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New evidence reveals the earliest use of cinnabar in the western Mediterranean: The Neolithic settlement of La Marmotta (Lazio, Italy)

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ABSTRACT

Numerous researchers point out the emergence of human symbolism is related to the evolution of the complexity of human cognition. Red mineral pigments have been used extensively, particularly with anatomically modern humans, for various purposes. However, the management and supply of these pigments during prehistoric periods remains poorly investigated. Still today, the limited application of physico-chemical analyses often leads to a simplistic attribution of these pigments as ochre. The studies of data from recent literature presented in our paper show a progressive introduction and exploitation of cinnabar ore, to achieve a red pigment, from the seventh millennium BC. In this panorama, the new data obtained from the analyses of samples of artefacts from La Marmotta (Italy) show a wide use of cinnabar in central Italy from the early Neolithic and attest to the earliest use of this ore in the western Mediterranean area.

1. Introduction

Natural pigments, especially red type, were widely used in prehistory. In recent years, several scenarios have been proposed to assess the use of these colours for cultural, medical, and artistic purposes. The emergence of human symbolism and a sense of art is usually related to the evolution of the complexity of human cognition (Watts, 2002, 2010; McBrearty and Brooks, 2000; d'Errico et al., 2003; d'Errico et al., 2010; d'Errico and Henshilwood, 2011; Henshilwood and Marean, 2003; Henshilwood and Dubreuil, 2009). Recent archaeological data testify to the earliest use of red pigments in Africa and the Near East during the Middle Stone Age (Watts, 1999, 2002; Rigaud et al., 2006; Jacobs et al., 2006; Bar-Yosef Mayer et al., 2009; Zilhão, 2012). At Blombos Cave (South Africa) in a layer dated circa 100.000 years ago, quartzite grinders used to produce powder of ochre, which was then voluntarily stored in shells (Henshilwood et al., 2009, 2011) were found. In Europe, a series of new dates obtained from carbonate crusts suggest attributing the artistic activities discovered in three Iberian caves (La Pasiega in Cantabria; Maltravieso in Extremadura; Ardales in Andalucía) to the last Neanderthals (Hoffmann et al., 2018). Other accounts bear witness to the interest of Neanderthal groups in the use of red colour. At Maastricht-Belvédèr, a layer referable to the middle Palaeolithic revealed the presence of non-local hematite infiltrating the sediment, following voluntary, not well-defined, human activity. Lumps of red pigment and a *Glycymeris* sp., having traces of ochre on the concave surfaces, were discovered in a burial dated 92.000 years ago at Qafzeh Cave (Israel, Walter, 2003).

During the upper Palaeolithic, the presence of red pigments in burials increased. The earliest evidence of this tendency includes the so-called red lady, a male burial from Paviland (South Wales, 26.000 years agoAldhouse-Green, 2000), the grave of a young prince at Arene Candide (Italy, 24.000 years ago Pettitt et al., 2003) and the double child burial at Sunghir (Russia, 24.000 years ago Formicola and Buzhilova, 2004). In all these cases, the pigment was sprinkled on the bodies. It is

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not completely clear if the Palaeolithic human groups used the mineral powder for a preservative purpose or a ceremonial function. The use of colour seems to assume a significant role in the social life of the Palaeolithic groups, as underlined by the considerable evidence of rock paintings from southwestern Europe. Despite our knowledge being restricted to preserved evidence, ethnographic observation on modern hunter-gatherer societies and experimental activities permits us to suppose a wider range of uses of natural pigments related to daily life. Mineral pigments can be used as tanning and preserving agents for leather and food, as substances added to adhesives to reinforce their adherent capacities, as medicinal substances, antiseptics, and insect repellents (Schmandt-Besserat, 1980; Velo, 1984; Rifkin, 2011). It certainly should not be ruled out the application of pigments to produce marks on objects or bodies, as later demonstrated by tattoos discovered on the mummy of Ötzi (Val Senales, Italy, Hedges et al., 1992), dated to the Copper Age (Table 1).

During the Holocene, the red colour maintained its importance, although we cannot attest to the same significance or symbolic value attributed to it in the previous period. Traces of red pigment are found in about 50% of the European Mesolithic burials (Grünberg, 2015).

From the Pre-Pottery period in the Near East, we recognise a large use of red pigment in architectural and funerary contexts. One of the first attestations dated to the Natufian site of Ain Mallaha (Israel, Perrot et al., 1988; Bocquentin, 2003; Valladas and Kalteneker, 2007; Valla et al., 2017) where the pigment was in the plaster covering the house walls. From the end of ninth millennium BC (Table 1), the habit of decoring the internal walls of dwellings became much more widespread, as testified by numerous sites, such as Ἀin Ghazal (Jordan, Rollefson, 1983; Banning and Byrd, 1987; Rollefson et al., 1992; Regagnon, 2001 Schmandt-Besserat, 2013; Grissom and Griffin, 2013), Aşikli Höyük (Turkey, Esin et al., 1991; Özbaşaran, 2011; Quade et al., 2014), Çatalhöyük (Turkey, Mellaart, 1967; Hodder, 2001, 2004, 2012, 2015; Carter, 2009; Orton et al., 2018; Bayliss et al., 2015; Schotsmans et al., 2022), and Arpachiyah (Jezireh irakienne, Stuckenrath and Ralph, 1965; Hijara, 1980; Campbell, 2000). At the latter site, the red colour was also used in depositional contexts and for pottery decoration (Anderson et al., 2014).

Although colours played a significant role in prehistoric human societies, there is still a lack of systematic studies on their management and supply modalities in different chronological and regional contexts (Upīte, 1987; Sulgostowska, 1990; Delibes de Castro, 2000; Salomon et al., 2008; Zilhão et al., 2010; Çamurcuoğlu, 2015; Domingo and Chieli, 2021).

Data from Çatalhöyük suggests the inhabitants used a wide range of pigments from the end of the eighth millennium BC. Ochre was the most relevant, while yellow ochre, cinnabar, blue azurite, and green malachite were less common (Schotsmans et al., 2022). It turns out to be cinnabar and not ochre also the red colour present in a Halaf burial at Tel Abu Huraira (Syria, Molleson et al., 1992; Moore et al., 2000; Molleson, 2016).

In central Europe, the use of cinnabar was first claimed by M.M. Vasić (Vasić, 1932) asserting to have found evidence of cinnabar in every layer of the site of Vinča (Serbia), ranging from the middle of the sixth to middle of the fifth millennium BC. Recent analyses (Gajić-Kvaščev et al., 2012) confirm the presence of this pigment in this site, although with a much lower incidence than previously assumed.

The earliest evidence of cinnabar in the Balkan region seems to relate to the settlement of Pločnik (Serbia), where a vessel containing red powder and red-painted figurines, dating back to the second half of the sixth millennium BC, were discovered. The results obtained from the analyses were not exhaustive to indicate the source area of the ore. The mineral could have been collected in the region of Mount Avala in the

Table 1
Radiocarbon dates of the principal Neolithic and Copper Age sites mentioned in the text (calibration with OxCal v4.4.2 using IntCal20, Reimer et al., 2020).

Ain Mallaha	Israel	GifA 100400	10540	90	10736-10527	Perrot et al. (1988)
		GifA 70013	8740	40	7932–7611	
Ἀin Ghazal	Jordan	AA 1164	9100	140	8530–8231	Rollefson et al. (1992)
		AA 5196	7670	100	6633–6432	
Çatalhöyük	Türkiye	PL 980525A	8340	90	7524–7199	Bayliss et al. (2015)
		OxA 11764	6707	38	5640–5569	Özbaşaran (2011)
Aşikli Höyük	Türkiye	GrN 28617	8980	40	8261-8220	
		AA 87976	8690	20	7735–7603	Quade et al. (2014)
Arpachiyah	Iraq	P 585	8064	78	7129–6839	Stuckenrath and Ralph (1965)
		BM 1531	6930	60	5887–5732	Palmisano et al. (2022)
Tel Abu Huraira	Syria	OxA 1227	8320	80	7508–7195	Moore et al. (2000)
		OxA 1931	7890	90	7031–6642	
Vinča-Belo Brdo	Serbia	Hd 16661	6353	66	5463–5221	Schier (1996)
		NOSAMS 67700	5890	45	4827–4714	Borić (2009)
		OxA 24923	5335	35	4247–4056	Borić (2009)
Pločnik	Serbia	OxA 14684	6354	36	5368–5231	Borić (2009)
		OxA 14678	4431	36	3263–3015	Borić (2009)
Casa Montero	Spain	Beta 206512	6410	40	5471-5324	Bustillo et al. (2009)
Cova de l'Or	Spain	OxA 10192	6310	70	5362–5214	García-Borja et al. (2006)
Montelirio	Spain	CNA 589	4400	60	3259–2916	Bayliss et al. (2016)
		OxA 30385	4151	30	2872–2635	Bayliss et al. (2016)
Campo de Hockey	Spain	CNA 833	5665	50	4544–4449	Vijande Vila et al. (2015)
		CNA 360	5020	50	3942–3711	
Peñacalera cave	Spain	Beta 491868	4620	30	3491–3364	Gleba et al. (2021)
Perdigões	Portugal	Beta 327750	4030	40	2579–2475	Emslie et al. (2016)
		Beta 308789	3840	30	2343–2207	
Carpignano	Italy	LTL 126A	5666	60	4547–4448	Tiberi and Dell'Anna (2013)
Grotta dei Cervi of Porto Badisco	Italy	LTL GCD2	5452	45	4346–4258	Quarta et al. (2018)
Catignano	Italy	Ly 2186 OxA	6455	60	5477–5367	Colombo (2006)
		LTL 1155A	5587	60	4488–4353	
Casale Somaini	Italy	LTL 3472A	5240	50	4217–3981	Barich et al. (1968)
Lunghezzina	Italy	OxA 80789	4740	45	3629–3382	Manfredini (2012)
Torre della Chiesaccia	Italy	LTL 3486A	4129	45	2869–2583	Anzidei et al. (2011)
La Vela VII	Italy	Utc 10555	5579	45	4452–4354	Dal Rì et al. (2002)
		Utc 10556	5555	48	4448–4347	
Mummy of Similaun (Ötzi)	Italy	OxA 3376	4450	80	3342–2928	Hedges et al. (1992)
		OxA 3371	4660	55	3519–3367	

Suplja Stena (Serbia) mine. The latter is close to the Vinča site, and about 300 km from Pločnik. Lithic tools and pottery have been found in the mine, testifying to its use since the late Neolithic. However, the earliest traces of mining may have been removed by later activities (Gajić-Kvaščev et al., 2012). Vessels decorated with cinnabar have also been found in Croatia, at the late Neolithic site of Grapčeva Cave (Kaiser and Forenbaher, 2016).

In the western Mediterranean, evidence of extensive exploitation of Neolithic cinnabar has been found in Almaden (Ciudad Real, Spain, Hunt-Ortiz et al., 2011). Other smaller mines in Spain are in Castellon, Valencia, and Murcia provinces, but the extraction activities in these latest areas seem to have started in medieval times (Domingo et al., 2012). Almaden mine was active from the Iberian early Neolithic, as witnessed by the evidence recovered at the flint mine of Casa Montero (Madrid, Spain). On this site, which was also a blade production centre, distributed all over the region (Castañeda, 2016), a blade was found covered with cinnabar. Radiocarbon dating of two charcoal samples collected in the flint shafts suggests its use from the second half of the sixth millennium BC (Bustillo et al., 2009).

Cinnabar was also found inside a *Glycymeris* sp. shell at Cova de l'Or (Alicante, Spain), an early Neolithic settlement, occasionally used as a burial place (García-Borja et al., 2006).

In Spain, the presence of this pigment in funerary contexts increased from the late Neolithic to the early Copper Age. Numerous burials, such as the dolmen de Alberite (Cádiz) and the dolmen of Casas de Don Pedro in the Guadiato Valley (Bueno Ramírez et al., 2020) dated to the end of the fifth millennium BC, show the widespread use of cinnabar to cover the bodies of the buried and parts of the funeral area. In the Cueva de Los Murciélagos de Zuheros (Cordoba, Martínez Fernández et al., 1999), in a similar chronological horizon, the pigment was used as a filler for the grooves of pottery decoration and to cover the surface of some lithic tools. Analyses (Emslie et al., 2022) of several burial contexts located in Portugal and Spain, ranging from the middle Neolithic to the Copper Age (i.e.: Campo de Hockey, Spain, Vijande Vila et al., 2015; Emslie et al., 2015; Cova da Moura Cave, Portugal, Silva, 2002; Perdigões, Portugal, Emslie et al., 2016; Montelirio and Montelirio Tholos, Spain, Emslie et al., 2016; García Sanjuán et al., 2018) have revealed high levels of total mercury (THg) in human bones from to the end of the fifth millennium BC. According to the researchers who conducted the study, this high presence of mercury is probably due to the use of cinnabar as body paint or as medicine, during rituals and social practices. By the end of the third millennium BC, THg levels in human bones became low, probably because of changes in funeral customs and human groups' social dynamics (Emslie et al., 2022). The first evidence for the use of cinnabar for dyeing textiles (Peñacalera cave, Cordoba) is also attributed to the Copper Age (Gleba et al., 2021).

This paper aims to present the results obtained from the analysis of artefact samples from the early Neolithic site of La Marmotta. The XRF analyses have underlined the choice of the inhabitants of the site to exploit two different substances to achieve a red colour: cinnabar and ochre. The presence of cinnabar will be discussed in relation to the different artefact types. It will also present an updated picture of the oldest evidence of the use and exploitation of cinnabar in Western Europe, among which, in the light of the new evidence, La Marmotta provide the earliest one.

In Italy, chemical analyses conducted on the Square Mouthed Pottery culture (SMP) burial of a child (Tomba 3, first half of the fifth millennium BC, Dal Rì et al., 2002) at La Vela di Trento in Northern Italy, revealed traces of cinnabar on the necklace's shell beads, bracelet, skull, and on a hemispherical bowl that the child held in his hands. In the late Neolithic grave of Grotta Patrizi di Sasso Furbara (Lazio, Grifoni Cremonesi and Radmilli, 2000), cinnabar was found on the skull of the buried person, on the bed, and in the grooves of the engraved decoration of a vase, placed near the body. Additionally, cinnabar residues were found on the skull of a man buried at Carpignano (Apulia, Tiberi and Dell'Anna, 2013). In a different context, a Serra d'Alto vessel with cinnabar painted decoration (Quarta et al., 2018) was discovered at Grotta dei Cervi of Porto Badisco (Apulia), a cave used for ritual purposes (Graziosi, 1980; Aprile et al., 2017). Traces of this pigment have also been attested on an animal bone handle in the settlement of Catignano (Abruzzi, Colombo, 2006).

During the Copper Age, numerous hypogeum burials, related to the Rinaldone *facies* (from the fourth to the beginning of the second millennium BC, Anzidei and Carboni, 2020), including Sgurgola, Ponte San Pietro, Bandita San Pantaleo (Barich et al., 1968), Casale Somaini, Lucrezia Romana, Ponte delle Sette Miglia, Lunghezzina (grave 3), Romanina (Anzidei and Carboni, 2020) and some Laterza *facies'* sites, as Torre della Chiesaccia 2 (grave 4, Anzidei et al., 2011), attest the use of this pigment to cover parts of the body, mainly the skull, and lithic artefacts, usually arrowheads and dagger. Lumps of the ore were also occasionally deposited near the buried.

2. The site of La Marmotta

The early Neolithic site of La Marmotta (Fig. 1) is located under the waters of Lake Bracciano (Anguillara Sabazia, Lazio). It was excavated from 1992 to 2006. In 2009 a small archaeological intervention was carried out under the supervision of the Soprintendenza speciale al Museo nazionale preistorico etnografico Luigi Pigorini (today the Museo delle Civiltà) (Fugazzola, 2002). The archaeological site lies approximately 300 m away from the modern shoreline, submerged at a depth of 11 m (8 m of water and 3 m of sediment), which has permitted exceptionally good conservation. The numerous and varied objects and implements made of wood, basketry and textiles reflect their importance for the Neolithic communities and the technical skill they reached to manufacture them. This technological know-how can undoubtedly be explained by a socioeconomic organisation based on the specialisation of particular artisans. Only people with specialised knowledge would be capable of building the large dwellings, but also many of the artefacts described below: bows, sickles, spindles, wooden recipients, baskets, fabrics and canoes (Fig. 2). That is why La Marmotta can be considered a *Pompeii-like* Neolithic site.

The radiocarbon dating of the village of La Marmotta was mainly carried out on wooden samples taken from poles deeply embedded in the lake. The site has been dated again more recently using short-lived samples (charred seeds of *T. dicoccum*) from Layers 1 and 2 (Fugazzola and Tinazzi, 2010; Mazzucco et al., 2022; Mineo et al., 2023). The radiocarbon calibrated dates obtained so far are distributed over a time interval between 5690 BC (Pole 214, Square A2) and 5260 BC (Pole 21, Square B29).

The stratigraphic sequence is composed of three main layers (Layer II, Layer I, Layer 'Chiocciolaio'), without any real interruption or sterile layer between them. Layers have been defined on the basis of the pedological characteristics, the radiocarbon dates and the type and style of the pottery assemblage associated. Layer II corresponds to the beginning of the settlement. The pottery assemblage belongs to the Tyrrhenian facies of the Impressed Ware Culture, and is characterized by the high-quality production of a diversity of vessels, including some unique shapes, like the pirogue-shaped vases (Fugazzola et al., 1993). Layer I is characterized by a greater number of findings of all types and represents the time of the most intense occupation of the village. This pottery is decorated with paintings and incised motifs (Sasso-Fiorano style). The last phase of Layer I, also called Layer 'Chiocciolaio', represents the abandonment of the settlement.

The settlement was formed by rectangular houses, 8–10 m long by 6 m wide, with internal compartments and a central hearth. Near some houses up to 5 canoes were documented (Mineo et al., 2023). The distribution of the houses seems to indicate that there was an organization of space.

La Marmotta was based on a consolidated domestic economy in which several animal and plant species were consumed: goats, sheep, cattle, pigs, wheat, barley, legumes (lentils, broad beans and peas), and

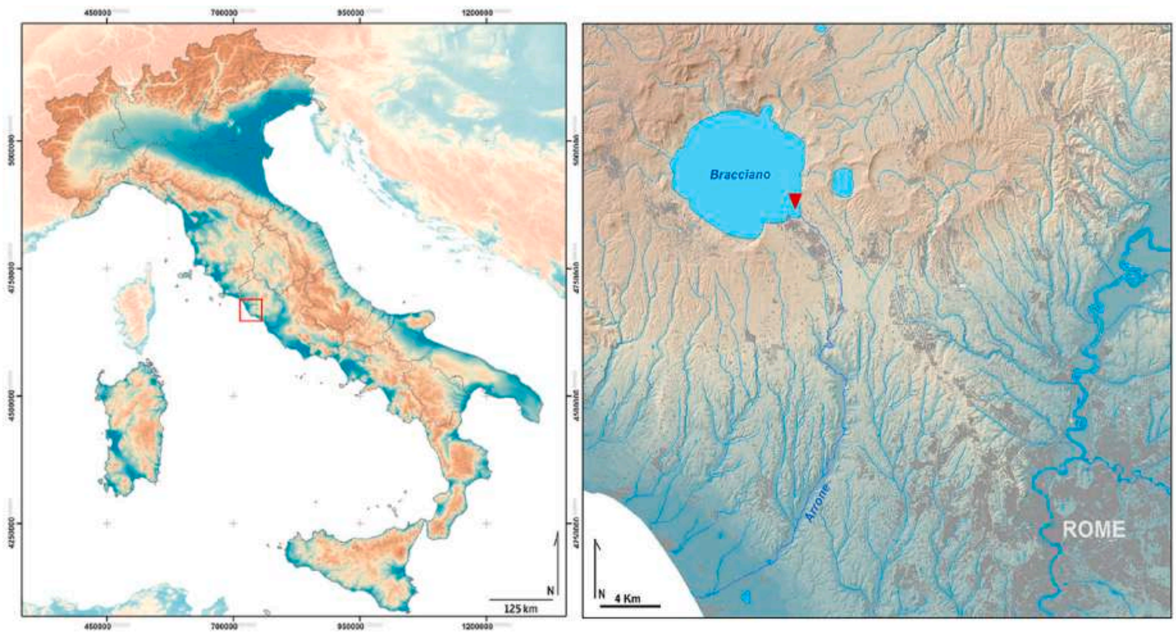

Fig. 1. Location of the settlement of La Marmotta (Anguillara Sabazia, Lazio, Italy).

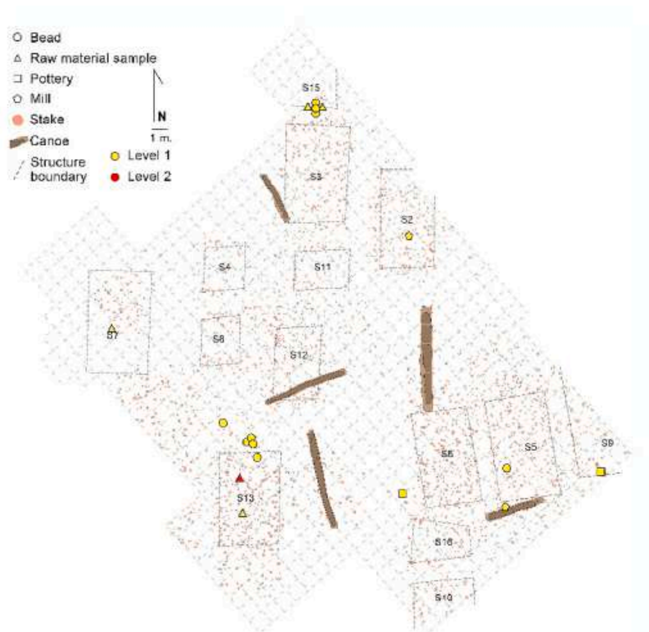

Fig. 2. Plan of the La Marmotta site with structures and canoes.

possibly also grapes and opium poppies (Rottoli, 1993; Tagliacozzo, 2005; Salavert et al., 2020).

The analysis of the lithic raw materials shows that the population of La Marmotta had a well-established contact network with communities from several parts of the central Mediterranean. This is demonstrated by the presence of different varieties of flint probably acquired locally or regionally, for example in the Apennines, whereas others came from distant locations, such as the Defensola mines in the Foggia region on the east coast of Italy (Radi and Danese, 2003; Pessina and Tiné, 2010; Muntoni et al., 2021), the obsidian from the Lipari and Palmarola islands (De Francesco et al., 2012), and some of the polished green axes and adzes probably originating from the Alpine territories (D'Amico, 2000).

3. Materials and method

The study was performed on eighteen artefacts showing red pigment traces (Figs. 3, 7 and 8, Table 2): ten ornaments, two stone tools, one pottery sherd, and five raw material samples.

The more extensive analysed set involves adornment items. Residual evidence of a thin, red-coloured substance was discovered on ten beads, mostly on lithic and shell beads and in traces on clay and seed beads. The seed beads are part of a necklace on display at the Museo delle Civiltà in Rome, which has been assembled for exhibiting reasons with heterogeneous pieces coming from different parts of the excavation. Two other clay beads from the same ensemble (unpainted and not listed in Table 2) were used for comparison with the XRF spectra obtained on red-painted clay bead 14504.

Further to these objects, similar traces were found on two stones, probably employed as querns. It was also decided to analyse samples of reddish raw materials recovered during the excavation, and a pottery fragment, where a white and a red substance to fill the incised decoration was clearly visible.

Ornaments and macro lithic tools were measured, weighed, and classified, according to their shape and size, based on typologies proposed in the literature (Ornaments: Barge, 1987; Bains, 2012; Micheli, 2009; Macro-lithic tools: Hamon, 2006; Adams et al., 2009; Lunardi and Starnini, 2013).

The surfaces of all the items were observed with the naked eye and with an optical binocular microscope (BMS 7458, with 10x to 50× magnification), connected to a digital camera (Canon EOS 550D). Finally, all the data and observations were recorded in a database.

In order to define the nature of raw materials of artefacts analyses and residues, archaeometric analyses were conducted. To the ED-XRF analysis (Mantler and Schreiner, 2000) the Elio XRF spectrometer produced by Bruker was used. The experimental conditions were set to 90 s acquisition time, voltage equal to 40 keV and current equal to 80 mA.

A confocal micro-Raman system linked to a Leica DLML microscope with a 0.75 numerical aperture 50× NPLAN objective was also used. The measurement was performed using the emission of a CW He–Ne laser at 633 nm. The spectral detection is realized using a single grating monochromator (1200 lines mm^{-1}), coupled with a Peltier-cooled CCD detector (578 × 400 pixels of 22 μm × 22 μm). The spectral resolution of the spectrometer is 2.0 cm^{-1}. Before the analysis, the instrument was

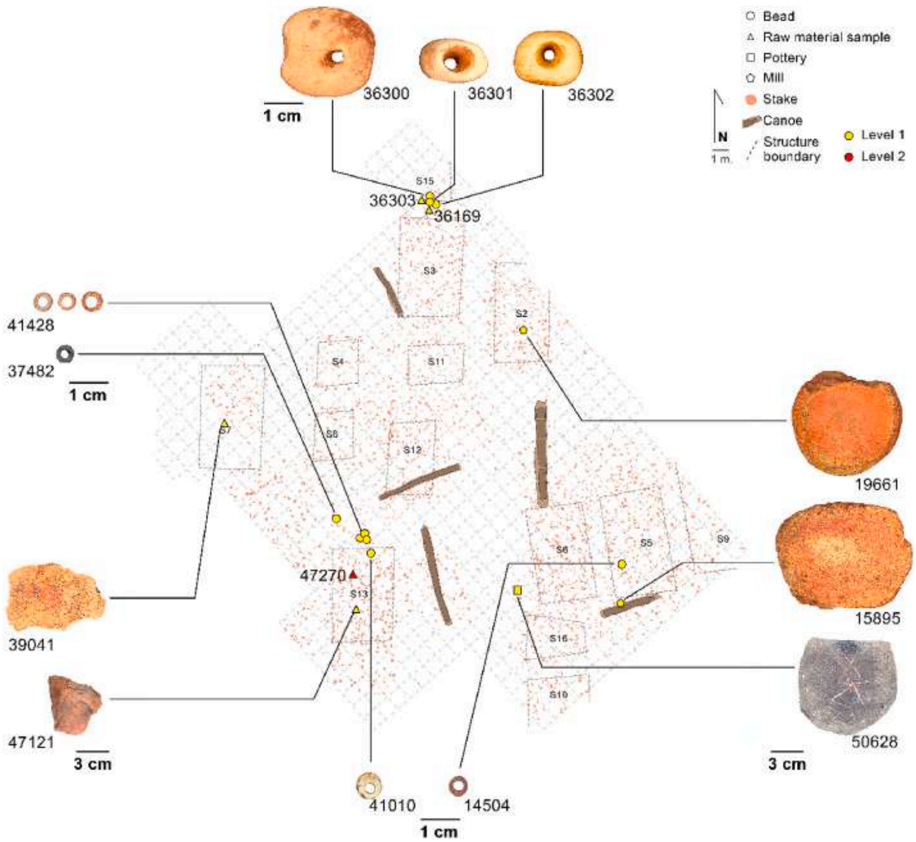

Fig. 3. Map of the distribution of artefacts with cinnabar residues.

wavelength-calibrated using the 520.0 cm^{-1} Raman band of a pure silicon crystal.

4. Archaeometric results

In Fig. 4A, the spectra of the raw materials 36303 and 47121 are compared. The first sample is dominated by the lines of mercury (L_a = 10.0 keV, L_b = 11.8 keV, L_g = 13.8 keV and the corresponding escape peaks at these energies minus 1.7 keV, resulting by the interaction with the silicon detector). The fluorescence lines of sulphur are also visible, very close to each other, at low energy (K_a = 2.3 keV, K_b = 2.5 keV) (Pérez-Diez et al., 2023). The presence of mercury and sulphur in a mineral indicates the presence of cinnabar (HgS).

The spectrum of the raw material 47121, on the other hand, does not show any fluorescence line at the energies of sulphur and mercury; however, the fluorescence lines of iron are prominent at K_a = 6.4 keV and K_b = 7.1 keV), hinting to its identification as red ochre.

The raw sample 36303 was also analysed using a laboratory confocal micro-Raman. The resulting Raman spectrum is shown in Fig. 4B. The two main Raman bands of cinnabar around 253 and 343 cm^{-1} are clearly visible (Pérez-Diez et al., 2023), thus confirming the ED-XRF results of the previous analysis.

The spectra of the raw materials 36169 and 39041 (not shown here) are very similar to 36303, while the one of item 47270 confirms that it is similar to 47121, thus suggesting that the first three are cinnabar (dominant lines sulphur and mercury), while the latter two are red ochre (dominant lines iron).

The ED-XRF spectra obtained on the lithic beads 41428a, 41428b, 41428d and 41010 are shown in Fig. 5A. The four beads were found in proximity (see Fig. 8).

The differences between the ED-XRF spectrum of the cinnabar layer and the one of the bead core are evident when comparing two zones on the 41010 sample, one covered in red and the other with the core exposed (see Fig. 5B). Mercury and sulphur lines are practically absent in the exposed region, the signal observed in the red region also shows the fluorescence lines of the iron present in the substrate. This evidence seems to exclude the possible mixing of cinnabar and ochre in the red pigment; in fact, the iron signal is mainly coming from the steatite bead core, while the sulphur and mercury signals come from the red cinnabar layer on the bead surface. Being the ED-XRF a volume technique (Angeli et al., 2019), the intensity of the iron lines observed is essentially the same in the two regions (covered and not covered in red).

The lithic bead 37482 gives an ED-XRF spectrum (not shown here) similar to the ones in Fig. 4A, although the colour of the cinnabar layer is darker than the others. In this case, the signal of mercury is slightly lower than for the 41010 sample (probably due to the lower thickness of the painted layer) but the iron signal is practically identical in both cases (Fig. 6). This evidence strengthens the hypothesis that cinnabar and ochre are not used together, at least in the samples that we have analysed.

The clay bead 14504 also evidences dark surface residues, but its fluorescence spectrum is much richer than the others (see Fig. 5C). Besides sulphur, potassium, calcium, titanium, mercury, and iron, the ED-XRF spectrum also shows clear fluorescence lines of rubidium, zirconium, and niobium. The fluorescence lines of these elements are typically found in volcanic sand and rocks (Tykot, 2017a, 2017b).

In Fig. 5D we show the comparison of the fluorescence spectra of the red-painted clay bead 14504 with two other clay beads from the exposed necklace (unpainted). Besides the sulphur and mercury signals, which are peculiar to sample 14504, the ED-XRF signals of Rb, Zr and Nb are practically identical in the three spectra. This might indicate the use of the addition of coarse sand of volcanic origin, collected along the shores of the lake, to prepare the clay mixture.

The spectrum of the decorated pottery sherd 50628 confirms that the red surface layer is made of cinnabar; the body of the shred shows the expected elements that are typically present in the clay (K, Ca, Ti, Mn,

Table 2
List of the artefacts analysed showing red pigment traces.

SAMPLE NO.	OBJECT	MATERIAL	LAYER	SQUARE	RESIDUAL TRACES
41428a	Bead	Steatite	I	A416	Cinnabar
41428b	Bead	Steatite	I	A416	Cinnabar
41428d	Bead	Steatite	I	A416	Cinnabar
41010	Bead	Steatite	I	A414	Cinnabar
37482	Bead	Steatite	I	A420/469	Cinnabar
36300	Bead	Spondylus Shell	I	A41	Cinnabar
36301	Bead	Spondylus Shell	I	A41	Cinnabar
36302	Bead	Spondylus Shell	I	A41	Cinnabar
14504	Bead	Clay	I	D191	Cinnabar
Exposed necklace	Bead	Seed	Und.	Und.	Cinnabar
36303	Raw Material Sample	Pigment	I	A41	Cinnabar
36169	Raw Material Sample	Pigment	I	A41	Cinnabar
39041	Raw Material Sample	Pigment	I	A488	Cinnabar
47121	Raw Material Sample	Pigment	I	A509	Ochre
47270	Raw Material Sample	Pigment	Post-holes	A464	Ochre
50628	Incised pottery sherd	Clay	I	D300 BIS	Cinnabar
15895	Quern	stone	I	D237	Cinnabar
19661	Quern	stone	I	A22	Cinnabar

Fe, Cu and Sr). Also in this case, the fluorescence lines of the volcanic sand elements Rb, Zr and Nb are visible, although weaker than the ones observed in the clay beads Fig. 7A).

The spectrum of the seed bead of the exposed necklace evidences the presence of lines of calcium, titanium, manganese, iron, copper, and strontium, on top of which we observe the ED-XRF fluorescence lines of sulphur and mercury from the surface residuals (Fig. 7B).

The ED-XRF spectra of the three *Spondylus* beads (36300, 36301, 36302) are dominated by the calcium lines since this element is the main component of the shell in the form of calcium carbonate. The lines of strontium are also very evident because Sr has the same chemistry of Ca and often substitutes it in the form of strontium carbonate. Also in this case, the characteristic lines of Hg, associated with the presence of cinnabar on the surface of the shell, are evident in the XRF spectrum (Fig. 7C).

The two querns (15895 and 19661) were used for processing the raw cinnabar, as evidenced by the red cinnabar residuals on their surfaces (Fig. 7D). However, the two spectra have significant differences. Besides the usual elements (K, Ca, Ti, Fe, Sr) and the lines of sulphur and mercury, the spectrum of item 19661 also shows the fluorescence lines of Rb, Zr, and Nb (see Fig. 7D). The reason for this difference is not clear; the two querns (15895 and 19661) seem visually similar, but only in one we observed the presence of Rb, Zr, and Nb, which would hint to a possible volcanic origin of the stone. Further mineralogical analysis will clarify the nature of the two stones.

5. Discussion

In most cases, the analyses have established that the brick-red substance detected in three of the five raw materials analysed (36303, 36169 and 39041 in Table 2) is mercury sulphide (HgS), known as cinnabar. The other two raw materials were identified as ochre (47121 and 47270).

As previously indicated, traces of red pigment were found on numerous ornamental items (Fig. 7), almost exclusively on lithic (5) and shell beads (3). The substance retains the original red pigment colour, although it has taken on a darker hue in some areas. Cinnabar is an unstable pigment in the long term and can turn black when exposed to light (Çamurcuoğlu, 2015; McCormack, 2000) or due to its long stay under water (Béarat et al., 2013). The latter seems the most likely reason given the particularity of the context of discovery.

Analyses have shown that a darker shade of red pigment, observed in some cases at Çatalhöyük (wall paintings and depositions) and in wall paintings from the Classical period, is caused by the use of a mixture of ochre and cinnabar, probably because the latter was in short supply. The preparation of the mixture seems to have been aimed at a desire to enhance the brilliance of the ochre and to accentuate its symbolic value (Çamurcuoğlu, 2015; Busacca, 2020). The hypothesis that this was the case at La Marmotta is less probable. The collected data show a deliberate choice to use cinnabar to decorate the ornaments. There is no evidence, at least in the samples analysed, of the possible use of a mixture of cinnabar and ochre since, as the XRF spectra show, the Fe detected in the red samples is compatible with the composition of the substrate material (i.e. Fig. 5B).

The five lithic beads analysed were obtained by processing steatite. One of these items (41010, Fig. 8A) can be considered a short cylinder

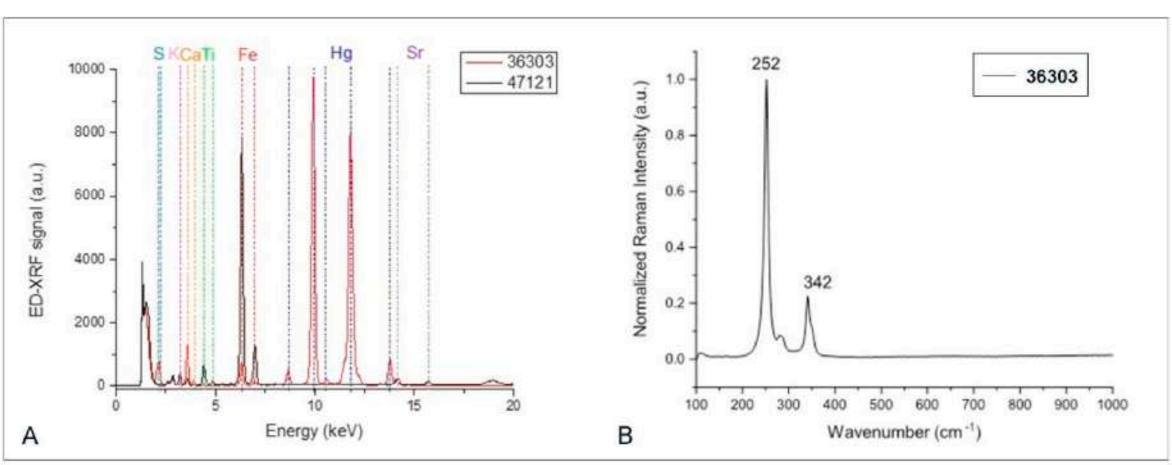

Fig. 4. ED-XRF spectra: A. Comparison between the ED-XRF spectra of two of the raw materials analysed (36303 – cinnabar and 47121 – red ochre); B. Raman spectrum of sample 36303.

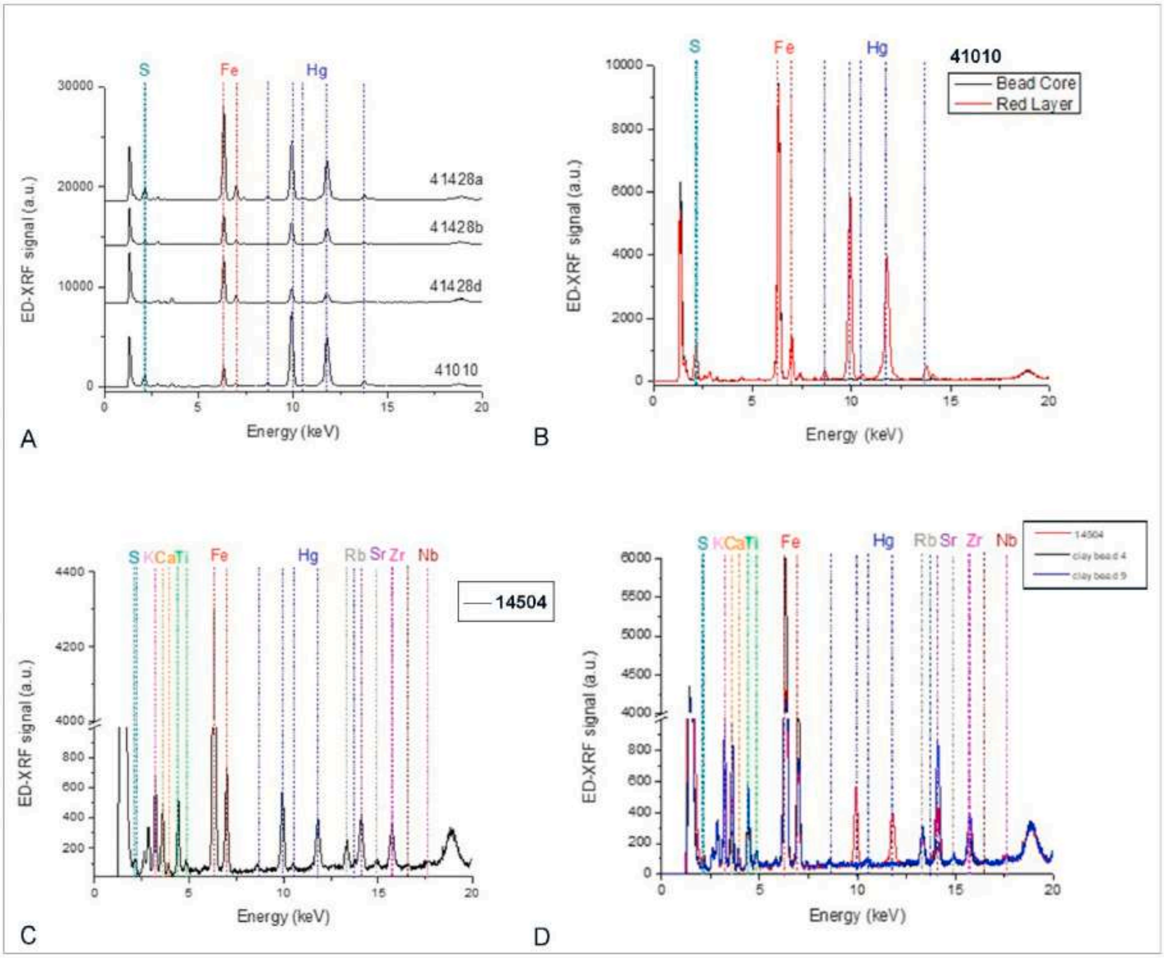

Fig. 5. A. Spectra obtained on the lithic beads 41428a, 41428b, 41428d and 41010; B. Comparison of spectra of two areas from the 41010 sample; C. Spectrum obtained from the clay bead 14504; D. Comparison of the fluorescence spectra of two clay beads (14504 and two beads from the exposed necklace).

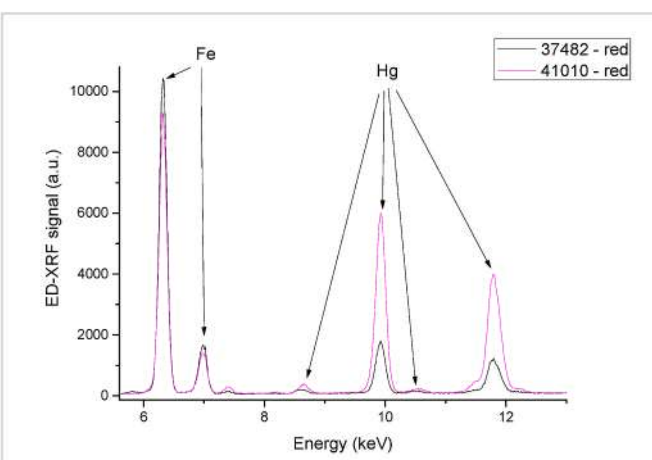

Fig. 6. Iron and mercury signals in samples 37482 (black curve) and 41010 (red curve).

based on the dimensions. The other four items (41428a, b, d, Fig. 9, and 37482) have a discoidal shape. Three of these last were part of a set of seven beads recovered together, probably belonging to the same jewel. One of these discoidal beads (41428d, Fig. 9d), in particular, shows a thick stratum of pigment covering its surface completely. This highlights a specific habit of the inhabitants of the village to cover the dark-coloured raw material of the lithic beads with a reddish pigment.

Conspicuous traces of cinnabar were also found on the surfaces of three ovoidal beads (36300, 36301, Figs. 8C, and 36302), with rather large dimensions. Based on the macroscopic inspection of the surface characteristics, these items are made in *Spondylus* sp. The use of these shells for making ornaments and their associated prestige value is known for the early Neolithic in the Mediterranean basin (Borrello and Micheli, 2004; Micheli, 2014; Windler, 2018). Some scholars claim that the Neolithic communities valued the *Spondylus* shell for its cultural significance and its white colour (Windler, 2019; Kurzawska and Sobkowiak-Tabaka, 2020).

At La Marmotta, on the contrary, the use of the red colour to cover their surfaces may reflect the intention to recall the intense purple colour, characterising the outer layer of certain *Spondylus* sp. (Dimitrijevic' and Tripkovic' 2006). It should be remembered that the red colour was used not only to cover the surface of these white-looking products but also for the dark lithic beads described above. Furthermore, the data highlights that only a small percentage of the total assemblage was dyed. This may indicate the intent to achieve an aesthetic effect by enhancing the appearance of only a few beads within a composition.

This habit probably did not concern only lithic and shell beads as evidenced by the coloured clay bead analysed (Fig. 8B). This last small bead with cinnabar traces on the surface is similar in dimension to the cylindrical lithic bead.

The seed bead analysed (exposed necklace, Fig. 8D:1; Petrinelli Pannocchia and Vassanelli, 2023) is small in dimension and preserves its

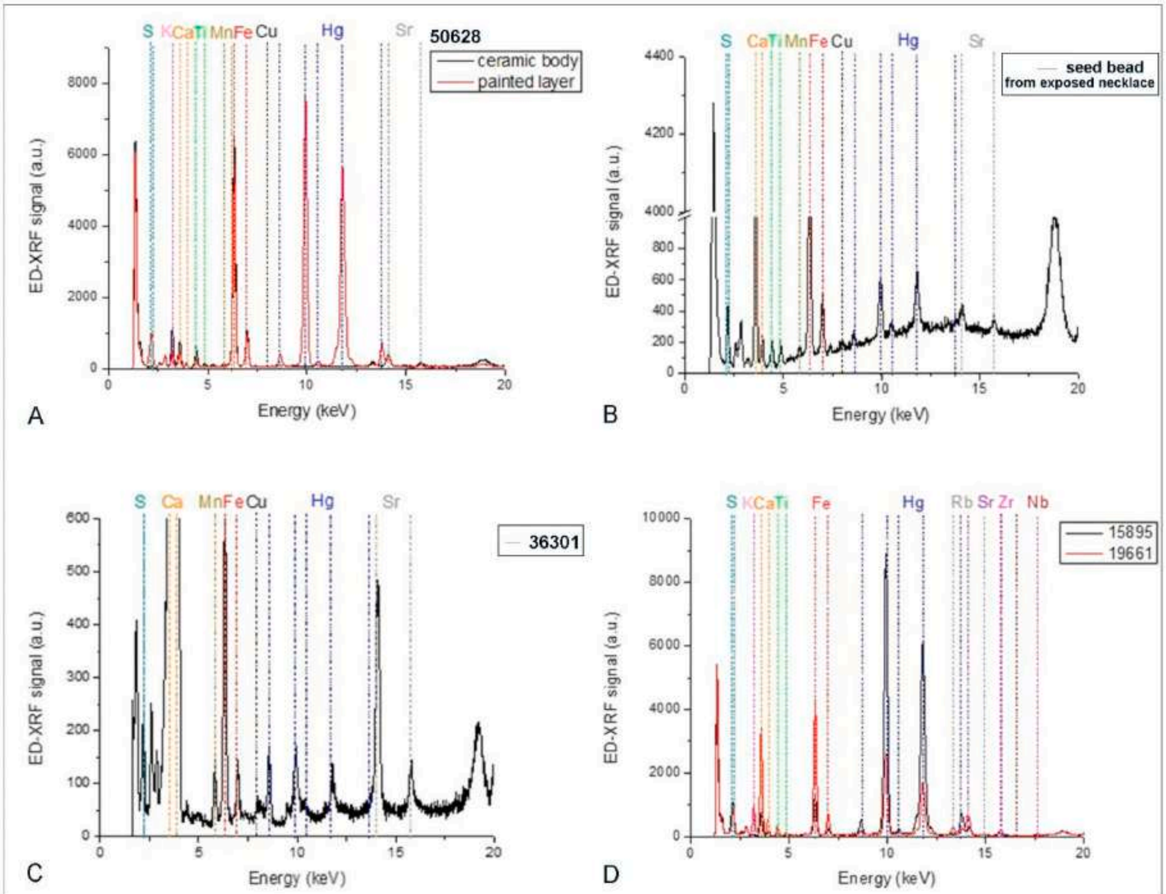

Fig. 7. A. Comparison between the ED-XRF spectra of the painted layer (red) and ceramic body (black) for sample 50628; B. ED-XRF spectrum of the seed bead in the exposed necklace; C. ED-XRF spectrum of the surface of *Spondylus* bead 36301. The other two (36300 and 36302) are very similar; D. Comparison of the ED-XRF spectra of the surface of the two querns 15895 (black) and 19661 (red).

automorphic ovoidal shape. Compared to the beads described up to now, the limited amount of cinnabar residue on this item makes it difficult to determine if it was originally coated with cinnabar or if it came into contact with pigment-coated materials.

In a few beads, traces of red pigment were found even inside the perforations. This was observed in two items, a *Spondylus* shell bead (36301, Fig. 8C) and a clay bead (14504, Fig. 8B). The presence of the pigment inside the perforations suggests possible dyeing of the thread with the red colour.

As mentioned above, the earliest use of cinnabar to treat textiles in the Mediterranean region is testified by the discovery at Peñacalera cave in the Sierra Morena hills, Southern Spain. The colouring of the thread used to tie the beads at La Marmotta could be considered indirect evidence of dyeing fabrics that could chronologically anticipate the appearance of this practice in the Mediterranean area.

Due to its nature, the ore must be ground into a fine red powder before being mixed with other components to make a spreadable paste or applicable liquid. The presence of three cinnabar samples of raw materials (36303, 36169, 39041) and two querns (15895 and 19661) indicate that the cinnabar was prepared at the site and was extensively used by this Neolithic community. Indeed, the analysis of the residues in the incisions of a ceramic sample (56628, Fig. 10) testifies to the use of cinnabar to heighten the chromatic effect of the pottery decoration.

As previously mentioned, similar use has been found in the late Neolithic levels at the Grapčeva site in Croatia. This is one of the few sites where mercury sulphide was used as a pigment to paint the ceramics after firing (Kaiser and Forenbaher, 2016). For the Italian peninsula, the use of cinnabar by the first agro-pastoral communities as a filler for impression and engravings has been only supposed until now (Grifoni Cremonesi, 2004). According to the data reached from the XRF analysis, the La Marmotta site represents the oldest analytically proven use of cinnabar for filling pottery decorations in the Italian peninsula.

The results of the analyses revealed that in addition to cinnabar, the craftsmen used ochre as a pigment. The use of both pigments was observed in other sites as well, where they were employed for different purposes (Gajić-Kvaščev et al., 2012). At La Marmotta, among the items analysed red ochre was detected only as raw material, therefore its use remains to be clarified.

Several vessels discovered at the site were described as having internal surfaces coated with ochre (Fugazzola, 2002). This could testify to the use of pottery to prepare or store red pigment, as testified by the vessels discovered in the Vinča sites (Mioč et al., 2004). Further analyses on a wider spectrum of archaeological materials could clarify how the villagers processed and used the different pigments, providing additional information on the possible role of the red colour in the site's life. However, the absence of ochre on the surfaces of the ornaments here analysed may suggest different uses of the two substances.

As previously mentioned, the lack of extensive and systematic archaeometric analysis in the Italian peninsula might have led to a misclassification of red pigments, which are often generically labelled as ochre. Consequently, information regarding the utilisation of cinnabar during the Neolithic is far from being complete. The Neolithic context accounts reveal similarities with the use of cinnabar at La Marmotta. Among the farming communities, this red pigment seems to have had a specific symbolic value tied to the body (La Vela and Grotta Patrizi) and to certain products: pottery (Grotta Patrizi, Grotta dei Cervi and La

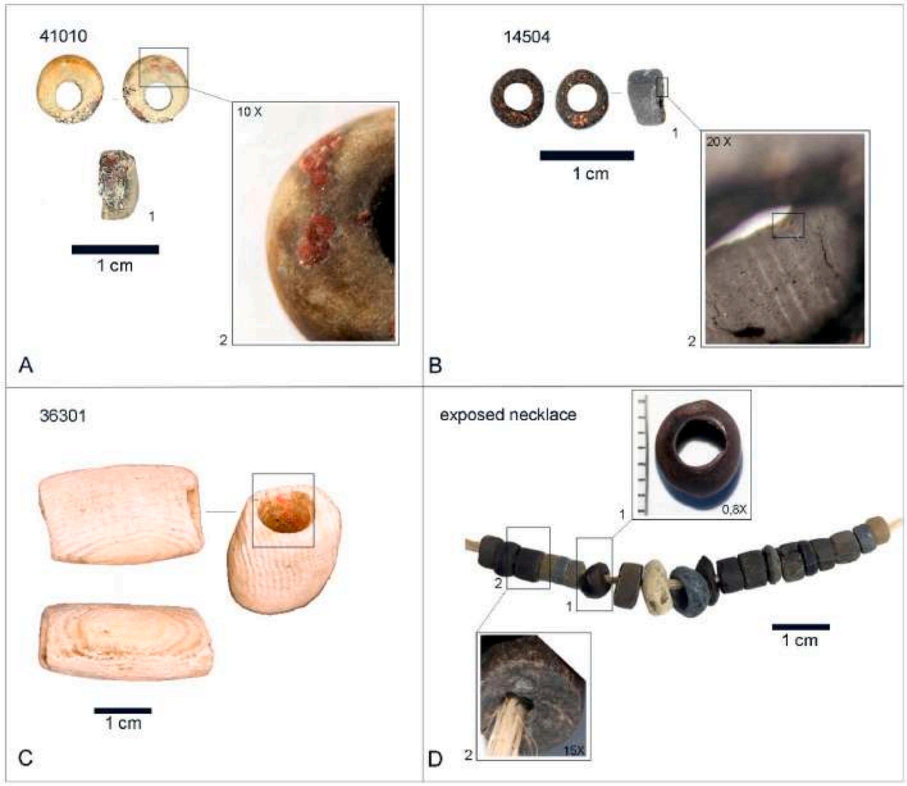

Fig. 8. A. Steatite short cylinder bead 41010 with cinnabar residue; B. Clay bead 14504; C. *Spondylus* shell bead (36301); D. Exposed necklace with detail of a seed bead (1) and a clay bead (2).

Fig. 9. The seven steatite beads recovered together (on top), and various images of bead number 41428d, where the cinnabar residues that almost entirely cover the surface are clearly visible.

Marmotta) and ornaments (La Vela and La Marmotta).

During the Copper Age, the exploitation became organised, as showed by the evidence of southern Tuscany (see next paragraph), and the intensive mining ensured a constant and secure supply of this red pigment. The practice of sprinkling the buried bodies with red pigment seems to have continued, often limited to the skull as if it was believed the representative part of the essence of an individual, as attested in numerous hypogeum burials in Latium. Among the grave goods, the lithic weapons seem to replace the previous connection between personal ornaments and red colour. Conversely, in Tuscany, the number of sites showing the use of cinnabar declines progressively, especially in the northern part, where a preference for the white colour seems privileged, as testified by the numerous marble beads recovered in the Copper Age burials (Vassanelli et al., 2023).

5.1. The presence of cinnabar in the Italian Peninsula: supplying areas

In Italy, several deposits (88) reveal the presence of cinnabar ore, mainly found in volcanic environments and hot spring deposits (mindat.org). These are almost exclusively along the central-northern Tyrrhenian coast, roughly from Lazio to Liguria region. Only two localities attest to the presence of cinnabar in the southern part of the peninsula: Monte La Mula, in the province of Cosenza, and Palinuro, in the Aeolian islands (Fig. 11).

The south of Tuscany is the closest region where cinnabar prehistoric deposits can be found. This mineral is present on Monte Amiata, in the inner part of the region, and on the Monti dell'Uccellina, along the coast. The distance between La Marmotta and both areas is about 100 km as the crow flies.

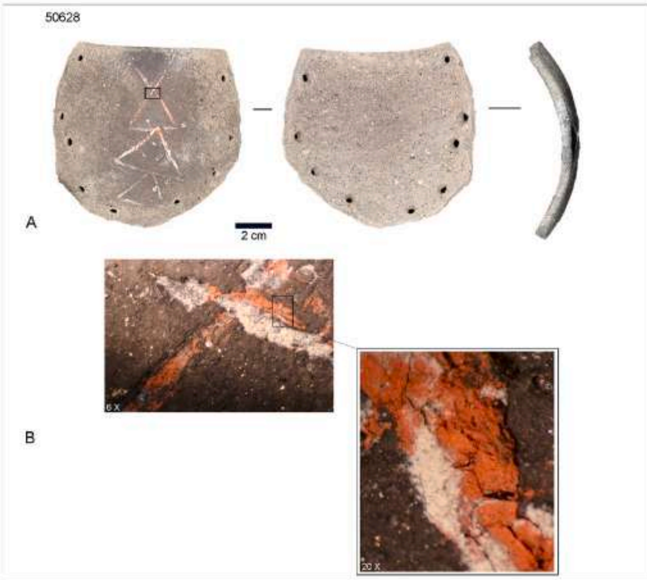

Fig. 10. A. The incised pottery sherd analysed (50628); B. Detail of the incised decoration filled with cinnabar and a calcareous whitish dough.

Until 1970, Monte Amiata was one of the largest mercury districts in the world (Rimondi et al., 2012). Numerous prehistoric artefacts recovered in several mines, located on the slopes of this mountain, attest to their exploitation from the Neolithic to the Etruscan periods, and then resumed from the Medieval age to the present. The radiocarbon dating of some oak wood mallets found at the Solforate mine, in the municipality of Piancastagnaio (Siena), traces the first exploitation of the Monte Amiata district to the second half of the fifth millennium BC (Volante et al., 2019).

Poggio di Spaccasasso, in Monti dell'Uccellina, was used as a burial site from the first half of the fourth millennium BC to the early centuries of the second millennium BC. Evidence has been discovered in a burial chamber beneath a level that contained a Copper Age burial, which suggests that the cavity was originally excavated for mining purposes (Volante, 2018).

Concentrations of calcareous debris and fragmentary heavy lithic tools identified during surveys on the flanks of the hills have confirmed cinnabar mining activities. Several fire-places at the base of mining fronts suggest the use of fire-setting techniques to cause the crumbling of the limestone that encased the red mineral. As several authors have pointed out, this is an inappropriate method of extraction because cinnabar becomes toxic when heated to temperatures above 600 °C. It is therefore likely that by the second half of the fifth millennium BC, the miners had already acquired impressive knowledge and skills in the extraction of ores. The radiocarbon date of (LTL15348A) 5565 $\pm$ 45 BP (Volante et al., 2019), obtained on heather charcoal found in one of the hearths, follows that of the Solforate mine of Monte Amiata. This indicates that the two supply areas were active simultaneously during the late Neolithic, suggesting a demand and, consequently, the existence of a trade network that distributed this mineral pigment along the Tyrrhenian area. However, it cannot be excluded the two districts were known and exploited, even if not systematically, in an earlier period.

Given the short distance, southern Tuscany is the favourite candidate

Fig. 11. Maps with cinnabar supply areas in central Italy (adapted from mindat.org) with some localities mentioned in the text. Hexagons: single area with correct (red) or estimated (blu) coordinates; Circles: areas with multiple supply zones (yellow: >10; green: <10).

as the area of origin of the recovered pigment. However, future isotopic analyses of the cinnabar found at La Marmotta will allow the precise identification of the supply areas. In any case, the import of cinnabar is another element that underlines the importance of the site within an extensive exchange network of raw materials, ideas, and traditions that crossed the Italian peninsula from the earliest phases of the Neolithic.

6. Final consideration and future perspectives

The exploitation of cinnabar is closely linked to specific technological know-how that foresees its detection, extraction, and processing. The framework available to us shows a progressive introduction of this pigment in the Mediterranean basin, from the east to the west. The use of cinnabar in the Near East (Çatalhöyük), in the seventh millennium BC, followed by its appearance in the Balkans in the sixth millennium BC (Vinča group), and then in the western Mediterranean (Italy and then Spain), during the fifth millennium BC, suggests a pattern of cultural diffusion and exchange that seems to follow the arrhythmic spread of the Neolithization wave (Guilaine, 2000). This aspect is relevant because the occupation of new territories by Neolithic communities did not only mean the presence of new human populations and the domestication of different animal and plant species. There are several objects, tools, techniques and even pigments that were transported or the knowledge of which was passed on from one generation to the next. Cinnabar seems to be one of the cases.

The widespread use of cinnabar at La Marmotta, as well as in other early Neolithic contexts on the Iberian peninsula, shows us the importance it had for those early agricultural and pastoral communities. Once the territories were occupied, it was necessary to identify potential sources of supply. This could be achieved through independent prospecting or establishing relationships with local hunter-gatherer groups. A recent study of genomic data focusing on the spread and migration of human populations across Europe (Olalde et al., 2015), suggests the likely derivation of Iberian Cardial people from an ancient Balkan population. This data may also be useful for understanding the spread of cinnabar in Western Europe. Indeed, the earliest Iberian site with traces of cinnabar is Cova de l'Or, one of the earliest Cardial sites in the Iberian region.

The site of La Marmotta was also established by settlers connected to the Impressed-Cardial ware (Level I, Delpino, 2020). This style of pottery lasts in the upper level (Level II), characterized by the painted and incised ware of the Sasso-Fiorano style (Delpino, 2020), from which artefacts with cinnabar traces come. The use of cinnabar may be part of a "traditional set of knowledge" that has continuity over time. Under this perspective, the presence of cinnabar could be considered a proxy for following specific Neolithic paths. The inhabitants of La Marmotta probably obtained this exotic element through the vast exchange network of raw materials, ideas and traditions that crossed the entire peninsula since the earliest phases of the Neolithic. However, further isotopic analysis will be required to clarify the source of this ore. Data suggest that cinnabar occurs throughout the Tyrrhenian area; therefore, it is plausible to assume that its use during prehistory is currently underestimated due to the lack of extensive and exhaustive application of archaeometric analysis. In the absence of specific analyses red pigments are often referred to as ochre.

In conclusion, our research shows that La Marmotta represents, to date, the earliest use of cinnabar in the western Mediterranean and stresses the need to increase the number of sites analysed in the future in order to define a more detailed chrono-cultural picture of the diffusion of red pigments in the Italian peninsula, as well as in the whole Mediterranean basin.

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CRediT authorship contribution statement

Cristiana Petrinelli Pannocchia: Conceptualization, Data curation, Formal analysis, Writing – original draft, Writing – review & editing. **Alice Vassanelli:** Data curation, Formal analysis, Writing – original draft, Editing of tables and figures of artefacts, Writing – review & editing. **Vincenzo Palleschi:** Formal analysis, Writing – review & editing. **Stefano Legnaioli:** Formal analysis, Writing – review & editing. **Mario Mineo:** archaeological fieldwork, Site presentation. **Gerard Remolins Zamora:** Distribution plan of the artefacts analysed. **Niccolò Mazzucco:** Site presentation, Writing – review & editing. **Juan F. Gibaja:** Investigation, Site presentation, Writing – review & editing, Funding acquisition.

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.quascirev.2024.108746.

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