/Pb/Zn system.

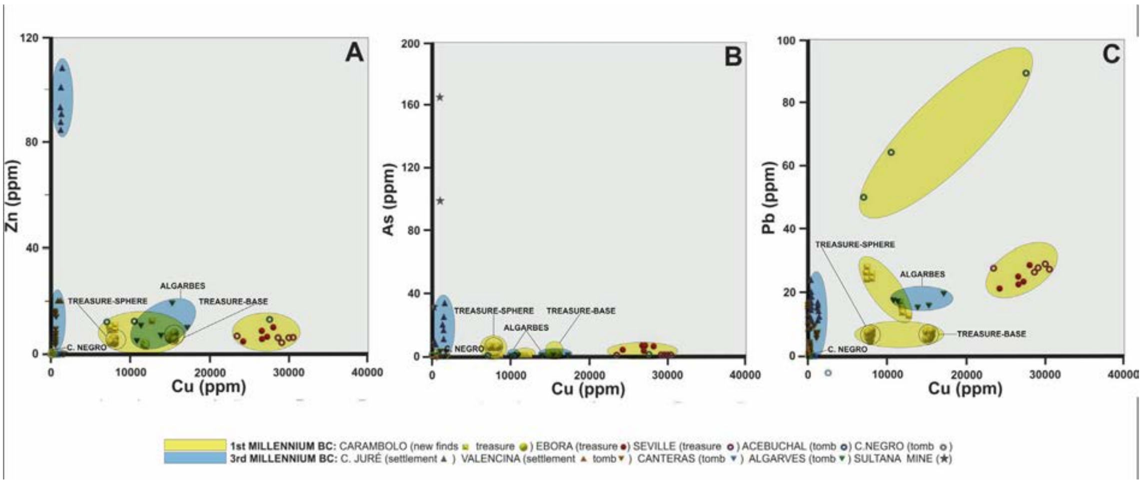

Fig. 11. Binary diagrams showing the composition of the samples analysed. A: Cu/Zn system; B: Cu/As system; C: Cu/Pb system.

this regard, such as lead isotopes. This is particularly advisable for the analysis of the Carambolo Treasure, as the addition of copper, which could have effects on the traceability of provenance of gold, does not appear to affect it in this case, since copper and lead show a negative correlation (Fig. 11:C).

3.2. Lead isotope analyses by MC-ICP-MS

The results of isotopic analysis (Table 4, Fig. 12) once again confirm those found in the elemental analysis: the base sheet and sphere from the Carambolo Treasure were manufactured with gold from the same source which, in common with the other samples analysed from the 1st and 3rd Millennia BC, included that of the earliest copper-rich sample in our study, that of the Algarbes, pointing to a regional origin.

The isotopic values of all the series analysed are markedly different from those supplied from the Atlantic (Ehser et al., 2011a, b; Standish et al., 2014) and Eastern Europe (Baron et al., 2011; Radivojević et al., 2010) and establish a clear link with the mining districts to the North and West of the Guadalquivir Basin. In the case of the Carambolo Treasure, the data suggest a direct relationship, both with the Ossa-Morena region to the north of the Lower Guadalquivir Basin and with the gold metallurgy supply systems from the 3rd Millennium B.C. in the settlement located next to it: Valencina de la Concepción.

The total agreement in the isotopic values of the Carambolo Treasure with those of a gold sample belonging to the settlement of Valencina de la Concepción and another in one of the main monumental tombs, Matarrubilla (Table 4: MIDAS 487 and 603, Fig. 12), suggests a historical continuity in the exploitation of the same source of supply and/or a supply resulting from possible plundering and subsequent recycling of gold types from the large tombs of Valencina de la Concepción in the 3rd Millennium BC during the Carambolo period. This last suggestion should be confirmed or discarded after future studies.

4. Conclusion

The results of the elemental analysis, by LA-ICP-MS of the base sheet and a semi-sphere of one of the plates from the Carambolo Treasure, show a homogeneous manufacture on a very pure gold (Au fineness 961–979 $^{\circ}/_{00}$) with low silver content (Ag: 1.29–2.39%). The proportion of copper (Cu: 0.75–1.58%) suggests a metallurgical alloying process and the presence of tin (Sn: 34–173 ppm) and platinum (Pt: 23–43 ppm), an origin in placer deposit in fluvial transport.

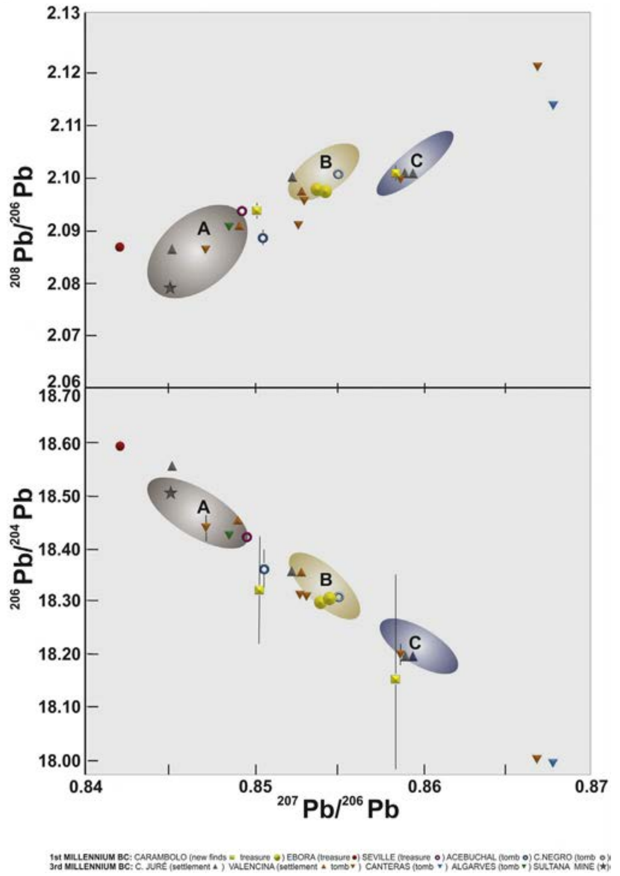

Fig. 12. Mirror plots of lead isotope ratios measured from the gold finds compared with the copper ores range from the mineralizations located in the group G3 from the Guadalquivir Depression and the Southwest (from Nocete et al., 2014): A) Los Pedroches Batolith, Cardeña (Ossa-Morena Zone); B) Variscan Hidrotermal Veins (South-Portuguese Zone) and Los Pedroches Batolith, Variscan granite-related Veins, Cerro Muriano (Ossa-Morena Zone); C) Massive Sulphides and Gossan (Iberian Pyrite Belt). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The results of isotopic analysis (Pb by MC-ICP-MS) once again confirm that the base sheet and sphere from the Carambolo Treasure were manufactured on gold from the same source of supply, which points to a regional origin. Their isotopic values suggest a direct relationship with the Ossa-Morena region to the north of the Lower Guadalquivir Basin.

Comparison of these results with those from a series of gold work samples dating from the 3rd Millennium BC, from the same geographical area (Lower Guadalquivir Basin), also suggest the origin of the gold need not be from thousands of kilometres away, in the Atlantic or Eastern Mediterranean, but rather from a very close source, which would have been used from 2000 to 1300 years earlier.

The results thus suggest a direct link with the gold metallurgy (technology, raw material and supply systems) from the 3rd Millennium BC civilization in the Lower Guadalquivir Basin and with a settlement located around 2000 m from the Carambolo, which acted as its main economic and political hub and a gateway for raw materials of regional and transcontinental origin and space for their artisanal transformation into products, including gold metallurgy: Valencina de la Concepción.

In the gold metallurgy of the 3rd Millennium BC not only we have indicators of technological process of copper addition, as documented later in the Carambolo Treasure documented (i.e. Algarbes), but also for the use of the same source of raw material (i.e. Valencina de la Concepción).

All in all, as regards the Carambolo Treasure, we would find ourselves not at the beginning, but rather at the end of a gold processing tradition that began in the Lower Guadalquivir Basin during the 3rd Millennium BC and to which ornamental techniques such as filigree or soldering were added at the turn of the 1st Millennium BC.

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