



# Use of cinnabar in funerary practices in the Central Pyrenees. Analysis of pigments on bones from the prehistoric burial of the *Cueva de la Sierra* cave in Campodarbe (Huesca, Spain)

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## ARTICLE INFO

### Keywords:

Archaeometry  
Burial caves  
Cinnabar  
Ebro Valley  
Funerary practices  
Late Neolithic/Chalcolithic  
Northeastern Iberian Peninsula  
Sulfur isotope analysis

## ABSTRACT

The appearance of red pigments in prehistoric burial sites is frequent. In the study presented herein, the composition and provenance of the pigment sprinkled in a burial dated between the Late Neolithic and Early Chalcolithic periods in the *Cueva de la Sierra*, in Campodarbe (Huesca, Spain), were characterized through X-ray fluorescence spectroscopy, scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy, Raman spectroscopy, X-ray powder diffraction, and sulfur ( $\delta^{34}\text{S}$ ) isotope analysis. The chemical composition of the pigment used in the burial could be unequivocally established as cinnabar, and the S isotope signature pointed to an origin in the ores of the Northwestern Iberian Peninsula, ca. 500 km far from the burial site, suggesting an alternative provenance to the Almadén outcrop, the main cinnabar source in the Iberian Peninsula. The presence of cinnabar in the Ebro valley, on the southern slope of the Central Pyrenees, forces us to rethink the complex processes of interaction between communities that populated the north of the Iberian Peninsula during the final moments of the 4th millennium BC.

## 1. Introduction

Between Campodarbe and Boltaña, on the so-called ‘Coasta’ hill, next to the ‘Pista de Los Rojos’ trail, is located the ‘Cueva de la Sierra’ cave, also known as the ‘Cueva de la Basa’ cave, at 1040 m above sea level (UTM coordinates: 31T 256310E 4701763N). Access from both villages is signposted and much of the path is marked for hiking and cross-country cycling (Fig. 1a,b).

The cave is cataloged in the inventory of the General Directorate of Heritage of the Government of Aragón with reference number 1-ARQ-HUE-003-051-026. Archaeological materials were discovered in the

1970s during successive visits by J. Gracia, A. Castán, V. Baldellou, and J.I. Lorenzo, who collected numerous ceramic and fauna fragments from the entrance and a series of human remains located in an inner chamber (Castán Sarasa, 2000). The findings were deposited at the Huesca Museum, where we were able to access them (Rodanés Vicente, et al., 2016). Subsequently, between 2016 and 2021, successive visits were made to the cave to confirm their provenance, contrast the oral or written information that had been transmitted to us, and take samples for dating and analysis. Thus, with this background, we herein propose a hypothesis of use of the cavity that best fits the available information, being aware of the great loss that has occurred due to the regular visits of

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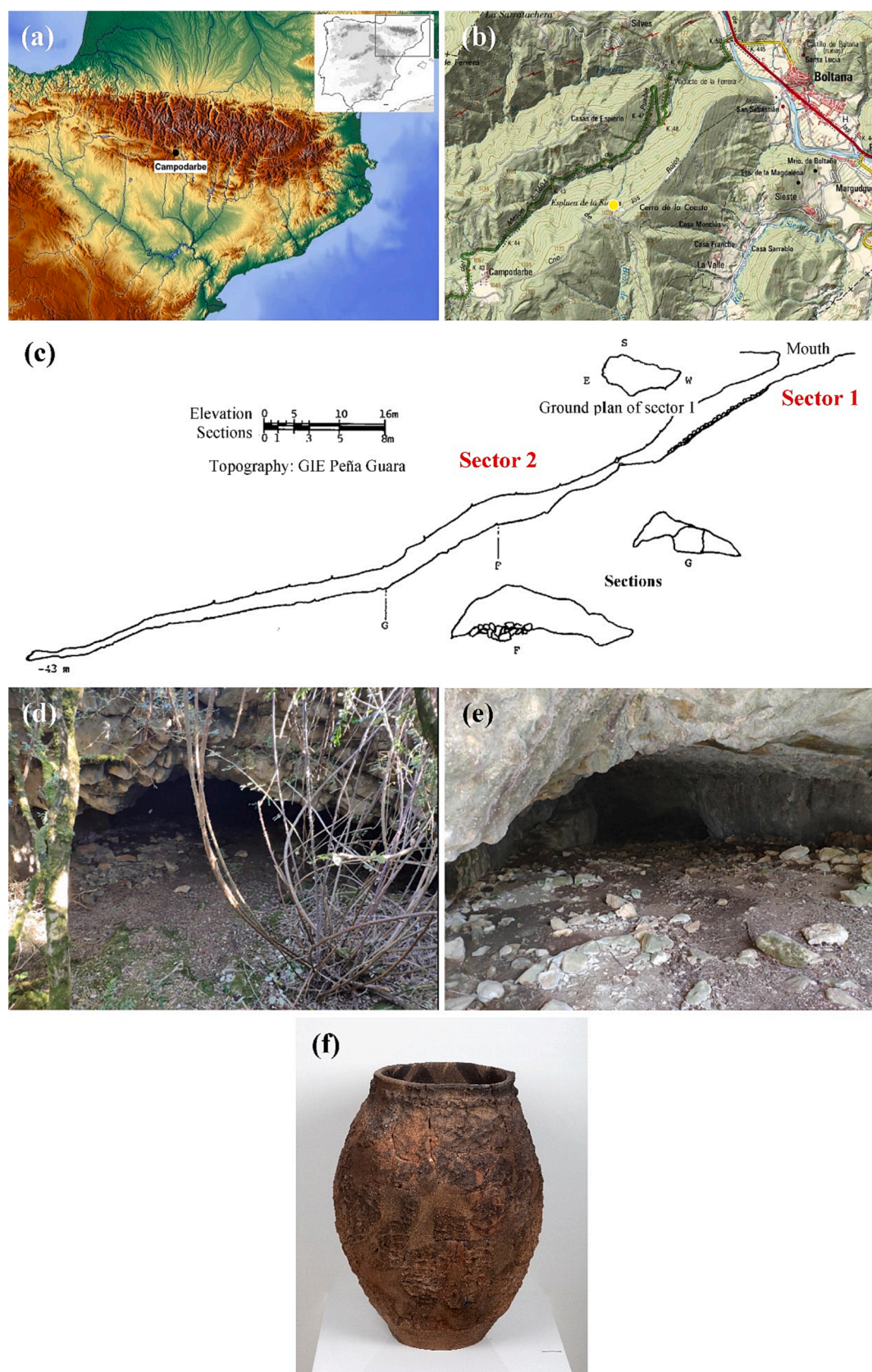
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<https://doi.org/10.1016/j.jasrep.2023.103849>

Received 22 October 2022; Received in revised form 15 January 2023; Accepted 15 January 2023

Available online 21 January 2023

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**Fig. 1.** (a,b) Location of the cave; (c) plan of the cave with the different sectors; (d) mouth of the cave; (e) entrance chamber (sector 1); (f) restored vessel from sector 1 (Photo: F. Alvira, Museum of Huesca).

hikers and caving enthusiasts who have passed through this unique site that has lacked adequate protection by the authorities.

The archaeological remains were located in two clearly differentiated areas (Fig. 1c). Sector 1 represents the entrance to the cave (Fig. 1d, e), where a small retaining wall was documented. Due to the steep slope, the first few meters have accumulated a large number of stones from the exterior. Here, and on the floor of this first room, numerous ceramic fragments were collected, characterized in some cases by smooth surfaces, with the presence of marked carination, together with others with rough surfaces with plastic applications from large vessels (Fig. 1f). The typology, decoration, and workmanship are remarkably similar to those found in many caves in the Pyrenees and Pre-Pyrenees, the most significant representation of which can be found in the excavated levels of the lower chamber of the 'Moro de Olvena' cave (Huesca), with a precise chronology corresponding to the Early and Middle Bronze Age (Ramón Fernández and Rodanés Vicente, 1996; Rodanés Vicente, 1992a). The

stratigraphic layout and the presence of ashes together with faunal remains allow us to hypothesize that it was a place of habitat, possibly occasional or seasonal, although further chronocultural precision is not possible due to the absence of systematic excavations.

Sector 2 covers an interior space that can be accessed from the entrance chamber through a narrow corridor. Here, in a room about 25 m wide by 7 m high, marked by large blocks detached from the ceiling, human bones were located in a small diverticulum along with the remains of a red dye that was undoubtedly related to funerary practices. Most of these bones have disappeared. Only a meager sample is preserved in the Museum of Huesca, included and studied in the doctoral thesis of Lorenzo Lizalde (1994), and some last vestiges that we were still able to collect in 2021, together with new samples of pigment that have been analyzed and discussed in this article.

The remains all come from the same diverticulum of the interior room, grouped in a small hollow at floor level (Fig. 2a-d). Their



**Fig. 2.** (a) Interior room (sector 2); (b-d) place from where the last remains were collected; (e) red-powder sprinkled soil sample; (f) red-stained human bones.

condition has substantially deteriorated as a result of being in an environment of high humidity and exposed to sediment with a large presence of calcium salts. They show a coloration between maroon and pinkish spread all over their surface (Fig. 2f). These are the preserved remains of a burial that the oral tradition of ancient visitors to Boltaña described as an “intact skeleton in a seated position with the legs extended and the back leaning against the right wall of the room” [tr.] (Castán Sarasa, 2000), which is unusual, although a Bronze Age skeleton sitting in an upright position was also reported in Humanejos (Madrid, Spain) (Herrero-Corral, et al., 2020).

At present, 18 human bones are preserved. Most of them come from the feet, among them a left talus, right and left calcaneus; three bones from the hand (left, fourth metacarpal; right, first phalanx of the fourth finger, right, first phalanx of the fifth finger), five fragments of ribs and a left eighth rib; and 3 vertebrae (two dorsal and a lumbar). Duplicate bones have not been identified, so they may belong to a single individual. They correspond to a gracile adult compatible with a female, although a rib and a lumbar vertebra point to a greater possibility that it is a male subject.

Except for the dye that is detected in circular accumulations, and is still appreciable (Fig. 2b-e), no element that indicates the existence of grave goods or the slightest vestige of artificial structures, as suggested at the time by Castán Sarasa (2000), has been found.

With these data, we propose the hypothesis, confirmed in other sites of the Ebro Valley, that it is an individual burial (Lorenzo Lizalde, 1994) in a mixed (b)-type funerary cave in which habitat and burial functions have been documented in different parts of the cave (Rodanés Vicente and Martínez Flórez, 1999).

In the opinion of Castán Sarasa (2000), the first phase of the ritual, coinciding with the defleshing of the corpse, would have taken place beforehand, possibly outside the cavity, later moving to the interior, where the impregnation of the bones with the red dye would have taken place. This would imply the acceptance of its secondary character, an assertion that is always difficult to demonstrate. However, this hypothesis cannot be ruled out, given that the process, although not the most common, is not exceptional and is sufficiently well-known in the Ebro valley and the Mediterranean coast (Rodanés Vicente and Martínez Flórez, 1999).

The selective preservation and arrangement of the bones in a small space where the entire skeleton could not fit, and especially the fact that they are dyed over their entire surface -including the inner part of the joints- allows us to suppose that the pigment may have been applied homogeneously because at this time the corpse was already clean and free of viscera and tissues. Such homogeneity in the distribution and conservation of the dye is difficult to achieve. If the impregnation takes place directly on the corpse, the opposite occurs: the accumulation of pigment is more random.

The absolute dating of one of the intensely colored human bones offered a date of  $4400 \pm 30$  BP (Beta-332608 in Table 1), which allows us to frame the burial during the final moments of the Neolithic and the beginnings of the Chalcolithic, a poorly known period in Aragon. We only find arguments to delimit the period and maintain its existence based on absolute dates from funerary contexts. Table 1 includes as a reference a series of shelters and caves with burials in Northern Aragón that cover more than half a millennium and that could have been contemporary at some point in time with the burial we are studying (Rodanés Vicente, 1992a; Rodanés Vicente et al., 2016; Nasarre Ortín et al., 2022; Rodanés Vicente and Lorenzo Lizalde, 2014).

## 2. Methods

### 2.1. Multielement analyses

Multielement analyses of red-stained bones and the red-sprinkled soil were performed *in situ* by X-ray fluorescence spectroscopy (XRF). A NITON XL3t GOLDD+ (Thermo Scientific, Waltham, MA, USA)

**Table 1**

Radiocarbon dating of other burial caves in Northern Aragón (Spain).

Archaeological site	Lab reference	Dating (BP)	2 $\sigma$ ranges* [start:end] relative area
Foz de Escalete 2	BETA-332606	4730 $\pm$ 30	[cal BC 3630: cal BC 3554] 0.411819
(Riglos)			[cal BC 3539: cal BC 3495] 0.231781
			[cal BC 3454: cal BC 3377] 0.3564
San Juan	GRA-38270	4620 $\pm$ 30	[cal BC 3513: cal BC 3426] 0.698115
(Loarre)			[cal BC 3409: cal BC 3396] 0.019549
			[cal BC 3383: cal BC 3347] 0.282335
Rambla de Legunova	GRA-24746	4545 $\pm$ 45	[cal BC 3486: cal BC 3472] 0.016486
(Biel)			[cal BC 3372: cal BC 3097] 0.983514
Trocs III	MAMS-16167	4512 $\pm$ 25	[cal BC 3353: cal BC 3264] 0.308748
(Bisaurri)			[cal BC 3246: cal BC 3101] 0.691252
Cueva de la Sierra	BETA-332608	4400 $\pm$ 30	[cal BC 3281: cal BC 3281] 0.000609
(Campodarbe)			[cal BC 3264: cal BC 3244] 0.022559
			[cal BC 3102: cal BC 2913] 0.976832
Cueva de los Cristales	GRA-38061	4370 $\pm$ 30	[cal BC 3089: cal BC 3056] 0.102989
(Sarsa de Surta)			[cal BC 3032: cal BC 2907] 0.897011
Forcas II	BETA-281899	4330 $\pm$ 40	[cal BC 3080: cal BC 3062] 0.03439
(Graus)			[cal BC 3028: cal BC 2886] 0.96561
Cueva de los Cristales	GRA-38062	4125 $\pm$ 30	[cal BC 2868: cal BC 2802] 0.286769
(Sarsa de Surta)			[cal BC 2776: cal BC 2614] 0.629257
			[cal BC 2613: cal BC 2580] 0.083974
San Juan	GRA-38268	4120 $\pm$ 30	[cal BC 2868: cal BC 2802] 0.279679
(Loarre)			[cal BC 2775: cal BC 2713] 0.195491
			[cal BC 2709: cal BC 2577] 0.52483
San Juan	GRA-3795	4110 $\pm$ 30	[cal BC 2867: cal BC 2802] 0.260841
(Loarre)			[cal BC 2774: cal BC 2714] 0.155074
			[cal BC 2708: cal BC 2573] 0.584086
Cueva Dróllica	GRA-38063	4105 $\pm$ 30	[cal BC 2865: cal BC 2803] 0.248122
(Sarsa de Surta)			[cal BC 2765: cal BC 2716] 0.127536
			[cal BC 2706: cal BC 2572] 0.610989
			[cal BC 2513: cal BC 2503] 0.013353
Plana de la Marquesa	BETA-598239	4080 $\pm$ 30	[cal BC 2855: cal BC 2808] 0.153058
(Uncastillo)			[cal BC 2749: cal BC 2723] 0.047988
			[cal BC 2700: cal BC 2564] 0.685815
			[cal BC 2534: cal BC 2494] 0.113138
Cueva Dróllica	GRA-33935	4000 $\pm$ 35	[cal BC 2622: cal BC 2595] 0.031727
(Sarsa de Surta)			[cal BC 2584: cal BC 2459] 0.968273

\* Calculated using CALIB radiocarbon calibration program (Stuiver and Reimer, 1993), and intcal20.14c calibration data set (Reimer, et al., 2020).

portable system, equipped with an Ag anode X-ray tube (6–50 kV, 0–200  $\mu$ A max) and a Geometrically Optimized Large Area Drift Detector (GOLDD), was used. The analyses were run with factory built-in standards. The beam size of the instrument was  $\sim 0.5$  cm<sup>2</sup> and a camera visualized the analyzed spot.

The multielement composition of the red pigment present on the bones was further analyzed by scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDX), using an EVO HD 25 (Carl Zeiss, Oberkochen, Germany) apparatus.

## 2.2. Raman spectroscopy

A high-resolution Horiba-Jobin Yvon LABRAM HR 800 UV Raman spectrometer, equipped with an Olympus BX41 microscope and a Symphony CCD detector, was used for the acquisition of Raman spectra from the surface of the same red-stained bone studied by SEM-EDS (microscopic backscattering mode with x15UV objective from Optics for Research). The  $\lambda = 325$  nm line of a He-Cd laser was used. The nominal laser power on the sample was 1 mW, with an approximate irradiance of 100 kW·cm<sup>-2</sup>. The acquisition time was set at 180 s with 10 accumulations, and the spectral slit aperture at 200  $\mu$ m, obtaining a spectral resolution better than 1 cm<sup>-1</sup>. The equipment was calibrated with the  $\nu(\text{Si-Si})$  mode at 520.7 cm<sup>-1</sup> of crystalline silicon from a bare (001) wafer.

## 2.3. Sulfur isotope analyses

The  $\delta^{34}\text{S}$  value (i.e.,  $^{34}\text{S}/^{32}\text{S}$ ) indicates the equivalence ratio between the red pigment sample found in the burial and the known reference standard, Vienna Cañón Diablo Troilite. It was determined according to the methodology described in Takahashi, et al. (2018) and Minami, et al. (2019), and was measured with the same equipment and conditions used in previous studies (Tsantini, et al., 2021, Tsantini, et al., 2018) to facilitate comparisons with a database of previously reported Iberian cinnabar isotope values. The standards mostly used in the laboratory are the IAEA-SO-5, IAEA-SO-6, and NBS-127 standard materials (+0.3, -34.1, and + 20.3 ‰, respectively), and reproducibility values are better than 3 ‰.

Red pigment samples for analysis were separated from other materials under a stereomicroscope and collected by gently pressing a sulfur-free polyester adhesive tape. A piece of folded tape was cut out and placed in an aluminum boat, which was introduced into a Vario ISOTOPE select (Elementar Analysensysteme GmbH, Langenselbold, Germany) equipped with a custom cryo-focusing device from Isoprime Trace gas (Elementar UK Ltd., Stockport, UK), and coupled to an IsoPrime100 (Elementar UK Ltd.) isotope ratio mass spectrometer (IRMS).

## 3. Results

### 3.1. Multielement analyses

#### 3.1.1. X-ray fluorescence analyses

The composition of two of the red-stained bones from sector 2 (Table 2) was compatible with that of bones in a mineral environment (quartz, aluminosilicates, and minimal amounts of sulfates). Calcium and phosphorus concentrations and Ca/P ratios were similar to those reported for contemporary human bones (19.3 %, 8.4 %, and 2.3, respectively) by Tzaphlidou and Zaichick (2003). The presence of mercury was appreciable and compatible with the presence of mercury

sulfide (vermilion). The presence of high amounts of HgS was also detected in the burial soil sprinkled with red pigment (Fig. 2e), whose composition is presented in Table 3.

#### 3.1.2. SEM image and elemental mapping

Given that the results of the elemental analysis obtained by pXRF (Table 2) were highly influenced by the underlying bone composition, an SEM-EDX study at low electron high tension (5 kV, to reduce the penetration depth of the X-ray beam) was also carried out to obtain more accurate information on the composition of the surface of the red-stained bones. This technique indicated the presence of significant amounts of mercuric sulfide, aluminosilicates, and quartz, mineral phases that are not part of the composition of bones (Fig. 3).

### 3.2. Raman spectrum

The Raman spectrum of the surface of the red-stained bone is shown in Fig. 4. The peak located at around 250 cm<sup>-1</sup> is characteristic of mercury sulfide and is attributed to the A1 stretching vibration of the Hg-S bonds of the vermilion mineral phase. The strong and well-defined peak at 315 cm<sup>-1</sup> is assigned to  $\nu_2$  PO<sub>4</sub><sup>3-</sup> bending mode (from bone hydroxyapatite). The strong peak at 458 cm<sup>-1</sup>, corresponding to the A1 symmetric  $\nu(\text{Si-O})$  stretching, appears alongside bands of medium intensities at 755 cm<sup>-1</sup> (Si-O-Si stretching) and 1079 cm<sup>-1</sup> (Si-O-(Si) stretching vibration), all from mineral components in a siliceous environment. The band at 1079 cm<sup>-1</sup> can be attributed to  $\nu_1$  symmetric stretching of CO<sub>3</sub> (bone carbonated hydroxyapatite).

### 3.3. X-ray powder diffraction analysis

A comparison between the X-ray powder diffraction patterns of the red-stained and non-stained regions of the bone (Fig. 5a and b, respectively) provided further evidence of the presence of cinnabar, confirming the material phase ( $\alpha$ -HgS, with P3<sub>2</sub>21 (154) space group) because of the presence of the characteristic reflections at 26.5° (101), 28.2° (003), 31.2° (012), 43.6° (110), 45.8° (104), 51.8° (021) and 52.7° (113). Other peaks correspond to hydroxyapatite and quartz.

### 3.4. Sulfur isotope analysis

Based on a comparison of the  $\delta^{34}\text{S}$  value for the cinnabar sample from the burial with the sulfur isotope compositions of cinnabar samples from different mining districts in the Iberian Peninsula (Table S1) (Domínguez-Bella, 2010, Higuera, et al., 1998, Jébrak, et al., 2002, Martín-Izard, et al., 2009, Rytuba, et al., 1989, Saupé and Arnold, 1992, Spangenberg, et al., 2010, Tsantini, et al., 2018), the most probable provenance for the mineral would be Northwestern Spain (i.e., León or Asturias), as depicted in Fig. 6.

## 4. Discussion

### 4.1. On the provenance

Since the presence of cinnabar in the bones has been confirmed without any doubt, it is appropriate to inquire about the origin of the mineral used.

Given the present knowledge of the cartography of cinnabar ore deposits in the Iberian Peninsula, possible sources may be located in the Aezkoa valley, Arribes / Auritz-Burguete / and Leitza, all in Navarra,

**Table 2**

Multielement composition (expressed as  $\% \pm 2\sigma$ ) of red-stained bones from the burial located in sector 2.

Bal (H,C,N,O)	Ca	P	Ca/P	Hg	Fe	K	Al	Si	S
65.37 $\pm$ 0.35	21.64 $\pm$ 0.27	6.86 $\pm$ 0.08	3.15	0.15 $\pm$ 0.01	0.24 $\pm$ 0.02	0.08 $\pm$ 0.01	0.50 $\pm$ 0.07	2.40 $\pm$ 0.06	2.52 $\pm$ 0.04
59.51 $\pm$ 0.40	22.68 $\pm$ 0.28	8.99 $\pm$ 0.09	2.52	0.19 $\pm$ 0.01	0.25 $\pm$ 0.02	0.07 $\pm$ 0.01	0.69 $\pm$ 0.08	3.13 $\pm$ 0.07	4.20 $\pm$ 0.05

**Table 3**  
Multielement composition (in %) of red-powder sprinkled soil from the burial located in sector 2.

Ba	Sn	Ag	Bal	Zr	As	Hg	W	Ni	Fe	Mn	Cr	Ti	Ca	K	Al	P	Si	S	Mg
0.09	0.01	0.00	19.11	0.01	0.03	7.67	0.35	0.09	3.02	0.16	<LOD	0.20	14.71	0.61	3.52	5.67	29.06	14.56	1.14
0.08	<LOD	0.01	23.35	0.01	0.02	7.29	0.51	0.04	2.44	0.13	0.01	0.23	8.33	0.63	3.12	4.65	31.21	17.94	<LOD

Bal (balance), i.e., the difference to 100 % of the sum of all measured elements, includes elements with atomic number  $Z \leq 11$  and mainly accounts for organic matter (H, C, N, O). <LOD indicates below the limit of detection.

located  $\approx 215$  km away as the crow flies. Somewhat more distant are the mineralizations of Gea de Albarracín, Teruel (333 km); Libros, Teruel (371 km); and Chóvar, Castellón (426 km), and much further away, the Leonese ores of Riaño (558 km), Lois, Riosol and Pedrosa del Rey (592 km) and the Asturian deposit of Mieres (621 km). The Almadén mines are 760 km away, and the Pulpí and Bayarque mines in Almería are 804 and 860 km away, respectively.

An initial reading of this brief inventory, taking into account the generally chosen criterion of proximity to the possible supply areas, would suggest that the logical origin would be the Aezkoa valley, followed by the outcrops of Teruel and Castellón and, finally, the most distant ones of León-Asturias and Almadén. However, the analyses allow us to propose another very different reading. The results of the analyses seem to indicate that the cinnabar found in Campodarbe could have its origin in the Asturian-Leonese mining area of Lois, according to the sulfur isotopic data. The  $\delta^{34}\text{S}$  values of the ‘Pedrosa del Rey’ mine (in León), and of the ‘Riosol’, ‘Eugenia’, ‘María de la Encarnación’, and ‘El Tarronal’ mines (in Asturias) are the closest to that of the cinnabar sample from the Campodarbe cave. Nevertheless, as can be seen in Fig. 6, some indicators may generate doubts. An origin in the environment of the Almadén mines cannot be discarded, with a mean value ( $n = 74$ ) of  $\delta^{34}\text{S}$  of  $+7.04$  ‰ (values ranging from  $-1.6$  to  $+13.6$  ‰ have been reported in the bibliography). However, if only the ore samples processed and analyzed under the same conditions (Tsantini, et al., 2018) are considered, the highest  $\delta^{34}\text{S}$  value (lower than  $+12.5$  ‰) would be further away from the target value than those provided by the León-Asturias ore deposits. In addition, sulfur isotopic analyses of other samples from the ‘María de la Encarnación’ mine (in Caunedo, Somiedo, Asturias) yielded a result of  $\delta^{34}\text{S} = +15.02$  ‰, almost identical to that obtained at Campodarbe ( $\delta^{34}\text{S} = +15.1$  ‰), so – with the currently available data – this may be put forward as the most probable origin.

#### 4.2. Cinnabar in funerary sites in the Iberian Peninsula between the 5th and the 2nd millennium cal BC

Cinnabar is detected more and more frequently. The so-called ‘red gold’ is the subject of new tests that demonstrate the importance this raw material had in antiquity (Zarzalejos Prieto, et al., 2020). Without taking into account its use in portable art, it has been identified in funerary contexts from at least the 5th to the 2nd millennium cal BC, with a spatial distribution that reaches most of the Iberian Peninsula, with less intensity in Galicia, the Cantabrian region, the Ebro Valley, and Catalonia (Bueno Ramírez, et al., 2020, Domínguez-Bella, 2010, Gómez-Merino, et al., 2011, Hunt and Hurtado, 2010, Hunt Ortiz, 2020, López Padilla, et al., 2012).

Red color is present in numerous burial sites contemporary to the Campodarbe cave. Its presence increases visibility in closed and darkened rooms and highlights the existence, importance, and significance of the burials. Both ochre and cinnabar were used as dyes. The choice of one or the other would depend on the intended purpose of the pigmentation, given that their properties and effects are very different (Delibes de Castro, 2000). Their joint use has been documented in the tombs of ‘Alberite I’ and ‘Montelirio’ in Cádiz and Seville, respectively, or ‘La Velilla’ in Osorno, Palencia (Delibes de Castro, 2000, Martín-Gil, et al., 1995, Martín-Gil, et al., 1994, Zapatero Magdaleno and Delibes de Castro, 1996), although “the reds associated with the corpses and funerary floors are mostly made of cinnabar, a foreign raw material in most of the known cases” [tr.] (Bueno Ramírez, et al., 2020).

Several hypotheses attempt to explain the presence of cinnabar. As it appears mixed with ochre and clay, it may be regarded as a suitable material for embalming or shrouding rituals (Martín-Gil, et al., 1995, Martín-Gil, et al., 1994), acting as an antiseptic to preserve the body (Delibes de Castro, 2000), although the possibility that its appearance in certain bones and different anatomical areas could have been transferred from the clothing or certain elements of adornment cannot be totally ruled out (Delibes de Castro, 2000, López Padilla, et al., 2012). Its

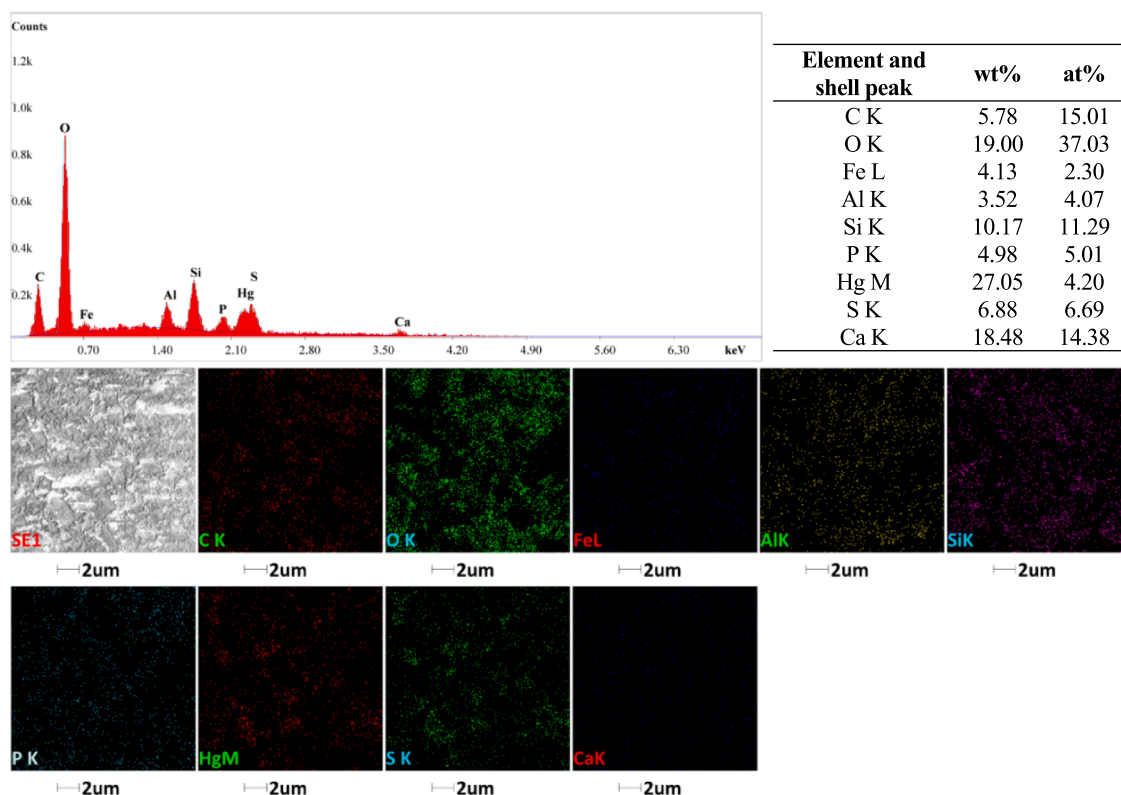


Fig. 3. SEM-EDX results for the red-stained surface of one of the bones: (top left) EDX spectrum; (top right) elemental analysis; (bottom) elemental mapping.

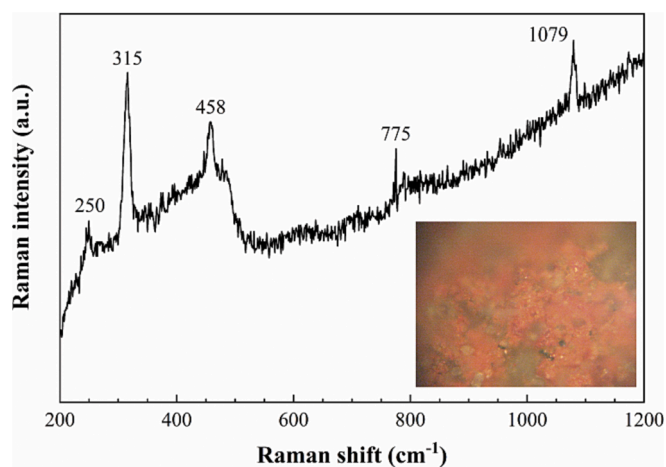


Fig. 4. Raman spectrum of the surface of a red-stained bone from sector 2. Inset: sampled area.

use has even been proposed as an indicator of the end of the burial activity, as a ‘closure’ (Blanco Vázquez and Carrocerá Fernández, 2013).

Finally, an interpretation that would explain the presence of intentional, postmortem pigmentation (Furgús, 1937) should be highlighted. In this case, staining is preceded by defleshing, hence the most colored areas coincide with phalanges, feet, and hands, a hypothesis that has been considered in some places such as the ‘Alberite’ dolmen (Gómez-Sánchez, 1996) and that – in view of the reasons put forward in the introduction section – is proposed as the most feasible in the case of the Campodarbe cave.

#### 4.3. Implications of the study

Without taking into account parietal art sites, the oldest non-natural pigmentation finding in Aragon is a Paleolithic pendant from the Badegulian level of the ‘Cueva del Gato’ in Épila (Zaragoza) impregnated with ochre (Rodanés Vicente, et al., 2021). In the Holocene, in the Neolithic, painted cobbles are very abundant in the ‘Chaves’ cave in Bastarás (Huesca) (Utrilla Miranda and Baldellou Martínez, 2007). Both examples are an example of their use in portable bone or stone art. It is, however, in funerary contexts in natural caves that we find a greater presence, a phenomenon also documented in other territories in the Iberian Peninsula (García Borja, et al., 2006). In Huesca, we have located two caves with burials in which the presence of ochre has been documented. The above-mentioned ‘Chaves’ cave presents a unique pit burial, whose inhumation shows reddish pigment remains in certain areas that could be caused by impregnation from a piece of fabric or shroud (Utrilla Miranda, et al., 2008); and the recently investigated ‘Moros de Alins’ cave, in which the dye is focused on a possible element of the grave goods, viz. a small container of vegetable fibers (Rodanés Vicente, 2017, Rodanés Vicente and Alcolea, 2017). So far, the occurrence of cinnabar has only been detected in Campodarbe.

Regardless of the practical or ritual sense that the impregnation with cinnabar of human remains could have played, summarized and commented on in the previous section, the symbolic content of the coloration is undeniable, although we cannot – with the information we possess – unravel its meaning. At the same time, it may be interpreted as an element of social prestige that, in addition, would have greater value and exclusivity (Liesau von Lettow-Vorbeck, et al., 2020) due to the difficulty of obtaining the material, since usually the archaeological sites are far from the outcrops (as occurs in the case of the present study).

We must admit to being surprised by its origin, given that the largest distribution center in the Iberian Peninsula – taking into account the analyses carried out so far – is Almadén, which has supplied raw material to most of the known sites in the center and south of the Peninsula

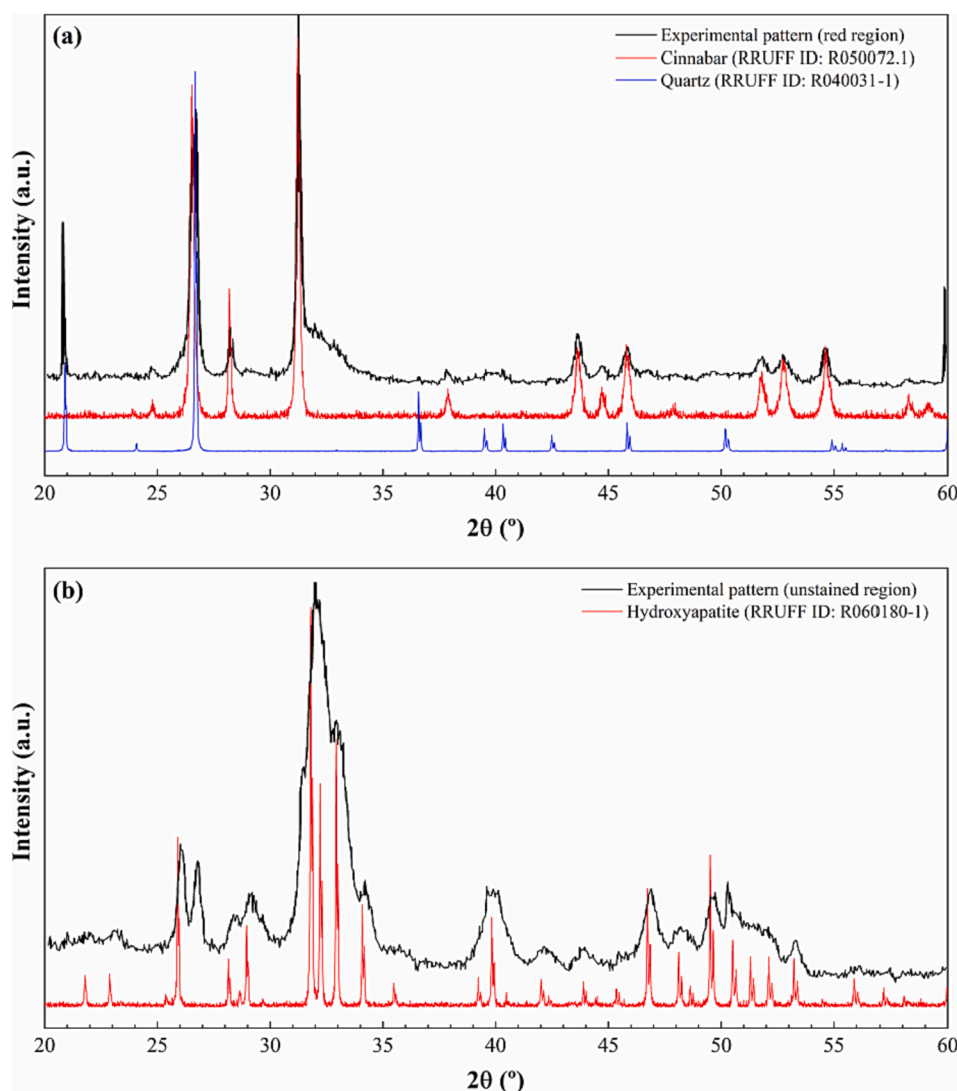


Fig. 5. Powder diffraction patterns of (a) red-stained region of a bone from sector 2; (b) non-stained region of the same bone.

(Hunt Ortiz, et al., 2011). This forces us to consider other possibilities of interaction with other territories where its presence is not unknown, but in which the findings are less frequent.

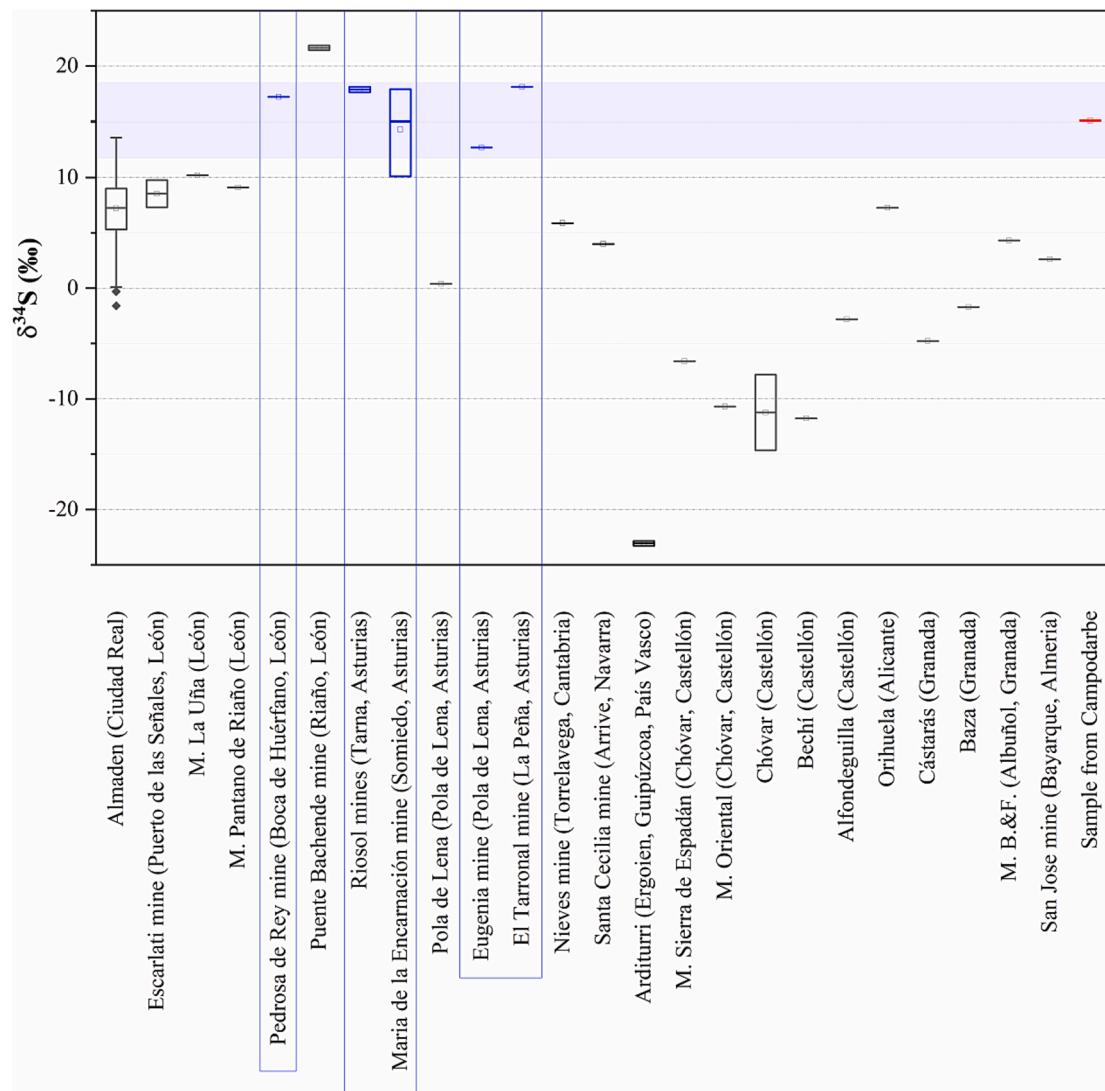
We start from the fact that the Ebro river has acted as an axis of north–south and east–west communication at different periods in pre-historic times (Rodanés Vicente, 1999). We must consider new exchange networks, cultural contacts, or small population movements, without ruling out any possibility, since – at least since the Neolithic period – the Ebro valley has served as a means of communication with the westernmost territories (Rodanés Vicente and Picazo Millán, 2005). Likewise, during the heyday of megalithism, without entering into the debate of its origin or diffusion, the regions, especially those of the upper Ebro valley, have been interconnected with the Northern Meseta. A good example is the distribution map of the so-called spatula idols (Delibes de Castro, et al., 2012), the Bell Beaker Culture (Rodanés Vicente, 1992a; Rodanés Vicente, 1992b), or different metallic objects characteristic of the Atlantic Bronze Age or Cogotas I (Rodanés Vicente and Picazo Millán, 2018). This dynamic would serve as an explanatory argument for the presence of cinnabar in the Central Pyrenees since it would be part of a reality already detected in other periods. In this case, the novelty would come from the origin of the raw material, which would unequivocally confirm the E/W relationships mentioned above.

## 5. Conclusion

The analyses of the pigment present on red-stained bones found in a burial in sector 2 of the *Cueva de la Sierra*, dated  $4400 \pm 30$  BP, were conclusive about the presence of cinnabar, which constitutes the first documented occurrence of cinnabar in Aragón (Spain). Sulfur ( $\delta^{34}\text{S}$ ) isotope analysis pointed to an origin in the ores of the Northwestern Iberian Peninsula, most probably from Caunedo (Asturias), ca. 500 km far from the burial site. Its presence on the southern slope of the Central Pyrenees corroborates the role of the Ebro valley as an axis of east–west communication in the Iberian Peninsula during the Late Neolithic and Early Chalcolithic periods.

## CRediT authorship contribution statement

**José María Rodanés-Vicente:** Validation, Investigation, Writing – original draft, Writing – review & editing, Visualization. **José Antonio Cuchí-Oterino:** Conceptualization, Investigation, Writing – original draft. **Takeshi Minami:** Methodology, Formal analysis, Investigation, Resources, Writing – original draft. **Kazuya Takahashi:** Methodology, Validation, Investigation, Writing – original draft. **Jesús Martín-Gil:** Methodology, Validation, Investigation, Resources, Writing – original draft. **José Ignacio Lorenzo-Lizalde:** Conceptualization, Investigation, Writing – original draft. **Pablo Martín-Ramos:** Formal analysis,



**Fig. 6.** Boxplot of the  $\delta^{34}\text{S}$  values, provided as ‰, of the cinnabar sample from Campodarbe (in red) and mineral deposits found in the literature (detailed in Table S2). The closest matches are highlighted in blue.

Investigation, Resources, Writing – original draft, Writing – review & editing, Visualization.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jasrep.2023.103849>.

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