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RESEARCH

Beautiful, Magic, Lethal: a Social Perspective of Cinnabar Use and Mercury Exposure at the Valencina Copper Age Mega-site (Spain)

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Abstract

Today, mercury is a matter of concern for health and environmental authorities across western countries, and legislation has been passed and programs have been implemented for its total elimination from human activity. But this was not always the case: mercury and its compounds have been highly appreciated and used since remote times all over the world with very diverse purposes ranging from decorative, medicinal, metallurgical and symbolic. In particular, cinnabar (HgS, mercury sulfide), a mineral of an intense red color, has been considered in many cultures as an exotic raw material, highly valued and associated with the elites and sacred practice. In this paper, we examine one such case, set almost 5000 years ago, in Copper Age Iberia, by investigating mercury exposure through human bone. The study presented here includes a total of 170 samples from 70 different human individuals and 22 animals (plus one soil sample) from the Copper Age mega-site of Valencina, south-western Spain. It is the largest ever single-site study of exposure to mercury based on human bone in combination with cinnabar use. Abnormally high values are recorded in some individuals dating between 2900 and 2650 BC, especially in those buried in remarkable tombs belonging to the social elite of this period, but high levels of mercury are also recorded in the rest of the population. Three lines of interpretation are used to explain these results, including the manipulation of cinnabar (grinding it into powder, mixing it with other substances, using it for the decoration of objects, buildings and the human body), its direct consumption through ingestion or inhalation by a 'special' social group and the contribution of environmental factors. Based on the currently available evidence, which is carefully reviewed, Valencina represents the most intense and prolonged case of exposure to mercury recorded in human history, which makes it an important site to assess the long and complex history of use of this substance.

Keywords Mercury · Cinnabar · Exposure · Copper Age · Iberia · Burial practices

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Introduction

Mercury and cinnabar (HgS, mercury sulfide) have a long and complex history of use by human societies across the world. Both substances are amply documented in the Americas, East Asia and Western Europe in societies occurring in the last 10,000 years. Their uses include a broad range of purposes, such as ceremonial and sumptuary display, pigment for painting, medicinal and magic drug, preservative and biocide and the extraction of precious metals such as gold and silver.

For millennia, cinnabar, often mixed with other mineral substances such as iron oxides and organic binders (for example, plant oils and egg yolk), has been used as a pigment to paint rocky surfaces, wooden and stone buildings, artefacts, textiles or even the human body itself (Siddal, 2018; Gliozzo, 2021). Cinnabar pigment was used to paint the clay-modelled human skulls dating to the 9th millennium BC found in the Pre-Pottery Neolithic site of Kfar HaHoresh (Israel), where no cinnabar is locally available (Goren et al., 2001). In the Neolithic town of Çatalhöyük (Turkey), dating between the 8th and 6th millennia BC, human bones buried underneath the houses were painted with pigment made with red ochre and cinnabar, as were the walls of the houses themselves (Schotsmans et al., 2022). In Europe, the earliest documented use of cinnabar goes back to the Vinča culture of the middle Danube basin, named after the eponymous site in Serbia and dating to the 6th millennium BC, in which it was used to decorate objects and, probably, other purposes (Gajic-Kvascev et al., 2012; Mioč et al., 2004).

In many cultures, cinnabar and mercury have also been used to provide health, prolong life or lead to immortality (Mahdihassan, 1987; Needham, 1976). Their use as medicinal or magic drugs is well attested. The use of mercury and cinnabar as a medicinal drug, as an elixir of life and to preserve cadavers, is well documented in China and India where these practices have continued until very recently (Liu et al., 2008; Needham, 1976). The alchemists of the court of Qin-Shi-Huang, first emperor of China (third century BC), sought the secrets of immortality in liquid mercury (Needham, 1976: 4). The trading of cinnabar along the 'Silk Route' is attested 2000 years ago (Sevillano-López & González, 2011). Before the twentieth century, Western medicine made use of oral doses of mercury salts, alone or mixed with other ointments, as a sedative and to treat lice, fever, insomnia, brain traumas, ulcers in the mouth and tongue or syphilis (Biehler-Gomez et al., 2022; Fornaciari et al., 2011, 2022; Lombardo et al., 2022; Panova et al., 2018; Rasmussen et al., 2008).

In the American continent, the use and exchange of cinnabar from the Huancavelica mines (Peru) are documented since the 3rd millennium BC (Prieto et al., 2016). In Mesoamerica, cinnabar was found in the graves of high-ranking individuals at Teotihuacan (México) (Sancho Mengod, 2013; Sugiyama & López Luján, 2006) and numerous Maya cities (Vázquez de Agredos, 2007; Batta et al., 2013; Ávila et al., 2014, Ochoa-Lugo et al., 2017; Cervini-Silva et al., 2018). Prominent among those is the tomb of the 'Red Queen' at Palenque (Cervini-Silva et al., 2013; González-Cruz, 2011).

The Iberian Peninsula has an old and extensive record of use of cinnabar and mercury, which is reflected in a substantial literature—for recent reviews see

Zarzalejos Prieto, 2019; Zarzalejos Prieto et al., 2020a. Without question, the prominence of both minerals in Iberian Prehistory and History is explained by the presence in Almadén (located in Ciudad Real central Spain) of the largest mercury mine in the world. This site was included in the UNESCO World Heritage List in 2012. The oldest evidence of cinnabar use in Iberia known to this date is represented by the pigments recorded inside a clam shell (*Glycymeris* sp.) found at Cova de l´Or (Valencia), a cave located in the Spanish Levant and occupied in the Early Neolithic (second half of the 6th millennium BC) (Domingo et al., 2012). At Los Murciélagos cave (Zuheros, Córdoba), dating to the same period, cinnabar was used as part of the infill applied to the grooves of ceramic vessels and marble bracelets and also to coat the inner side of a pot (Martínez et al., 1999; Gavilán Ceballos et al., 2020). Cinnabar-dyed textiles of similar date have been recently published for a small cave located at Peñacalera, also in Córdoba (Gleba et al., 2021). At the flint mine of Casa Montero (Madrid), also dating to the second half of the 6th millennium BC, a knapped flint blade was found with traces of cinnabar, which were sourced to the Almadén mines (Hunt-Ortiz et al., 2011).

Cinnabar has been found in numerous burial contexts dating to the 4th and 3rd millennia BC (Late Neolithic and Copper Age) across Iberia, either sprayed over the floors, bodies and grave goods of tombs or used as a pigment to paint the uprights and capstones of megaliths. Such is the case of a number of burial monuments (megaliths and hypogea) including Alberite (Cádiz) (Domínguez Bella & Morata Céspedes, 1995; Domínguez Bella, 2010), La Velilla (Palencia) (Zapatero Magdaleno, 1990; Martín Gil et al., 1995), Valle de las Higueras (Toledo; Bueno Ramírez et al., 2019), Getsemaní-Cerro del Ojo (Sevilla) (Bascón Mateos et al., 2016), Peñacalera (Córdoba) (Gleba et al., 2021), the Montelirio tholos and other burials located at the Valencina Copper Age mega (Sevilla) (Rogerio-Candelera et al., 2013; Bueno Ramírez et al., 2016; Emslie et al., 2022; etc.) as well as several others in southern Portugal (Inácio et al., 2013; Rocha et al., 2018) and central Spain (Garrido-Pena et al., 2019).

Clear vestiges of the use of cinnabar have also been found in Bell-Beaker (Liesau et al., 2020) and Bronze Age burials (López Padilla et al., 2012) dating to the late 3rd and early 2nd millennium BC. In the Iron Age, the use of cinnabar pigment is well documented in high-end sculptures (Chapa Brunet et al., 2021) and pottery (Barrio Martín, 2020). Cinnabar and mercury were highly-valued commodities in Roman-time Hispania, when the Almadén mines were exploited extensively, as reported by Pliny in his 'Universal History' (Gil Bautista, 2015: 59–60). Cinnabar was used to produce the 'Pompeian Red' pigment applied to paint the walls of aristocratic houses and works of art across the Roman empire (Guiral Pelegrín & Íñiguez Berrozpe, 2020), while mercury was used to help extract gold at the mines of Las Médulas (León) (Zarzalejos Prieto, 2019; Zarzalejos Prieto et al., 2020b; Rodríguez García et al., 2021). Mercury was also well-known in Medieval Spain. The Almadén mines were mentioned by geographer Al Idrisi, in the twelfth century AD (Gil Bautista, 2015: 60), while in the Medina Azahara palatial complex located outside Córdoba, capital of the Spanish Caliphate between the tenth and eleventh centuries AD, there was a famous mercury 'basin' or 'fountain' that impressed visitors (Molina, 2004). The exploitation of the Almadén mines peaked between the sixteenth and nineteenth centuries AD, when mercury was extensively used for gold mining in colonial Mexico and Peru, which involved a vast system for its transportation by land and sea set up by the Spanish Crown (González Tascón et al., 1996; VVAA, 1996; Gil Bautista, 2009, 2013, 2015).

Because of its pervasiveness, and because of its long and complex history of exploitation, use, consumption and exposure, mercury is currently a topic of great scientific interest. This is largely explained by the implications of its presence in the environment and its negative impact for human health. Mercury occurs naturally in the environment. Half the emissions in the atmosphere come from primary sources (such as volcanic eruptions). In nature, it is found as metallic or elementary mercury (Hg°), organic mercury and inorganic mercury (Hg^{2+}, Hg_2^{2+}), while cinnabar is one of its natural inorganic forms (mercury sulfide, HgS). However, human exploitation, particularly in the last 2000 years, but also quite earlier, has significantly increased its environmental occurrence and its impact.

Until recently, mercury and cinnabar have been widely used for a multiplicity of industrial purposes and are therefore present in the daily lives of millions of people (Park & Zhen, 2012). Contemporary small-scale mining accounts for 37% of mercury emissions, while carbon combustion accounts for 24% (WHO, 2017a; Bell et al., 2017; European Commission, 2017: 13). However, a great deal of the mercury currently present in the environment was released in pre-industrial times. A linear increase has been postulated starting c. 2000 BC (or earlier) until c. 1570 AD, followed by 300 years of rapid increase caused by the exploitation of the Almadén mines for the smelting ('amalgamate') of silver and gold in the Spanish Empire and a constant level of emissions after c. 1850 AD (Amos et al., 2013: 413). Pre-industrial emissions would account for as much as 39% of the mercury present in today's environment, the remaining 61% having been caused by emissions post c. 1850 AD (Streets et al., 2011).

Although the toxicological effects of mercury exposure in the human body are not known in detail, scientific research in the last century has led to a greatly improved knowledge and awareness of its dangers for human health. In turn, this has favoured the generalization of measures to reduce exposure (Bellanger et al., 2013; Risher, 2003). The current contemporary positioning of western science and public health policies against mercury is certainly well-grounded in solid science but also runs in stark contrast with its long history of use. Western medicine made extensive use of mercury as a medicinal drug until barely a century ago, as noted above. Indeed, in some regions of the world, cinnabar is still consumed today (ingested or inhaled), either for magic and religious reasons, as happens in the Caribbean (Wendroff, 2005), or as a medicinal drug, as in South Africa (Street et al., 2015) and Tibet (Gerke, 2021).

Regardless of its past history and specific uses in the contemporary world, today, mercury is widely considered as a toxic metal, non-essential for the human body. It is currently listed by the World Health Organization as one of the ten most threatening chemical substances for public health worldwide (WHO, 2017b). The manifestation of its effects is closely related to the means of exposure. In turn, this is largely conditioned by the specific form of the mercury causing the exposure. Well-known effects of mercury on human health include cardiovascular toxicity, reproductive and developmental toxicity, neurotoxicity, nephrotoxicity, immunotoxicity and carcinogenicity.

Within the context of the long-standing and on-going interest in the scientific study of mercury and cinnabar use and exposure in human society, recent research based on the geochemical analysis of human bone has highlighted the social and cultural pervasiveness of both substances in Iberian Late Prehistory—see Emslie et al. (2015, 2016, 2019, 2022). As noted above, there is ample evidence of cinnabar use in Iberia during the 4th and 3rd millennia BC. One recent paper in particular has provided an overview on mercury exposure, based on the largest sampling of human bone ever undertaken, including 370 samples from 50 different burial structures (both individual and collective) of 23 different sites dating to the Late Neolithic (c. 4200–3200 BC), Copper Age (c. 3200–2300 BC) and Bronze Age (c. 2300–850 BC) (Emslie et al., 2022). Although this study showed a high variability of results (as was expected), unusually high levels of mercury were noted for some of the sampled human bones, particularly those dating to the Copper Age. While a major study centred on the site of La Lanzada (Pontevedra, Spain), a necropolis dating to the first to seventh centuries AD, which included 143 samples of 76 individuals (Álvarez-Fernández et al., 2020; López-Costas et al., 2020) analysed atmospheric mercury as the source for Hg in the bone, the approach followed by us looks at both environmental mercury and cinnabar as sources of exposure. In general, most previous studies of mercury in human bone recovered from archaeological contexts were based on a much smaller number of samples—see for example Cockburn et al., 1975: 1159; Yamada et al., 1995: 254–255; Torino et al., 2015; Bocca et al., 2018; Rasmussen et al., 2008, 2012, 2013a, b, 2015, 2019; Walser et al., 2019. This topic is further discussed below.

The paper presented here is intended to further expand this line of research and, by focusing on a single site, namely Valencina de la Concepción-Castilleja de Guzmán (henceforth Valencina) located in Sevilla, south-west Spain, extend our knowledge of the use and exposure to cinnabar and mercury in Copper Age Iberia (Fig. 1). For reasons that are briefly outlined below, Valencina currently stands as one of the most intensely researched and debated sites of 3rd millennium Europe. The study presented here includes a total of 170 samples from 70 different human individuals and 22 animals (plus one soil sample), which, together with above-mentioned La Lanzada research, is the largest ever single-site study of exposure to mercury based on human bone, although centred on much older material. Some of the results included in this study (64 samples) were discussed in previous papers (Emslie et al., 2016, 2022), but the majority (106 samples, 62% of the total) are presented here for the first time. The contribution of this study lies not only in the high number of unpublished analytical results, which are combined with already-published ones from the same site, but also in that the sampled individuals come from well-contextualised and well-dated burial structures occurring within a short time span of no more than 250 years (c. 2900–2650 BC). This allows, for the first time in prehistoric studies, to assess cinnabar use and mercury exposure within the frame of a short number of human generations, leading to more precise conclusions in terms of the social and cultural background for which the substances were used.

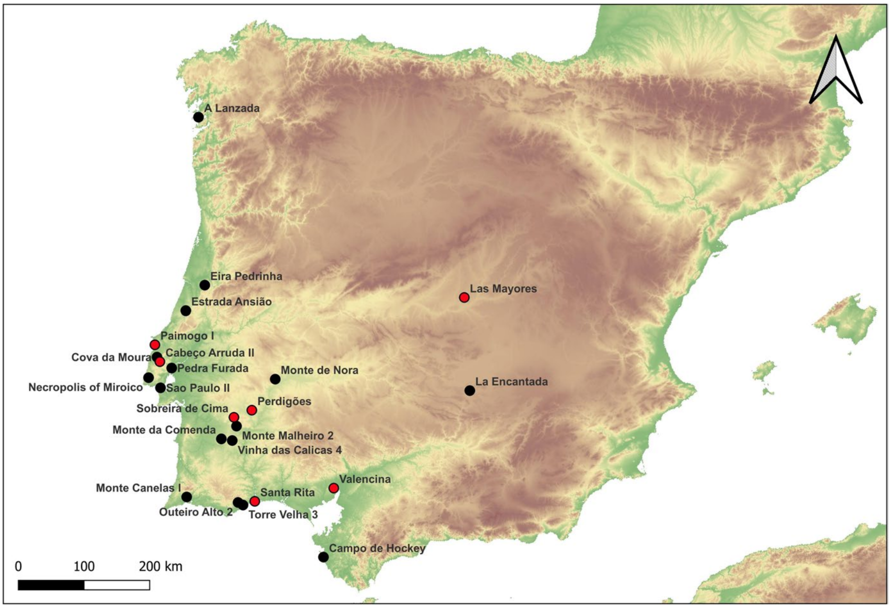


Fig. 1 Main Iberian sites with THg measurement in human bone mentioned in the text (in red, sites with evidence of cinnabar). Design: Manuel Eleazar Costa Caramé and Raquel Montero Artús

Thus, the main aim of this paper is to present and assess all the results available for total mercury (THg) in human bone at Copper Age Valencina and to discuss possible hypotheses regarding the sources of exposure. We depart from the assumption that unexpectedly high levels of mercury in human and animal bone can stem from (i) environmental pollution, (ii) the pre-mortem use and manipulation of mercury and associated compounds (cinnabar), and (iii) post-mortem diagenetic processes. That at Valencina cinnabar was used in some high-ranking burials, coupled with evidence that it was thoroughly ground and sieved to produce a fine powder and then mixed with other red ochres to extend its availability (Rogerio-Candelera et al., 2013), very much suggests that it was valued as a high-end commodity. In turn, that some individuals reveal very high levels of mercury in their bones raises several questions regarding its use and its social and ideological significance.

Contexts and Sampling

After the first discoveries in the 1860s and then gradual, if slow, advances throughout the twentieth century, research on the Valencina Copper Age mega-site has accelerated in the last two decades. The site is remarkable because of its large size (c. 450 hectares), monumentality, with massive ditches and some of Iberia's largest Chalcolithic megaliths (including remarkable *tholoi* such as Montelirio, La Pastora and Matarrubilla), number and density of features (estimated in the tens of thousands, many of them funerary), as well as the amount and quality of the material culture found in them, which boasts some of the finest and most accomplished examples of 3rd millennium BC craftmanship across western Europe. Studies published in recent years include general overviews of the site (García Sanjuán et al., 2017), high-resolution approaches to some of its most important megalithic monuments and burials (Cáceres-Puro et al., 2014, 2019; Fernández Flores et al., 2016; García Sanjuán et al., 2019; Vargas Jiménez, 2021; etc.), extensive radiocarbon dating (García Sanjuán et al., 2018a), as well as detailed analyses of metallurgical (Murillo-Barroso et al., 2015; Nocete Calvo et al., 2008, 2014), lithic (Martínez-Sevilla et al., 2020; Nocete Calvo et al., 2005) and ivory technology (García Sanjuán et al., 2013; Nocete Calvo et al., 2013; Luciañez Triviño et al., 2022). On account of these advances, Valencina has featured prominently in recent reviews of Neolithic and Copper Age Europe—see for example Guilaine, 2018; Whittle, 2018: 11–12; Gaydarska & Chapman, 2022: 51–62.

Previous studies have suggested the extent and importance of cinnabar use (Hunt Ortiz & Hurtado Pérez, 2010; Hunt-Ortiz et al., 2011; Rogerio-Candelera et al., 2013; Bueno Ramírez et al., 2016) and mercury exposure (Emslie et al., 2022) at Valencina, a body of evidence consistent with the role the site probably played as a central place of high social and religious significance across a large territory. This paper is intended to expand on that previous work. It is based, as noted above, on a total of 170 samples obtained over the last 5 years. Figure 2 shows the location of the various Valencina and PP4-Montelirio sectors and features sampled in this study. In total, 53 samples from 23 different individuals come from the Montelirio Copper Age tholos (22 individuals of Chalcolithic age and one individual, buried outside the tholos, of Early Neolithic chronology) (Table 1 and Supplementary Data SD 1). Another 94 samples come from the adjacent PP4-Montelirio sector, including 89 samples from 42 Copper Age individuals found across 22 different burial structures (Table 2 and Supplementary Data SD 2), as well as five samples from as many Roman-time individuals found in as many graves (Hg results published in Emslie et al., 2022). Regarding to the sampling of animals, 22 samples were taken, six from PP4-Montelirio sector and 16 from three features located in others sectors of the Valencina mega-site (Pabellón Cubierto, Calle Mariana de Pineda and Calle Huelva) (Table 3). Finally, also one soil sample was taken. This represents the largest single-site sampling program of mercury in human bone ever undertaken for a prehistoric site and is aimed at exploring the social use and cultural significance of cinnabar at the Valencina mega-site and in Copper Age Iberia at large.

Among the numerous structures sampled for this study (see full list in Table 1 and Table 2), two stand out on account of their architectural sophistication as well as the wealth of the grave goods laid in them: The Montelirio tholos (Fernández Flores et al., 2016) and Structure 10.042–10.049 (García Sanjuán et al., 2019), both located on the south-eastern quadrant of the site (Fig. 2). Structure 10.042–10.049 and the Montelirio tholos stand next to each other and are part of the same temporal and cultural dynamics of burial use of the south-eastern sector of the site between c. 2900 and 2650 BC. However, they were excavated by different teams at different times, and for that reason, their study has progressed in part separately.

According to the available radiocarbon chronology, Structure 10.042–10.049 (a semi-destroyed double tomb) was likely built between 2900 and 2800 BC (García

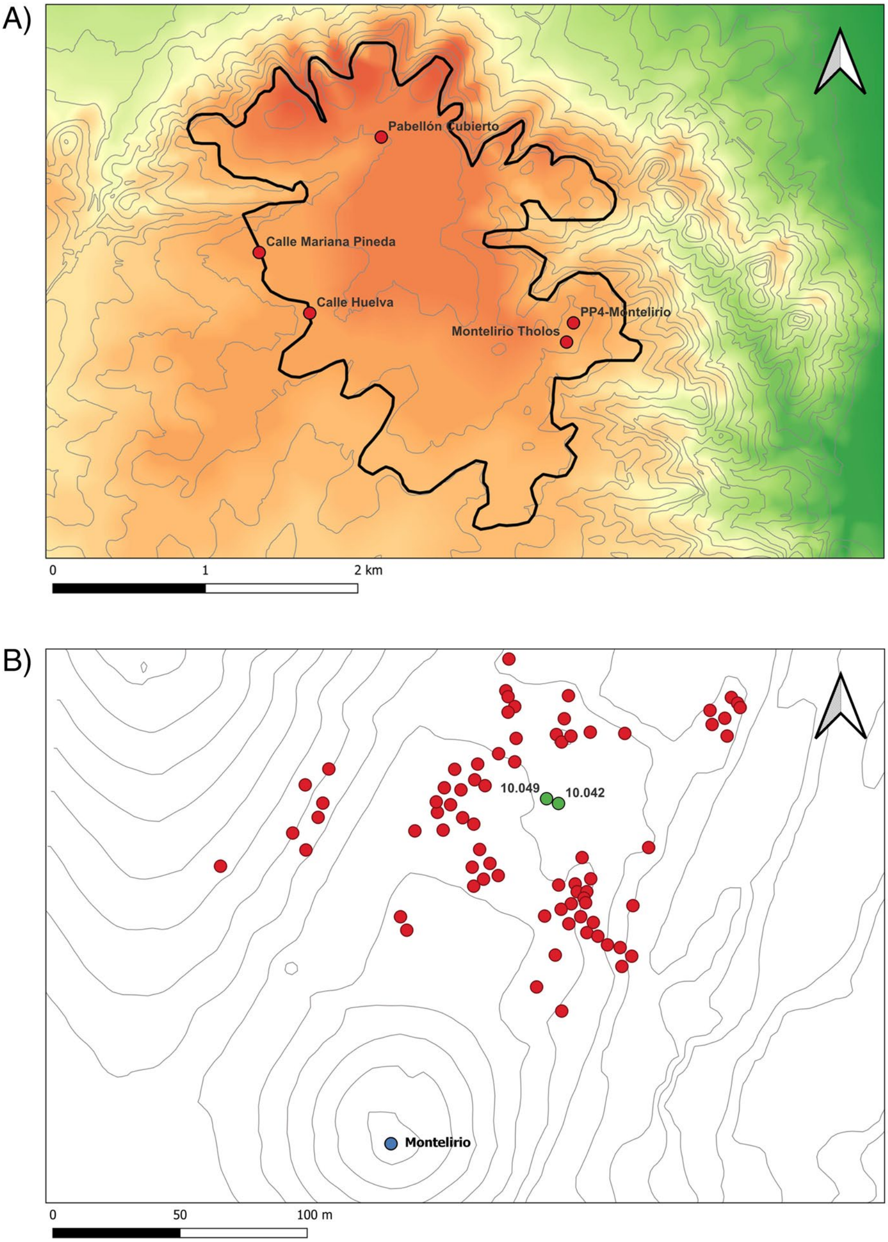


Fig. 2 **A** Location of Valencina sectors with THg measurement. **B** Location of the Montelirio tholos and 10.042–10.049 structure within the PP4-Montelirio sector. Design: Manuel Eleazar Costa Caramé and Raquel Montero Artús

Table 1 THg (μg/g) content (max) in bones for 23 individuals buried in the Montelirio tholos (the SC contained a MNI = 2, but three samples were analysed)

Structure	Individual	THg (μg/g)	sd (μg/g)	Bone	Age	Sex	Chronology	Pathology	MSSM[a]	Garment	Other	Source
Large Chamber	UE 115	483.97	102.94	F	Adult	¿?	-	NO	-	NO	Cut by UI64	This paper
	UE 112	273.52	38.32	H	30–35	F	-	NO	HRG	NO	Cut by UI64, PA diet	Emslie et al., 2016
	UE 346	255.88	43.17	H	25–29	F	-	Osteoarthritis	HRG	NO	Cut by UI64	Emslie et al., 2016
	UE 104	207.15	31.72	F	25–34	F	-	Osteoarthritis	-	NO	-	This paper
	UE 103	156.01	5.73	F	25–29	F	4220 ± 17	Osteoarthritis	HRG, FAT	Skirt / Belt	-	This paper
	UE 113	132.21	38.29	F	30–35	F	4116 ± 24	Osteoarthritis	-	NO	Lower level	Emslie et al., 2016
	UE 107	123.02	34.84	H	35–39	F	-	Osteoarthritis	-	NO	PA diet	Emslie et al., 2016
	UE 108	116.14	12.31	H	30–45	F	-	Osteoarthritis	-	NO	-	This paper
	UE 114	110.30	10.58	F	18–23	F	-	NO	-	Skirt / Belt	Cut by UI64	This paper
	UE 101	92.26	2.19	H	25–29	F	-	Periostitis	HRG, FAT	Skirt / Belt	Polydactyly	This paper
	UE 109	84.14	15.26	H	18–25	F	-	Periostitis	IOF	NO	-	Emslie et al., 2016
	UE 105	55.23	2.09	H	18–25	F	4164 ± 36	NO	-	NO	-	This paper
	UE 102	52.83	6.97	T	25–34	F	4195 ± 22	NO	HRG	Full costume	To the east of stela	This paper
	UE 111	50.21	6.48	H	20–29	F	-	NO	-	Beads under shoulder ¿rug?	By stela	Emslie et al., 2016
	UE 396	30.55	3.84	FIB	Adult	¿?	-	NO	-	NO	Cut by UI340	This paper
	UE 110	10.67	0.93	F	22–24	F	4129 ± 33	NO	-	NO	Cut by UI340	Emslie et al., 2016
	UE 343	8.77	0.36	H	24–32	F	4168 ± 30	NO	-	Full costume	Cut by UI340	This paper

Table 1 (continued)

Structure	Individual	THg (µg/g)	sd (µg/g)	Bone	Age	Sex	Chronology	Pathology	MSSM[a]	Garment	Other	Source
Small Chamber	UE 80	38.10	2.39	UND	Adult	¿?	4180 ± 30	-	-	-	-	This paper
	UE 88	28.75	1.86	UND	Adult	¿?	4002 ± 31	-	-	-	-	This paper
	UE 73	19.61	1.50	UND	Adult	¿?	-	-	-	-	-	This paper
Corridor	UE 251	4.89	0.32	T	Adult	F?	-	-	-	-	-	Emslie et al., 2016
	UE 229	3.82	0.22	H	30–40	M	4125 ± 30	-	-	-	-	Emslie et al., 2016
	UE 232	2.00	0.44	T	25–35	¿?	4151 ± 30	-	-	-	-	Emslie et al., 2016
Exterior	UE 273	4.32	0.82	T	35–55	M	5802 ± 34	-	-	-	-	Emslie et al., 2016

[a]MSSM, musculo-skeletal stress markers; HRG, hyper-development of the first gluteal tuberosity of the femur; FAT, squatting facets in the tibiae; IOF, tendon insertions in hands

Table 2 THg (µg/g) content (max) in bones for the 42 Copper Age individuals buried in the PP4-Montelirio Sector

Structure	Individual	THg (µg/g)	sd (µg/g)	Sex	Bone	Source	Description
10.074	UE 923	480.50	19.58	U	R	This paper / Emslie et al. 2022 (415.88)	Negative structure, irregular plan (1.60 × 1.40 m), NMI: 1, 1 marine malacofauna shell (*Pecten maximus*), faint stains of cinnabar
10.049	UE 667	185.54	10.65	F	T	This paper / Emslie et al. 2022 (44.74)	Ivory Lady, burial with cinnabar
10.035	UE 503	52.29	4.68	U	H	Emslie et al. 2022	Negative structure, irregular plan (2.15 × 1.68 m), NMI: 2, no grave goods, no cinnabar
10.042	UE648	28.10	6.23		F	Emslie et al. 2022	Passage grave separate from Ivory Lady, burial with cinnabar
	UE 211	19.70	1.62	M	F	Emslie et al. 2022	
10.073	UE 866–1	28.08	2.50	F?	H	This paper	Negative structure with stone elements, quadrangular plan (1.19 × 1.13 m), NMI: 3, 1 marine malacofauna (*Cymbula nigra*), copper awl with bone sleeve, no cinnabar, IND-1: stress marks in the form of enthesopathies on the left radio
	UE 866–2	14.01	1.27	F?	H	Emslie et al. 2022	
10.044	UE 632–2	17.13	2.67	M	H	This paper	Negative feature with stone elements, rectangular plan corridor (L 2.21 × W 1.36–0.55 m) and circular chamber (1.82 × 2.07 m), NMI: 3, ceramic (1 vessels and fragments), small rough stone, no cinnabar
	UE 632–1	2.48	0.05	F	H	Emslie et al. 2022	
	UE 632–3	0.38	0.01	U	UNK	This paper	

Table 2 (continued)

Structure	Individual	THg (µg/g)	sd (µg/g)	Sex	Bone	Source	Description
10.028	UE 189	16.31	0.96		H	Emslie et al. 2022	Negative structure, circular plan (diameter 1.70 m), NMI: 9; 1 ceramic vessel, no cinnabar; (189.2 y 189.3 females, peptides)
	UE 189	7.69	0.17		H	Emslie et al. 2022	
	UE 189	4.62	0.15		H	Emslie et al. 2022	
	UE 189	1.54	0.08		H	Emslie et al. 2022	
	UE 189	1.14	0.07		H	Emslie et al. 2022	
	UE 189	0.86	0.01		H	Emslie et al. 2022	
	UE 189	0.75	0.03		H	Emslie et al. 2022	
	UE 189	0.69	0.02		H	Emslie et al. 2022	
10.075–10.078	UE 938–2	10.18	0.66	F?	H	Emslie et al. 2022	Negative structure with stone elements, rectangular plan corridor (L 1.40 × W 1.08 m) and circular chamber (2.31 × 2.56 m), NMI: 4, ceramics vessels and fragments, flint blade, bone/ivory needle or awl, no cinnabar
	UE 938–3	0.38	0.01	U	UNK	This paper	
	UE 875	0.34	0.02	U	H	This paper	
10.005	UE 30 A	2.74	0.24	U	T	This paper	Negative structure, irregular plan, NMI: 2 no grave goods, no cinnabar
	UE 30 J	0.74	0.01	U	T	This paper	
10.060	UE 670–2	1.76	0.02		T	This paper / Emslie et al. 2022 (0.48)	Negative structure, irregular plan (2.60 × 1.43 m), NMI: 2, ceramic vessels, lithic halberd, no cinnabar
	UE 670–1	0.18	0.002		F	This paper / Emslie et al. 2022 (0.09)	
10.084	UE 868	1.54	0.20	F?	T	This paper / Emslie et al. 2022 (1.38)	Negative structure with stone elements, irregular plan (L 1.94 × W 1.58 m), NMI: 1, ceramic vessels, flint blade, no cinnabar

Table 2 (continued)

Structure	Individual	THg (µg/g)	sd (µg/g)	Sex	Bone	Source	Description
10.034	UE 499	1.33	0.14		H	Emslie et al. 2022	Megalithic structure, rectangular plan corridor (L 1.32 × W 0.53–0.55 m) and circular chamber (diameter 2.13 m), NMI:7, ceramic vessels, axe, halberd, arrowheads, lithic blade, small rough stone, bone/ivory objects, flint fragment, 2 marine shells (*Pecten maximus* and *Pecten jacobeus*), no cinnabar
10.059	UE 730	0.98	0.01	M?	T	This paper / Emslie et al. 2022 (0.60)	Negative structure, irregular plan (1.65 × 0.94 m), NMI: 1, ceramic vessel and fragment, flint blade, lithic knapping debris, rare stone, no cinnabar
10.031	UE 453-M	0.94	0.28	M	T	This paper / Emslie et al. 2022 (0.11)	Negative structure, circular plan (2.40 × 1.94 m), NMI: 3, ivory 'staff', no cinnabar
	UE 453-F	0.86	0.16	F	T	This paper / Emslie et al. 2022 (0.42)	
10.055	UE 718	0.94	0.82	U	T	This paper / Emslie et al. 2022 (0.40)	Negative structure with stone elements, rectangular plan (L 3.34 × W 1.79 m), NMI: 1, ceramic vessels and fragments, flint blade, bone/ivory awl or needle, small rough stone, no cinnabar
10.013	UE 294	0.76	0.97	F?	H	Emslie et al. 2022	Negative structure with stone elements, irregular plan (L 1.65 × W 0.99 m), NMI: 1, no graves goods, no cinnabar
10.040	UE 564–2	0.53	0.07		T	This paper	Negative structure, circular plan (L 1.42 × W 1.11 m), NMI: 3, Roman lamp, worked bone fragment, 1 marine malacofauna shell (Pecten sp), no cinnabar; 564–1 (male, peptide)
	UE 564–1	0.40	0.01	M	F	This paper / Emslie et al. 2022 (0.23)	

Table 2 (continued)

Structure	Individual	THg (µg/g)	sd (µg/g)	Sex	Bone	Source	Description
10.071	UE 698 Unk	0.48	0.01	F	F	This paper / Emslie et al. 2022 (0.33)	Negative structure, circular plan (D 1.73 m), NMI: 9, 2 ceramic vessels, no cinnabar
	UE 698-F	0.46	0.06	F	T	This paper	
	UE 698 J	0.29	0.001	F	F	This paper	
10.002	UE 43	0.35	0.02	U	T	This paper / Emslie et al. 2022 (0.29)	Negative structure, irregular plan (L 1.09 × W 0.63 m), NMI: 1, ceramic vessels, no cinnabar
10.012	FN 246–2	0.32	0.02	U	H	Emslie et al. 2022	Negative feature, irregular plan (L 1.88 × W 1.40 m), NMI: 5, 1 marine malacofauna shell (*Pecten jacobeus*), no cinnabar
	FN 246–1	0.19	0.01	U	H	Emslie et al. 2022	
10.080	UE 950	0.29	0.03	F?-U	C	This paper	Negative structure with stone elements, irregular plan (L 1.95 × W 1.30 m), NMI: 2, 6 ceramic vessels and fragments, 2 marine shell, 1 flint blade, 1 arrow head, no cinnabar
10.077	UE 936	0.19	0.01	U	T	This paper	Negative structure, circular plan (L 1.38 × W 1.32 m), NMI: 1, 1 ceramic vessel and fragments, no cinnabar

Sanjuán et al., in preparation). Structure 10.049 contained the remains of an individual first thought to be a young man (García Sanjuán et al., 2019: 1017). Later, through the analysis of sexually dimorphic amelogenin peptides in tooth enamel by nanoflow liquid chromatography-tandem mass spectrometry, this individual was revealed to be a woman (Cintas-Peña et al., 2023). This woman was laid to rest with a large set of grave goods, including several distinguished ivory objects (among them a full tusk of African elephant), which is unparalleled in the Iberian Pre-Beaker Copper Age (Garcia Sanjuán et al., 2018b). For that reason, she was nicknamed 'The Ivory Lady'. The high social and political standing of this woman is revealed not just by the rich grave goods she was buried with,but also by two additional facts: (i) around her grave, up to 134 other features were later made in relatively quick succession (c. 170 years), including 61 graves in what is a formal burial area (referred to as the PP4-Montelirio sector), but they were all made at a distance of at least 23 m; the presence of this 'respect area' suggests there was an inalienable space (probably delimited by physical means not verified archaeologically—wooden fence or similar) around 'The Ivory Lady' that underlined her importance and standing as a prominent ancestor; (ii) when the Montelirio tholos was built and used some two or three generations later, between c. 2800 and 2700 BC (García Sanjuán et al., 2018a: 235), a fresh set of offerings as lavish and sumptuous as the original one was laid for 'The Ivory Lady'—right above her. A meticulous analysis of this material culture reveals a clear connection between the offerings made inside Montelirio and the ones laid above 'The Ivory Lady', suggesting the builders of Montelirio acknowledged her as a prominent and respected ancestor (García Sanjuán et al., 2018b).

Both Structure 10.049 and Montelirio stand as a fair representation of the highest social positions ever reached by 'elite' groups and individuals in Copper Age Iberia. In both tombs, cinnabar was extensively used: it was sprayed over the bodies and grave goods of both tombs, and in Montelirio, it was used to coat and paint the black-slate slabs used to line the corridors and the chambers (Fig. 3).

The initial target of the study was to obtain mercury samples from three of the bones of each human individual, including humerus (H), femur (F) and/or tibia (T), depending on the availability. At the end, for preservation reasons, samples of all these three bones could only be taken for 28 individuals. For 16 individual samples were taken from two bones and for another 17 individuals from just one. Some other nine bone samples (ulna, rib, radio, fibula, unknown) were also analyzed. The 22 samples of animal bone each come from different animals, including domestic (dog, oxen, sheep/goat) and wild ones (wild boar and deer) which were not found in primary position and for which, therefore, samples were taken only from one bone. A soil sample from inside an animal bone was taken to factor in environmental mercury.

Samples were taken and analysed according to the procedure already explained in previous papers (Emslie et al., 2016, 2022). Most of the samples were taken from compact bone located in the medial area of the diaphysis. The surface (cortical) area of the bones was cleaned and not used for analysis to prevent as much as possible the contamination with particles of the cinnabar sprayed over some of the bodies. Samples were analysed in a Milestone Tri-Cell Direct Mercury Analyser (DMA-80) or in a Nippon MA-3000 analyser (Nippon Instruments Corp., Japan). In most cases,

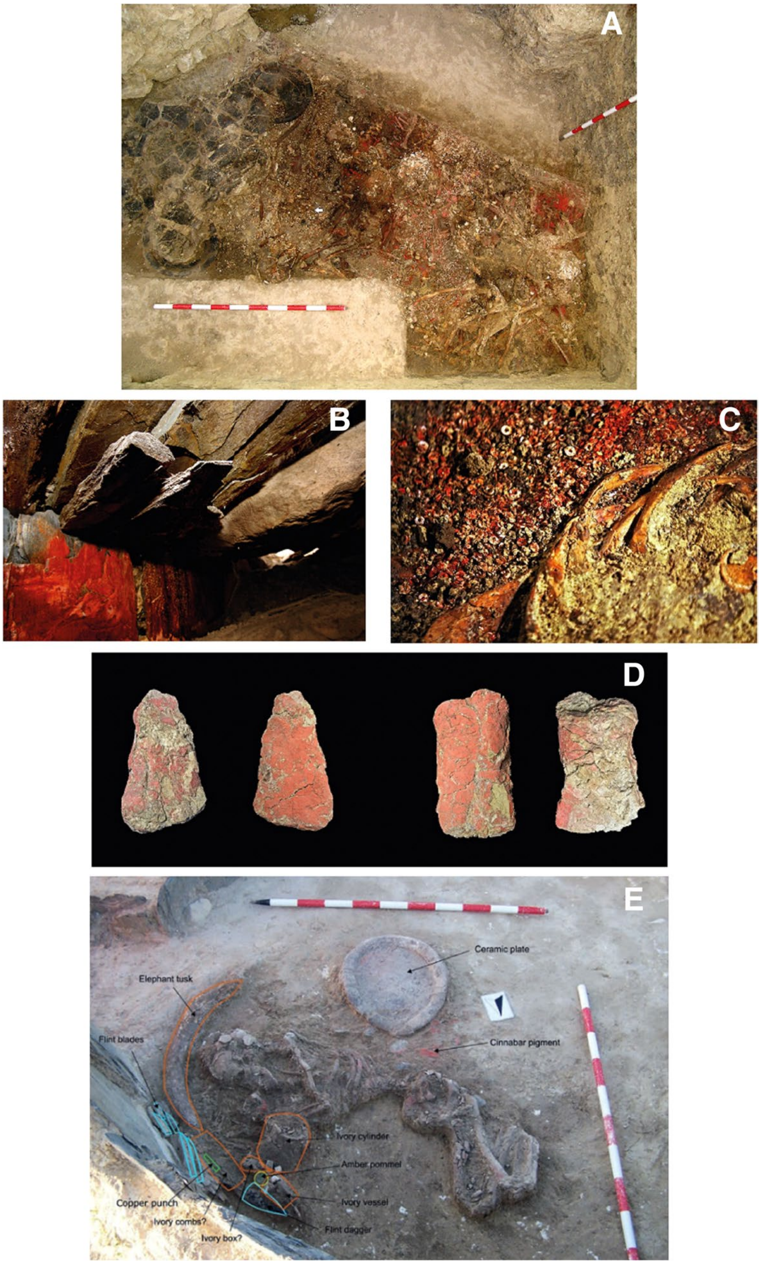


Fig. 3 Examples of the use of cinnabar in Valencina. **A**, **C**, **D** Montelirio Large Chamber: cinnabar powder on human bones and beads, painted baetyls. **B** Montelirio corridor: decorated slabs. **E** Cinnabar powder sprayed on Structure 10.049. Photos: A: Álvaro Fernández Flores and C: Antonio Acedo García (from Fernández Flores & García Sanjuán, 2016); B: Antonio Acedo García (from García Sanjuán et al., 2016); D: adapted from Rodrigo de Balbín Behrmann (from Bueno Ramírez et al., 2016); E: photo: José Peinado Cucarella; drawing: Miriam Lucianez Triviño (from García Sanjuán et al., 2019)

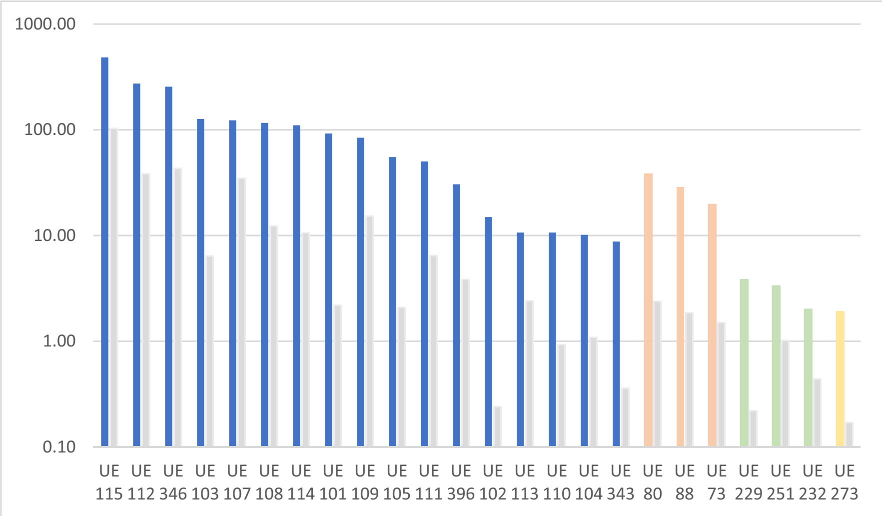


Fig. 4 THg concentration (±sd) in human bone (µg/g) for 23 individuals buried in Montelirio Tholos (17 in LC, 2 in SC (3 SUs), 3 in corridor, 1 outside). The SC contained a MNI = 2, but three samples were analysed)

samples were analysed twice of thrice to obtain an average THg in µg/g (ppm). Each of the measurements was preceded and followed by two samples of two standard reference materials (DORM-4 and DOLT-5 or DORM-4 and TORT3). The results are presented as means ± standard deviation of the values obtained. For individuals with more than one sampled bone, the highest THg value is used.

The results were analysed quantitatively and comparatively to assess potentially significant differences in the occurrence of mercury among different individuals in different contexts and regarding a series of variables. To achieve this, a Shapiro–Wilk normality test, Mann–Whitney's U and χ^2 significance tests were carried out at various times, as described below. To better understand the data, intervals were established to distinguish between 'very high' (> 100 µg/g), 'high' (10–100 µg/g), ´moderate´ (1–10 µg/g) and `low' (below 1 µg/g) THg values.

Results

Montelirio Tholos

A human contingent of 26 people was found in Montelirio, including 20 individuals in the Large Chamber (LC), two in the Small Chamber (SC), three in the Main Corridor (MC), which leads to the LC from the exterior, and one outside the megalith (Pecero Espín, 2016: 410). The individual buried outside the megalithic structure yielded a much older radiocarbon age (OxA-X-2535–32, 4727–4547 cal 2σ BC) and therefore represents an earlier burial episode at the site (García Sanjuán et al., 2018a, b: 208; Table 1). In total, 50 values of total

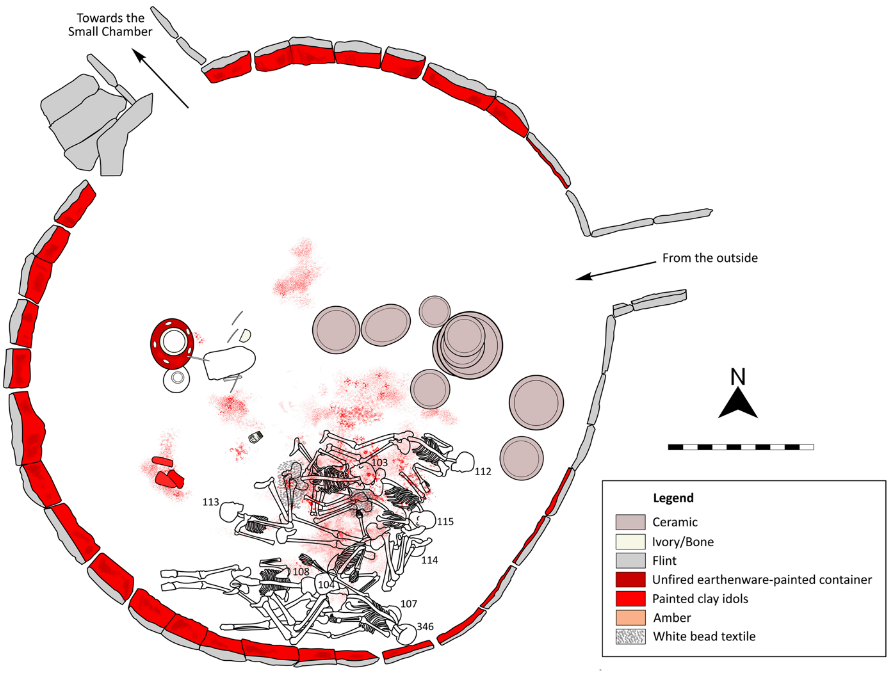
Fig. 5 Graphic recreations of the distribution of individuals in the LC of Montelirio according to THg ▶ levels. Above, THg > 100 ppm; Below, THg: 1–100 ppm. Drawings: Miriam Luciañez Triviño (adapted from Pecero Espín, 2016)

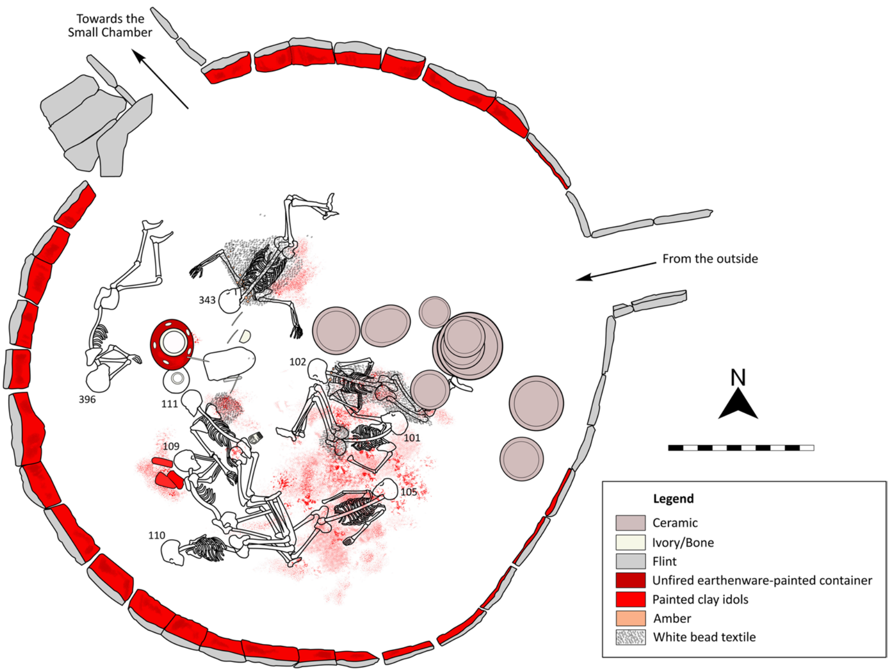
mercury THg (μg/g) are available for the Montelirio tholos, covering 23 of the 26 individuals buried in it (the ones excluded are UE116, UE356 and UE398, all from the LC; Fernández Flores et al., 2016; Fig. 4). While 26 of those values have already been published (Emslie et al., 2016), the remaining 24, corresponding to 12 individuals (10 in the LC and 2 in the SC), are presented here for the first time.

In general, mercury values recorded in the Montelirio human bones are very high. While they all rise above 1 μg/g, 83% (excluding the individual buried outside the megalithic structure) are above 10 μg/g, with average total mercury concentrations of 81.92 μg/g, 95.20 μg/g and 18.79 μg/g in humeri, femora and tibiae, respectively. Recorded values are particularly high for the 17 individuals from the LC, with values ranging from a maximum of 483.97 ± 102.94 μg/g (individual UE115, femur) to a minimum of 8.77 ± 0.36 μg/g (individual UE343, humerus). In the SC, both individuals present levels higher than 10 μg/g (19.61 ± 1.50 b–38.10 ± 2.39 μg/g). The lowest values are recorded in the three individuals buried in the MC (2.00 ± 0.44 μg/g–4.89 ± 0.32 μg/g).

The human remains analysed from the LC belong to adult women except in two cases (individuals UE115 and UE396), for which sex could not be established through conventional anthropological examination (Pecero Espín, 2016: 427). The bodies were in anatomical connection, occupying the whole space of the LC, except the area leading to the SC. It is important to note that these bodies were carefully arranged around a stela or altar made in unfired clay that was found at the centre of the LC, with some ceramic plates and other objects surrounding it (Fig. 5). Two subgroups of individuals can be discerned according to total THg levels.

The first subgroup includes nine individuals (eight women, one sexually undetermined) with maximum values between 110.30 ± 10.58 and 483.97 ± 102.94 μg/g (four each in femora and humeri). That means that up to 53% of the individuals sampled in the LC have total mercury values above 100 μg/g (Fig. 5a). Generally, the individuals of this subgroup were between 25 and 45 years of age at the time of death, with only one exception (individual UE114, who was 18–23 years old). They were mostly laid in the south-eastern sector of the LC and according to their relative vertical position (showing the sequence of deposition) were the first to be deposited in the tomb with the only exception of individual UE104, who was one the latest to be buried (García Sanjuán et al., 2016:519). This subgroup includes all the women buried in the tholos (six in total) who all exhibit a degenerative condition (arthrosis in the dorsal vertebrae, shoulders and lower limbs) caused by ageing or functional wear (Pecero Espín, 2016: 434–435). In addition, three of these women showed an anomalous development of the first gluteal tuberosity of the femur in connection with higher muscle work and blood flow, in turn connected with sustained physical activity involving the legs (Pecero Espín, 2016: 432). Individuals UE112 and UE107


Towards the Small Chamber
From the outside
N
103
112
113
115
114
108
104
107
346
Legend
Ceramic
Ivory/Bone
Flint
Unfired earthenware-painted container
Painted clay idols
Amber
White bead textile


Towards the Small Chamber
From the outside
N
343
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Legend
Ceramic
Ivory/Bone
Flint
Unfired earthenware-painted container
Painted clay idols
Amber
White bead textile

showed higher levels of animal protein consumption, which could be explained by their enjoying a higher social status (Fontanals Coll et al., 2016: 446), possibly also in connection with their older age.

The second subgroup includes seven adult females and one adult of undetermined sex with THg levels below 100 µg/g (Fig. 5b) and maximum values between 8.77 ± 0.36 and 92.26 ± 2.19 µg/g (five of them measured in humeri). In general, these individuals were younger, between 18 and 34 years of age at the time of death. They were buried partly above the previous subgroup, in two different areas of the chamber: individuals UE110, UE396, UE111, UE109 and UE343 were laying on the western side, closer to the clay stela/altar, while the other three (UE102, UE101, UE105) were laid closer to the entrance of the LC, next to a set of large ceramic plates and clearly above the first subgroup. In general, the individuals of this subgroup with lower THg levels do not display any pathologies, while those two with comparatively higher THg levels (UE109 and UE101) present periostitis of the femora and tibiae, respectively. In addition, one woman (UE101) had squatting facets in both tibiae and polydactyly (six toes) in both feet (Pecero Espín, 2016: 436). The latter condition was undoubtedly a very visible and remarkable physical trait that probably made this woman quite special in her time. Women UE101 and UE102, who were laid closely together, also showed hyper-development of the first gluteal tuberosity of the femur.

In the SC, which was badly altered by a re-use in Roman times, only a small number of poorly preserved human remains was found. They revealed two adult individuals, one female and one sexually indeterminate (Pecero Espín, 2016: 423), dating to the same period as those in the LC (Bayliss et al., 2016). Mercury was measured on anatomically-unspecific bones from stratigraphic units UE80 and UE88, revealing THg concentrations ranging between 19.61 ± 1.50 µg/g and 38.10 ± 2.39 µg/g, which are similar to those of the second subgroup of the LC described above.

The central section of the MC yielded three inhumations, two of which were secondary (individuals UE229, a male adult, and UE251, a probably female adult) and one primary (individual U232, a sexually undetermined adult) (Pecero Espín, 2016: 423–425), as well as numerous offerings, including plants, animals and artefacts. Mercury levels measured in the bones of these three individuals were significantly lower than those from both chambers, ranging between 2.00 ± 0.44 and 4.89 ± 0.32 µg/g. Individual UE232, showing the lowest THg concentrations, was buried with a small ceramic vessel placed under his hand.

Individual UE273, who was between 35 and 55 years old at the time of death, was found outside the MC, on the left-hand side of the MC (as one enters). Both his stratigraphic position and one radiocarbon date show this individual predates the construction of the tholos by several centuries. The bones of this subject had mercury levels at 4.32 ± 0.82 µg/g.

PP4-Montelirio Sector

As described above, the PP4-Montelirio sector, comprising 134 features that surround 'The Ivory Lady' grave, lies adjacent to the north to the Montelirio tholos (Mora Molina et al., 2013). In total, 94 bone samples belonging to 47 different

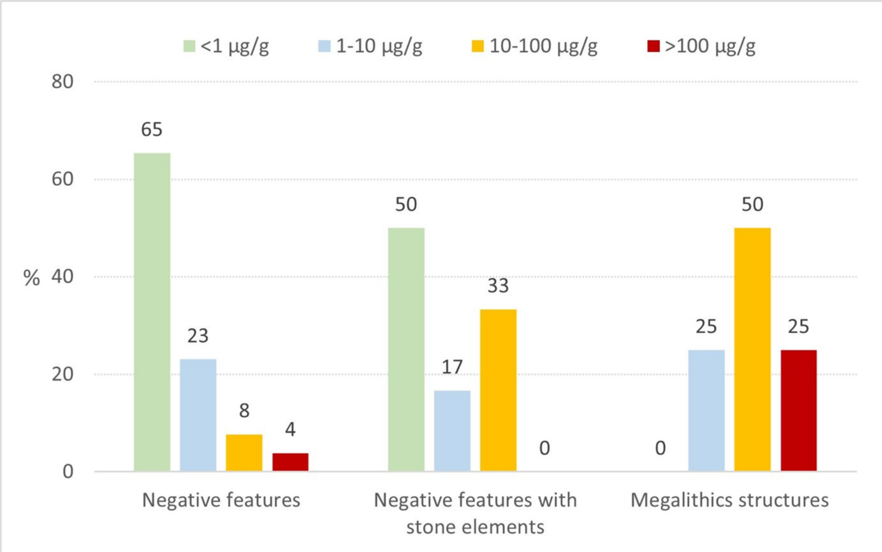

Fig. 6 Percentage of individuals according to THg levels for each type of funerary structure in PP4-Montelirio sector

human individuals (89 Chalcolithic and five Roman ones) from this sector have been analysed (Table 2), Emslie et al., 2022). Only for 18 of them was it possible to measure THg concentrations for all three target bones (humerus, femur and tibia). The sampling for this sector encompasses 42 Copper Age individuals (89 results) buried in 22 different burials that break down as follows: three megalithic structures, 12 negative features (pits or basins) and seven negative features with stone elements (Fig. 6). Seven of those features are individual burials, while 15 were collective. In this paper, 59 measurements of THg concentrations for 31 of those Copper Age individuals (found in 20 different structures) are presented for the first time). The sampling of the PP4-Montelirio also includes five Roman-time individuals from as many graves found on the northern limit of the sector. The results for the Roman burials were presented in a previous paper (Emslie et al., 2022) but are also discussed here as a control group for the 3rd-millennium population. Cinnabar powder was only observed in two of the 27 burials (22 dating to the Copper Age, five to Roman time), namely structures 10.042–10.049 and 10.074, which, as is discussed next, are the ones with the bones showing the highest mercury values.

The results for the Copper Age population of the PP4-Montelirio sector presents a high variability of THg, ranging from 0.19 to 480.50 µg/g. Up to 45% of the individuals of this sector having mercury levels above 1 µg/g, in particular those buried in megalithic structures. Half of the 22 burials analysed include at least one individual with mercury levels above 1 µg/g, and in eight of them (36%), measurements are above 10 µg/g (10.18 ± 0.66–480.50 ± 19.58 µg/g).

The second highest recorded mercury levels correspond to 'The Ivory Lady' (THg value of 185.54 ± 10.65 µg/g), while the two individuals found in Structure 10.042

(indeterminate sex) returned values of 19.70 ± 1.62 and 28.10 ± 6.23 µg/g. Substantial amounts of cinnabar powder were sprayed in both chambers. Interestingly, however, the highest mercury value was not recorded in Structure 10.042–10.049, but in an adult person of indeterminate sex found in Structure 10.074, a single inhumation in a simple pit of small size (1.60 × 1.40 m), irregular plan and no stone elements (Robles Carrasco et al., 2017:106). The extremely high value recorded for the person buried in Structure 10.074 (480.50 ± 19.58 µg/g in the radius) is on par with the highest recorded at the Montelirio tholos (individual UE115, 483.97 ± 102.94 µg/g). Mercury measurements for a rib of this individual, however, yielded much smaller values (16.55 ± 2.29 µg/g). Cinnabar powder as well as a scallop shell (*Pecten maximus*) were identified inside Structure 10.074 (Liesau et al., 2014: 96). In principle, although the configuration of the grave (simple pit with no stone elements) would not lend the person buried inside Structure 10.074 a very high social standing, the presence of cinnabar and a scallop shell suggest some kind of social prominence.

Scallop shells must have had a high symbolic value within the Copper Age world view[1] as they were used as grave goods in 16 other graves in the PP4-Montelirio sector (Liesau et al., 2014: 82–83) and to manufacture the perforated beads of the garments worn by some of the women found in the LC of Montelirio (Díaz-Guardamino et al., 2016). Mercury values are available for the bones of six of the persons who received scallop shells as grave goods. Three of them recorded high (> 10 µg/g) or very high values (> 100 µg/g, Structure 10.074), while the others showed much lower ones: Structure 10.034, a single inhumation, returned a THg value of 1.33 ± 0.14 µg/gµg/g, and Structures 10.012 and 10.040 yielded values < 1 µg/g (although data are not available for all the individuals buried in these two graves).

It should be noted that in the analysed PP4-Montelirio burials, whether individual or collective, where mercury levels > 10 µg/g are recorded, no evidence of cinnabar powder has been found. Structure 10.035, a simple pit with no grave goods and no cinnabar powder, contained two individuals. One of them, an adult of undetermined sex, exhibited osteitis in an ulna (Robles Carrasco et al., 2017: 107) and returned a THg value of 52.29 ± 4.68 µg/g. The other individual is an infant between 7 and 10 years of age, the only surviving elements being the teeth (Cintas-Peña et al., 2018: 98).

Structure 10.073 is a negative feature with some stone elements that yielded a minimum number of three individuals as well as grave goods consisting of three ceramic vessels, a marine shell (*Cymbula nigra)* and a copper punch with a handle made in worked osseous material (probably animal bone). Two of these individuals, between 26 and 40 years of age and likely female, returned high THg concentrations (28.08 ± 2.50 µg/g and 19.70 ± 1.62 µg/g). One of them presented stress marks in the form of enthesopathies in the left radius bone (Robles Carrasco et al., 2017: 107).

Structure 10.044 is also a negative feature with some stones in which a minimum number of three individuals was recorded accompanied by very simple set of grave goods consisting of some fragmented ceramic vessels, one whole vessel and a small unworked stone. All three individuals were analysed for mercury, yielding values

[1] The earliest known symbolic use of these shells in Iberia goes back to c. 35,000 years BP and is attributed to late Neanderthals (Zilhão et al., 2010).

of 17.13 ± 2.67 μg/g, 2.48 ± 0.05 μg/g and 0.38 ± 0.01 μg/g. Individual #2 presented stress marks in the form of enthesopathies in the femur and collarbone (Robles Carrasco et al., 2017: 110).

Structure 10.028 is a simple pit 1.7-m across, containing a MNI of 9 individuals distributed in two levels: the lower level contained five individuals, while the upper level yielded four individuals arranged radially. Eight of them were analysed for mercury. Five returned values above 1 μg/g (between 16.31 ± 0.96 and 1.14 ± 0.07 μg/g) while the other three yielded values between 0.86 ± 0.01 and 0.69 ± 0.02 μg/g.

Structure 10.075–10.078 is a negative feature with stone elements. It presents a chamber circular in plan, an access corridor and several slate slabs at the entrance. Two depositional levels were recorded; in the lower level (10.075), a single individual was accompanied by a whole ceramic vessel and fragments of a second, incomplete one next to the hands; in the upper level (10.078), three primary inhumations were recorded alongside three ceramic vessels, a needle or punch of worked bone and a flint blade. Mercury concentrations were measured for the individual found in the lower level and for two from the upper level. All THg values were < 1 μg/g except for one of the individuals in the upper level which yielded relatively high concentrations (THg 10.18 ± 0.66 μg/g) compared to all other individuals from this feature.

PP4-Montelirio Sector: Roman-Time Structures

Alongside the Copper Age features, five Late Roman burials were found at the PP4-Montelirio sector, including some simple pits and small chambers made with brick walls (García Sanjuán et al., in preparation). In none of them was cinnabar powder recorded. Mercury contents were analysed for the humeri of five individuals (structures 10.020, 10.045, 10.057, 10.009 and 10.058), showing average values of 0.32 ± 0.52 μg/g, in all cases below 1 μg/g (0.04–0.20 μg/g), except for the individual found in Structure 10.020 (THg 1.24 μg/g; Emslie et al., 2022). As will be discussed below, these values are in line with two other studies of mercury in human bone of Roman chronology in Iberia.

Faunal Remains and Soil Samples

As well as human bone, as part of this study, mercury concentrations were measured in 22 samples of animal bones from the PP4-Montelirio (6 samples) and other Valencina sectors (16 samples; Table 3).

From Structure 10.042, six poorly preserved animal bones from stratigraphic units UE640 and UE648 were analysed. The first group of samples (UE640) yielded very high values, between 32.57 ± 8.44 and 120.68 ± 8.06 μg/g, the latter corresponding to a deer antler that showed signs of thermal stress and had been probably used as a tool (Liesau et al., 2014: 90). In turn, all THg values for UE648 were below 1 μg/g (0.46 ± 0.18–0.79 ± 0.04 μg/g). The Pabellón Cubierto animal bone samples yielded high values for sheep/goat, with THg concentrations of 14.44 μg/g and 11.17 μg/g. Animal bone samples from Mariana Pineda Street and Huelva Street

Table 3 Comparison of mercury levels (μg/g) in animal bones from different prehistoric sites and recent times (*included prehistoric sites near Almadén mines, Ciudad Real)

Period	Site	*n*	Media	sd	Min	Max	Source
Chalcolithic	Sector PP4- Valencina (10.042, UE640)	3	69.42	45.79	32.57	120.68	This paper
	Sector PP4- Valencina (10.042, UE648)	3	0.649	0.171	0.457	0.787	This paper
	Valencina (Pabellón Cubierto)	13	4.727	4.289	1.540	14.44	This paper
	Valencina (others)	3	0.270	0.144	0.110	0.390	This paper
	Perdigões	5	0.016	0.014	0.008	0.041	Emslie et al., 2015
	Los Cercados, Las Quintanas, Melgar de Abajo, El Castillo (Valladolid)	10	0.018	0.016	0.007	0.050	Logemann et al., 1994
Bronze Age	El Balconcillo (Soria), Cueva de Pedro Fernández and Arenero Soto (Madrid), Cerro de la Encantada and Motilla de Sta. María del Retamar (Ciudad Real), Cerro de la Campana (Murcia)	34	0.161	0.261	0.008	1.373*	Logemann et al., 1994
Iron Age	El Castillejo (Soria)	17	< 0.006	-	-	-	Logemann et al., 1994
Recent time	Almadén (Ciudad Real), Teruel	15	0.079	4.472	0.005	0.887	Logemann et al., 1994

Table 4 Total mercury concentrations (µg/g) at Valencina

	n	Average	sd	Min	Max
Montelirio tholos					
Large chamber	17	131.93	119.69	8.77	483.97
Small chamber	2	28.82	9.25	19.61	38.1
Corridor	3	3.57	1.46	2.00	4.89
Exterior	1	4.32	0.82	4.32	4.32
PP4—Montelirio sector					
Chalcolithic burials	42	21.17	78.42	0.19	480.5
Roman burials	5	0.32	0.52	0.04	1.24
Animals	6	35.03	47.53	0.46	120.68
Other sectors					
Animals	16	3.89	4.24	0.11	14.4
Soil	1	14.5	-	14.5	14.5

yielded lower values (0.11–0.39 µg/g). In none of the features from which these 16 samples were retrieved was cinnabar powder observed.

A soil sample from inside the shaft of an animal long bone (the radius of an ovicaprid with THg: 2.73 µg/g) located in the Pabellón Cubierto sector was also analysed, yielding a THg concentration of 14.50 µg/g.

Discussion

Exceptionally High levels?

In the context of evidence available for Iberian Late Prehistory, with 23 sampled sites (Emslie et al., 2022), the first observation that stands out from the data described above is the exceptionally and pervasively high values of mercury in Copper Age human and animal bone (and even in the only soil sample available) at Valencina—a full synthesis of the THg values is presented in Supplementary data SD 1 and 2. Copper Age Valencina has yielded the highest values of mercury in human bone recorded in Iberia to date, with concentrations above 400 µg/g at Montelirio and Structure 10.074 and several occurrences of values above 100 µg/g (Table 4, Fig. 7). Notably, about 65% of all Valencina individuals produced concentrations of THg higher than 1 µg/g while the individual with the lowest level had only 0.19 µg/g. It is also important to underline that this appears to be strictly a Copper Age phenomenon, as the values obtained for the PP4-Montelirio individuals of Roman age are much lower, with an average concentration of 0.32 µg/g (Tables 4 and 5).

But how high are the values obtained for Copper Age Valencina relative to other sites? A main source of comparison is the data currently available for Neolithic and Copper Age Iberia, between the 5th and 2nd millennia BC (Table 6; Emslie et al., 2022). This evidence shows a non-homogeneous distribution of mercury values over

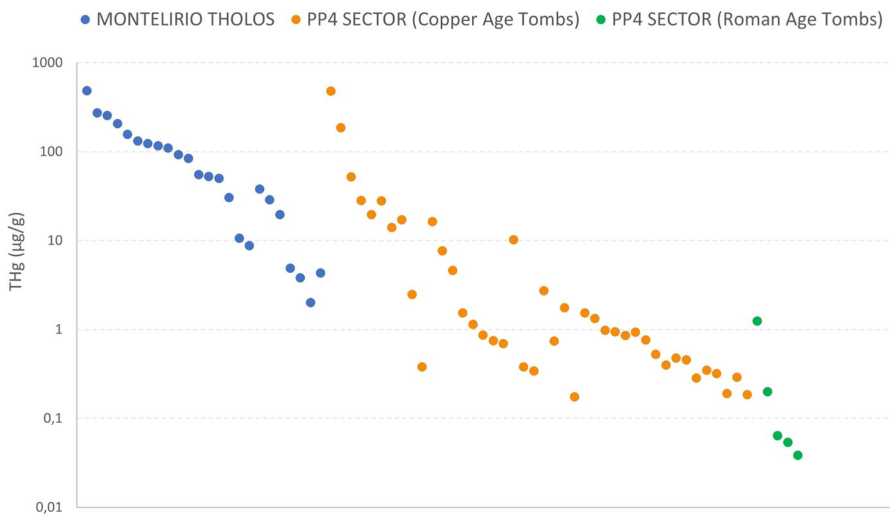

Fig. 7 THg concentration (µg/g) in human bones from archaeological site Valencina de la Concepción—Castilleja de Guzmán (Sevilla)

Table 5 χ^2 test results

Concentrations intervals (µg/g)	Results	H_0 = no difference in different types of bone
4 intervals: > 100, 10–100, 1–10, < 1	8.062 *p* (0.05): 12.5916	H_0 accepted
3 intervals: > 100, 10–100, < 10	7.66 *p* (0.05): 9.4877	H_0 accepted
2 intervals: > 1, < 1	0.86 *p* (0.05): 5.9915	H_0 accepted
2 intervals: > 10, < 10	5.58 *p* (0.05): 5.9915	H_0 accepted

time, with the highest results observed for populations living between the late 4th and late 3rd millennia (or the very end of the Late Neolithic, and the Early Copper Age). Out of 12 sites within this chronology, nine (75%) show THg values above 10 µg/g.

The earliest site for which THg measurements are available is Campo de Hockey (San Fernando, Cádiz), a burial site that includes a proto-megalithic grave (Vijande-Vila et al., 2022), one of the earliest of its kind in Iberia, dated to the late 5th millennium BC (García Sanjuán et al., 2022). Some of the burials at this site yielded high-end objects made from exotic materials such as amber and turquoise. The average THg value for the individuals analysed (n = 18) is 0.62 µg/g, with a maximum value of 2.45 µg/g.

Of the sites dating to the late 4th and early 3rd millennium, or roughly contemporary with Montelirio and the PP4-Montelirio burials, a great majority (75% of them) show maximum mercury values falling in the range we have designated as 'very high'

Table 6 Iberian archaeological sites for which analysis of total mercury in human bone is available (modified from Emslie et al. 2022)

Site	MN	LN/EC	C	BA	IA/RE/ME
Campo de Hockey	0.62				
Outeiro Alto 2		229.23 (Pits 5 and 14)		0.14 Hypogeum	
Monte Malheiro 2		28.85/164.72 (Hypogeum)			
Valencina de la Cuesta - Castilleja de Guzmán*		185.54 (PP4-10.049)	131.93 (Montelirio LC)		
			17.16 (PP4-Sector Chalcolithic burials)		
			28.82 (Montelirio (SC)		
			3.57 Montelirio (Corridor)		
		4.32 (Exterior)			
					0.32
Santa Rita*		130.49			
Dolmen Estrada Ansi~ao		18.71			
Natural Cave Cova da Moura		14.65			
Sobreira de Cima*		51.87 (Tomb I)			
Monte Canelas I		10.96 (Hypogeum)			
Monte da Comenda		22.65			
Tholos of Cabeço Arruda II*		7.35 (Tholos)			
Natural Cave of Pedra Furada		0.13			
Perdigoes*		0.26 (Pit 7)	36.56 (Tomb I Chamber)		
		0.06/0.12 (Pit 11)	99.16 (Tomb II Chamber)		
			2.10 (Tomb III Chamber); 0.41/1.7 (Pit 16)		
			0.36/0.41 (Tomb IV Chamber)		
Las Mayores*			226.7 - 497.7 (Tomb with cinnabar)	0.06	
			7.39 (Tomb without cinnabar)		
Paimogo I*			1.54 (Tholos)		
Sao Paulo II			9.89 (Hypogeum)		
Natural Cave of Eira Pedrinha			0.19		
Torre Velha 3			0.026 (Pit burials)	0.021 (Hypogeum); 0.26 (Pit burials)	
La Encantada				2.41	
Vinha das Calicas 4					0.09
Necropolis de Miroico					0.044
Monte de Nora					0.041
A Lanzada					0.264

*Site with cinnabar

MN Med Neolithic, *LN/EC* Late Neolithic / Early Chalcolithic, *C* Chalcolithic, *BA* Bronze Age, *IA/RE/ME* Iron Age, Roman Era, Middle Age

THg values (above 100 µg/g), including Outeiro Alto 2, Monte Malheiro 2, Sobreira da Cima and Santa Rita, all of them in southern Portugal. In turn, 'high values' (between 10 and 100 µg/g) were found at Estrada Ansimao (a dolmen), Cova de Moura (a natural cave), Monte Canelas I and Monte da Comenda (hypogea), also in southern Portugal. Of this chronology, only Perdigões (pit burials) and Pedra Furada (natural cave), again in southern Portugal, have 'low' values (below 1 μg/g). Sites dating to a later phase of the Copper Age (c. 2800–2200 BC) also show high THg values, akin to those found in the LC at Montelirio. The recorded levels are 'very high' (> 100 μg/g) at Las Mayores (Numancia de la Sagra, Toledo), in central Spain, and Tomb II of Perdigões, and 'high' (10–100 μg/g) at Perdigões Tomb I. One exception is Tomb IV at Perdigões, dating to the latest Copper Age (Valera 2020). Two individual burials in this tomb had low THg values (0.31 and 0.41 μg/g; Emslie et al., 2022). It is also interesting to note that most of the Copper Age sites with human bone showing high or very high mercury values are located within the Guadiana River basin and therefore easily connected with the Almadén mines in central Spain (Emslie et al., 2022: 209).

In turn, of the four sites dating to the Bronze Age for which THg data in human bone are available, only in one, La Encantada, were values reported above 1 μg/g. La Encantada (Granátula de Calatrava, Ciudad Real) is located very near the Almadén mines and the individuals analysed ($n = 37$) yielded average mercury values of 2.41 μg/g, with a maximum value of 12.43 μg/g. No cinnabar was found in any of those burials (Emslie et al., 2019: 4), which could suggest exposure via environmental mercury or via processing of the material (mining, grinding, etc.).

This summary shows that, among the Late Neolithic, Copper Age and Bronze Age sites of Iberia sampled to this date, Valencina ranks the highest in mercury exposure. The results obtained for the PP4-Montelirio Roman-time individuals are quite valuable as corroboration of how exceptionally high these Valencina mercury values are. Indeed, the validity of the results for the Roman burials at Valencina is demonstrated in that they are fully in line with what is known for the two only other Roman-time Iberian human contingents for whom mercury analysis has been published. At La Lanzada, located in Galicia, north-west Spain and dating to the first to fourth centuries AD (López-Costas et al., 2020), mercury levels were measured in cortical tissue (mostly from femurs) in 26 individuals of both sexes, who showed average values of 0.136 ± 0.114 μg/g with a maximum of 0.516 μg/g. At La Lanzada, levels of mercury in human bone are two and a half orders of magnitude above the concentrations measured in individuals of Early Medieval/post-Roman date (fifth to seventh centuries AD) from the same site, with average values of 0.039 ± 0.043 μg/g ($n = 18$), which in itself again demonstrates how high the Valencina Copper Age values are within the Iberian context. The conclusions of the study suggest that the population of La Lanzada, who lived in an area where no major industrial activity took place in Antiquity, was exposed to environmental mercury, regardless of specific dietary, social or cultural practices AD (López-Costas et al., 2020: 6; Álvarez-Fernández et al., 2020). The results obtained from samples of the late Roman necropolises of Miroiço (Sintra) and Monte da Nora (Elvas) in southern Portugal (Emslie et al., 2022) are even lower than those of La Lanzada and one order of magnitude below those measured in the Roman bones of the PP4-Montelirio. At Miroiço ($n = 16$), the THg values ranged from 0.02 to 0.11 μg/g (average THg of 0.044 ± 0.029 μg/g), while at Monte da Nora ($n = 15$), they ranged from 0.01 to 0.11 μg/g (average THg of 0.041 ± 0.028 μg/g).

A comparison with the evidence available outside Iberia also corroborates that values observed at Valencina are anomalously high. Measurements of mercury in bone from two Egyptian mummies of Ptolemaic age (c. 170 BC) yielded results of 0.43 and

0.1 µg/g (Cockburn et al., 1975: 1159). A study of mercury in human bone from the sites of Tokushima and Matsuyama (Shikoku, Japan), dating to different periods (to the sixth–seventh and twelfth–seventeenth centuries AD; Yamada et al., 1995) yielded some interesting results. The average mercury value from both sites for the most recent individuals was 8.2 ± 3.3 µg/g, while the individuals of the earlier period showed abnormally high values above 1.000 µg/g at Tokushima and between 100 and 200 µg/g at Matsuyama. In this earlier period, the tombs of males belonging to the elite were probably painted with mercury pigments, as were the bodies themselves (Yamada et al., 1995: 254–255). By comparison, the Hg average concentrations in the bones of more than 100 individuals of Medieval and Modern age in Europe have a general background level of ca. 0.050–0.100 µg/g (Rasmussen et al., 2008; 2032). In Sardinia, the mean level in human bones from four burial sites dating to the twelfth and eighteenth centuries AD oscillates between 0.13 and 0.74 µg/g (Bocca et al., 2018). In Iceland, where mercury in human bone was measured to ascertain its possible medicinal use in the treatment of infectious diseases during the sixteenth century, as well as to assess the influence of the island's volcanic activity on the island on the mercury content in bone, record values of 13.059 ppm were detected (Walser et al., 2019: 9).

A recent study of mercury in bone based on 95 present-day Polish hospital patients between 25 and 91 years of age (Zioła-Frankowska et al., 2017: 549) showed average total THg values in femora of 0.037 ± 0.035 µg/g, including maximum values of 0.177 µg/g for men (n = 38) and 0.170 µg/g for women (n = 57). Another paper published in recent years (Miculescu et al., 2012: 39) reported average levels of 0.9 µg/g in human bone from individuals older than 65 years. Comparatively, these values are between two and three orders of magnitude (or between 100 and 1000 times) below the average total mercury in the bones of the people buried in the LC of Montelirio.

Other evidence not directly comparable with that of Valencina, but of significance, comes from studies of contemporary populations based on samples of blood, urine or hair, which are, for obvious reasons of accessibility, the preferred biomarkers in current medicine. These data cannot be directly compared with those based on bone for various reasons, including different accumulation rates, as well as growth and remodelling patterns (Bernhof, 2012). It is important to note that accumulations in bone and hair reflect variable times of exposure: while hair samples reflect Hg exposure for, approximately, the last 2 months (assuming a growth rate of 10 mm per month), bone samples reflect average exposures over periods of several years (in the femur, probably for the last 5 to 10 years; Rasmussen et al., 2012: 1191). In principle, there are grounds to assume that mercury levels observed in bone tissue might significantly underestimate accumulations in other bodily organs to between one and two orders of magnitude. Mercury appears to concentrate on bones between 10 and 100 times less efficiently that in hair. Individuals with mercury levels of 2.0–16.4 µg/g in hair returned values of 0.054–0.092 µg/g in their bones (Rasmussen et al., 2012: 1192–1193). A recent study of animals exposed to MeHg revealed that THg concentrations in the kidneys (32.5 µg/g) were 100 times higher than those in bones such as humeri and femora (0.45 ± 0.43 and 0.37 ± 0.38 µg/g) (Halffman, 2009: 103). Such differences could indicate different accumulation rates; low-intensity chronic exposure may cause a slow incorporation of mercury in an organism leading to higher 'final' values at death (Rasmussen et al., 2012). Despite the difficulty of comparing data from bone samples and from other biomarkers, the information derived from modern biological or health studies provide an interesting background to better assess the extent and scale of mercury exposure in Copper Age Valencina.

A recent study of THg in hair samples of 1044 women between 18 and 44 years of age in 25 different countries (Bell et al., 2017) found that 42% had values above 1 µg/g which, according to the US Environmental Protection Agency (EPA) is a reference value below which mercury would not represent a significant health threat. Another 55% of these women showed values above 0.58 µg/g, which is a threshold set recently in a more conservative approach to public health (Bell et al., 2017: 6). According to this study, the scenarios with higher exposure are those involving the inhalation of mercury vapours, including small-scale gold mining and gold craftwork with substances that contain mercury. Individuals falling in the first group from Indonesia showed maximum values of 90.84 µg/g ($\bar{X}$: 9.405 ± 18.87, 94% > 1 µg/g), whereas others from Kenia returned maximum levels of 81.12 µg/g ($\bar{X}$: 5.26 ± 15.663, 64% > 1 µg/g). In the second group, the maximum value of 28.46 µg/g ($\bar{X}$: 3.62 ± 12.382, 75% > 1 µg/g) was recorded in Nepal.

Since the Minamata catastrophe (Japan) in the 1950s, several studies have focused on the analysis of the relationship of dose–response between the exposure to the organic form of mercury, methylmercury (CH_3Hg), and its toxic effects. Exposed women recorded concentrations of 40 µg/g in hair (27 times higher than reference areas). In the USA, average Hg concentration in the hair of women between 1999 and 2000 was 0.2 µg/g (McDowell et al., 2004), while in the Faroe Islands between 1986 and 1987, it was of 4.27 µg/g (Driscoll et al., 2013), and in Valencia (Spain) in 2016, it ranged from 0.07 to 6.87 µg/g (Yusà et al., 2017).

The international health community considers as acceptable concentrations of mercury in hair of 1–6 µg/g (ATSDR, 1999), 1 µg/g (Bellanger et al., 2013) or 2.3 µg/g (Den Hond et al., 2015). There is widespread agreement that values above 10 µg/g in hair are damaging for human health. Values between 200 and 800 µg/g measured in hair have been reported in cases of moderate mercury exposure (Rasmussen et al., 2012: 1192). These levels, derived from hair samples, are typically used as an index of methylmercury exposure because inorganic forms of mercury do not accumulate in significant amounts in hair, which makes it an unsuitable marker of exposure to inorganic substances (Risher, 2003: 15), such as cinnabar.

In addition to comparisons with studies based on human bone or hair samples, another potential interpretation of the results for Copper Age Valencina is to consider the results obtained for samples of animal bone and soil. In this sense, the results of animal bone samples from Structure 10.042 (UE640) and Pabellón Cubierto at Valencina are much higher than those obtained from other Iberian sites (Emslie et al., 2015; Logemann et al., 1994).[2] Although there is a degree of regional variability, mercury concentrations in animal bones are generally below 1 µg/g, even at sites in the vicinity of the Almadén mines such as Cerro de la Encantada

[2] It is worth noting that while Logemann et al. (1994) express Hg values in ng/g, a later paper by Sánchez Meseguer & Galán Saulnier (2004: 140) reproduced the same table with the same numeric values, but changing the units to mg/g instead of ng/g.

(Granátula de Calatrava) and Motilla de Santa María del Retamar (Argamasilla de Alba), which date to the Bronze Age.

At the Pabellón Cubierto sector in Valencina, a soil sample yielded THg concentrations of 14.50 µg/g. This is in line with soil samples from the Copper Age site of Perdigões, in southern Portugal, which returned average values of 10.85 ± 14.57 µg/g (Emslie et al., 2015). Generally, it has been established that maximum allowable concentrations for mercury in agricultural soils should be between 0.5 and 5 µg/g (Kabata-Pendias, 2010: 24). In Andalusia, the general reference level (NGR)[3] used in environmental science is 6 µg/g for elemental mercury and 25 µg/g for inorganic mercury in soils of inhabited areas. In the Almadén mining district, varying total mercury values are recorded in soils, depending on the study area (Higueras et al., 2012). Thus, agricultural soils from Dehesa de Castilseras provided values between 14 and 21 µg/g, while at the old train station of Chillón, used for the transportation of the mineral, concentrations of up to 350.9 ± 68.6 µg/g were recorded. The highest values were recorded at the mining and metallurgical area of Almadenejos, ranging between 50 and 40.000 µg/g (Lominchar et al., 2010: 21).

Although the Pabellón Cubierto value is moderately high, more soil samples will be needed to adequately assess the presence of mercury in the Valencina soils. In addition, it is important to note that THg values in soil do not reflect the amount of mercury available to enter the trophic chain, which depends on the type of soil and the form of the mercury itself. In this sense, studies in Almadén mercury mining district suggest that the mercury fraction easily available is less than 0.15% of the total mercury measured in the soil samples (Lominchar et al., 2010).

Overall, the information on mercury exposure in humans available for comparative purposes is far from ideal, with few studies based on bone tissue or based on other biomarkers (hair, urine, blood) which cannot be directly compared with those based on bone. The review made here, based on currently published data for the last six millennia, including contemporary populations, suggests that the magnitude of the mercury exposure among the population of Copper Valencina was quite exceptional. In Valencina, mercury concentrations in human bone are between 10 and 1000 times above what would be expected in a healthy person today. In addition, animal bone and soil samples also suggest a strong and pervasive presence of mercury, or associated compounds such as cinnabar, in the natural environment and/or social life of Copper Age Valencina.

Intra-skeletal Variability

An important question is whether mercury concentrations are statistically different in different bones, i.e. whether they display intra-skeletal variability. If all 65 Valencina individuals of Neolithic/Copper Age studied here (23 from Montelirio and 42 from the PP4-Montelirio sector) are considered, the majority of high

[3] In Spanish law, the NGR refers to the concentration of a polluting substance in soil not implying a risk for human health or ecosystem preservation. In Andalusia, this is defined legally by Decree 18/2015 passed on 27 January 2015 (BOJA 38, 25/02/2015).

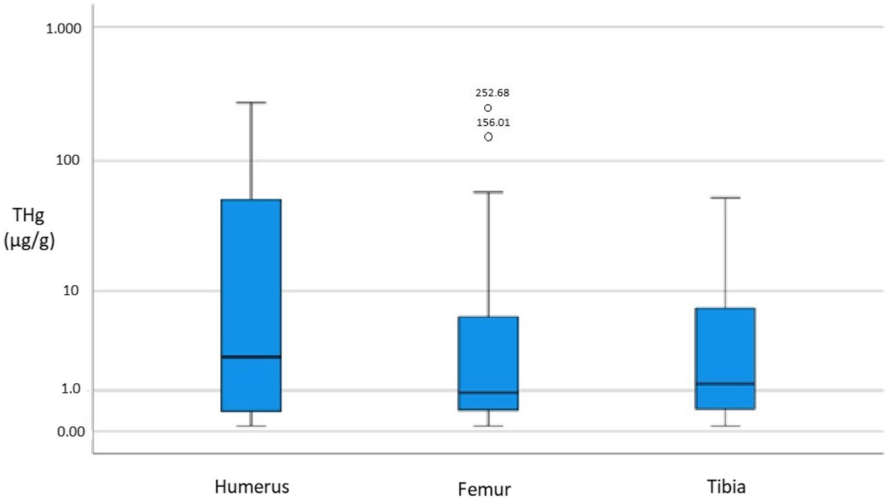

Fig. 8 Total mercury content (µg/g) at Valencina site according to bone type

mercury values (> 10 µg/g) are recorded in humeri, followed by femora and tibiae, which is in line with observations made in previous papers (Emslie et al., 2015: 2; 2019). This higher accumulation of mercury in one type of bone (humerus) would directly disprove the diagenetic contamination hypothesis, as the post-mortem incorporation of Hg from the cinnabar sprayed in the tombs would not 'select' which bones to accumulate in. This has been further discussed in a previous paper by Álvarez-Fernández et al. (2022).

As noted above, only for 28 out of the 65 Valencina individuals was it possible to measure mercury in all three target bones (humerus (H), femur (F) and tibia (T)), totalling 84 THg values. Considering that measurements in different bones are not always available for the same individuals, the results presented in Tables 1 and 2 show the maximum levels recorded for each individual, regardless of the sampled bone. Figure 8 shows the total mercury content according to bone type for those 28 individuals. While femora and tibiae appear to show roughly similar values, the figures for humeri are higher, which was noted by Emslie et al. (2019). To determine whether these apparent differences are statistically significant, various tests were applied. First, a Shapiro–Wilk normality test (< 50 observations) with a 95% significance level, yielded a value of $p > 0.05$ which, entails the rejection of the null hypothesis according to which mercury would be normally distributed in human bones. In addition, non-parametric Mann–Whitney's U significance tests ($p < 0.05$) were calculated for various combinations of bones in that subsample, including H-F, H-T and F-T. Values of $p > 0.05$ ($p = 1$ (H-F), $p = 0.403$ (H-T) and $p = 0.623$ (F-T)) were obtained, and therefore, H_0 stating that no significant difference exists in mercury concentrations among different bones must be accepted. Additional χ^2 tests were used to look at possible statistically significant differences in the distribution of THg in the three target bones, with an α value of $p < 0.05$. While different tests were carried out at different concentration intervals (< 1 µg/g, 1–10 µg/g, 10–100 µg/g, > 100 µg/g; Table 5), no differences were found. Therefore, for this particular group of samples, no statistically significant differences were found in the Mann–Whitney's *U* and χ^2 tests, which, in principle, would run counter to previous assumptions that mercury values in humeri were higher than in femora and tibiae (Emslie et al., 2016, 2019: 7–8). No effect of type of bone has been noted in previous studies (Álvarez-Fernández et al., 2020: 5).

The distribution of mercury in bone tissue has been examined in laboratory studies that suggested higher accumulations in the skull than in humeri and femora (Halffman, 2009). The reason for this may be physiological, such as variations in blood flow, but also 'external', such as differences in the pre-treatment of bone samples. Among the latter, it is worth noting the inclusion of trabecular tissue in skull samples but not in those on long bones, or the elimination process of the 'fatty part' of femora, where mercury could potentially accumulate (Halffman, 2009: 154). The presence of higher mercury values in trabecular versus cortical tissue has also been noted in a study of long bones of Medieval individuals (Rasmussen et al., 2013a). In our study, samples were always taken from cortical bone, and therefore'external' variations can be ruled out to explain potential differences in mercury values across different bones.

Studies investigating the concentration of other metals showed variable concentrations in different bones, but the pattern is also related to the age factor. Thus, in a sample of 134 individuals who had experienced a 'normal' exposure to lead, adults showed higher concentration in their tibiae than in the crania, whereas younger subjects (between 21 and 35 years of age) showed more uniform distributions of the metal across the skeleton (Wittmers et al., 1988). A suggested explanation for this phenomenon is that, in the long term, bones with a higher proportion of cortical tissue, such as tibiae, display higher lead concentrations than bones with a higher proportion of trabecular bone, such as the cranium. This study has important implications for the study of mercury exposure among past societies as the only available samples are bones (normally, unless mummies exist) and underlines the need to examine intra-skeletal variability, while at the same time controlling other biological factors such as age.

In the case of the Valencina population, the subgroup for which measurements in three bone types are available ($n = 28$) is too small. In addition, there is no evidence pertaining to factors such as time, frequency and duration of exposure or, in some cases, age and/or sex of the individual (but see below). Therefore, the analysis of the intra-skeletal variability of mercury at Valencina will have to wait until the sampled population is expanded, and more data are available. But it is important to note that the negative results of the significance test described above does not allow the use of differential pre-mortem rates of accumulation as an argument against potential diagenetic factors. Mercury behaviour in graves is complex, and many different aspects must be taken into account to analyse the interaction between mercury from the bones and the surrounding soil (Álvarez-Fernández et al., 2021, 2022).

Variability by Sex and Age

At Copper Age, Valencina there appears to exist a correlation between high mercury levels and the sex of the individuals, at least in the two main megalithic burials included in this study, namely Structure 10.042–10.049 and the Montelirio tholos, in both of which women display very high THg levels. However, the sample available appears to have a severe demographic bias. While in Structure 10.042–10.049, only one individual, 'The Ivory Lady', was sexed; in Montelirio, all the individuals for whom sex could be established were females, with only one exception. Clearly, the Montelirio tholos did not house a 'natural' representation of the society that built it, as no elderly individuals, children and virtually no men were buried in it. However, it is interesting to note that the only confirmed male subject, a secondary inhumation found in the Main Corridor, showed the lowest mercury values for the whole of the monument (3.82 μg/g). A similar concentration (4.32 μg/g) is recorded for the individual buried outside the megalithic structure, also a male, who, nevertheless dates to the Early Neolithic and therefore cannot be compared with the contingent buried inside the tholos.

For the burials of the PP4-Montelirio sector, sex could only be established for a little over 70% of the individuals involved in this study, including 12 indeterminate, 13 females or possible females and five males (Robles Carrasco et al., 2017). Only two male individuals yielded THg values around 20 μg/g (17.13 and 19.70 μg/g), while the other three showed values ranging between 0.40 and 0.98 μg/g. Among the most exposed individuals (> 10 μg/g), females have much higher THg values than males (40% vs 20%).

As for age, it is worth noting that at Montelirio older individuals tend to have higher THg levels. Individuals with values above 100 μg/g showed ages between 25 and 45 years (except UE114, who was 18–23 at the time of death), while individuals between 10 and 100 μg/g are younger, ranging between 18 and 34 years of age. There could be two reasons for this: (i) older people had greater lifetime exposure and bioaccumulation; (ii) on account of their seniority, older people were assigned tasks more directly connected with the handling, use and/or consumption of cinnabar more often than younger people.

Intra-site Variability

At Valencina, cinnabar has been reported in a small number of structures, including Structures 10.042–10.049 and 10.074 in the PP4-Montelirio sector and the Montelirio tholos. Red pigments, often vaguely referred to as 'ochre', are mentioned in other excavation reports, but there is no analytical evidence indicating that they included cinnabar.[4] And yet, THg values are generally high in human bone for the whole of Valencina. This contrasting evidence (small number of features with cinnabar vs. pervasive presence of mercury in human bone) raises a number of questions; what was the social dimension of cinnabar? Was there

[4] A case in point of particular interest is the tholos-type monument recorded in the early 1990s barely 50 km to the south of Montelirio, often referred to as the Depósito de Agua ('Water Cistern') tholos. Although this monument was wantonly destroyed by order of the Castilleja de Guzmán municipality, a crime never investigated by the heritage authorities, the available photos and descriptions suggest a presence of cinnabar not unlike that of Montelirio (Santana Falcón, 1991).

significant variability of its use across the site? Did the intensity of its usage change through time?

A Mann–Whitney *U* significance test ($p < 0.01$) revealed statistically significant differences in the amount of THg in bone between the individuals buried in the Montelirio tholos and those buried in the neighbouring PP4-Montelirio sector (*p*: 5.4×10^{-15}), and also between those buried in megalithic vs non-megalithic burials (*p*: 1.6×10^{-17}). Departing from the assumption that cinnabar was a high-end commodity, probably only accessible to individuals of certain social standing, these differences support the (otherwise apparent) comparatively higher social status of the group buried in Montelirio and, generally, of those buried in megalithic graves.

In general, when analysing the intra-site variability of cinnabar use and mercury exposure at Valencina, the following can be noted: (i) the values recorded in the individuals buried in Montelirio are the highest for the entire site, while at the same time, in no other burial, a group so large had such consistently high THg values; (ii) the Montelirio contingent was endowed with finely crafted high-end artefacts, in many cases manufactured from foreign raw materials which, within the whole of Valencina, are only matched by those found next to and above 'The Ivory Lady'; (iii) cinnabar has a pervasive presence at Montelirio (in the form of coated and painted slabs as well as sprayed-on bodies and grave-goods), but at the PP4-Montelirio sector, it was only recorded in two features (structures 10.042–10.049 and 10.074); (iv) while Montelirio is a collective burial, the two individuals with the highest THg values (> 100 µg/g) at the PP4-Montelirio sector were buried in single graves; (v) while all individuals buried in Montelirio showed Hg exposure and had therefore experienced some degree of interaction with cinnabar in their lives; at the PP4-Montelirio sector, only one part of the population (45% of individuals) shows Hg exposure (THg > 1 µg/g)]; (vi) the variability of mercury levels within the Montelirio contingent is lower (one order of magnitude) than that of the PP4-Montelirio (two orders of magnitude).

It is also worth noting that the individual most exposed to mercury within the PP4-Montelirio sector (on par with the highest recorded at the Montelirio tholos) was in Structure 10.074, a single inhumation in a simple pit of small size. Persons buried in other graves where scallop shells were also found (namely structures 10.040, 10.012 and 10.080) appear to have very low THg levels. Therefore, neither in terms of the architecture of this grave nor in terms of the grave goods (a scallop shell and some sprayed cinnabar), this individual appears to have a social standing akin to that of 'The Ivory Lady' or those buried in the two chambers of Montelirio.

In this regard, it is also worth remarking that not all individuals exposed to mercury in Valencina were buried in graves where cinnabar was used as part of the funerary ritual. In fact, only in the two features mentioned above (structures 10.042–10.049 and 10.074) was cinnabar observed. The same is found in other Late Neolithic and Copper Age burials across the Iberian south-west. At sites such as Sobreira da Cima, Santa Rita and Perdigões (Tombs I and II), where high Hg values in human bone were detected, cinnabar was found. At the sites Cabeco Arruda and Paimogo I, cinnabar was found in the burials, but the analysis for mercury revealed no exposure. Inversely, at Outeiro 2 and Monte Malheiro 2, cinnabar was not found in the tombs, but some people showed very high mercury values, above > 100 µg/g.

These are important observations for two reasons, first, because they support the hypothesis of substantial pre-mortem exposure to mercury, as opposed to a post-mortem, diagenetic hypothesis—see discussion in Emslie et al. (2019). Bones buried without cinnabar in association cannot have experienced diagenetic absorption of mercury. Second, because they suggest that there was a broad range of factors and circumstances influencing how people interacted with cinnabar. Why were non-elite members of the community, for example the person buried in Structure 10.074 at Valencina, so seriously exposed to a high-end commodity which at that site was only used in elite graves? What was their connection with cinnabar? Were they involved in the extraction (mining) or manipulation of the substance? Undoubtedly, a number of people must have been involved in the procurement and preparation of cinnabar and must have experienced some degree of exposure to it. Alternatively, some people may have used it in a variety of ways (body paint, consumption, etc.) but were not able to use it in their funerals. Indeed, some people appear to have been able to make funerary use of the red powder, although the analysis of their bones revealed no mercury exposure, as suggested at Cabeço Arruda and Paimogo I. Therefore, the patterns of interaction between people and cinnabar must have been complex and organised by numerous principles of varying nature, including, probably, biological (age and sex), social, ethnic and cultural. This topic will be further discussed below.

An additional point of possible relevance is that at Valencina, the graves without cinnabar in which the highest THg levels are observed are collective (between two and nine individuals), whereas THg values observed in individual graves are rather low (between 0.19 and 1.54 μ/g). Were persons buried in individual graves not attached to larger kin or social groups? And, if that was the case, did they have less interaction with the red substance (or were they even 'segregated' from it)?

Finally, there is an important element to consider in terms of the intra-site variation of mercury exposure at Valencina: time. As is shown graphically in Fig. 9, with the exception of three slightly earlier features (structures 10.042–10.049, 10.071 and 10.044), all the Montelirio tholos and PP4-Montelirio individuals analysed here died within a relatively short period of time, between 2900 and 2650 BC (a full discussion of this chronology is available in García Sanjuán et al., in preparation). This is important because this 'short' chronology shows that both the remarkable use of cinnabar and high exposure to mercury this people experienced were part of relatively short-termed social and cultural dynamic, lasting between two and two and a half centuries. Cinnabar 'flowed' extensively during this period in Valencina, as it did in other contemporary sites of south-western Iberia as discussed above and was intensely present in social life, ritual performances and burial ceremonies. It was, in a way, a period of 'cinnabar rush'.

Cinnabar Use

The next obvious question is: how did these high accumulations of mercury occur among the Valencina population? What were the environmental conditions and/or cultural practices that led to what appears to be, on the basis of the currently available evidence, the most intense exposure to mercury in human history, with the

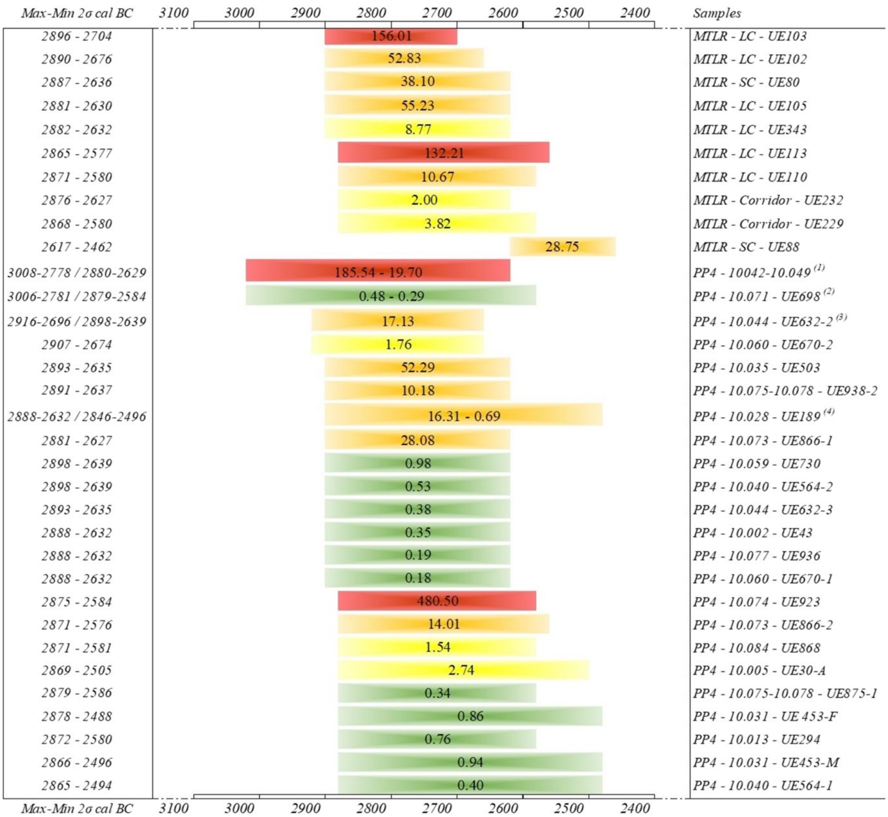

Fig. 9 Synthesis of total mercury levels (μg/g) over time in Valencina

possible exception of Tokushima (Shikoku, Japan), during the sixth–seventh centuries AD (Yamada et al., 1995). Cinnabar is not frequent in nature, which makes it a raw material hard to procure, with a high cost of acquisition and a demonstrable history of use as a high-end commodity. Its bright red colour, of spectacular visual effect, afforded it a high symbolism as a sacred, magic and exclusive material, only accessible to the initiated or to the elites.

In Structure 10.042–10.049 at Valencina, cinnabar was sprayed on the corridor, covering some perforated beads, the body of 'The Ivory Lady' (10.049), and some of the grave goods surrounding her (especially a large almond-rim type ceramic plate of large size placed at her back; García Sanjuán et al., 2019; Rogerio-Candelera et al., 2013). In the remarkable Montelirio monument, cinnabar had a pervasive presence. It was used to coat and paint the large-sized slate slabs that line both corridors and both chambers. Cinnabar-based pigments were sprayed abundantly in the LC, above the bodies and the grave goods, and in the hearth located at its centre; red pigment was also used, perhaps burned (Fernández Flores & García Sanjuán, 2016: 108). Next to the unbaked-clay stela that presided over the LC, three small anthropomorphic figurines also made of unbaked clay had been painted with cinnabar (Bueno Ramírez et al., 2016: 391). For all other Valencina features examined in this study, no cinnabar was reported with the only exception of Structure 10.074, where small amounts of it were noted.

Undoubtedly, the Copper Age population of Valencina attached great symbolic and ritual significance to cinnabar. The individual from Structure 10.074 with very high THg values, who does not appear to be an elite member, may have been involved in the procurement or processing of the raw material. In addition, in Valencina cinnabar was strictly associated with death as no cinnabar has ever been identified in non-funerary features, unlike the site of Gramalote (Peru), dated to the 2nd millennium BC (Prieto et al., 2016), where cinnabar is found in both burial and domestic contexts. Considering the biocidal potential of mercury compounds,[5] which may act against the proliferation of the micro-organisms involved in the decomposition of flesh; cinnabar may have also been aimed at slowing down or counteracting the decay of the bodies or even to achieve partial mummification (Delibes de Castro, 2000: 230; Cervini-Silva et al., 2013, 2018).

Cinnabar does not occur naturally in the surroundings of Valencina and therefore was brought from further afield. The presence of this substance is consistent with the broad spectrum of foreign raw materials that made their way into burial and votive structures at the site (including abundant elephant ivory, a sperm-whale tooth, ostrich eggshells, various types of marine shells, flint, mylonite, rock crystal, amber, variscite and, probably, jadeite). Although initial studies provenanced Valencina cinnabar from the Almadén mining district (Hunt Ortiz & Hurtado Pérez, 2010; Hunt-Ortiz et al., 2011), located some 250 km to the north, later research suggested a more complex picture in which establishing the precise origin of the substance is not straightforward (Rodríguez et al., 2020). In Perdigões, isotopic analysis suggests that cinnabar was brought from Almadén but also from one other unknown source (Emslie et al., 2015). Other cinnabar deposits exist in southern Spain, including Las Alpujarras (Tímar-Cástaras, Granada), about 350 km to the east of Valencina, and Usagre (Badajoz), 120 km to the north-west. There is evidence of the latter being exploited in the sixteenth century AD (Bleiberg, 1984).

Unlike other minerals, such as flint, variscite, or copper, nothing is known about the mining of cinnabar in Neolithic and Copper Age Iberia. The long history of exploitation of the Almadén mercury mines (Gil Bautista, 2013, 2015; Zarzalejos Prieto et al., 2012, Zarzalejos Prieto, 2019) shows the deployment of a substantial workforce. In his twelfth century AD *Description of Spain* geographer Al-Idrisi mentioned over 1000 men working at the Almadén mines to obtain mercury and cinnabar, which were then shipped to all countries of the world (Gil Bautista, 2015: 61). After the mining, there was the problem of transportation from Almadén to Sevilla, across the Sierra Morena range. Since the sixteenth century onwards, the transportation of mercury to Sevilla for its shipment to Peru and Mexico is well-documented.

[5] Their use as biocides has been banned by the European Union since 1 January 2021 because of their negative effects on the environment and human health (UE Regulation 2017/852 of the European Parliament and European Council directive 17 May 2017 on mercury use, which supersedes CE Regulation #1102/2008).

The distance was covered, preferably in April or May, either by oxen-pulled carts, which could carry weights of up to 460 kg between 20 and 30 days, or by mules in 8–10 days (Gil Bautista, 2015: 313). It has been suggested that the presence of several Bronze Age stelae along some of the pathways, tracks and roads used at that time point to their probable prehistoric origin (Gil Bautista, 2015: 58). In fact, the spatial, GIS-based analysis of Late Neolithic and Copper Age megalithic monuments in Sierra Morena does indeed reveal a high degree of correlation with historic pathways (such as drovers), which in turn follow the layout of the most naturally-transitable ('least-cost') routes (Murrieta Flores, 2012; Murrieta Flores et al., 2011).

The variability of THg levels in human bone and the variable occurrence of cinnabar with and without high THg bone levels suggests there were varying degrees of exposure to the substance. This in turn could be explained by diverse forms of interaction between people and the raw material, including its processing and application as well as its consumption. Among the former, several activities were needed: (i) repeated grinding and sieving of the material to obtain nanometric particles which could then be mixed with other red pigments, a labour intensive procedure; both in the Montelirio tholos (Hunt Ortiz & Hurtado Pérez, 2010: 124) and in Structure 10.042–10.049 (Rogerio-Candelera et al., 2013: 286), cinnabar was found to have been mixed with more easily available iron oxides, in all probability with the aim of increasing the amount of material available for use; (ii) mixing of the red powder with natural binders (water, egg yolk, oils, etc.) to create liquid pigments that could be applied as paint on the walls of megalithic burials and on the bodies (presumably of both the dead and the living); (iii) spraying of powder on the burial areas.

People working on the processing of the cinnabar ore into powder would have been constantly exposed to the involuntary inhalation and ingestion of the fine dust, which would have also fallen on foodstuffs and on the surfaces of surrounding living and working areas, likely causing it to enter the food chain (herbivores eating grass, etc.). Indeed, if cinnabar was more or less frequently used for bodily decoration, some people must have been exposed to mercury through the skin for substantial lengths of time or even constantly. Heating cinnabar powder, as is done even today in some regions of the world to elaborate medicinal drugs (Gerke, 2021), would have produced mercury vapours which, if inhaled, would have led to immediate health issues (alternations of the nervous system and sensory organs, etc.). If transported by the wind, these vapours could have transported mercury towards the surrounding environment, where it would have been easily deposited (Poulin & Gibb, 2008), leading to ulterior forms of exposure—see discussion below.

In addition, it is likely that some 'special' people intentionally consumed cinnabar (or products that included it), either via inhalation of the vapours produced after heating it, or via direct ingestion (for example in liquid, or semi liquid form). There are several cross-cultural examples of the consumption of mercury and associated compounds. Cinnabar powder has been used until recently in Chinese and Indian traditional medicine and is still used today (Huang et al., 2007, 2012; Liu et al., 2008, 2018). Approximately 40 traditional Chinese drugs contain cinnabar (Liu et al., 2008, 2018: 3). In recent years, advances have been made in the understanding of the molecular-level mechanisms underlying the pharmacological and toxicological effects of cinnabar, which is of key importance to distinguish therapeutic from toxic doses (Lu et al., 2020; Yang et al., 2020). In the Copper Age, the consumption of cinnabar by a group of initiated or special people would have been made on the belief that cinnabar had healing and/or magic properties. Indeed, it is quite possible that certain effects caused by the exposure to the substance (such as shakes, convulsions, loss of consciousness) that are akin to those verified in trance states were sought-after. As mentioned above, the group of women buried in the LC of Montelirio display some very specific biological and cultural traits suggesting they were religious specialists.

However, even if some specific forms of use were restricted to a small group of people, at some point, cinnabar powder must have been, on account of its processing and manipulation, a relatively common, even pervasive occurrence at Valencina not just in the domain of the dead, but also in daily lives of the people living in and frequenting the site. Having said that, it is worth noting that at Valencina, no cinnabar powder has ever been recorded in non-funerary features.

Environmental Exposure

The discussion made above suggests two major forms of interaction with cinnabar at Copper Age Valencina: processing and consumption. By their very nature, both of them must have affected a small number of people. And yet, the presence of mercury in human bone is quite pervasive, to which it must be added that it is also found in animal bone and in the only soil sample analysed to date. This suggests a possible third vector of interaction: the environment.

Today, mercury occurs naturally in the environment, mainly on account of volcanic eruptions and because of several thousand years of human activity (Sundseth et al., 2017). Mercury presence is also quite persistent. Most of the mercury found in Earth's atmosphere today (up to 60%) is anthropogenic Hg 'inherited' from re-emissions from surface deposits, while natural emissions and primary anthropogenic emissions account for 13% and 27%, respectively (Amos et al., 2013). Studies carried out in Iceland show the accumulation of mercury in the bones of individuals living in volcanic environments (Walser et al., 2019). A recent study (Freire et al., 2010) found that the infant population of present-day Granada (Spain) shows higher mercury exposure (1–2 μg/g MeHg in hair) than populations from more industrially polluted areas of northern Spain and Europe. The explanation for this, it has been suggested, could lay in fish consumption through the incorporation of environmental mercury to water and subsequent entry in the trophic chain (Freire et al., 2010: 100–101).

Upon its deposition from the atmosphere, aquatic organisms biotransform environmental mercury into organic mercury (methylmercury, MeHg), a more easily available form that is incorporated into the trophic chain through water and food, mainly fish and molluscs. Most organic mercury compounds are easily absorbed through the lungs, gastrointestinal tract or skin. Exposure to MeHg can have various types of effects on the central nervous system, including motor and sensory alterations, visual impairment, and migraines, some of which may disappear if the exposure is discontinued. Severe effects include lasting cognitive damage: a linear relationship has been established between the loss of 0.18 IQ points and a 1 μg/g increase of mercury in hair (Axelrad et al., 2007).

Obviously, in the Copper Age there was very little or no history of previous anthropogenic mercury in the environment, so one relevant question is, were there significant levels of naturally occurring environmental mercury in the Valencina region during the 3rd millennium BC? Isotopic data suggest that the diet of the Valencina population studied here was not rich in marine resources (Fontanals Coll et al., 2016: 446). Basically, the same applies to other Copper Age sites for which data are available (Beck et al., 2018: 37). Although the real extent of fish and seafood consumption during the Copper Age is yet to be established through more robust sampling and a broader examination at the evidence, mercury intake through that vector appears to have been low. For the four individuals of Perdigões and Sobreira da Cima for whom the proportion of MeHg (CH_3Hg) with regards to THg could be established, the resulting values were lower than 0.05% (Emslie et al., 2015: 6). This suggests that virtually all measured mercury is in inorganic form, again ruling out large intakes through diet.

Other sources of environmental mercury must be considered. Mining unrelated to cinnabar (for example copper) was important in the Copper Age. It has been suggested that early copper mining in the Huelva 'pyritic belt', barely 30 km to the West of Valencina as the crow flies, may have caused significant environmental pollution at distances of up to 100 km from the mines (Leblanc et al., 2000). Long-term atmospheric data do not suggest that lead levels in the Copper Age were significantly higher than those of the pre-metallurgical era (García-Alix et al., 2013), but copper mining and smelting activity may have caused some small-scale impact on the environment in which the Valencina population was living.

Could the numerous cases of medium-to-high levels of THg in human bone at Valencina be explained through environmental mercury? This could be potentially relevant for individuals with bone concentrations around 1 μg/g in whose burials no remains of cinnabar were observed. This explanation could apply to the Neolithic individual found in the Montelirio mound (pre-dating the construction of the tholos by almost 2000 years), who yielded THg values of 4.32 μg/g, or to the Roman-time burials of the PP4-Montelirio sector, with average THg values of 0.32 μg/g and a maximum of 1.32 μg/g. Indeed, environmental mercury was invoked to explain the presence of medium-to-high THg levels at the necropolis of La Lanzada (Galicia) (Álvarez-Fernández et al., 2020; López-Costas et al., 2020). Environmental mercury, whether because of natural occurrence, mining of other metals in the 'pyritic belt' or deposition caused in the surrounding area of the site by the processing of cinnabar, could also explain the medium-to-high THg levels found in animal bones at Valencina, which are higher than the only comparable sample, dating to the Bronze Age (Logemann et al., 1994), as well as those found in soil samples from the Pabellón Cubierto (14.5 μg/g) where no cinnabar was reported. Future studies will have to place more emphasis on animal and soil samples to clarify the extent of mercury in the environment of 3rd-millennium BC Valencina.

Altogether, although environmental mercury may have played a part in the medium-to-low mercury levels found in the bones of most of the Valencina Copper Age population, the highest levels must have occurred in connection with specific cultural practices as described above.

Health Effects

Whether through exposure to environmental mercury, involuntary inhalation/ingestion while grinding or processing cinnabar, or because of its deliberate consumption (with medicinal or ritual/magic purposes), it may be assumed that the population living at Valencina in what was the peak period of what we have termed here the 'cinnabar rush' (c. 2900–2650 BC) suffered, to a variable degree, the effects of mercury on human health. Regrettably, the data on total mercury in human bone currently available do not permit inferences regarding the means of exposure or what specific form of mercury the exposure was derived. The toxicology of mercury is very complex, and each of its chemical forms can cause different health effects (Zahir et al., 2005; ATSDR, 1999, WHO, 2007; Risher, 2003; Poulin & Gibb, 2008). It seems obvious, however, that the pervasively high THg levels recorded across the population would have implied significant health effects, which in some cases may have been severe and even lethal (ICSC, 2019).

The main pathway of mercury into the human body is through the inhalation of elemental mercury vapours (Hg^0), approximately 80% of which are absorbed via the respiratory tract, spreading to the entire body. The most affected organs are the brain and the kidneys, leading to damage in the nervous, respiratory, immune and digestive systems (Liu et al., 2008). Elemental mercury can reach the brain simply via the nasal cavity (Park & Zhen, 2012). Individuals exposed this way can suffer numerous health problems, including neurological (tremors, insomnia, memory loss, neuromuscular damage, headaches, loss of reflexes, polyneuropathy and loss of cognitive and motor functions), respiratory (cough, asthma, chest pain) and cardiac (increased blood pressure and palpitations) (Barregard et al., 1990; Boffetta et al., 2001; Kobal et al., 2004). Children inhaling mercury vapours can develop a skin condition known as acrodynia (Liu et al., 2018: 6). While some of those problems can disappear upon termination of the exposure, any increase in the duration and concentration of the exposure will lead to more severe, even irreversible damage.

Written records produced in connection with the mining of cinnabar at Almadén from the sixteenth century AD onwards provide substantial evidence of the consequences that continued inhalation of mercury vapours and other toxic substances involved in the metallurgical process, such as sulphur dioxide (SO_2), had for the miners (Almansa Rodríguez et al., 2011; Gil Bautista, 2013; Menéndez Navarro, 1993, 2012). Those included uncontrollable shaking, excessive salivation (ptyalism) and 'somnolence', which could lead to dementia. At times, the tremors were so intense and prolonged (over months) that some individuals were tied to their beds. In Mateo Aleman's *Informe Secreto* ('Secret Report'), written in 1593 AD after his tour to the mines, he stated that many of the miners interviewed by him were 'shaky, silly and out of their wits' on account on their work in the 'firing' of the minerals (Gil Bautista, 2015: 239). It is worth noting that those conditions were not always lethal and depended largely on the duration of the exposure. Between 1841 and 1850, out of 7748 Almadén miners who developed poisoning symptoms, only 7% died (Hernández Sobrino, 2007: 355), although most of them saw their life expectancy reduced by as much as one quarter (Hernández Sobrino, 2007: 360). Mercury exposure was treated through 'sweating' in sauna-like chambers that were specifically designed for that purpose (Hernández Sobrino, 2007: 371).

Anthropological studies focusing on the contemporary processing of cinnabar for medicinal purposes in Tibet mention some of the health effects observed in the individuals involved (Gerke, 2021), describing how cinnabar processing practices are still carried out in an 'artisanal' way for medicinal purposes. During the process, individuals are exposed to both mercury and sulphur vapours, also present in the combustion. The persons who carry out this manipulation have been doing it for years and they know well the process and its empirical variables. They suffer chronic exposure since this preparation is usually done once or twice a year. Unfortunately, the levels of mercury in the body of these people have not been analysed. On the other hand, it is also evident that people outside the mineral heating process, who circumstantially are relatively close to the source of emission, immediately perceive the external symptoms such as irritation and coughing when the cinnabar vapours are released (Gerke, 2021: 205). Instead, in the Caribbean (Wendroff, 2005) and Africa (Street et al., 2015), cinnabar, or even metal mercury in USA (Riley et al., 2001), is heated and its vapours inhaled deliberately as the neurological and motor effects (tremors) are sought-after as part of rituals involving trances, divination and revelations.

In addition, the manipulation and use (for example as body paint) of cinnabar powder leads to exposure to inorganic mercury (Hg^{2+}), which can cause irritation of the skin and eyes. Upon ingestion, it will have a bioavailability of between 7 and 15% (Park & Zhen, 2012), the kidney being the critical target organ and causing gastrointestinal symptoms (EFSA Panel on Contaminants in the Food Chain (CONTAM), 2012). Inorganic salts are absorbed by the skin through the transportation of mercury in the epidermis and sweat glands, sebaceous glands and hair follicles, causing adverse renal effects (Chan et al., 2020) as well as neurological damage (Weldon et al., 2000).

Undoubtedly, the Valencina population studied here must have suffered some, if not most, of the symptoms described above. The exceptional concentrations of mercury found in the bones of the women buried in the Montelirio LC, 'The Ivory Lady' and the person buried in Structure 10.074 may have resulted from inhalation, whether accidental or deliberate, of mercury vapours upon the heating of cinnabar. The case of the Montelirio women is particularly intriguing. It has been suggested that these women were religious specialists in charge of a famous sanctuary or temple, which would partly explain the high degree of connectivity seen in the Valencina mega-site (with exotic raw materials coming from long distance, and a high percentage on non-local individuals) and the scale and originality of some of its monuments (García Sanjuán et al., 2016: 547). This characterisation of the Montelirio women is partly based on their health condition which, as well as the very high mercury exposure included as noted above, one case of polydactyly (to our knowledge, the earliest documented in Europe to this date), and degenerative problems (arthrosis in the dorsal vertebrae, shoulders and lower limbs) caused by ageing or functional wear. But it is also based on other kinds of evidence, such as their 'choreographed' arrangement around the stela that presided over the LC and the impressive beaded attires decorated with pendants made of ivory and amber they wore when they were buried. The neuro-cognitive, motor and behavioural disorders caused by mercury poisoning, in many respects not unlike those caused by hallucinogenic drugs, may have been a sought-after feature among a highly specialised group of people involved in mystic practices, occasional, carefully staged and dramatic ritual appearances and, more generally, political governance. The consumption of cinnabar as a magic or sacred substance would have made sense in the context of ritual performances involving divinations, predictions and oracles, as was the case in famous ancient sanctuaries such as Dodona in Greece (Parke, 1977) or Chavín de Huántar in Perú (Rick, 2017). It is important to note that those sanctuaries had religious as well as political significance and acted as central places attracting major congregations of people, much as may have occurred at Valencina in the Copper Age (Stanish et al., forthcoming).

Even if the period of prevalence of such an institution was relatively short (as the radiocarbon chronology appears to suggest), and the number of people devoted to such 'extreme' specialized religious practices was small, the processing of cinnabar would have demanded a relatively significant workforce devoted to the procurement (mining and transportation), processing and manipulation of the ore, including thoroughly grinding it, mixing and sieving it (perhaps also heating it) and, eventually, applying it onto the specified surfaces (megalithic walls and sacred artefacts) and spraying it on specific locations. This would have led to exposure by a substantially higher number of people. Indeed, the fine cinnabar dust would have been carried around the workplaces, sacred spaces and monuments, causing it to settle on the surrounding soils and vegetation, subsequently entering the food chain. Leaving aside its possible use as body paint (which cannot be ruled out either), which would not have been as toxic and damaging as inhalation (Fornaciari et al., 2022: 46), the inevitable inhalation and ingestion that cinnabar processing would have affected an even wider segment of the Valencina Copper Age population, who, most likely, developed some of the ailments described in detail in the historical sources known for the Almadén mines.

Corollary

The study presented in this paper is based on the largest sampling of mercury in prehistoric human bone ever attained. The main aim was to examine the patterns of mercury exposure and cinnabar use at the Copper Age mega-site of Valencina. The main conclusions can be outlined as follows.

Valencina was a central place of great religious, social and political significance over several centuries in the 3rd millennium BC. Cinnabar was used intensively and in a variety of ways, although largely restricted to very special monuments such as the grave of 'The Ivory Lady' or the Montelirio tholos. Extensive cinnabar use is also documented at other Copper Age sites in Iberia, particularly in the south-western quadrant.

At Valencina, cinnabar use appears to have been particularly intense in the period c. 2900–2650 BC, which is the first of the two major phases of development that the site appears to have experienced, according to a recent radiocarbon analysis (García Sanjuán et al., 2018a). In this phase, Valencina witnessed intense monumentalism as part of sophisticated ritual and burial practices often involving finely crafted artefacts manufactured in exotic raw materials. In the second phase (c. 2600–2350 BC), the use of cinnabar appears to have been far less intense.

Cinnabar use at Valencina between the twenty-ninth and twenty-seventh centuries BC appears to have caused a significant exposure of its population to mercury. Various types of exposure can be discerned. The first would have affected a rather small group of religious specialists and elite members who perhaps inhaled or consumed cinnabar as part of their lifestyle. The second exposure would have affected a larger contingent of people, but still small, involved in the procurement and processing of cinnabar for the designated uses. A third kind of exposure, affecting a much larger group of people (perhaps the entire population inhabiting or frequenting Valencina), would have been indirect, resulting either from the presence of mercury in the natural environment or by the pollution caused by the sustained manipulation of cinnabar over several decades. This third type of exposure would appear to have also affected wild and domestic animals living at or around the site.

Altogether, the data compiled for this study reveal that Copper Age Valencina witnessed one of the most dramatic cases of human exposure to mercury known in human history. On-going research expanding on the type of samples analyzed, with more soil and faunal samples from a wider range of sites (both functionally and chronologically), and also including experimental work, will contribute to a more robust assessment of the conclusions reached here, and, more generally, to increase our knowledge of the relationship that human beings have established over several thousand years with one of the most complex and fascinating natural substances.

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Data Availability The primary dataset used in this study is included as Supplementary Material.

Declarations

Competing Interests The authors declare no competing interests of a financial or personal nature.

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