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REVIEW

Pigments — Mercury-based red (cinnabar-vermilion) and white (calomel) and their degradation products

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Abstract

This article summarises the history of cinnabar, from its first uses in burials to modern oils on canvas. After a brief introduction on mercury and contamination issues, the article gets to the heart of the topic. First, mercury-based minerals significant for studying pigments, *i.e.* cinnabar, metacinnabar, hypercinnabar and calomel, are presented. Structural information and properties precede an overview of the geographic distribution of cinnabar deposits. The following section addresses the multiple uses of cinnabar, divided into funerary use, decorative use, lustre and Chinese lacquer production. The use of cinnabar for writing (ink), medicine and cosmetics is briefly described, and a shortlist of uncommon finds is further provided. The following section approaches inherent but less known topics such as cinnabar procurement, trade, production technology, application and alteration. An entire section is dedicated to calomel before concluding with an overview of the analytical methods for the characterisation and provenance investigation of cinnabar.

Keywords Cinnabar-metacinnabar-hypercinnabar · Vermilion and pigment analysis · White mercury and calomel, corderoite and terlinguaite · Archaeometry and archaeology

Premise

This Topical Collection (TC) covers several topics in the field of study, in which ancient architecture, art history, archaeology and material analyses intersect. The chosen perspective is that of a multidisciplinary scenario, capable of combining, integrating and solving the research issues raised by the study of mortars, plasters and pigments (Gliozzo et al. 2021).

The first group of contributions explains how mortars have been made and used through the ages (Arizzi and Cultrone 2021; Ergenç et al. 2021; Lancaster 2021; Vitti 2021). An insight into their production, transport and on-site organisation is further provided by DeLaine (2021). Furthermore, several issues concerning the degradation and conservation of mortars and plasters are addressed from practical and technical standpoints (La Russa and Ruffolo 2021; Caroselli et al. 2021).

The second group of contributions is focused on pigments, starting from a philological essay on terminology (Becker 2021). Three archaeological reviews on prehistoric (Domingo Sanz and Chieli 2021), Roman (Salvadori and Sbrolli 2021) and medieval (Murat 2021) wall paintings clarify the archaeological and historical/cultural framework. A series of archaeometric reviews illustrate the state of the art of the studies carried out on Fe-based red, yellow and brown ochres (Mastrotheodoros et al. 2021); Cu-based greens and blues (Švarcová et al. 2021); As-based yellows and reds (Gliozzo and Burgio 2021); Pb-based whites, reds, yellows and oranges (Gliozzo and Ionescu 2021); Hg-based red and white (this paper) and organic pigments (Aceto 2021). An overview of the use of inks, pigments and dyes in manuscripts, their scientific examination and analysis protocol (Burgio 2021) as well as an overview of glass-based pigments (Cavallo and Riccardi 2021) are also presented. Furthermore, two papers on cosmetic (Pérez-Arantegui 2021) and bioactive (antibacterial) pigments (Knapp et al. 2021) provide insights into the variety and different uses of these materials.

Introduction

Mercury is the only metal (chalcophile) that occurs in liquid form at room temperature. Due to its colour and form, it has

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also been called liquid silver, *hydrargyrum* and, especially, quicksilver, a term, this latter, mainly used by geologists involved in the study of its occurrence and mode of transport (especially in the 1940s/1950s, *e.g.*, Dreyer 1940a, 1940b; Ross 1942; Krauskopf 1951).

Mercury is classified as a rare element as it is present in low concentrations in the Earth's upper (~0.05 ppm), middle (0.0079 ppm) and lower (0.014 ppm) crust (Rudnick and Gao 2004).

Its multiple uses ranged from measurement tools such as thermometers, barometers and pressure-sensing devices to dental amalgam, batteries, lubrication oils, lamps and other industrial processes and products.

Despite its varied and widespread use, mercury is toxic, and, for this reason, it has been banned for paints since 1990, while mercuric chloride is still used as a pesticide and a disinfectant. The WHO identifies mercury "*as one of the top ten chemicals or groups of chemicals of major public health concern*", that "*may have toxic effects on the nervous, digestive and immune systems, and on lungs, kidneys, skin and eyes*" (https://www.who.int/news-room/fact-sheets/detail/mercury-and-health). However, natural cinnabar is less toxic than organic mercury (methyl mercury or dimethyl mercury) or liquid mercury. At the same time, the process for mercury extraction can be highly toxic (esp. mercury vapours) as well as the accumulation of methylmercury ($[CH_3Hg]^+$), which is mainly formed by the action of anaerobic bacteria[1] (see, *e.g.*, Manceau et al. 2015 and Supplementary materials Appendix 1). Environmental issues have stimulated the creation of an extensive body of literature both focused on specific case studies related to the mining exploitation of geological deposits and of a more general nature, aimed at unravelling the main processes (Burkstaller et al. 1975; Rytuba 2000, 2003; Horvat 2005; Holley et al. 2007; Levin 2014; Chen et al. 2017).

The first category includes such a large number of research papers that it is impossible to mention them all in this contribution; however, it is useful to point out two important aspects. Firstly, mercury pollution has an extremely vast range. Therefore, it is possible to find clear signs of pollution in soils, waters, flora, fauna and atmosphere at considerable distances from the extraction area, especially in correspondence of river courses. Secondly, ancient and modern working places are often mentioned and investigated in these studies, near and far from the mining district; therefore, they represent "a mine of information" for archaeometric studies on provenance and technology.

Indeed, mercury poisoning is thoroughly documented by ancient literary sources and archaeometric investigations. For example, cases are reported for the Late Neolithic/Chalcolithic (5400–4100 BP) population of southern Portugal (Emslie et al. 2015), for miners buried at the archaeological site of Ranas in the Sierra Gorda Querétaro (Mexico; Serrana culture)[2] and for Southern Denmark and Northern Germany medieval population (Rasmussen et al. 2015).

This review will tackle the "cinnabar" theme from multiple perspectives, based above all on its characteristics and use. The main objective is to provide the broadest possible framework for the study of cinnabar rather than address each topic in detail. While the dimensions of a single article would not allow a comprehensive discussion, it is possible to provide an overview of the state-of-the-art to guide both the study and the bibliographic search.

Mercury in naturally occurring minerals

Mercury compounds mainly include sulphides (*e.g.*, cinnabar, HgS), chlorides (*e.g.*, mercuric chloride, $HgCl_2$ and mercurous chloride Hg_2Cl_2 known as calomel), halides (*e.g.*, terlinguaite, Hg_2ClO; corderoite and kenhsuite, $Hg_3S_2Cl_2$), oxides (*e.g.*, montroydite, HgO), along with natural alloys (*e.g.*, leadamalgam, $HgPb_2$), phosphates (*e.g.*, artsmithite, $Hg_4Al(PO_4)_{1.74}(OH)_{1.78}$), silicates (*e.g.*, edgarbaileyite, $Hg_6Si_2O_7$), arsenates (*e.g.*, chursinite, Hg_2AsO_4), carbonates (*e.g.*, peterbaylissite, $Hg_3(CO_3)(OH)•2(H_2O)$) and acetates (*e.g.*, mercuric acetate, $HgC_4H_6O_4$).

The sulphides are the most abundant, and, among them, the three HgS polymorphs—namely cinnabar, metacinnabar and hypercinnabar—prevail. Among chlorides and halides, calomel, terlinguaite and kenhsuite are significant in the Cultural Heritage studies because they are the only ones found in artworks to date. Among natural alloys, the amalgams have a technological and historical interest (Giumlia-Mair et al. 2014) that, however, go beyond the objectives of this review.

A shortlist of research articles that may help study Hg-bearing phases is provided in Supplementary materials Appendix 1.

Cinnabar, metacinnabar, hypercinnabar and calomel

Cinnabar (α-HgS) is bright red mercury sulphide (HgS; Fig. 1), also named κιννάβαρι (*kinnabari*) in Greek, *minium cinnabaris* in Latin and *šangarf* or *sīm- šangarf* in Persian (the Arabic *zinjifra* should correspond to red lead). On the etymology of cinnabar and the corresponding names in Latin, Persian and Arabic, the reader is referred to the detailed study by Rosół (2018) and Becker (2021 in this TC). On the distinction between inorganic cinnabar and dragon's blood, or

[1] For example, the danger of high levels of mercury in fish is a sadly known phenomenon.

[2] A well-known cinnabar Mesoamerican mining area between the 2nd and 14th century AD (Ávila et al. 2014).

Fig. 1 Macrophotos of **A** cinnabar from the Almadén Mine, Ciudad Real, Castile-La Mancha, Spain (specimen and photo: fabreminerals.com – Mindat.org Photo ID: 56508); **B** cinnabar from the Tongren Mine, Bijiang District, Guizhou, China (Arkenstone specimen. Photo credits: Rob Lavinsky, irocks.com – Mindat.org Photo ID: 206087); **C** cinnabar from Pozo de San Teodoro, Almadén district (specimen and photo: fabreminerals.com – Mindat.org Photo ID: 942728); **D** cinnabar crystals on pyritic matrix from Las Cuevas Mine, Almadén district, Spain (photo credits: Carlos Gonzalez Bargueño – Mindat.org Photo ID: 867406); **E** metacinnabar with pyrite from El Entredicho Mine, Almadén district (photo credits: Borja Sainz de Baranda Graf – Mindat.org Photo ID: 937144); **F** distorted cluster of cubo-octahedral metacinnabar crystals, intermixed with hypercinnabar, on quartz matrix, from the Mount Diablo Mine, Clayton, CA, USA (photo and specimen, Jeff Weissman - ID 1035520); **G** intermixed, highly modified and distorted trigonal/hexagonal appearing hypercinnabar crystals with cubic metacinnabar crystals, with pale yellow flaky copiapite crusts on crystalline quartz (photo and specimen, Jeff Weissman - Mindat.org Photo ID: 1035520); **H** calomel from Terlingua Mining District, TX, USA (Arkenstone specimen. Photo credits: Rob Lavinsky, irocks.com – Mindat.org Photo ID: 716834). The CC-BY license does not supersede previously copyrighted material; therefore, these images remain under owners' copyright

organic cinnabar, the reader may consult both Rosół (2018) and Trinquier (2013).

Studied since the early decades of the 1900s (Buckley and Vernon 1925; Olhausen 1925; Ramsdell 1925), its structure has been further provided by Berry and Thompson (1962), Auvray and Genet (1973) and Schleid et al. (1999). Cinnabar is a trigonal phase (Table 1; Fig. 2), constituted by helical chains of -S-Hg-S-Hg-.

Metacinnabar (β-HgS) is an isometric black mineral with a zincblende structure (Table 1; Figs. 1–2), whose structure has been investigated by Lehman (1924), Wyckoff (1963) and Ballirano et al. (2013).

Table 1 Space groups and structural parameters of Hg-minerals mentioned in text

	Space group	*a*	*c*	V	*Z*	Reference
Cinnabar (trigonal)	$P3_221$ (154)	4.16	9.54	142.977	3	Buckley and Vernon 1925
		4.15	9.51	141.843	3	Ramsdell 1925
		4.14	9.49	-	3	Berry and Thompson 1962
		4.148	9.492	-	-	Mikolaichuk and Dutchak 1965
		4.145	9.496	141.293	3	Auvray and Genet 1973
		4.1489 (2)	9.4947 (5)	-	-	Schleid et al. 1999
		4.1489 (2)	9.4947 (5)	-	3	Ballirano et al. 2013
Metacinnabar (isometric)	*F* -43*m* (216)	5.858	=	201.024	4	Lehmann 1924
		5.8517	=	200.376	4	Wyckoff 1963
		5.850 (4)	=	-	-	Mikolaichuk and Dutchak 1965
		5.8461 (4)	=	-	-	Ballirano et al. 2003
Hypercinnabar (hexagonal)	-	6.86 (1)	14.07 (7)	-	-	Mikolaichuk and Dutchak 1965
		7.01 (3)	14.13 (7)	601.32	12	Potter and Barnes 1978
Calomel (tetragonal)	*I*4/*mmm* (139)	4.47	10.89	217.592	2	Havighurst 1926
		4.464	10.9	217.208	2	Hylleraas 1926
		4.478	10.91	218.773	2	Wyckoff 1963
		4.482 (2)	10.910 (3)	-	-	Dorm 1971
		4.4795 (5)	10.9054 (9)	218.83 (8)	2	Calos et al. 1989

Hypercinnabar (γ-HgS) is a hexagonal phase (Table 1; Fig. 2), first identified by Mikolaichuk and Dutchak (1965). Later, it was found by Protobyakonova et al. (1971) in Russia and approved by the IMA commission in 1978 (Potter and Barnes 1978) as a new HgS polymorph. This black mineral -which may contain minor Fe- has an undefined hexagonal structure (Table 1; Fig. 2). An intermediate pseudocubic phase (XHgS) has been further observed by Bell et al. (2010) between 467 and 552 K, *i.e.* below the metacinnabar → cinnabar transition temperature.

Calomel (Hg_2Cl_2) is likely the most important compound in which Hg is univalent. From white to yellowish-grey, grey and brown, its tetragonal structure (Figs. 1 and 3) was first investigated by Havighurst (1926) and Hylleraas (1926) and then later by Wyckoff (1963), Dorm (1971) and Calos et al. (1989).

It is curious to note that the English word calomel (καλομέλανο, calomelano in Greek) derives from the Greek καλός (kalos, beautiful) and μέλανος (melanos, black). Swiderski (2008) narrates that according to Pereira (1849-1850) the name is "*referred to Dr. Theodore de Mayerne's black servant, who was so skilled in preparing the drug that Mayerne called it "beautiful black" in praise of both drug and servant*". Soon after, Swiderski reconstructs the history of the name's assignment and, above all, traces the significant correlation between the term and the production of mercury sublimates. Considering the medicinal use of sublimates, the author observes the convenience of a term recalling beauty and honey (μέλι, meli, honey) compared to one that explicitly mentions mercury. The latter would undoubtedly have been more frightening for a patient than the former. In any case, while the name calomel does not refer to the natural whitish mineral, it reflects the characteristic blackening obtained with exposure to light (photosensitivity) or ammonia.

Apart from the literature mentioned above, the phase relations in the mercury–sulphur system have been chiefly investigated by Dickson and Tunell (1954, 1959), Kullerud (1965), Ohmiya (1974) and, above all, by Potter and Barnes (1978), Mel'chakova and Kiseleva (1990), Sharma et al. (1993) and Ballirano et al. (2013). Stable under ambient conditions, cinnabar converts to metacinnabar between 315 and 345±2° C (depending on Hg%; see Potter and Barnes 1978) or even higher at 673° K in an oxidising atmosphere (=399.8° C in Ballirano et al. 2013). In turn, metacinnabar converts to hypercinnabar between 470 and 481° C. Above these temperatures, hypercinnabar is stable up to 788° or 804° C, still depending on Hg%. The diagrams in Fig. 4 show that the stability of the various phases is directly linked to the stoichiometry (*i.e.* purity) of the HgS. Moreover, the variation in temperatures is also due to different experimental setups (*e.g.*, *in situ*, *ex situ*, under vacuum, in an oxidising atmosphere, etc.).

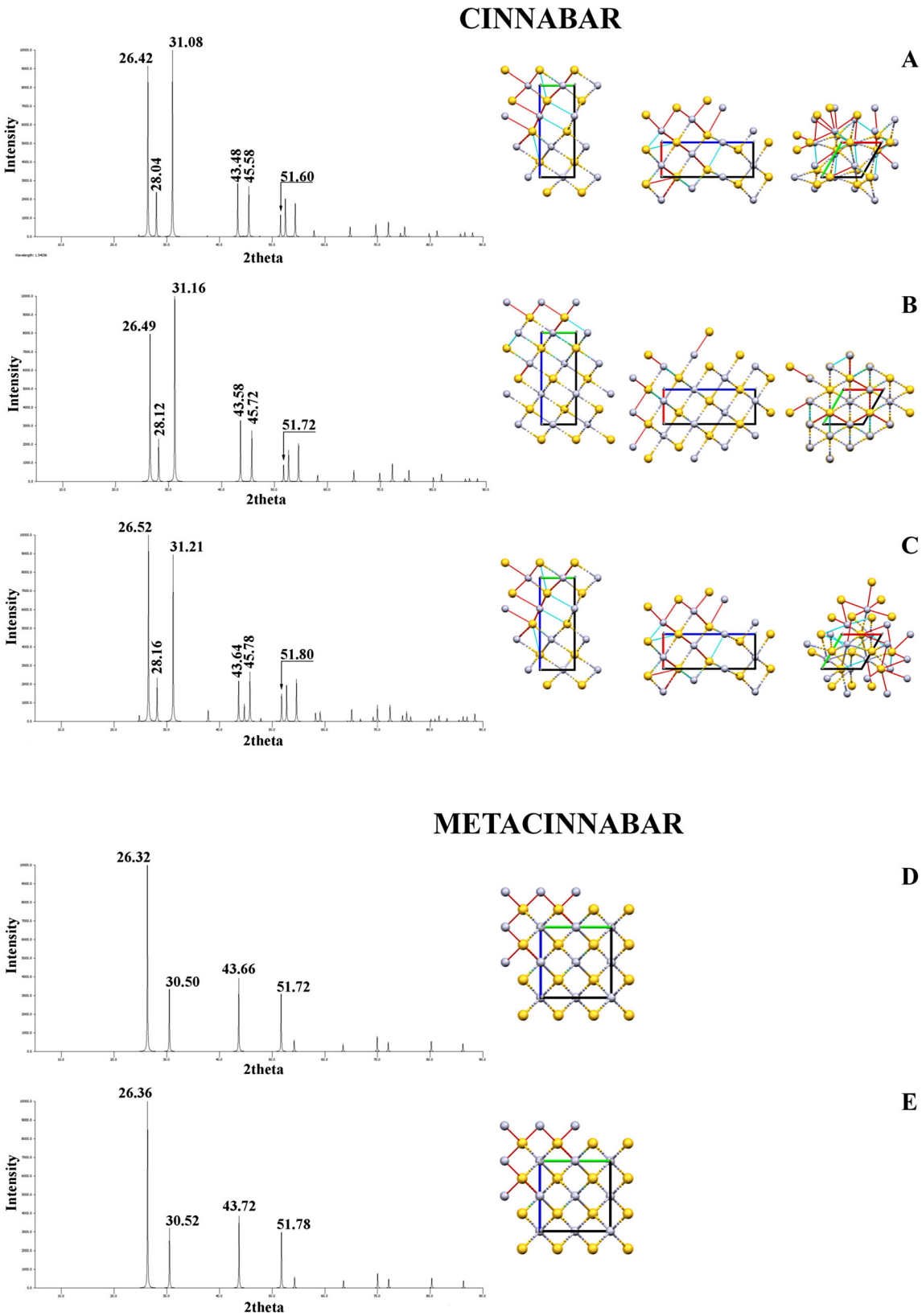

Fig. 2 XRD patterns (Cu-Kα) and (ball-stick) packing along a, b and c (unit cell shown) of cinnabar and metacinnabar. **A** Buckley and Vernon (1925); **B** Ramsdell (1925); **C** Auvray and Genet (1973); **D** Lehmann (1924); **E** Wyckoff (1963)

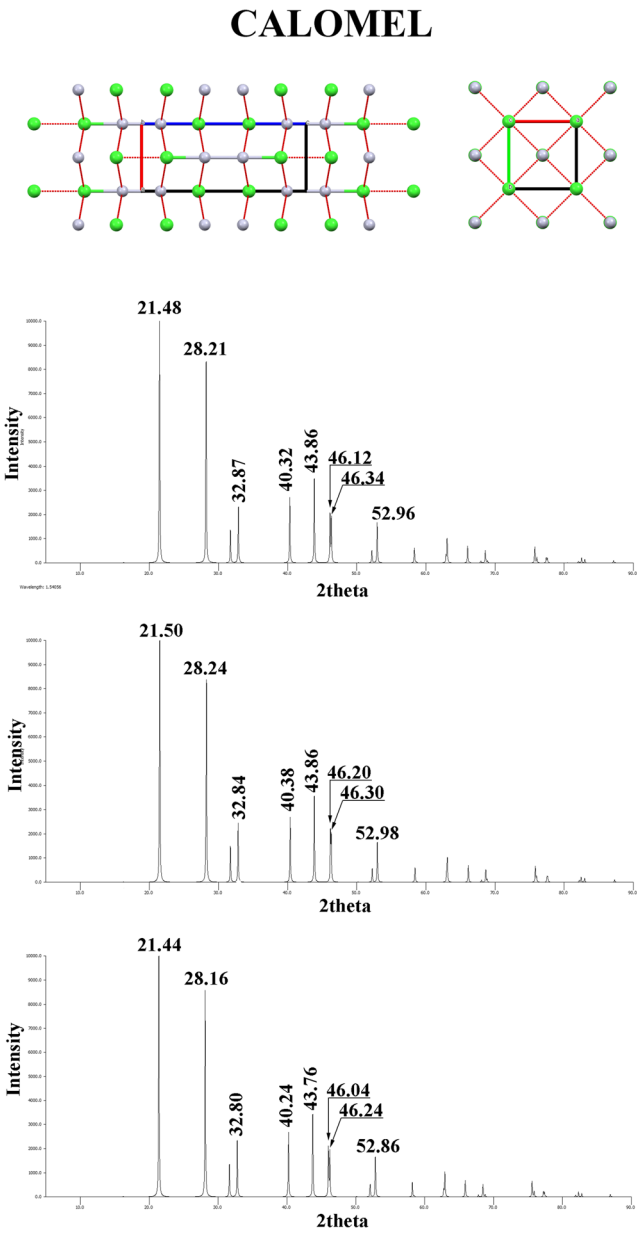

Fig. 3 XRD patterns (Cu-Kα) and (ball-stick) packing along b and c (unit cell shown) of calomel. **A** Havighurst (1926); **B** Hylleraas (1926); **C** Wyckoff (1963)

Beyond these thermal transformations, it is worth underlining that metacinnabar can also form at room temperature from mercury iodide dissolution in an aqueous solution of potassium sulphide (Ballirano et al. 2013).

The geographic distribution of cinnabar deposits

The geographic distribution of cinnabar is worldwide. Mindat database enlists 2694 localities in 66 countries, including both outcrops and mines: Afghanistan (27), Argentina (2), Australia (4), Austria (118), Azerbaijan (1), Belgium (9),

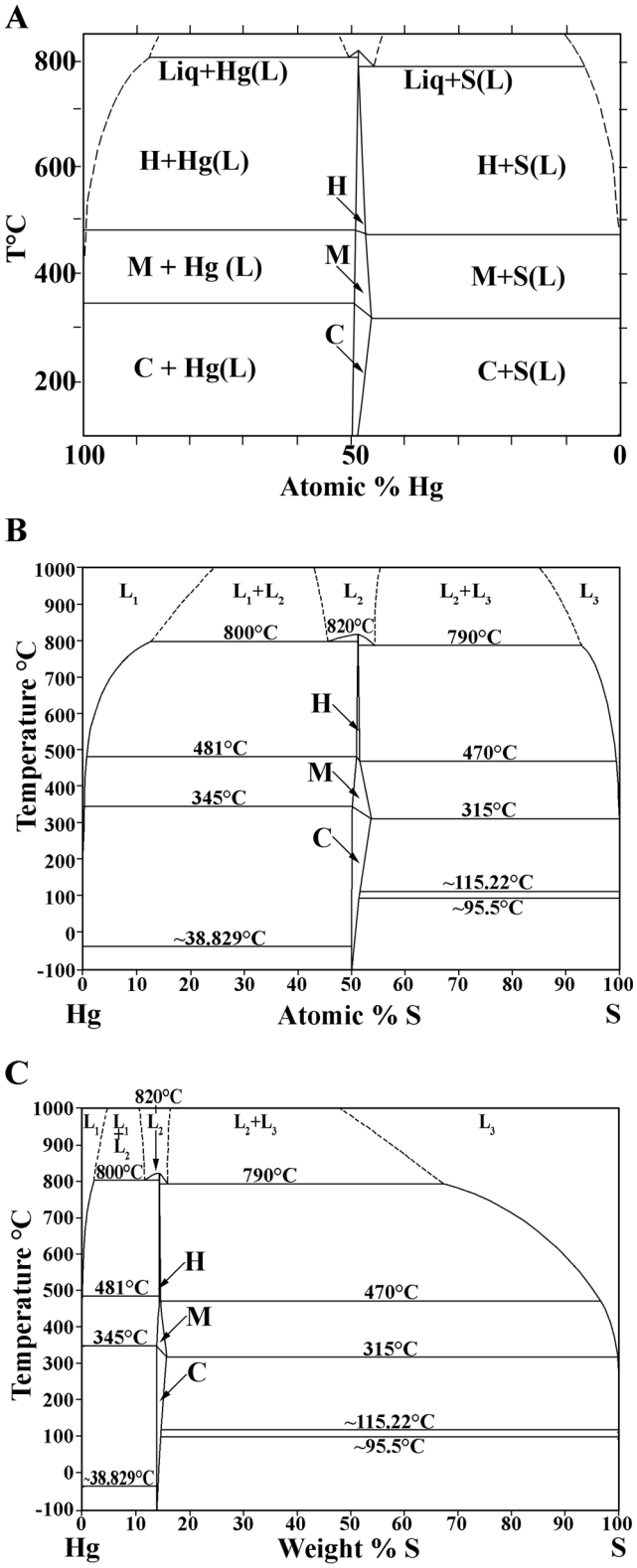

Fig. 4 Phase relations. **A** Modified from Potter and Barnes (1978); **B**–**C** simplified after Sharma et al. (1993) (C=cinnabar; M=metacinnabar; H=hypercinnabar)

Bolivia (12), Bosnia and Herzegovina (4), Brazil (3), Bulgaria (5), Canada (20), Chile (28), China (202),

Colombia (2), Croatia (1), Czech Republic (7), Ecuador (1), Fiji (1), France (21), Georgia (6), Germany (89), Greece (4), Hungary (35), India (1), Indonesia (7), Iran (3), Ireland (1), Italy (88), Japan (34), Kazakhstan (2), Korea (1), Kyrgyzstan (6), Malaysia (3), Mexico (83), Mongolia (5), Montenegro (1), Morocco (3), Myanmar (1), Namibia (1), New Zealand (27), North Macedonia (5), Norway (1), Pacific Ocean (1), Papua New Guinea (1), Peru (5), Philippines (3), Poland (8), Portugal (3), Romania (6), Russia (56), Serbia (4), Slovakia (96), Slovenia (3), South Africa (6), Spain (77), Sweden (1), Switzerland (9), Taiwan (5), Tajikistan (4), Tunisia (2), Turkey (10), UK (22), Ukraine (8), USA (1481), Uzbekistan (6) and Zimbabwe (2).

These occurrences and relative quantifications must serve exclusively as an example as they reflect the state of the art of the studies carried out in each country. On the other hand, the numbers account for more modern exploitation than the actual extent of the deposits. Moreover, some attestations are missing, while others refer to the same mining district.

The list is not exhaustive but it gives an idea of the distribution of cinnabar. In support of what can be easily found online, Fig. 5 illustrates the distribution of the main geological deposits of cinnabar. The occurrences have been drawn based on >250 papers indexed by Scopus; therefore, some information is missing but can be recovered on the mindat.org database. In Supplementary materials Table 1, the deposits shown in Fig. 5 are listed together with the corresponding bibliographic reference.

As for the associations, cinnabar is frequently found in gold/silver/arsenic/antimony deposits. Apart from native elements, the most typical association is with stibnite, followed by pyrite, sphalerite, chalcopyrite, galena, arsenopyrite, realgar, marcasite, orpiment, tetrahedrite and Hg-bearing halides. Scheelite, cerussite, pyrrhotite, acanthite, siderite, anglesite, chalcocite, covellite and pyromorphite are other minor phases frequently found in association with cinnabar. Ag-Hg amalgam and rare phases such as tellurides and sulphobismuthides have also been found associated in Hg-deposits.

Worthy of mention are the numerous (but apparently rare) Hg-bearing phases discovered in Clear Creek mine (San Benito County, CA, USA; Dunning et al. 2005), such as the deanesmithite (Roberts et al. 1993), edoylerite (Erd et al. 1993), hanawaltite (Roberts et al. 1996), clearcreekite (Roberts et al. 2001), tedhadleyite (Roberts et al. 2002), vasilyevite (Roberts et al. 2003) and aurivilliusite (Roberts et al. 2004).

Quartz/chalcedony and barite are the most common gangue minerals, followed by calcite/aragonite and kaolinite.

To summarising, cinnabar is mainly found in volcanic environments and hot springs deposits. It is often associated with stibnite and other Hg-, Sb- and Pb-based phases, as well as with gangue minerals as quartz, barite and calcite. In some archaeometric publications, we read that a cinnabar deposit's limited size may have prompted local populations to obtain supplies elsewhere. While this assumption is undoubtedly

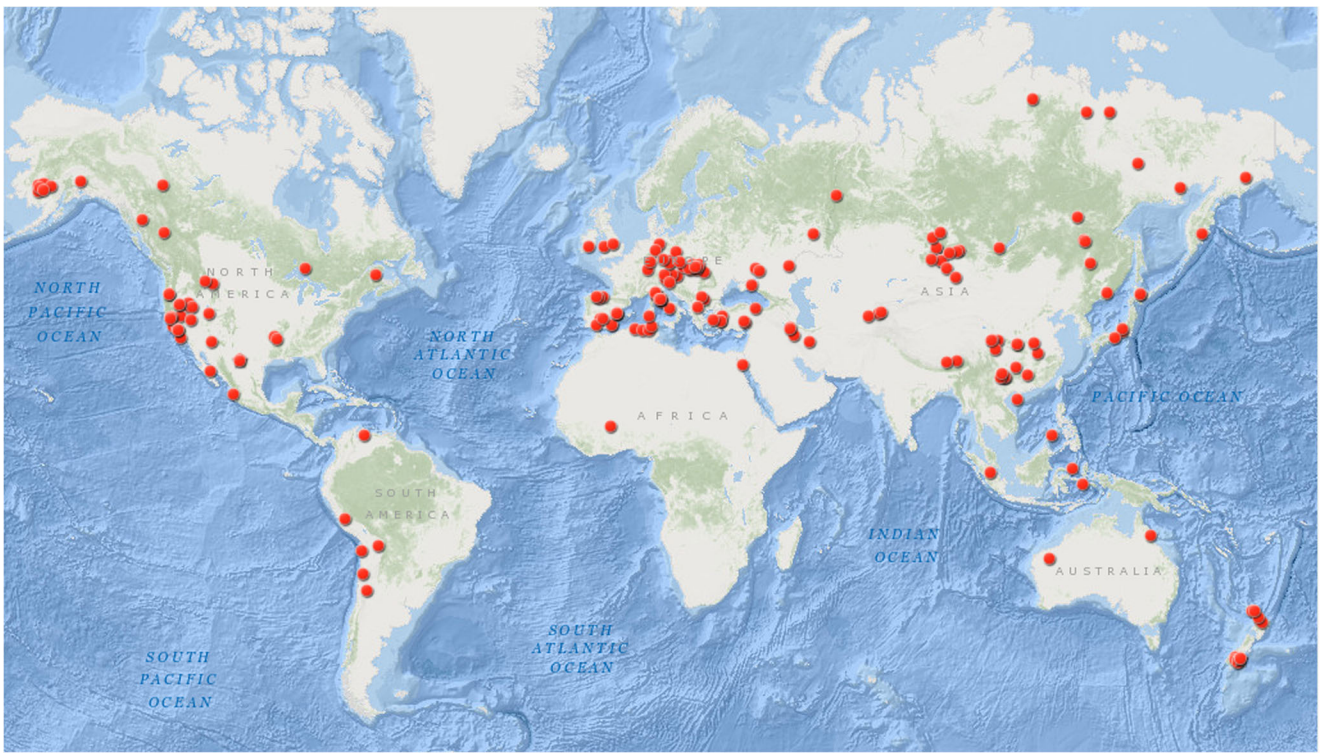

Fig. 5 Main cinnabar deposits. Data from Supplementary Table 1

valid for industrial applications and thus related to the modern world, it seems less effective when applied to ancient civilizations.

The multiple uses of cinnabar

Cinnabar has been mainly used

- To paint human bones and offerings (funerary use). Based on the archaeometric evidence available so far, the first use of cinnabar appears related to funerary practices and ceremonial activities. To the best of my knowledge, the 9th millennium BC painted plaster skull from Kfar HaHoresh in Israel (Goren et al. 2001) represents the oldest attestation of the use of cinnabar as a colouring pigment. Similar examples increase when dealing with the Late Neolithic and especially the Chalcolithic periods. The following section "Funerary use: esoteric power, social self-representation and archaeological evidence" is entirely dedicated to this topic.
- To decorate architectural structures and mobile objects (decorative use). Based on archaeometric data, the decoration of ceramic vessels, sculptures and cartonnages precedes that of wall paintings. Over time, the use of cinnabar to decorate structural elements increases considerably while its use for ceramics apparently disappears. This topic is discussed in the section "Decorative use: cinnabar over time and space".
- To write (ink in manuscripts). The use of cinnabar as ink is frequently attested, especially during the Middle Ages (see, *e.g.*, Vlad et al. 2011; Serhrouchni et al. 2019; Safronov and Sozontov 2020). For the discussion of this topic, the reader is referred to Burgio (2021) in this TC.
- To enhance the reduction in lustre production, addressed in the section "Cinnabar for ceramic lustre".
- To colour red lacquers, addressed in the section "Cinnabar in lacquered objects".
- To serve for medicinal and cosmetic purposes (pharmacological use). Long-lasting use of mercury compounds is attested in ancient and oriental medicine, for example, in Indian Ayurveda (Murthy 1983), Oman (Hardy et al. 1995), Japan (Yamada et al. 1997), China (Anonymous 1967; Wu et al. 2002; Huang et al. 2007; Liu et al. 2008; Jain et al. 2019) and ancient Tunisia (Huq et al. 2006). The numerous therapeutic indications of cinnabar (esp. mercurial salts and other Hg-based compounds mixed with ointments) included fever, insomnia, lice, stoke, trauma, mouth ulcers and syphilis (Liu et al. 2018a). A further indication of the importance of mercury compounds in oriental tradition may be represented by the translation of the *tanden* (now better known as dantien) as the "cinnabar field", which is situated "two inches below the navel" (Ahn 2008) and represents the centre of gravity of the human body. The discussion of this topic is limited to some hints functional for the general discussion. For the details, the reader is referred to Pérez-Arantegui (2021) in this TC.

Other "particular" findings and occurrences are also reported in the last section of this brief roundup on cinnabar occurrences (namely, "A (very) shortlist of other particular findings and occurrences of cinnabar").

An indispensable premise for the following sections concerns the bibliographic collection. Firstly, the publications collected are limited (almost always) to those in which the authors identified cinnabar through analytical techniques. Secondly, I am sure many publications were overlooked for various reasons ranging from accession difficulties to the language in which they were written. Consequently, the collection presented is not exhaustive but may offer a sufficiently articulated panorama of the state-of-the-art of the studies. The literature search was carried out by setting "cinnabar" as the key term in the search field (title and abstract) of Scopus. Eastaugh et al. (2004) and Siddall (2018) are also recommended on the uses of cinnabar and its characteristics.

Funerary use: esoteric power, social self-representation and archaeological evidence

The wide occurrence of Hg-based compounds in funerary contexts has long attracted the interest of researchers. Already in 1927, Peabody compiled a list of numerous occurrences in which "red paint" could be associated with the direct or indirect colouring (transfer from the burial) of the bones (Peabody 1927). He focused his review on ochres at a time when archaeometric analyses were not yet performed. Therefore, the achieved conclusion is also of interest for cinnabar: "*We have seen that the use of color, especially red, in mortuary ceremonies is practically universal in time and space over the earth among prehistoric and primitive peoples*". Peabody wondered, "*what is the meaning of red?*" and, after having warned the reader that "*color symbolian is anything but obvious*", came to the conclusion that "*the most obvious suggestion of red is blood*" (thesis argued and expanded in the following lines of his text).

Several authors have returned to this concept in the following years and the debate has opened numerous hypotheses and reconstructions. The common opinion is still focused on the correlation between red colour and blood and, consequently, the symbolism of life, death and sacrifice. In this regard, one of the anonymous reviewers rightly invites me to recall a concept

dear to ancient philosophy, such as the "cosmic *sympatheia*". According to this Stoic thought, all beings on earth and in the heavens are closely connected[3]. However, other aspects such as the preservative properties of cinnabar, its use for body painting and tattooing and its magical aura are equally interesting and must be considered.

As for the conservative properties, cinnabar can delay the decomposition of the body thanks to its powerful bactericide and insecticide properties (Martín-Gil et al. 1995; Cervini-Silva et al. 2013). This intent is usually highlighted by the location of burials in underground environments and/or far from sunlight and humidity. In this case, its use would have been functional rather than symbolic; consequently, determining whether cinnabar was applied soon after death or after decomposition of the soft tissues can undoubtedly be of crucial importance (see also Domingo Sanz and Chieli 2021in this TC).

As for its use for body painting and tattooing, the discussion becomes more complicated and intriguing. In this case, the use of cinnabar may directly connect the deceased to her/his life. Body painting could probably have had both an aesthetic and a social self-representation purpose during life. This practice would have followed a social dynamic that does not seem so distant from today's reality (with 60 million tattooed in Europe and more than 20% in the USA. Percentage considerably reduced in those countries like Japan where it is considered a social stigma — Harris Poll).

In this regard, the studies presented by Carter (2008) on EBI Cycladic society, Padilla et al. (2012) on argaric civilisation and Burger and Leikin (2018) on prehispanic Central Andean communities are exemplary. Carter (2008) connected the geographical and social expansion of the cultural and commercial exchanges of Early Bronze Age I Cycladic society (late 4^{th}–3^{rd} millennium BC) with a new way or a renewed interest in personal adornment (from tattooing to hair removal and jewellery). Quoting the authors, cinnabar "*may have been reserved for special occasions (and people), limited body decoration and tattooing*", thus "*embodying*" the ongoing political change. To support this intriguing hypothesis further, Carter also recalled the votive use testified by red-painted marble figurines and vessels found in Early Cycladic burials (references therein).

Padilla et al. (2012) observed that the diffusion of cinnabar in Argaric funerary contexts was more widespread among female individuals than in males and, therefore, deduce that cinnabar was linked to face and body makeup.

Burger and Leikin (2018) underlined that the prehispanic Central Andean communities used cinnabar for facial and body paint and its use went far beyond a vain desire for beauty to become an "*expression of social identity*" (Burger and Leikin 2018).

Observing the use in life for both beautification and social self-representation, it follows that the use after death (for the painting of human bodies) may achieve multiple values, from the desire to preserve one's natural appearance for the afterlife world to the desire to externalise one's social rank also through one's own burial or even, more simply, to give the deceased the appearance of a leaving body for the benefit of the living.

Even the magical aura to which cinnabar was likely associated could have played a role in all this. Still, several distinctions would become necessary as it is not obvious to assign a unique meaning to a pigment that has gone through very different cultures and chronological periods. Finally, it is good to include among all these likely hypotheses the possibility that the bodies of the deceased and the offerings were sprinkled with cinnabar to keep thieves and profaners away. The poisonous nature of the pigment was already well known in ancient times. This aspect may have represented both an intended use and an added value of cinnabar compared to red ochre.

Other aspects concern analytic practice more closely, such as assessing whether mercury entered bones either biogenically or diagenetically. In the former case, mercury derives from direct exposure; while in the latter, the process involves a transfer from the soil. On this topic, the research conducted by (a) Emslie et al. (2015, 2019) on skeletal material from Middle and Late Neolithic and Early Bronze Age Iberian and Portuguese necropolis and (b) Cervini-Silva et al. (2013, 2018) on Mexican Red Queens and other funerary contexts must indeed be cited for both their intrinsic and methodological value.

As anticipated above, a significant body of literature exists on the contamination of land and rivers following mining activities and valuable information can also be drawn from these publications. In this regard, two case studies clearly explain how complicated or controversial it may be to correctly interpret the presence of mercury/cinnabar in archaeological sites. The first case regards the mobilisation of cinnabar during diagenesis. This naturally occurring phenomenon has been recognised by García-Alix et al. (2013) as the alteration process responsible for the red colour of Miocene fossil mammals (bones and teeth) found in the Otura section (Granada Basin, Spain).

The second case regards the black mercury alteration detected on the surface of numerous jades found in "high-grade" Chinese tombs. This alteration has been recognised as a human-related phenomenon by Bao et al. (2019), who found that the alteration was due to the fire ("*Liaoji*") ceremonial activity (burning oblations) during which the jade was placed over cinnabar. The latter turned into mercury vapour (> 800°C) and thus caused the superficial alteration of jade.

[3] The concept recurs in medical writings (*e.g.*, Hippocrates, *De alimento*, 23.1) to indicate how soul and body or the different parts of the body are related to the body as a whole.

As for the occurrences of cinnabar in burial contexts, the attestations are many; however, the archaeological findings are not accompanied by archaeometric analyses in many cases. Therefore, it is not always possible to know whether it is ochre or cinnabar or a mixture of both or if there are associated aromatic compounds. Here, only a few verified examples are presented to show the spread of this practice broadly. The number of studies performed on this topic also appears unbalanced as most refer to Spanish, Portuguese and South American archaeological sites.

The occurrences in Spain are listed in Table 2 and shown in Fig. 6. Those reported in Meso- and South America (Belize, Chile, Guatemala, Honduras, Mexico and Peru) are provided in Table 3 and shown in Fig. 7.

In Meso- and South America, the occurrences are mainly concentrated in the Mayan territory and scholars partially agree in stating that cinnabar was used above all in the funerary contexts of the ruling elite (Schele and Mathews 1999; Vázquez de Ágredos Pascual 2007, 2018; Fitzsimmons 2009; Quintana et al. 2015). A comprehensive work on the use of cinnabar and other pigments for body paint is provided in the recent volume edited by Vázquez De Ágredos Pascual and Dupey García (2018), in particular, by Vázquez De Ágredos Pascual (2018) within the volume. The numerous contributing authors examined the phenomenon from all its points of view (material and cultural) and in the various social and cultural contexts of Mesoamerica, from pre-Columbian to modern times.

Apart from these two large areas that have returned most of the evidence (or in which the researchers carried out the highest number of archaeometric investigations), we must finally also remember the few occurrences around the Mediterranean basin:

- Israel — On the 9th millennium BC (Pre-Pottery Neolithic B) plastered skull of the KHH-Homo 8 from Kfar HaHoresh (Goren et al. 2001; Goring-Morris and Horwitz 2007 for an insight on the site);
- Syria — On the painted Pre-Pottery Neolithic B skull inv. No. 73.2772 found at Tell Abu Hureyra (Trench A, level 212) (Molleson et al. 1992);
- Turkey — In the 8th millennium BC phase of Çatal Hüyük, where cinnabar was used to paint the skull of a "*woman with a necklace of sliced dentalium beads*" and for wall paintings (Mellaart 1967);
- Russia — In the 34th–30th century BC burial 1 (individual 1) at the Maikop-Novosvobodnaya settlement of Chekon-2 (Taman Peninsula), cinnabar was found in the fragments of the upper cranial vault (Korenevskiy et al. 2015).

To conclude, it is also worth underlining that the use of cinnabar in burials is also attested in prehistoric and historical China and Japan (Bao et al. 2019; Liu 2004).

In China, a comprehensive study on textiles and cinnabar decorated leather objects (1st millennium BC– 1st/3rd century AD) associated with mummies from the Xinjiang Uyghur Autonomous Region was performed by Kramell et al. (2016). The leather scabbard of a bronze sword covered by cinnabar was also found in the tomb M4 of the Qiaojiayuan site (Yun country, Hubei province; Luo et al. 2011).

Another example is represented by the cinnabar powder covering the surface of a set of the mid-11th–mid-9th century BC jade artefacts related to burial ceremonies from a tomb of the Ying State's Cemetery (Pingdingshan, Henan; Zhao et al. 2014). Particular use is also testified by the Shang epoch (1766–1122 BC) oracle bones (turtles plastrons and bovine scapulas used in divination) painted with cinnabar from Yin Hsü (Anyang, Honan; Benedetti-Pichler 1937).

In Japan, the funerary use of cinnabar lasted from the mid-Yayoi period till the late Kofun period (4th century BC–6th century AD). It was aimed at decorating (coffins), preserving (antisepsis) and staining (textiles), as well as having a ritual function and representing a symbol of power (Kawano et al. 2014). An example is provided by the *Takamatsuzuka* tumulus (7th–8th centuries) at Asuka (Nara prefecture, Kinki region), where cinnabar was identified in the murals (Kitada et al. 2015).

As a very last example, it is also worth mentioning the cinnabar painted skull found in Idaho that, dated to 600–700 years BP, was assessed to belong to one of four major Native American mitochondrial DNA lineages (Watkins et al. 2017).

Decorative use: cinnabar over time and space

Cinnabar was used for the decoration of mobile objects and probably later for the decoration of architectural surfaces. This trend seems valid both for the Mediterranean world and the Near East as well as for Meso- and South America. The most ancient attestations include mobile objects decorated with cinnabar found worldwide in funerary, religious and, to a lesser extent, residential contexts.

In South America, to give a representative example, the enormous number of 1500–1200 BC objects related to funerary practices and daily life found at Gramalote in Peru were painted with cinnabar or hematite or a mixture of both (Prieto et al. 2016).

In the Mediterranean area, particularly ancient attestations are sporadic. The likely oldest attestation on wall paintings dates to the level VIII (6700 cal. BC) onwards of the archaeological site of Çatalhöyük in Turkey (Çamurcuoğlu 2015). Out of a total of fifty-nine red pigment samples taken from wall paintings and burial contexts, the author found cinnabar in only 3 of them. Cinnabar was identified by Raman spectroscopy, either used alone or in mixture with red ochre, and was tentatively traced back to the mercury deposits "*in the southwest and north of Konya region as well as around Niğde in the east*". Mellaart (1967) hypothesised a provenance

Table 2 Cinnabar in funerary contexts in Spain and Portugal (the numbers in the second column refer to Fig. 6)

Spain	1. 5300–5000 cal BC **Cova de l'Or** (Beniarrés, Alicante). Cinnabar was found as a fine and pure red powder inside a *Glycymeris* sp. shell.	Roldán et al. 2008; Domingo et al. 2012
	2. Early Neolithic (likely between 5300 and 5200 cal BC) flint mine of **Casa Montero**. A thin cover of cinnabar was found on a flint blade.	Hunt et al. 2011
	3. Fifth millennium BC **dolmen de Alberite** (Villamartín, Cádiz). Finely chrushed cinnabar (silt and clay as prevailing fractions) mixed with hematite was covering bony remains and structural elements, also forming a ~10-cm-thick layer in a portion of the gallery's floor.	Domínguez-Bella and Morata Céspedes 1995; Ramos et al. 1997; Domínguez-Bella 2010
	4. Chalcolithic (4th–3rd millennium BC) collective hypogean graves found in the **Higueras Valley** (Toledo) returned bones and human figurines coloured by cinnabar. Interestingly, this was the only use that was made of cinnabar onsite while hematite was used for the decoration of the structural elements. The authors also draw a correlation between the use of light-reflecting materials such as cinnabar, amber, quartz and gold and lighting that was to play an important role in the ceremonial.	Bueno-Ramírez et al. 2019
	5. Chalcolithic (3800–4100 BP) hypogeum of **Carrer Paris** (Cerdanyola del Valles, Barcelona). Cinnabar (together with quartz) was found on the left mandibular fragments of individual no. 5 and on loose fragments recovered from the cleaning of the cranium.	Gómez-Merino et al. 2011
	6. Chalcolithic burial cave of **Getsemaní-Cerro del Ojo** (Pedrera, Seville).	Bascón-Mateos et al. 2016
	7. Neolithic (3000 BC) dolmenic burial of "**La Velilla**" (Osorno, Palencia). Hundreds of pure and finely grained kilograms of cinnabar were covering human bones.	Martín-Gil et al. 1995
	8. **Cueva de los Murciélagos** (Cordoba). Cinnabar was found in a Neolithic container.	Barrios et al. 1999; Martínez-Fernández et al. 1999
	9. **Montelirio** megalithic tomb within the Valencina de la Concepción settlement (Copper Age, early 3rd millennium BC). Cinnabar of likely Almadén provenance was mixed with minor iron oxides contents and spread over human bones, ivory and pottery along with coating a large slate slabs that formed the walls of the mortuary chambers.	Vargas Jiménez 2004; Hunt Ortiz and Hurtado Pérez 2010; Hunt et al. 2011; Rogerio-Candelera et al. 2013; Fernández Flores et al. 2016
	10. Artificial cave of **La Molina** (Lora de Estepa, Seville). Cinnabar was found on several (but not all) inhumations, especially in zone no. 1. The authors suggested a baetic provenance and intepreted the different distribution of cinnabar as an evidence of unequal social practices among the burials as its selective use appeared limited to men and women who must have had a preponderant role in the community.	Juárez Martín 2010
	11. 3rd–early 2nd millennium BC necropolis of **Paraje de Monte Bajo** (Alcala de los Gazules, Cádiz). Cinnabar mixed with iron oxides was found on human bones.	Lazarich et al. 2010a, 2010b; Briceño et al. 2012
	12. Bell-Beaker Chalcolithic period "Tumba 1" (tholos) of **La Pijotilla**. Cinnabar was found over a sheet of slate.	Hurtado Pérez 1999; Hunt Ortiz and Hurtado Pérez 2010; Hunt et al. 2011
	13. Late Neolithic (4th–3rd millennium BC?) **Dolmen de las Casas de Don Pedro** (Belmez, Córdoba). Remains of the funerary equipment consisting of arrowheads, necklace beads and cinnabar were found in sector I and a rock crystal arrowhead and some cinnabar nodules were found in sector II.	Gavilán 1987; Gavilán and Vera 2004
	14. Chalcolithic settlement of **Camino de las Yeseras** (San Fernando de Henares, Madrid). Cinnabar was used to paint an inhumed body placed in lateral decubitus with bent legs.	Liesau et al. 2008
Portugal	15. Late 4th–half 3rd millennium BC megalithic tomb **Santa Rita** (Cacela, Vila Real de Santo António) where a cinnabar-bearing quartzite, porbably from the Almaden mining district, has been found.	Inácio et al. 2013
	16. **Dolmen of Marcela**, where two big pieces of cinnabar were found.	Obermaier 1919
	17–20. 4th–3rd millennium BC dolmens of **Zambujeiro** (17), **Bola da Cera** (18), **Horta** (19) and **Cabeçuda** (20) and the Neolithic hypogeus necropolis of **Sobreira de Cima** (21).	Oliveira 1997; Dias et al. 2011; Barrocas Dias and Mirão 2013; Rocha et al. 2018

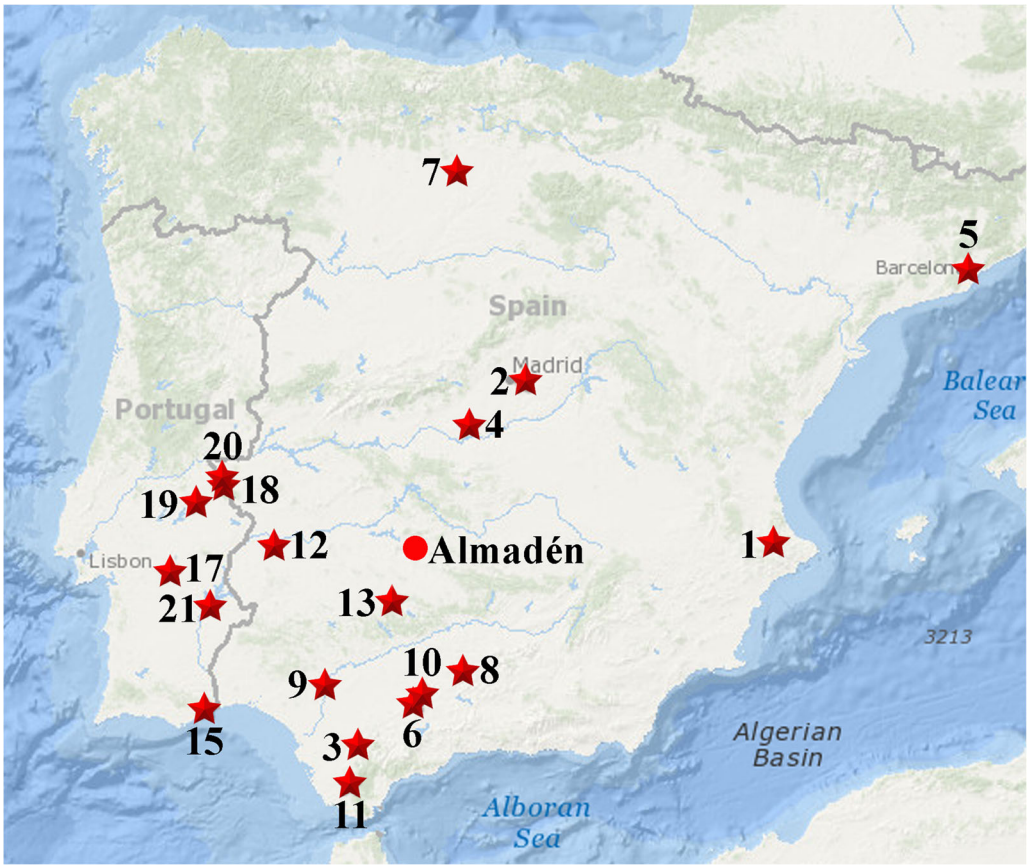

Fig. 6 Cinnabar in funerary contexts — Spain and Portugal. The geographic location of the sites listed in Table 2

from the Sizma deposits but no supporting archaeometric investigations are available in this regard (Doherty 2011). In any case, the Sızma deposit (not reported in the Mindat.org database) is about 60 km away as the crow flies from Çatalhöyük and it is, therefore, the closest deposit known so far (Fig. 8).

The Vinča settlement provides another example. Cinnabar appeared ubiquitous in layers dated from the mid-6th to the mid-5th millennium BC (Vasić 1932–1936, I as quoted by Gajić-Kvaščev et al. 2012). A cinnabar powder was also stored inside Neolithic pottery (inventory no. C-417, dated to 5200–4200 BC) (Mioč et al. 2004). Similarly, at Pločnik, a Gradac sub-phase ceramic vessel (end of the 6th–early 5th millennium BC) was found to contain a cinnabar powder (cinnabar mixed with quartz, illite, kaolinite and other clay minerals); moreover, cinnabar was also used to decorate figurines (Gajić-Kvaščev et al. 2012).

In the 1st millennium BC, ancient examples (based on archaeometric evidence) are present on ceramic pottery and sculptures from Spain, Italy and Greece. As for ceramic decoration, it is possible to mention:

(a) the 6th–5th century BC small vessel found in tomb no. 233 (burial mound H) of the cemetery of La Noria (Fuente de Piedra, Málaga; Tuñón et al. 2016);
(b) the 5th millennium BC Serra Alto pottery found in the Grotta dei Cervi (Porto Badisco, southern Italy; Quarta et al. 2018); and
(c) the 4th–3rd century BC ceramic vessels found in the Iberian cemetery of Tutugi (Galera, Granada, Spain; Sánchez et al. 2012).

Regarding sculpture, the Early Cycladic II (3100–2400 BC) marble figurines (Hendrix 1998; Carter 2008) provide a representative example.

Moving forward in time, cinnabar has been frequently used for the decoration of numerous Egyptian coffins. The cartonnages dated between the 21st and the 25th Dynasty (1070–525 BC) show a somewhat standardised palette with slight variations. Six basic pigments are constantly found: cinnabar, Egyptian blue, yellow and red ochres, orpiment and carbon black. Other copper-based pigments such as atacamite have been found less frequently; conversely, the presence of azurite, Prussian blue and lead white are generally attributed to retouches and restorations of a later period. The study performed by Gard et al. (2020a, 2020b) on a Ptolemaic cartonnage (305–30 BC) deserves a special mention because—besides the typical pigments—pararealgar, bonazziite and/or alacránite, uzonite and Egyptian green ($(Cu,Ca)SiO_3$) were also found (for the As-containing phases

Table 3 Cinnabar in funerary contexts — Meso and South America (the numbers in the second column refer to Fig. 7)

Mexico	1. At **La Tronera** site (Chupicuaro culture), in 400–200 BC burials, where cinnabar was found both on human bones and on a funerary mask.	Vázquez de Ágredos Pascual et al. 2019
	2. At **Teotihuacán** in burials from the Tzacualli (1–150 AD) to the Metepec (550–650 AD) phases. Cinnabar has been archaeometrically identified in the burials and the relative offerings (balls of pigment in pigment balls in miniature ceramic vessels) of Teopancazco.	Gazzola 2009; Vázquez de Ágredos Pascual et al. 2018
	3. At the 3rd–9th century Mayan site of **Jaina**, in burial no. 23 where both hematite and cinnabar were used for painting the buried male body.	Batta et al. 2013; Ávila et al. 2014
	4. In the ancient Maya town of **Palenque**. In the "Reina Roja" temple XIII (Red Queen), cinnabar has been found on her body, on the sarcophagus and several obects constituting the funerary equipment. In the tomb of Anab Pkal cinnabar was used to paint the human bones. In the temple XVIII-A of Palenque, cinnabar was found on the skeleton PAL-44 of a male of the ruling elite dated between 250 and 420 cal AD as well as on his offerings.	Schele and Mathews 1999; Tiesler and Cucina 2006; Gómez-Merino et al. 2011; Couoh 2015; Quintana et al. 2015
	5. At **Calakmul**: (a) in the tomb of *Yuknom Yich'ak K'ak*, where cinnabar was used to paint both the tomb and the body of the king and (b) in the Classical period structure II—Burials 5a and 9 where it was identified on a bone fragment (where animal fat was also found) and dust, respectively. Other occurrences from Calakmul and Dzibanché have also been investigated.	García-Moreno and Granados 2000; Vázquez de Ágredos Pascual 2007, Vázquez de Ágredos Pascual 2009; Quintana et al. 2015; Rigon et al. 2020
	6. In the Classic Period (250–900 AD) burials from (a) the small settlement of Xcambó; (b) the remains of king *Okit Kan L'ek Tok* from **Ek Balam**; (c) the elite contexts at Oxkintok; (d) the ancient capital of the 'Snake Empire' of Calakmul and **Dzibanché**.	
	7. In the Classic Period (250–900 AD) burials of **Toniná**.	Fitzsimmons 2009
	8. At the Olmec site of **La Venta**, cinnabar was found on human bones and among some offerings (*e.g.*, on figurines), together with basalt, asphalt and salt.	Pool 2007
Belize	9. A late 9th or early 10th century offering consisting of 19g of cinnabar and 100g of hematite, along with other objects (jade, shell and pearl), were found at the **Lamanai** archaeological site in Belize (central Maya lowland) attesting the use of cinnabar for cerminial activity.	Pendergast 1982
Guatemala	10. At **Kaminaljuyu**, cinnabar was found amoung the offerings of several tombs (A-V. A-VI, B-I and B-III) covering jade (frequently found in Spondylus shells) and/or sting-ray spines. In this site, cinnabar was also used to paint miniature vessels as well as on a pyrite encrusted plaque, a cilindrical trypod with plano-relief carvings filled by cinnabar, a pair of thin orange jars, and a textile-impressed effigy. Cinnabar was sometimes mixed with calcite or diatomaceous earths and liquid mercury was also found.	Kidder et al. 1946
	11. At the underwater site of **Contreras Alto – Los Jicaqnes** (in the Lake Amatitlan), fragments of cinnabar and graphite were found together with about 400 fragments of jade ear-spools.	Borhegyi 1959
	12. At the archaeological site of **Rio Azul**, the men in the tombs 19 and 23 were covered by cinnabar.	Carlsen 1986, 1987
	13. At the Early Classic (250–550 AD) archaeological complex of **El Diablo** (San Miguel La Palotada Biotope in the Municipio of San José, Department El Petén), in the Royal Maya Tomb of **El Zotz** where cinnabar (and hematite) was found on human bones and in (spondylus) shells.	Cheung et al. 2013
	14. In the Classic Period (250–900 AD) tomb burials of **Tikal** and those interred in the ancient city of **Sacul**.	Fitzsimmons 2009; Quintana et al. 2015
Honduras	15. At **Copán**, in the excavation of the mound in centre of the Plaza, a pot containing cinnabar powder and bones covered by cinnabar were found(1). At the same site, bones painted by cinnabar has been found in several tombs: the 400–450 AD Hulan tomb of the Copán founder *K'inich Yax K'uk' Mo'*; the Margarita tomb (burial 93-2), presumably the wife of the previous, the mid-6th century burial 92-3 and the ~450 AD disarticulated burial 92-1. Other and/or same occurrences are reported in Gorokhovich et al. (2020).	Maudslay 1889-1902; Schele and Mathews 1999; Sharer et al. 1999; Sedat and López 2004; Bell et al. 2004; Ashmore 2015
Peru(2)	16. In the necropolis of **Ancón**, cinnabar was found on one 11th–13th century AD mummy's hair.	Degano and Colombini 2009
Chile	17. In the 1399–1475 AD burial of two girls at **Cerro Esmeralda** (close to Iquique) likely testifying an Inca *capacocha*, *i.e.* a human sacrifice, cinnabar was found on the mantos (textile) of one of the mummies.	Arriaza et al. 2018

(1) "In the centre of the mound, about 6 feet from the top, an earthern pot (Plate XXL, a) was found containing a bead- shaped piece of greenstone, pierced, diameter two and three quarter inches (b), the jade whorl of a spindle (c), the remains of a necklace of nine jade beads (d), four pearls (e), some small rough figures and other ornaments cut out of pearl oyster-shell (f), and other irregular pieces of roughly carved pearl-shell (g). At the bottom of the pot was some red powder (which proved to be finely ground cinnabar), and several ounces of quicksilver. Eighteen inches above this pot some traces of bone were found mixed with sand. At the level of the ground, more traces of bones were found mixed with red cinnabar powder and sand, and one large pierced bead-shaped stone, diameter three inches (h). About eight to nine feet below the level of the plain, a skeleton of a jaguar was found lying under a layer of charcoal."

(2) Further occurrences in Peruvian pre-Hispanic period are also reported in Burger et al. (2016).

and the Egyptian blue and green pigments see Gliozzo and Burgio 2021 and Švarcová et al. 2021, respectively in this TC).

Going further in time, the evidence of pictorial decorations on architectural elements begins to take on consistency. The fragments of decorated walls, floors and columns recovered in Persepolis and Pasargadae (6th–4th century BC), the wall paintings of the Tomb IV in Phoinikas in Greece (end of 5th–4th century BC; Avlonitou 2016), the second tomb of Vergina in Greece (4th century BC; Filippakis et al. 1979), the Etruscan 'Tomba dell'Orco' at Tarquinia in central Italy (4th century BC; Sodo et al. 2008) and, probably also the first Pompeian style House in Pella (400–168 BC; Calamiotou et al. 1983) are among the earliest attestations. However, it will be necessary to wait until the 2nd and 1st century BC before finding cinnabar constantly used to decorate architectural elements (based on sure evidence verified through archaeometric analyses).

Between the 3rd and 1st centuries, verified evidence testifies the use of cinnabar on both mobile objects and wall paintings in Europe and Asia. In the first case, some examples are found:

- in China, on the polychrome terracotta soldiers found in the Tomb of Jing Di Emperor — Xi'an (221–140 BC; Chiavari and Mazzeo 1999), in the Royal tomb in the Qingzhou County (Shandong; Wei et al. 2012) and on the Jian Hu polychrome terra-cotta jar found at the Xi'an Airport (206 BC–23 AD; Chiavari and Mazzeo 1999);
- in Europe, on the terracotta figurines found in Hellenistic tombs at Thessaloniki and Demetrias (Greece, 3rd–2nd century BC; Fostiridou et al. 2016; Tsatsouli and Nikolaou 2017).

Testimonies on wall paintings are instead found:

- in Italy, in the House of the Golden Bracelet at Pompeii and the villa of Papyri at *Herculaneum* (2nd century BC; Durán et al. 2010a, 2010b), in the painting layer on mortar under the mosaics of a house located under the church of St. Susanna in Roma (2nd–1st century BC; Boschetti et al. 2008), and in wall paintings found at the archaeological sites of Torre (Pordenone), Crosada (Trieste) and Montegrotto (Padova) (1st century BC; Mazzocchin et al. 2004);
- in Spain, in the Roman necropolis of Camino Viejo de Almodóvar (Córdoba, 1st century BC; Cerrato et al. 2020) and in the Roman villa of Baños de Valdearados (Burgos, 1st century BC–1st century AD; Villar and Edwards 2005);
- in Palestine, in the Palace of King Herod the Great at Jericho and Massada (1st century BC; Edwards et al. 1999b; Porat and Ilani 1998).

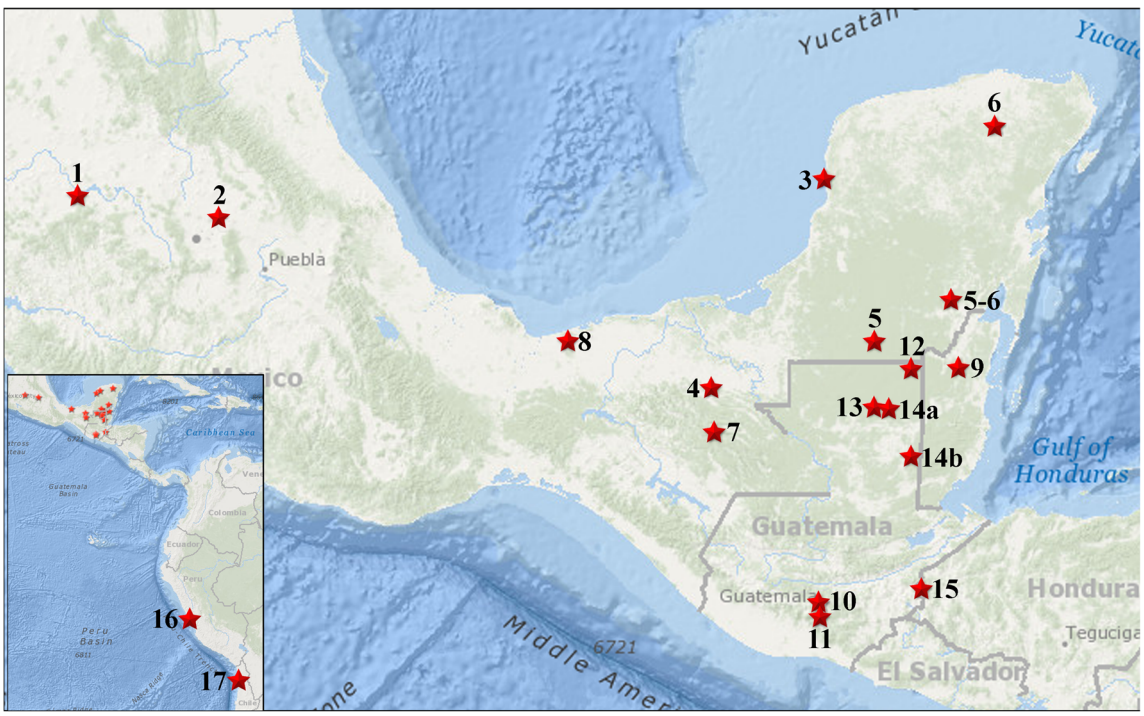

Fig. 7 Cinnabar in funerary contexts — Meso- and South America. The geographic location of the sites listed in Table 3. Sites 14a and 14b correspond to Tikal and Sacul, respectively. Site 5-6 indicates the location of Dzibanché

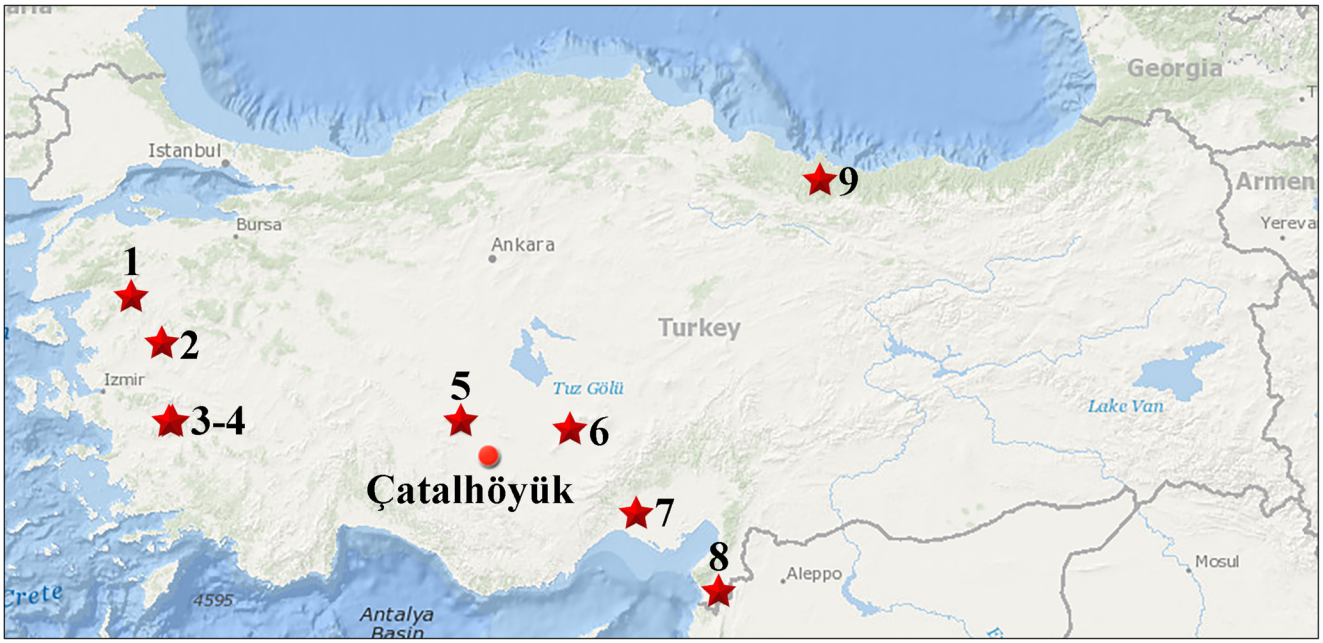

Fig. 8 Cinnabar deposits and occurrences in Turkey. (1) Kuçukyenice, Ivrindi, Balikesir, Marmara; (2) Baglar Hill, Mumcu, Balikesir, Marmara; (3) Emirli Sb-Au deposit, Ödemiş, Izmir; (4) Halıköy Hg deposit, Ödemiş, Izmir; (5) Sızma, Selçuklu, Konya; (6) Rasih-thsan occurrence, Niğde; (7) Madsan Sb deposit, Çamardı, Niğde; (8) Kisecik Au deposit, Antakya, Hatay; (9) Akoluk, Ulubey, Ordu. Data from Mindat.org except for Sızma. The archaeological site of Çatalhöyük is indicated with the red filled circle

The numerous bowls containing powdered pigments found in Pompeii and analysed by Aliatis et al. (2010) must also be referred to an unspecified time before 79 AD.

Up to this period, the association with inorganic pigments is limited to carbon black and sporadically to madder lake while animal glue prevails among the binders. Other organic pigments such as indigo and other binders such as oils will be more frequent in the following centuries.

Between the 1st and 3rd centuries, the testimonies relating to the wall paintings gradually increase. However, most of the investigations are relevant to Italian archaeological areas:

- *Domus Aurea*, Rome (64 AD; Clementi et al. 2011);
- *Domus* at *Liternum* (1st century; Corso et al. 2012);
- *Domus* of *Octavius Quartio*, Pompei (1st century; Germinario et al. 2018);
- Tomba dei Pesci e delle Spighe, necropolis of Tuvixeddu, Cagliari (1st century; Solla et al. 2015);
- House of Diana, *Cosa*-Ansedonia (after 80 AD; Damiani et al. 2003; Fig. 9A–B);
- *Domus* below the Basilica of SS. John and Paul on the Caelian Hill, Rome (end of 1st–4th centuries; Fermo et al. 2013);
- Roman *villa* in Vicenza (Mazzocchin et al. 2003);
- Tomb 75 — Necropolis at Tenuta Boccone D'Aste, Roma (2nd century; Aurisicchio et al. 2002);
- *Thermae* of *Iulia Concordia*, Venezia (second half of the 2nd century; Mazzocchin et al. 2010).

Other case studies regard wall paintings found in Morocco (*Thamusida*, 1st–3rd centuries by Gliozzo et al. 2012), Switzerland (Gallo-Roman villa at Dietikon, 1st–3rd centuries by Béarat 1996) and Turkey (Sinop Balatlar Church Complex, 2nd–4th centuries, by Bakiler et al. 2016).

In Egypt, most findings relate to paintings on wood. In this regard, famous examples are represented by the Roman mummy portraits of approximately the 2nd century found at Tebtunis and investigated by Salvant et al. (2018).

From about the 4th century up to the Middle Ages, the evidence relating to manuscripts (see Burgio 2021 in this TC), scroll painting and, overall, to painting on parchment, paper and various types of textiles is added to mobile objects (Table 4). In this regard, it is worth adding that, in manuscripts, cinnabar was used both for illuminations and as ink (see also above).

Among mobile objects, I have not found any evidence of possible use in ceramic decoration. On the contrary, I have collected numerous attestations relating to the sculptural decoration and, above all for the clay or sandstone sculptures of Buddha present in many Chinese regions such as Sichuan (618–907 — Caves no. 512 and 689, Guangyuan Thousand-Buddha Grotto; He et al. 2012), Shaanxi (11th century — Main cave, Zhongshan Grottoes; Egel and Simon 2013; ≥1016 AD — Jizo Hall, Chongqing Temple; Wang et al. 2014), Datong (1038 AD — Hua Yan Temple; Wang et al. 2020) and

Chongqing (late 12th–mid 13th centuries, retouched until the 1850s — Dazu Rock Carvings; Li et al. 2020b).

Among proper mobile objects, there are also three uncommon finds. In chronological order, the first is represented by the early 11th-century beeswax seals from the documents of the Order of St. John of Jerusalem, sc. Knights of Malta (National Library of Malta, Valetta) investigated by Szczepanowska and Fitzhugh (1999). The second concerns the late 12th–early 13th century Lewis chessmen in ivory preserved in the Collection of the National Museums Scotland and investigated by Tate et al. (2012). The third ones are the 14th–15th century alabaster panels produced in the Midlands (UK) and analysed by Mounier et al. (2020) and Pereira-Pardo et al. (2019).

As far as architectural elements are concerned, the attestations are many and geographically widely distributed. The list provided in Table 5 shows that (a) the *a fresco* and the *a secco* techniques continue to be attested, although the authors frequently specify that some pigments as cinnabar were applied over an ochre layer (Fig. 9C–D) or on dried lime; (b) the evidence is not chronologically continuous. The gaps recorded for the 5th and 8th–9th centuries may be due to the lack of archaeometric studies on wall paintings of that time, or a lack in the bibliographic collection, or an actual decrease in the use of cinnabar in these periods.

Another interesting aspect is that at least until the 4th century, the use of cinnabar remained for the realisation of mosaics' sinopias. In fact, to the 2nd–1st century BC examples found in the house below the Santa Susanna church in Rome (Boschetti et al. 2008), it is possible to add the 4 th-century sinopia found under the mosaic of the villa of Lod in Israel (Piovesan et al. 2014; wall paintings of the villa of Lod in Piovesan et al. 2016).

Lastly, it is worth mentioning the copious use of cinnabar for the decoration of icons. The examples are concentrated between the 13th and 19th centuries and are almost always prepared with the tempera technique (Table 6).

As for Meso and South America, apart from the wide use of cinnabar to paint human bones and funerary/ceremonial practices, the attestations regarding the decoration of mobile objects and wall paintings gradually increase. In the first case, the objects are still primarily linked to the funerary sphere; in the second case, painted walls are concentrated in tombs and monumental complexes. The documentation appears still scarce regarding residential buildings.

To give just some examples, evidence of the use of cinnabar for the decoration of mobile objects has been provided for:

- a funerary golden mask on tumbaga belonging to the Sicán culture (750–1375 AD) and preserved at the Museum of Sicán at Ferrañafe in Peru (Cesareo et al. 2010);
- a Red Jaguar Throne sculpture (800–1250 AD) found in 1936 in the upper part of the sub-structure of the Kukulkan's pyramid at of Chichén Itzá, Yucatán, Mexico (Juárez-Rodríguez et al. 2018);
- the 500–1000 AD wooden Pachacamac Idol (Sepúlveda et al. 2020).

As for wall paintings, based on Magaloni et al. (1993), Magaloni 1998and Vázquez de Ágredos Pascual (2007), "*the earliest use of cinnabar in Mayan wall painting is documented at Bonampak, around the mid Late Classical period*" (*i.e.* 250 and 900 AD). However, further research may lead to identifying this pigment in more ancient Classic contexts (*i.e.* Early Classic or Late Preclassic).

The testimonies are limited in number for the most ancient periods while they increase in the following centuries. Apart

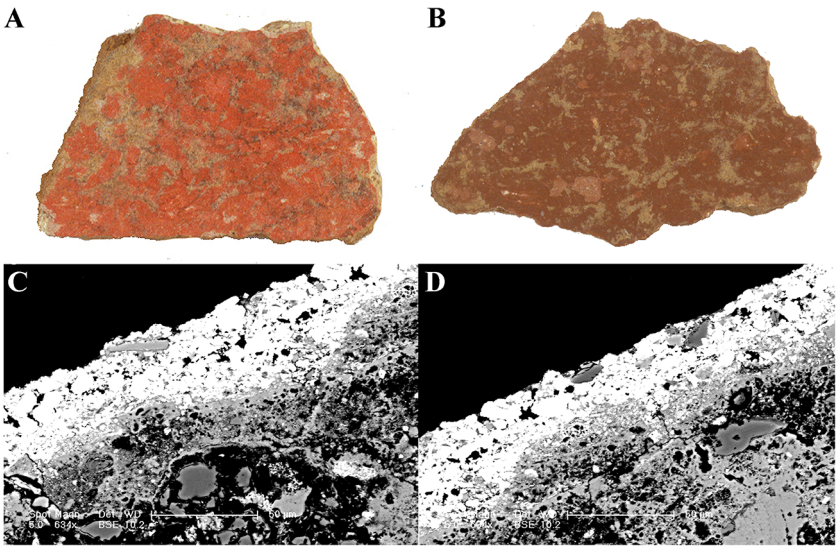

Fig. 9 The visible difference between **A** cinnabar and **B** red ochre in two fragments from the House of Diana at *Cosa*. **C**–**D** SEM-BSE image: it is possible to observe an upper layer of cinnabar (white) overlaying an irregular layer made of yellow ochre (light grey) above the plaster (contrast enhanced to emphasise layering)

Table 4 Cinnabar on parchment, paper and textiles (some examples). Conservation sites in square brackets

Artwork	Chronology	Reference
Codex Eusebii Evangeliorum (A Codex) by Saint Eusebius, first Bishop of Vercelli (?) [Cathedral Treasure Museum of Vercelli, Italy]	345–371	Aceto et al. 2008
Vienna Dioskurides (*Codex Vindobonensis* Med. gr. 1) — Copy of the Herbarium of the Greek doctor Pedanios Dioskurides (parchment) — Written in Constantinople [Austrian National Library at Vienna]	1st quarter 6th	Aceto et al. 2012
Mahamayuri Vidyarajni Sutra (bamboo and bark/textile) found in the Nanmen Buddhist pagoda, Anhui, China	618–627	Liu et al. 2019
Tours Gospel, "*Evangelia Quatuor*" [British Library (Add. MS. 11848), UK]	ca. 825	Clark and Van Der Weerd 2004
Koran (parchment MEK-MS-7LP and MEK-MS-6LP) [Cultural Complex Lamnouni of Meknes, Morocco]	9th	Oubelkacem et al. 2021
Beato (parchment/vellum) [Monastery of Santo Domingo de Silos, Burgos, Spain]	10th–11th	Carter et al. 2016
Commentaries of the Apocalypse (parchment/vellum) [Santa María la Real Monastery in Nájera (AMS-N1), Spain]	10th	Carter et al. 2016
Found near Dunhuang, Khara-hoto or Tujuk, China [Oriental Institute of the Russian Academy of Sciences in St Petersburg, Russia]	pre-10th AD	Clark et al. 1997
Arabic manuscripts (paper) [Archive of the Sacromonte Abbey in Granada, Spain]	11th–17th	Espejo Arias et al. 2008
Medieval miniatures on paper of Saint Matthew and an eagle (symbolic of the evangelist John the Theologian) [Holy Monastery of Simonopetra on Mount Athos, Greece]	12th	Daniilia and Andrikopoulos 2007
Folios on *The Book of Curiosities of the Sciences and Marvels for the Eyes* [The Bodleian Library, Oxford, UK]	Early 13th	Chaplin et al. 2006
Byzantine/Syriac Gospel [British Library Oriental and India Office Collection, UK]	1216–1220	Clark and Gibbs 1997, 1998
Diplomatic documents of the Ottoman Empire (parchment/paper) [Ottoman Archives in Istanbul, Turkey]	13th–20th	Kantoglu et al. 2018
Manuscript D.I.21 better known as Messale Rosselli by different artists. Made in Avignon for the Aragonese Cardinal Nicolas Rossell (1314–1362) [Biblioteca Nazionale Universitaria in Torino, Italy]	Mid-14th	Calà et al. 2019
Lubab al-Ta'*wilfi ma'ani al-tanzil by* Al-Jazin	14th	Durán et al. 2009
Book 814 (1R, 5V, 11R, 26V) by Evangelista della Croce Paper (parchment) [Old library of the Certosa di Pavia]	15th	Bonizzoni et al. 2016
Book 814 (16R, 21V; parchment) by Girolamo dai Libri [Old library of the Certosa di Pavia]	15th	Bonizzoni et al. 2016
Book 818 (1R; parchment) by Benedetto da Bergamo [Old library of the Certosa di Pavia]	15th	Bonizzoni et al. 2016
Book 819 (35R; parchment) by Benedetto da Bergamo [Old library of the Certosa di Pavia]	15th	Bonizzoni et al. 2016
Book 820 (1R; parchment) by Benedetto da Bergamo [Old library of the Certosa di Pavia]	15th	Bonizzoni et al. 2016
Book 821 (15V, 25R; parchment) by Guarnerio Beretta [Old library of the Certosa di Pavia]	15th	Bonizzoni et al. 2016
Book 822 (1R; parchment) by Evangelista della Croce [Old library of the Certosa di Pavia]	15th	Bonizzoni et al. 2016
Book 823 (21R, 39R; parchment) by Guarnerio Beretta [Old library of the Certosa di Pavia]	15th	Bonizzoni et al. 2016
Illuminated manuscript (parchment) [State Archives of Milan, Italy]	1450s	Bruni et al. 1999
The *Cofre no. 31* [Library of the National Palace of Mafra, Portugal]	15th	Carvalho et al. 2018
King George III copy of the *Gutenberg Bible* [British Library, UK]	Mid-15th	Chaplin et al. 2005
Privilegio rodado by King Enrique IV (parchment) [Archive of the Royal Chancellery in Granada, Spain]	15th	Durán et al. 2014
Paper currencies *Da ming bao chao*, *da qing bao chao*, *hu bu guan piao* - China	15th, 19th	Shi and Li 2013
Book of Tides (illuminated parchment) [Private collection]	15th/16th	Vanmeert et al. 2018
A choir book (MAR-97, 98) and a liturgical book (MAR-68) [Collection of Canon Jean Marcadé donated to the French State in 1947, France]	1520–1530	Mounier et al. 2016
Illuminated foral charter of Setubal (parchment) [Portuguese National Archive, Portugal]	1515	Guerra et al. 2016
Persian Herati lacquered manuscript (paper). Produced in the city of Herât, Afghanistan	1530	Hayez et al. 2004
Manueline foral charter of Sintra (parchment). Attributed by D. Manuel I of Portugal to the village of Sintra, Portugal	1514	Manso et al. 2013
Coaching inn books (parchment/vellum) [Monastery of Santo Domingo de Silos, Burgos, Spain]	16th–18th	Carter et al. 2016
Printed book *Osorio* (paper) [Museum of Slavonija, Osijek, Croatia]	16th–19th	Lukačević et al. 2013
Map of Vicenza (paper), Italy - Atlas Major	1640	Castro et al. 2008
Lengjinjian (gold-dusted) paper [China Printing Museum, Beijing, China]	1662–1722	Li et al. 2020c
Japanese painting on paper (horizontal scroll) - Bamo Dōi-zu attributed to Kanō Sansetsu (1589–1651). [Stibbert Museum in Florence, Italy]	17th	Quattrini et al. 2014
Javanese and Thai manuscripts [British Library Oriental Department]	ca. 1738	Burgio et al. 1999

Table 4 (continued)

Artwork	Chronology	Reference
Portrait of Bazalibudala Arhat (woven silk net and paper) by Ding Guanpeng [Palace Museum, Beijing, China]	1756	Li et al. 2020a
Chinese scroll paintings - Portrait of Bazalibudala Arhat by Ding Guanpeng [China]	1756	Li et al. 2020d
Volume *Ornitologia dell'Europa meridionale* by Clemente and Rosalba Bernini. Printed in Parma. [Biblioteca Palatina at Parma, Italy]	1772–1793	Zannini et al. 2012
Volume *Beytraz zur Naturgeschichte der Vogel* by Joachim Johann Nepomuk Spalowsky. Printed in Vienna [Biblioteca Palatina at Parma, Italy]	1790–1795	Zannini et al. 2012
Cover of a cantoral book (parchment/vellum) [Monastery of Santo Domingo de Silos, Burgos, Spain]	18th	Carter et al. 2016
Drawings and sketches by Szymon Czechowicz (1689–1775) [Jagiellonian University Museum, Krakow; National Museums in Krakow and Warsaw, Poland]	18th	Doleżyńska-Sewerniak et al. 2020
Thangkas (cotton cloth) likely from Tibet [private collection]	18th (?)	Brocchieri et al. 2020
Palm leaf manuscript from India	18th–19th	Singh and Sharma 2020

from the previously mentioned findings of Bonampak, the testimonies mainly regard the following sites:

- Teotihuacán, Mexico — Flourished between the 1st and 6th century, returned several occurrences: (a) the altar stone from the Conjunto Xolalpan (not analysed in Linné 1942); (b) the mural painting from the Calzada de Los Muertos analysed by Torres Montes (1972); (c) the structure 52F in the Complejo Calle de Los Muertos analysed by SEM in Gazzola (2009); (d) the Xalla building, analysed but doubtfully in López Puértolas et al. (2019); and (e) the Temple of the Feathered Seashells and Quetzalpapalotl Palace analysed by Argote et al. (2020);
- Monte Albán, Oaxaca, Mexico — In the Late Classic period (790–792 AD) tombs 104 and 105 (Magaloni 1998);
- Tajín, in southern Mexico — In the buildings nos. 1 and 42 (site chronology: 100–1023; Gazzola 2009 reporting personal communications and Ladrón de Guevara 1999);
- Calakmul, Campeche, Mexico — In the Royal Tomb Garra de Jaguar, dated to the Classical Period (Vázquez de Ágredos Pascual 2004);
- Palenque, southern Mexico — In the Temple of La Cruz (site chronology: 3rd century BC–8th century AD; Gazzola 2009 reporting a personal communication);
- Ek'Balam, Mexico — In the room 23 at the Acropolis (~900 AD; Vandenabeele et al. 2005);
- Huaca Tacaynamo, part of the Chan Chan complex in Perú (1412–1614 AD; Brooks et al. 2008).

Cinnabar for ceramic lustre

The lustre (or better "reduced-pigment lustre") is a particular decoration with a metallic sheen (coppery, golden or silvery) and iridescent colours (yellow, amber, ruby red, brown, green, blue, violet), used in Medieval and Renaissance times for the decoration of ceramics (Caiger Smith 1985; Mason 2004). First produced in Iraq (esp. Baghdad) around the 9th century, it was then introduced in Persia (*e.g.*, Kashan in Iran) and in some Mediterranean countries such as Egypt and Spain (esp. Almeria and Malaga, followed by Manises, Muel, Paterna and Valencia). Later, the technique arrived in Italy (esp. Gubbio and Deruta followed by Cafaggiolo, Faenza and Naples) probably thanks to the mediation of Pinturicchio. It is likely that this Umbrian painter became aware of this technique during the works carried out for Pope Alessandro VI Borgia at Rome (Caiger Smith 1985; Padeletti and Fermo 2003a, 2003b; Hess 2004).

Each region developed its own recipes and procedures so much so that the final products differ in decoration and visual appearance. For example, metal reflectivity is a characteristic of Spanish products not found in Syrian or Egyptian lustre (Molera et al. 2007). At a smaller scale, Deruta and Gubbio (at 43 km as the crow flies) developed two different styles, respectively characterised by "*a copper-oxide luster and a brassy, straw-colored sheen*" and a "*silver-oxide luster and a more golden reddish gloss*" (Hess 2004).

As reported in the literature, several recipes were handed down by several authors such as:

(1) Muhammad al-Jowhar al-Neyhapuri in his treatise *Jowhar-name-ye Nezami* in 1196 Iran (Pradell et al. 2004; Pérez-Arantegui and Pardos 2008);
(2) Abu'l Qasim of Kashan in 1301 (Pérez-Arantegui and Pardos 2008);
(3) Gaetano Milanesi in 1864, publishing three treatises on glass manufacture (*Trattato secondo di Benedetto di Baldassare Ubriachi* presumably of the 14th century in Milanesi 1864; Pérez-Arantegui and Pardos 2008);
(4) Nicolau de Reyner of Barcelona in his *Libre de les Fornades* in the years 1514–1519 (Pradell et al. 2004; Pérez-Arantegui and Pardos 2008);

Table 5 Cinnabar on architectural structures from the 4th century AD onwards (some examples for Europe, Africa and Asia)

Type	Location	Country	Chronology	References
Wall painting (fresco and secco retouches)	Roman villa of Lod	Israel	4th	Piovesan et al. 2016
Wall (secco)	Maiji Mountain Grottoes, Tianshui	China	6th–19th	Liu et al. 2016
Wall painting	Barone and Leonesse Tomb, Tarquinia	Italy	6th BC	Barone et al. 2018
Walls, reliefs and Buddha statues	Caves no. 512 and 689. Guangyuan Thousand-Buddha Grotto, Sichuan	China	618–907	He et al. 2012
Wall (fresco)	12 Byzantine churches, Mani Peninsula	Greece	10th–15th	Hein et al. 2009
Wall painting	Feng Hui tomb, Bin County, Shaanxi Province	China	907–960	Wang et al. 2004
Wall painting (fresco)	Monastery of San Baudelio de Casillas, Soria	Spain	11th	Edwards et al. 2001
Wall painting	Mosque of al-Qarawiyyin, Fez	Morocco	11th–12th	Fikri et al. 2018
Wall painting (fresco, mezzo fresco)	St. Maria Veterana, Triggiano, Bari	Italy	11th–16th	Fioretti et al. 2020
Wall (fresco-secco)	Yemrehanna Krestos Church, Mount Abuna Yosef	Ethiopia	Early 12th	Gebremariam et al. 2013
Wall	Fortress at Mount Sofeh, Isfahan	Iran	12th	Holakooei et al. 2020
Wall (fresco and lime painting)	By Manuel Panselinos. Protaton Church, Mount Athos	Greece	13th	Daniilia et al. 2000
Stalactite vaults, plasterwork	Hall of the Kings, Alhambra, Granada	Spain	13th–15th	Dominguez-Vidal et al. 2012, 2014
Wall painting (fresco)	Islamic style. Convento de la Peregrina, Sahagun	Spain	13th	Edwards et al. 1999a
Wall (fresco, secco)	Church of St. Gallus in Kuřívody, northern Bohemia	Czech Republic	2nd half 13th	Hradil et al. 2014
Sculptures and reliefs	West and south portals, Parma baptistery	Italy	13th(?)	Pinna et al. 2020
Wall painting (fresco)	Convento de la Peregrina, Sahagun, Léon	Spain	13th	Rull Perez et al. 1999
Marble capitals	Alhambra complex, Granada	Spain	14th(?)	Arjonilla et al. 2016
Wood ceiling	Hall of the Abencerrages, Hall of the Two Sisters - Alhambra, Granada	Spain	14th	Arjonilla et al. 2019a
Plasterwork - scalloped arches	Salón de Embajadores, Mudéjar Palace of the Real Alcázar, Seville	Spain	14th–17th	Blasco-López et al. 2016
Woodwork - carved polychrome carpentry (tempera grassa)	Hall of the Mexuar Palace, Alhambra, Granada	Spain	1314–1325	Cardell et al. 2009
Wall painting and stuccoes	Alhambra (Granada): east wall of the Hall of the Mexuar, façade of the gate of the Mexuar, east pavilion in the Lions Courtyard (Lions Palace), Gonz lez Pareja House in Partal Palace	Spain	14th and beyond	Cardell-Fernández and Navarrete-Aguilera 2006
Wall painting (fresco-secco)	Panagia Church at Patsos and Church of Theotokos in Meronas, Amari, Crete	Greece	14th	Cheilakou et al. 2014
Wall painting (fresco, secco)	By Ambrogio Lorenzetti. St. Augustine church, Siena	Italy	1335–1338	Damiani et al. 2014
Wall painting	Patio de las Doncellas, Reales Alcazares, Sevilla	Spain	14th–16th	Durán-Benito et al. 2007
Wall painting (fresco)	Church of SS Cosmo and Damian at Basconcillos del Tozo, Castille y Léon	Spain	14th?	Edwards et al. 1999a, 1999c
Red pigments on stone (moulded architectural fragments)	Augustinian friary at the Magistrates Court Site, Kingston upon Hull, England	UK	14th–16th (?)	Edwards et al. 2010
Vaulted ceiling	Alhambra's Hall of the Kings, Alhambra, Granada	Spain	14th	Gómez-Morón et al. 2016
Wall (fresco, secco)	Dominican Monastery in Ptuj	Slovenia	3rd quarter of the 14th	Gutman et al. 2014
Wall painting	Princely church of Curtea de Arges	Romania	14th	Ionescu et al. 2004
Wall (fresco-secco)	Churches of Sts. Georgios Vounou, Nikolaos Kyritzi and Nikolaos Magaliou at Kastoria	Greece	14th–17th	Iordanidis et al. 2011
	Dagaoxuan Taoist Temple, Beijing	China	1368–1912	Lei et al. 2017

Table 5 (continued)

Type	Location	Country	Chronology	References
Walls, ceiling, architrave, etc.				
Wall painting	Façade of the King Pedro I Palace, Alcázar of Seville	Spain	1356–1366	López Cruz et al. 2011
Wall painting (fresco)	Royal Wawel Cathedral in Krakow	Poland	14th–15th	Rafalska-Lasocha et al. 2010
Stuccoes	Oratory room, Islamic University Madrasah Yusufiyya, Granada	Spain	14th (original)–19th retouches	Romero-Pastor et al. 2011a
Wall painting	Dazhao Temple, Hohhot, Mongolia	China	1368–1644	Wei et al. 2010
Wall painting	Longju Buddhist temple, Guanghan, Sichuan	China	~1466–1644	Chen et al. 2019
Wall (fresco)	Saint Stephen's chapel, Val Venosta, Bozen, Italy	Italy	~15th	Costantini et al. 2020
Wall (fresco-secco)	Monastery church Christ Antiphonitis, Kalogrea, Kyrenia	Cyprus	End of 15th	Daniilia et al. 2008a; Daniilia and Minopoulou 2009
Wall painting (tempera)	Abuna Yemata Guh church	Ethiopia	2nd half 15th	Gebremariam et al. 2016
Wall (fresco, secco)	Minorite church of St. Francis of Assisi, Koper	Slovenia	15th	Levstik et al. 2019
Wall painting	Forbidden City, Yanxi Hall, Bejing	China	15th	Liu et al. 2018b
Wall painting (fresco)	Church of Santa Maria de Hermo, Asturias	Spain	15th	Pérez-Alonso et al. 2004
Wall painting (fresco)	Mirador de la Reina, Baños de Doña María de Padilla, arch of the gypsum palace; Alcazar of Seville	Spain	? from 15th	Pérez-Rodríguez et al. 2014
Wall painting (fresco)	San Isidoro del Campo Monastery	Spain	15th	Pérez-Rodríguez et al. 2020
Wall painting	Chapel of the Ponthoz Castle	Belgium	15th?	Vandenabeele et al. 2005
Wall painting (fresco and mezzo fresco?)	By the Workshop of S. Lorenzo de Skofja Loka? Church of Anunciación de María at Crngrob	Slovenia	1400–1410	Kriznar et al. 2007
Wall	By Luca Longhi (1507-1580). Sala Dantesca, Biblioteca Classense in Ravenna	Italy	ca. 1580	Fiorillo et al. 2020
Mudéjar Ceiling and Doors	Casa de Pilatos Palace, Seville	Spain	16th	Garrote et al. 2017
Wall painting (fresco)	Assumption Cathedral, Sviyazhsk	Russia	Late 16th–early 17th	Khramchenkova et al. 2018
Wall painting (fresco)	Vaults of the Sala Vaccarini Library, Benedictine Monastery, Catania	Italy	17th	Barone et al. 2016
Wall painting (secco)	Katholikon of St Stephen's monastery at the Meteora	Greece	Early 17th	Daniilia et al. 2008b
Wall painting	Imperial Taidong Tomb (wood), Hebei Province	China	1644–1912	Fu et al. 2020
Ceiling	By Peter Paul Rubens (1577-1640). The Banqueting House - Palace of Whitehall, London	UK	1630–1636	Vlachou-Mogire et al. 2020
Wall painting	Fatih Mosque, Instanbul (Architect Mimar Mehmet Tahir)	Turkey	1767 and 1771	Akyuz et al. 2015
Wall painting	Silsangsa temple and Bulyoungsa temple	Korean	18th	Ha and Lee 2015
Wall (fresco, secco, egg tempera)	Catholicon of Saint Demetrius, Stomion, Larissa	Greece	2nd half 18th	Malletzidou et al. 2019
Wall painting (fresco and tempera)	By Konstantinos and Athanasios Zografi. Church of St Athanasius, Moschopolis	Albania	1745	Pavlidou et al. 2008
Stuccoes	Presbytery, Church of Sant'Andrea, Mantua (SAM)	Italy	18th–19th	Sansonetti et al. 2010
Wall painting	Room of the Beds (Royal Bath of Comares), Alhambra, Granada	Spain	19th century redecoration	Arjonilla et al. 2019b
Wall paintings	Drăguțești wooden church, Argeș County	Romania	1813–1814	Dinu et al. 2020
Wall painting	Church of St. Peter and St. Paul at Upton, near Newark, Nottinghamshire	UK	Early 19th	Edwards et al. 2005

Table 5 (continued)

Type	Location	Country	Chronology	References
Wall painting	Five Northern Provinces' Assembly Hall, Wafangdian	China	1861–1874	Hu et al. 2013
Painted plafond (ceiling)	By Antonio Vighi (1765–1844). Red Living Room, historical house, St.-Petersburg	Russia	ca. 1830	Petrova et al. 2019
Wall painting	Jokhang temple, Lhasa, Tibet	China	Mid to late 19th	Song et al. 2018
Wall (tempera)	Catholicon of the Rila Monastery	Bulgaria	1st half 19th	Stamboliyska et al. 2021
Wall painting	The Cistercian Abbey of Stična and The Manor of Novo Celje	Slovenia	?	Škapin et al. 2007

(5) Cipriano Piccolpasso from Casteldurante in his treatise *Li Tre Libri dell'Arte del Vasaio*, reporting in 1558 the recipes of Mastro Giorgio Andreoli (Padeletti and Fermo 2003a-b; Pérez-Arantegui and Pardos 2008);
(6) Henry Cock in his *Relación del viaje hecho por Felipe II, in 1585, in Zaragoza, Barcelona y Valencia, in Morel Fatio and A. Rodríguez Villa* in 1585 (Pradell et al. 2004; Pérez-Arantegui and Pardos 2008).

Other ancient recipes were also found in other types of documents, such as those reporting the recipe of Jacinto Causada from Alcora (1765), the *Ordinaciones de la Cofradía y Gremio de Alfareros y Vajilleros* (Ordinances of the Fraternity and Guild of Potters) and a formal request emitted by the chief magistrate of Valencia in 1785 (Pérez-Arantegui and Pardos 2008). In this regard, also the distinction made by Caiger Smith (1985) between passive and active recipes is worth mentioning considering that the active ones are those using cinnabar.

All these recipes are discussed in detail by Pérez-Arantegui and Pardos (2008) and reported in Padeletti and Fermo (2003a, 2003b), Pradell et al. (2004) and Roqué et al. (2008) to which the reader is referred for details. Here, it is interesting to note that only the treatises at points 1, 4, 5 and 6 and the

Table 6 Cinnabar on icons (some examples from Europe and Africa). Conservation sites in square brackets

Artwork	Findsite [Conservation site]	Chronology	Reference
Life of the Virgin and Christ	St. Mercurius Church, St. Mercurius monastery, Old Cairo, Egypt	13th	Abdel-Ghani et al. 2009
St. Nicholas (tempera)	Church of Ascension in Mborje, Korçë, Albania [Museum of Medieval Art of Korçë]	14th	Franceschi et al. 2011
Pietà, Virgin and Child, Virgin unfolding Rose and Saints, St. Panteleimon, Deesis, St. Menas (modified egg tempera)	Byzantine painting art of northern Greece	14th–19th	Lazidou et al. 2018
St. George with scenes (tempera)	Church of St. Nicholas in Boboshtica, Korçë, Albania [Museum of Medieval Art of Korçë]	15th	Franceschi et al. 2011
Cretan painters	[Benaki Museum, Athens, Greece]	Mid 15th–17th	Karapanagiotis et al. 2009, 2013
Saint Theodore El-Shatby by Anstasy Al-Rumi (tempera)	Painted at Jerusalem (inscribed) [Church of Saint Theodore El-Shatby, Deir EL-Sankoria, El Minia]	1549/19th	Abdel-Maksoud et al. 2020
Our Lady, the Life-giving Spring; Saint Athanasios the Athonite (egg tempera)	[Saint Modestos's Church in Kalamitsi, Chalkidiki]	16th	Daniilia et al. 2002
Ethiopian icons	[National Museum of African Art, Smithsonian, USA]	17th–19th	James 2005
Daniel and Jeremiah prophets and Aaron and Gideon prophets (tempera)	Greek school style [Romania?]	Early 18th	Bratu et al. 2016
Last Judgement by Greek master Ioannis from Kapesovo (egg tempera)	[Byzantine Museum of Ioannina, Greece]	1771	Kovala-Demertzi et al. 2012
Saint Nicholas; Saint Basil the Great; Saint John Baptist; Our Lady of seven sorrows; The Lord Christ entrance into Jerusalem (egg tempera)	Antim Monastery, Bucharest - One-wood Monastery, Valcea county, Romania	1st half of the 19th	Serafima et al. 2019
Minoan icon by Nevjanska school	[Manastir Pokrova Presvete Bogorodice, Lešje, Paraćin, Serbia]	19th	Stojanović et al. 2015

recipe of Jacinto Causada inform about the use of cinnabar. Therefore, its use is to be considered widespread but not mandatory to produce lustre.

The numerous archaeometric analyses allow the main characterising feature of lustre to be described as a few hundred nanometres thick layer, including silver and/or copper nanocrystals dispersed in a glassy matrix.

The production required three firings:

(1) firing of the biscuit at about 900–1000°C;
(2) glazing (likely by immersion) and subsequent firing;
(3) painting of the lustre decoration (dissolved in vinegar) and subsequent firing in a 480–600°C temperature range and a reducing atmosphere (but not fully reducing, see Molera et al. 2007), able to reduce Cu and/or Ag compounds into the metal state.

The materials used and the procedures adopted in the three phases all affect the final appearance of lustre and characterise specific products. For example, among all possible types of glazes, (a) the tin-opacified lead-alkali were the preferred ones (Tite et al. 1998), (b) lead-containing glazes were necessary to obtain a metal-like reflectivity (Molera et al. 2007) and (c) the Italian glazes were often characterised by higher Na_2O contents than those measured in Spanish products (Padeletti and Fermo 2003a).

Among the key factors responsible for the appearance of the end product, it is possible to enlist:

– the (nanometric) dimension, composition and distribution of the particles (for iridescent metallic sheen),
– the firing and annealing temperatures (influencing the size of the final particles),
– the atmosphere conditions (influencing the reduction of Cu and Ag compounds), and
– the relative ratio of Cu and Ag and their absolute amounts (for colour variations).

The key factors mentioned above have all been investigated in detail and discussed in Molera et al. (2001), Pérez-Arantegui et al. (2001), Padeletti and Fermo 2003a, 2003b, 2004), Padovani et al. (2003, 2004), Pérez-Arantegui et al. (2004), Pradell et al. (2004), Pradell et al. (2005), Padovani et al. (2006), Smith et al. (2006), Cepriá et al. 2007, Molera et al. (2007), Pérez-Arantegui and Pardos (2008), Pradell et al. (2008a–b), Roqué et al. (2008) and Fermo and Padeletti (2012).

These milestone papers represent the essential starting point for tackling a study of lustre and I, therefore, refer to these authors for the necessary insights. Here, I will focus on why cinnabar was used, although it decomposes completely during firing and, therefore, there is no trace of it in the finished products.

Experimental archaeometry performed by Pradell et al. (2004, 2008b), Cepriá et al. (2007), Molera et al. (2007) and Roqué et al. (2008) has clearly shown that the presence of cinnabar is relevant during firing rather than in the characteristics of the final product.

The decomposition of metacinnabar creates a sulpho-reducing atmosphere that reduces tenorite (CuO) to cuprite (Cu_2O). In a temperature range between 400 and 600°C, the released sulphur forms copper and silver sulphite (*e.g.*, Ag_2SO_3), sulphate and sulphide (*e.g.*, $CuSO_4$, Ag_2S, $3Ag_2S{\cdot}Ag_2SO_4$) that prevent the alloying of Ag with Cu while mercury vapours ensure that silver compounds are not reduced to metallic silver before entering the glaze.

Undoubtedly, lustre production implies a delicate process in each phase and accidents may occur along the way. For example, the formation of Ag-Hg compounds such as luanheite (Ag_3Hg) can inhibit the formation of metallic silver. Furthermore, it has been proved by voltammetry of immobilised microparticles (VMP) studies that both iron and cinnabar promote the reduction of Ag and Cu, provided that only Ag or only Cu are present. When both Cu and Ag are present, the reduction process of Ag is made more difficult (Cepriá et al. 2007). Consequently, the role of cinnabar varies with the variation of the present quantities of Cu and Ag because it triggers different chemical processes, induces a variation in the temperatures at which the processes take place and, inevitably, leads to products with distinct characteristics.

Despite evidence mainly obtained on Hispano-Moresque pottery, further studies on Italian lustre have also suggested the use of cinnabar as a pigment in ruby red lustres from Gubbio (Padeletti and Fermo 2004)

Among the archaeometrically investigated lustre productions, the use of cinnabar has been assessed for both Hispano–Moresque (13^{th} century *Les Olleries Xiques* workshop at Paterna, Valencia, Spain; Molera et al. 2001) and Italian lustres (15^{th}–17^{th} century majolicas from Gubbio; Padeletti and Fermo 2004); however, the quantification of Hg is not a common routine.

Cinnabar in lacquered objects

The lacquer is a natural resin chiefly composed of catechol derivatives that "*polymerizes by the oxidation of urushiol with catalysts of laccase during the drying process*" (Ma et al. 2017). The composition of the lacquers varies according to the tree from which the sap is extracted, and, on this basis, it is possible to draw meaningful geographical distinctions:

– the *Rhus vernifera* grows in China, Japan and Korea and urushiol is the characteristic component of the lacquer it produces;

- the *Rhus succedanea* grows in Vietnam and Taiwan (Formosa) and laccol characterises the composition of this lacquer;
- the *Melanorrhoea usitate* grows in Laos, Myanmar, Tailand and Cambodia and thitsiol identifies its lacquer.

The production technology, hardening and stability of these three types of lacquers have been studied, among others, by Kumanotani (1995), Niimura et al. (1996a, 1996b), Niimura and Miyakoshi (2006), Lu et al. (2007), Frade et al. (2009) and Ma et al. (2014).

Lacquerware has been and still is popular throughout Asia. It has been used for multiple purposes, ranging from the decoration of objects such as tableware and furniture to the decoration of inscribed plaques, jewellery and coffins.

Indian, Burmese, Thai and Japanese and, finally, European decorations are well known. The production period probably begins in the Stone Age and reaches up to the present day. The oldest examples are likely represented by Japanese (*e.g.*, the Stone Age arrowheads; see Niimura et al. 1999 and references therein) and Chinese objects (see below). Vietnamese products also include particularly ancient lacquer decorations, dated to the Dong Son culture (*i.e.* more than 2000 years ago) (Naziree 2013). Conversely, Burmese lacquer—the so-called *thitsi*—appears chronologically later basing on archaeological evidence. Nevertheless, Tamburini et al. (2019) pointed out that it "*has probably been used for more than a millennium*".

Unfortunately, most of the studies concerning Asian lacquers did not concern the pigment palette identification; therefore, the archaeometric case studies reporting on cinnabar use are few and mainly concern Chinese products.

In China, the oldest examples date back to the Chinese Neolithic age; however, the authors disagree on which is the oldest lacquered object found so far. Based on Ma et al. (2017), the oldest example is represented by a black lacquered bowl dated to 8000 years BP, found at Kuahuqiao (Xiaoshan, Zhejiang). Based on Li et al. (2009) and Wang et al. (2018), the oldest example is represented by the cinnabar-containing lacquer-painted wood bowl found at the Hemudu site (Yu-yao, Zhejiang) and dated to more than 7000 years BP. Li et al. (2009) also propose a map of the lacquerware distribution between the Qin (5000–206 BC) and the Han (206 BC–220 AD) Dynasties.

Starting from the Tang Dynasty (618–907 CE), it is possible to follow the development of the "carved lacquer", which reached its peak in the Qianlong Period (1736–1796 CE) and was particularly popular until the Qing Dynasty (1636–1912 CE) (Wang et al. 2018). This technique has its basic raw material in lacquer and consists of obtaining the decoration by carving a thick lacquer layer. Among the colours of carved lacquered objects (red, yellow, green and black or polychrome), the red one was the favoured and cinnabar was the preferred colouring pigment used to prepare it.

Archaeometric studies were conducted on various types of Chinese objects and provided comparable results.

- In the wood-based lacquer painting screen discovered in the Northern Wei (386–534 AD) tomb of Si-ma Jin-long (Shi-jia-zhai village, Datong, Shanxi), Li et al. (2009) identified cinnabar along with numerous other pigments such as gypsum, carbon black, orpiment and realgar.
- In the imperial lacquer plate (202 BC–8 AD) found in the Luozhuang Han tomb (Zhangqiu, Shandong), Ma et al. (2017) demonstrated that the first ground layer (over the hemp canvas) is made of organic materials (*urushi*, tree oil, amorphous carbon, quartz, albite and K-feldspar). The lacquer layers are made of *urushi* and perilla or tallow tree oil. The final red-coloured layer is made of *urushi* and cinnabar.
- In the lacquer objects (mostly fragments of a chariot dated to the 1st century) found in the 22nd and the 31st Noin-Ula barrows in Mongolia, Karpova et al. (2017) found cinnabar, umber, iron oxides, orpiment and charcoal in the red and brown lacquers, made of urushiol, drying oil (probably tung oil) and colophony.
- In the carved lacquers (1772 AD) sampled from a decorated panel in the Forbidden City, Wang et al. (2020) identified cinnabar and orpiment as the main colourants of the red and yellow lacquers, respectively.
- In the coffin of Emperor Qianlong (1711–1799 AD) brought to light at Zunhua, in the Eastern Imperial Tombs of the Qing Dynasty, Hao et al. (2017) found a complex stratigraphy including (a) the wooden layer; (b) the lacquer ash layer (9 stucco layers and 10 fibres layers; the so-called *wan* lacquering technique) and (c) the lacquer film pigment layer, mostly consisting of organic materials (lacquer sap from *Rhus vernicifera* ➔ urushiol, animal gelatin, drying oil and proteinaceous materials) and four layers of inorganic pigments: calcite, carbon black, cinnabar and gold (the so-called *Jin Jiao* technique, *i.e.*, the technique applied to mix pigments and lacquer and "*painting lacquer above the gold technique*").
- In a birthday inscribed lacquer plaque (1866 AD), Zheng et al. (2020) found a primer lacquer layer made of calcite, cinnabar, minium and Chinese lacquer, followed by a second plaster lacquer layer made of gypsum and Chinese lacquer and a third and last layer made of Chinese lacquer.

Aside from China, a few examples of cinnabar-lacquered objects from the Japanese and Vietnamese areas have been analysed by Colomban and Mancini (2013) and Kamiya et al. (2015). Burmese lacquered objects kept at the British

Museum in London (UK) have been the object of the archaeometric study performed by Tamburini et al. (2019).

In Europe, lacquerware began to be imported in the mid-16th century. Around the late 17th century, the practice of combining local furniture with lacquered Asian panels spread throughout Europe. The example presented by Bösiger (2019) well testifies this practice and the advent of French lacquer. The history of the red lacquered chest of drawers (18th–20th centuries), presumably realised by Charles Chevallier dit Le Jeune and François Rübestück[4], has a long and complicated history that not even the many analyses were able to clarify fully. The only incontrovertible results concern (a) the use of cinnabar in the Chinese lacquer, (b) the use of minium in the French lacquer and (c) a reworking in the 20th century confirmed by the presence of the red pigment PR3 (beta-naphthol-toluidine).

A special mention of the South American lacquers decorated with the technique called *barniz de Pasto*[5] is deemed necessary at this point. This technique uses a native South American resin named *mopa mopa*. This phenolic resin is obtained from the *Elaeagia pastoensis*[6] tree, which grows "*in mountainous regions of western South America from Colombia to Ecuador*" (Newman et al. 2015). Although initially used as an adhesive (for example, glue feathers to ceremonial plumes; see Gomezjurado Garzón 2008), its later use mainly included decorative purposes. Once soaked, heated, kneaded and masticated, the *barnizadores* spread in wide and thin sheets and coloured with the addition of mineral pigments (*e.g.*, ochre) or animal (*e.g.*, cochineal) and vegetable dyes (*e.g.*, achiote—*Bixa orellana*) (Mora-Osejo 1977; Gomezjurado Garzón 2008). When a particular brilliance was sought, a layer of metal leaf — typically silver — was "*sandwiched between two layers of coloured and/or uncoloured Pasto varnish*" (Portell 1992). Depending on the materials used and the technique, various types of *barniz* could be obtained (*e.g.*, bright, matt and chinesco) to waterproof and decorate (carved) wooden surfaces of domestic or religious materials (Fig. 10). The analyses carried out on this type of object are very limited. Consequently, also the claims of cinnabar are numerically small and concentrated on the *qeros*, *i.e.* the typical Andean drinking vessels:

- on a series of Inka *mopa mopa* carved and painted wooden *qeros*, the most common pigment was cinnabar (Pearlstein et al. 2000);
- in the pink samples taken from three *qeros* dated to the Transitional Inka/Early Colonial period, cinnabar was found together with cristobalite, anatase and α-quartz[7] or with gypsum and anhydrite[8] (Howe et al. 2018);
- in Inka-Colonial *qero* cups kept in several Museums in the USA[9] (Newman and Derrick 2001);
- in an Inka *qero* excavated at Moqi (southern Peru), cinnabar and orpiment were used for the red and yellow inlays, respectively (Newman et al. 2015).

Regarding these *mopa mopa* objects, it is also interesting to note that not only cinnabar but also calomel has been found (see the following section on Calomel).

Lastly, it is also worth adding that the studies performed by Strahan and Tsukada (2016) revealed that cinnabar-containing lacquered objects emit small amounts of mercury vapour and may represent a source of contamination.

A (very) short list of other uncommon finds and occurrences of cinnabar (chronological order)

- The second century — Traces of cinnabar were found on stones deemed to have served as touchstones. Together with gold and silver weapons and vessels, these stones were found in the rich burial of two males aged 40 and 60, uncovered near the Roman army camp at Mušov (south Moravia, Czech Republic). In particular, the stone no. 12 bore traces of cinnabar as well as streaks of gold, silver, tin, lead and various types of gold and silver or gold, silver and copper alloys (Ježek et al. 2018).
- The fifth–seventh centuries — Cinnabar has been found on textiles at Samdzong in Nepal. The archaeological context includes ten shaft tombs containing 105 individuals. The collection of textiles analysed by Gleba et al. (2016) were coloured with Indian lac, munjeet, turmeric and knotweed/indigo. Those coloured with cinnabar likely recall a technique (dyeing with mineral pigments) that "*was well developed in China already during the Shang (1600–1046 BCE) and Zhou (1046–256 BCE) Dynasties*" and whose products were intended for the elite (Gleba et al. 2016).
- High Middle Ages — Cinnabar, mercury and antimony were used to produce counterfeits of natural bezoars. These last are masses, sometimes mixed with food, formed in the digestive tracts of some ruminants and

[4] Preserved at the Musée d'Art et d'Histoire de Genève, Switzerland.

[5] Barniz (or barniz de Pasto) is the name used to indicate the resin in today's Colombia.

[6] The further use of the *Elaeagia utilis* (Goudot) Wedd by the Incas, for example, for the decoration of the *qeros* (wooden ceremonial drinking vessels), has been hypothesised by Newman et al. (2015).

[7] Samples Private Collection 2 and Smithsonian National Museum of the American Indian no. 10/5860 in Howe et al. (2018).

[8] Sample Smithsonian National Museum of the American Indian no. 15/2413 in Howe et al. (2018).

[9] Brooklyn Museum of Art, National Museum of the American Indian/Smithsonian Institution, Metropolitan Museum of Art, and American Museum of Natural History.

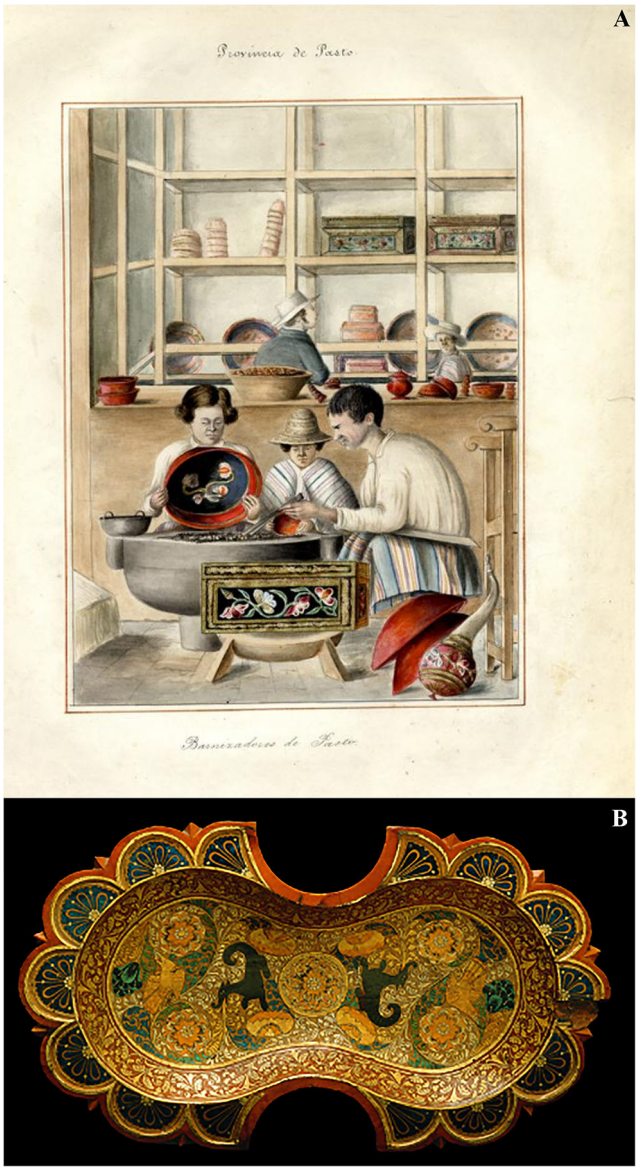

Fig. 10 **A** "Barnizadores de Pasto" — A watercolour made in 1853 by Manuel María Paz (1820–1902) showing three people decorating mobile objects in a shop at Pasto in Colombia. Photo from the World Digital Library (https://www.wdl.org/en/item/9074/). **B** Barniz de pasto: Batea lacada from Pasto in Colombia (inv. no. 12242). Dated to the 17th century. Dimensions 5.50 (H) × 43.5 (L) × 24.5 (W) cm (Photo credits: Joaquín Otero Úbeda, Museo de América. CER.es (http://ceres.mcu.es), Ministerio de Cultura y Deporte, Spain). The CC-BY licence does not supersede previously copyrighted material; therefore, these images remain under owner's copyright.

humans. Introduced to Europe by the Crusaders — together with unicorn horns — they were believed to be magical antidotes against arsenic poisoning (Barroso 2013).

– The sixteenth century — In the wreck of Gnalic various kinds of goods were found: glassy materials, semi-finished metals, white lead in wooden barrels and cinnabar. The ship "Gagliana Grossa" set sail from Venice in the autumn of 1583 was headed for Constantinople (Auriemma 2018).
– The eighteenth century — The frigate HMS Pandora was dispatched (1790) to intercept mutineers on the HMS Bounty. Returning from Tahiti (March 1791), it struck the Barrier Reef and sank. Cinnabar powder associated with keratotic material (*e.g.*, collagen) was found in a chest found in an officer's cabin (Edwards et al. 2003).
– The nineteenth–twentieth centuries — Cinnabar has been frequently used in reverse glass painting in Chinese artworks (Steger et al. (2019a), as well as in the artworks of the famous Russian artist Wassily Kandinsky (1866–1944; Steger et al. 2019b) and the German artist Carlo Mense (1886–1965; Steger et al. 2019c).
– Undetermined age — Human blood later reinforced by sub-micron particles of cinnabar and red ochre were found in The Turin Shroud (Fanti and Zagotto 2017 with references therein).
– While I cannot support this with proper literature, I have also found that in India, cinnabar was typically used for the Hindu practice of making the red dot on the forehead.

Cinnabar procurement and trade

I warn the reader that while the investigations carried out on finished products are consistent, those relating to mines, processing and trade are much smaller. The discussion of the various topics is therefore limited and geographically unbalanced.

In Europe, apart from the Almaden district's mines in Spain, the information on which mines were exploited in ancient times is still scarce or scattered in hardly accessible publications. Perhaps the exception is the case of the mines of Mt. Altai in Serbia, where the archaeological excavations brought to light several findings of great interest. Several authors claimed that the exploitation of the Šuplja Stena mine (Mt. Avala, Belgrade), was already running as early as the 4th millennium BC (Jovanovic 1978; Shepherd 1980 reporting the opinion of Childe 1957). This statement is likely true also considering that, before 1968, it was the only mercury mine in the whole territory of Serbia (Gajić-Kvaščev et al. 2012). As for Spain, a map of the primary cinnabar deposits probably exploited since ancient times is provided by Domínguez-Bella (2010). Other isolated cases seem deduced more from cinnabar discovery in the neighbouring archaeological sites than from investigations aimed at verifying ancient exploitation. In other cases, the information is linked to modern exploitation. Therefore, one remains in doubt about when the exploitation began (*e.g.*, Monte Amiata in Tuscany).

In China and Japan, the Wanshan mine (Guizhou, China), the XunYang mine (Shaanxi, China), the Niu mine (Mie, Japan), Yamato-Suigin mine (Nara, Japan) and Sui mine (Tokushima, Japan) were identified as ancient mines (Kawano et al. 2014).

In Meso- and South America, the Peruvian-Ecuadorian case is perhaps the one that has received the greatest attention because a real controversy has arisen over the exploitation of cinnabar from Huancavelica (Peru) and that of Azogues (Cañar, Ecuador). As far as it is possible to reconstruct, the *casus belli* is represented by Truhan et al. (2005) paper. During an excursion to Loma Guaschon, they found cinnabar deposits already reported in previous documentation. After providing the reader with various information about the mining exploitation of some Ecuadorian, Colombian and Peruvian deposits, they hypothesised that "*la productividad del cinabrio azogueño durante la temprana colonia fue de tal magnitud que la misma Fuente puede haber tenido importancia en las redes de intercambio precolombinas*"[10].

Burger et al. (2016) responded by claiming the primacy of Huancavelica in prehispanic times. These authors argued about the correctness of the interpretation of the documents presented by Truhan et al. (2005) and strongly opposed the "Ecuadorian cinnabar hypothesis". For clarity, Burger et al. (2016) did not object that there was a trade network between Ecuador and Peru (witnessed by other types of goods such as *spondylus*, *strombus* and *conus*) as to the fact that cinnabar was part of these trades. Burger and coworkers seriously questioned the very existence of mercury mines in Azogues. They pointed out that cinnabar had not been unequivocally identified "*in any prehispanic archaeological context or on any prehispanic object in an Ecuadorian museum or private collection*". To further support their theory, they recalled the isotope (Hg) investigations obtained in 2003 (Cooke et al. 2013). The Hg-isotopic analyses were carried out on geological ores from Peru (Chonta and Huancavelica), Honduras (Jalaca), Colombia (Antioquia and Quindio), Bolivia (Cerro Colorado and Mina de Pedernal) and Chile (Algarrobo Mine) and on archaeological artefacts from Peruvian archaeological sites and/or referring to the Early Horizon and Late Intermediate period of Peruvian culture. The results assigned almost all archaeological finds to Huancavelica except for samples A15-A17, corresponding to three wooden digging boards from private collections (preserved at Metropolitan Museum of Art, New York and the National Museum of the American Indian, Washington, DC, USA). As for the latter, the authors stated that further analyses of raw materials were necessary to establish their provenance.

The answer was not long in coming. Bruhns et al. (2017) refuted the objections raised by Burger and co-authors on the documentation concerning the Azogues mines and replied with an indication that seemed fundamental to me: "". The quarrel continued but on closer inspection, this seems a case in which only archaeometry can write the ending. Indeed, while there is no archaeological evidence to support the "Ecuadorian cinnabar hypothesis", there is not even to discard it definitively.

Other studies are available for the exploitation of sources in Mexico and Honduras sources.

In Mexico, Manzanilla (2005) claimed that the Teotihuacans likely exploited the Sierra Gorda of Queretaro sources and, possibly, also those of San Luis Potosi in the Classical period (1^{st}–9^{th} centuries). Moreover, the ancient and modern exploitation of Queretaro mines has been the object of the study performed by Campos and Muñoz (2013) to map the archaeological sites and evaluate the Hg contamination issues from mining.

In Honduras, Gorokhovich et al. (2020) investigated the provenance of cinnabar found in the pre-Columbian site of Copan. They carried out a field survey that led them to detect the presence of mercury in the valley. However, they believed that the field was of such limited size/extension to make it more likely to import this material rather than exploiting the local sources (*e.g.*, those present in the Quebrada Sesesmil watershed close to the settlement). On the other hand, they believe that future research in El Tablón mountain may instead be considered promising about the possible identification of cinnabar and As-based pigments mines.

Cinnabar production technology, application and alteration

Production technology

Theophrastus informs us about the old way of producing cinnabar (*De Lapidibus*, 58–60) and indicates two types of cinnabar, one natural and one artificial.

In the first category, he included the cinnabar from Iberia (Ἰβηρίαν), very hard and stony, and that found in the Colchis (ἐν Κόλχοις), which was found on mountain cliffs and was brought down by shooting arrows.

In the second category, he included cinnabar coming only and exclusively (εξ ενός τόπου μόνον) from the region located just above Ephesus (ὑπὲρ Ἐφέσου). According to Theophrastus, a particular sand that shone like the scarlet dye (λαμπυρίζουσαν καθάπερ ὁ κόκκος; Kermes) was collected in this area. This sand was first pulverised in stone

[10] Translated: the productivity of the Azogues cinnabar during the early colony was of such magnitude that the same source might have been important in pre-Columbian exchange networks.

vessels until it reached the desired grain size and then washed by decanting in copper vessels. The worker then repeated the process until the achievement of the desired product. Theophrastus also added that workers' skills were essential in determining the amount of finished product (from nothing to much). The cinnabar remained at the bottom while the fraction remaining suspended in the washing water was used mainly for cosmetics.

The discovery of the procedure is attributed to Kallias, an Athenian involved in the activities of the silver mines (probably those of the Laurion) who, associating the shiny appearance of that sand with the presence of gold, would have collected and processed it, about 90 years before Praxiboulos was archon at Athens.

According to Theophrastus, this was the sign of how art imitates nature while producing its own substances. After a short digression, he describes the procedure for obtaining quicksilver (χυτὸν ἄργυρον): grinding cinnabar with a copper pestle together with vinegar in a copper mortar.

Since it is beyond the subject of this review, I will omit to discuss the production technology regarding quicksilver and the evident differences between the procedure handed down by Theophrastus and that reported by Dioscorides (*De materia medica*, 5, 94; through heating cinnabar). However, it is necessary to specify two critical aspects of the text of Theophrastus.

Firstly, the territory indicated with the term *Iberia* by Theophrastus and later disclosed as *Hispania* by *Plinius* (*Naturalis Historia* 3, 4, 30; 33, 38, 113–114) [11], perhaps does not correspond to Spain as to the "*eastern part of the present Transcaucasian Georgia*" as pointed out by Caley and Richards (1956) and Rosół (2018). On the other hand, the possible error of Pliny is well understood in the 1st-century Roman world, that is when "*the most famous cinnabar mine for the revenues of the Roman nation being that in the region of Sisapo in Baetica, no item being more carefully safeguarded: it is not allowed to smelt and refine the ore upon the spot, but as much as about 2000 pounds per annum is delivered to Rome in the crude state under seal, and is purified at Rome, the price in selling it being fixed by law established at 70 sesterces a pound, to prevent its going beyond limit. But it is adulterated in many ways, which is a source of plunder for the company*" (Plinius, *Naturalis Historia* 33, 40, 118–119, translated by Rackham 1952).

Secondly, according to Theophilus (*De diversis artibus*, first half of the 12th century) the second "factitious" type was not cinnabar but, likely, minium.

Soon after, Theophilus does not fail to describe "*the method of making the best vermilion, 'Vermiculum optimum'*" handed down by Petrus, of St. Audemar (MS. 6741. Bib. du Roi, Paris. Art. 174. Le Begue):

> "*If you wish to make the best vermilion, take a glass bottle and cover it with a lute outside; and take one part of quicksilver, by weight, and two, by weight, of white or yellow coloured sulphur. Put it into the above bottle, which you afterwards place on four stones, and, laying a very slight fire of coals round the bottle, cover its mouth with a tile, and when you see the smoke come white from the mouth of the bottle, close it, but when a smoke as red as the vermilion shall come out, take it from the fire and you will have the best vermilion. Similar recipes are found amongst the medical writers of the thirteenth and fourteenth century, but are mostly repetitions*" (reported from Hendrie 1847).

Other recipes have been handed down in manuscripts such as the ancient *Compositiones ad tingenda* or "Lucca manuscript" (8th–9th centuries), the *Mappae Clavicula* (from the 9th century) and the Bologna manuscript 2861 (15th century).

> "*If you wish to make vermilion, take a glass flask and coat the outside with clay. Then take one part by weight of quicksilver and two of white or yellow sulphur and set the flask on three or four stones. Surround the flask with a charcoal fire, but a very slow one, and then cover the flask with a tiny tile. When you see that the smoke coming out of the mouth of the flask is straw-colored, cover it; and when yellow smoke comes out, cover it again; and when you see red smoke, like vermilion, coming out, then take away the fire, and you have excellent vermilion in the flask.*" (*Mappae Clavicula*, Chapter i. Vermilion; reported from Smith and Hawthorne 1974, p. 26)

> "*The recipe for cinnabar. Take 2 parts of clean quicksilver and 1 part of native sulphur, and put them in a flask, and, cooking them without smoke and over a slow fire, make cinnabar. Wash it properly*" (*Mappae Clavicula* corresponding to Lucca manuscript 223v.10; reported from Smith and Hawthorne 1974, no. 105, p. 42)

> "*A recipe for cinnabar. A recipe for true, clean cinnabar. Take 2 parts of quicksilver and 1 part of native sulphur, and 1 part of clean urine. Take a very clean strong flask that will endure heat without smoke. Put into the flask the sulphur, ground and mixed with the quicksilver, 2 ounces short of filling it; but if it is a larger flask, it should be short 3 ounces. Mix and shake. Get*

[11] This indication was omitted by *Vitruvius* (*De Architectura* 7, 8).

ready a smaller glassworker's furnace, which should amply hold the flask, leaving a place where the flask may enter. Split reeds and with them light the furnace. Leave another window so that the flames may breathe out all round the flask. The sign of [completion of] the cooking is this: when you see that the flask has less purplish smoke and is making a color like cinnabar, stop adding fuel, for the flask gives a crashing sound from the great heat. When the cinnabar is thoroughly cooked leave it to cool." (*Mappae Clavicula* corresponding to Lucca manuscript 229.24; reported from Smith and Hawthorne 1974, no. 221C, p. 61)

"*M263. To make cinnabar. Take quicksilver and two parts of white or yellow sulfur. Incorporate the finely ground sulfur with quicksilver, put it in a bottle well luted with lutum sapientiae and let dry. Then put it on the fire over a low heat and cover the mouth of the vase with a tile. Cover and uncover it frequently. When you see the yellow smoke coming out, you will know it is nearly done; let it remain and keep the fire until the smoke becomes almost peacock red. At that point you take it off the heat and let it cool and so you will have fine cinnabar*"[12] (Bologna Manuscript, very similar to *Mappae Clavicula*, Chapter i. Translated).

Ultimately, the ancient procedures to obtain cinnabar were all relatively simple. They started from raw materials that were already well recognisable and relatively pure. They then mixed a few ingredients and let the heat do the rest, just taking care to observe the colour of the fumes. Therefore, the process did not require particularly high temperatures or complex tools. Also, the *lutum sapientiae* was a compound typically used for distillation and its production required raw materials that were simple to find, such as strips of linen or wool, flour, egg white, ash, dung and clay (Biringuccio, *De la pirotechnia*, 1540).

The production of synthetic cinnabar, on the other hand, seems to derive from a subsequent technological development, probably transferred from Arabic (Gettens et al. 1972; Miguel et al. 2014) or Chinese (Gettens et al. 1972; Franquelo and Pérez-Rodríguez 2016) artisans.

[12] Non-literal translation. Original text: "M263. *Affare cinabrio. Abbi arge(n)to vivo et doi pa(r)te de solpho bia(n) / co o giallo et i(n)corporalo lo solpho b(e)n(e) tri / to cu(m) large(n)to et pollo i(n) una boccia alu / tata b(e)n(e) de luto de sap(ient)ia et lassa sciu / tar(e) poi la pon(e) nel fornello et fallj / foco ligiero et cop(ri) la boca del vaso cum / una tegola et spesso lo scop(ri) et ricop(re) et / q(ua)n(do) tu vedj vuscire el fumo giallo / s(e)ra ap(re)sso ch(e) facto et lassalo tanto sta / re et dallj lo foco ch(e) facia lo fumj ro / sso q(uas)i pavonazo ahlora toli via lo / foco et lassa fredar(e) e de fact(o) fino / cinabrio*".

The feasible processes are essentially two: the dry one and the wet one. Gettens et al. (1972) reported numerous manuscript sources which, starting from the 8th century, describe the dry process and its subsequent modifications. It consists of producing black β-HgS and heating it until it turns red α-HgS.

Melo and Miguel (2010) and Miguel et al. (2014) reproduced one of the dry process recipes contained in "The book on how to make colours" and verified that the complete grinding of S with Hg produces black β-HgS. Based on the process described by Franquelo and Pérez-Rodríguez (2016), it would simply consist of stirring Hg with S and successively heating at 235°C. These last authors also discuss the possible addition of small amounts of Sn.

As for the wet process, Gettens et al. (1972) seems to lead it back to Gottfried Schulz based on Kopp 1843-1847). In 1867, Schulz would have discovered that metacinnabar heated in a solution of ammonium or potassium sulphide transforms into vermillion. From here, it would have resulted in a production destined to become the favourite in Germany and England.

For further information on production technology, the reader is referred to the aforementioned publications. Still, it may be helpful to add that these authors have also provided optical and microscopic images and descriptions to favour the discrimination of the three types of cinnabar (*i.e.* natural, obtained with the dry process and obtained with the wet process) without hiding the objective difficulties. The distinction between natural and artificial cinnabar is not simple. The presence of other phases to which cinnabar may be associated in the supply source (see the associations described in the first section) undoubtedly represents the first clue but may prove insufficient.

On this topic, Franquelo and Pérez-Rodríguez (2016) discussed how the presence of impurities, the size and morphology of grains could guide the determination of natural cinnabar (generally heterogeneous in size and shape irregular) from a synthetic one (presence of K, S and Sn). However, they also note objective difficulties in distinguishing natural cinnabar from that produced through the dry process.

Cinnabar application

This topic requires two critical aspects to be addressed separately: (1) cinnabar and the *fresco* technique and (2) cinnabar and organic materials.

The first point is well known: cinnabar is unsuitable for painting *a fresco*. This incompatibility has been known since ancient times but has not limited its use to decorate wall structures. Cinnabar is poorly soluble and therefore when it comes into contact with fresh plaster (*i.e.* not yet dried = opposite of *secco*), it reacts and transforms into black metacinnabar. To

overcome this problem, it was common to apply a red ochre base *a fresco* and finish with cinnabar once the fresco had dried. According to fresco and lime-painting techniques, an example of the yield of cinnabar was experimentally made by Piovesan et al. (2012).

As for the second issue, the cinnabar interaction with ovalbumin and casein has been experimentally tested by Duce et al. (2012) in both unaged and aged tests. The authors observed that while cinnabar forms stable complexes with ovalbumin and promotes oxidation, it modifies the elution pattern of casein and promotes hydrolysis. The interactions with protein-based binders and fatty acid esters from egg yolk have also been investigated by Romero-Pastor et al. (2011b). They observed how the interaction of cinnabar with the protein causes a shift in the spectral region where the polyunsaturated fatty acid esters of the egg yolk appear. Further information is also provided in the study of the degradation processes involving several types of binders due to ultraviolet (UV) radiation and ageing by Ropret et al. (2007) and Romero-Pastor et al. (2012). The interaction with eastern and western drying oils was the object of the experimental study performed by Wang et al. (2015). These authors demonstrated that, compared to azurite, malachite and ochre, cinnabar is more effective in accelerating the ageing of drying oils and their hydrolysing process.

Cinnabar alteration

The colour variation of HgS has intrigued scholars from various fields so much that it has inspired fine philosophical discussions on the reproducibility of representations and empirical imagination in the *Critique of Pure Reason* by Kant (Westphal 1997).

As described above, the cinnabar (α-HgS) → metacinnabar (β-HgS) conversion implies that the colour changes from red to black and occurs above temperatures that may vary from 315 to 400 °C depending on the surrounding conditions and the degree of purity of the mineral.

However, it is common knowledge that high temperatures are not strictly necessary to observe cinnabar's blackening on artworks. Therefore, numerous researchers studied the phenomenon more thoroughly to trace all the possible causes and discovered that metacinnabar formation is only one (rare) cause.

Firstly, McCormack (2000) highlighted that chlorine contents are decisive for cinnabar blackening. His research focused on natural deposits containing photosensitive cinnabar (generally associated with calomel, corderoite, terlinguaite and kleinite, eglestonite, comancheite, mosesite, radtkeite and kenshuite). The chemical analysis of some photosensitive cinnabar samples showed chlorine contents ranging from 0.04 to 0.96 wt%. This evidence led the author to conclude that the "*darkening of cinnabar in sunlight is caused by the presence of contained chlorine or other halogens*". Furthermore, McCormack experimentally observed that non-photosensitive cinnabar blackens to light when exposed to halogens.

Spring and Grout (2002) used McCormack's conclusions to explain the visible blackening of some artworks. They realised that the sodium chloride in the dirt particles on paints could trigger the same reaction that, in nature, induces the vermilion → corderoite transformation. This, in turn, would have led to the formation of calomel + S + black HgS upon light exposure. However, Spring and Grout further highlighted how even the painting technique is a crucial factor in the pigment's discolouration. They observed that the alteration was more prominent where cinnabar was used alone while absent or lower when applied together with red lake or red lead.

A few years later, the study by Cotte et al. (2006) returned to this topic, focusing on Pompeian frescoes. Their investigations confirmed the heterogeneous nature of the degradation products and the possible coexistence of corderoite (ascertained by spectra XANES), terlinguaite, corderoite and calomel. The authors indicated Punic wax — often used to protect frescoes and produced using seawater — as a possible chlorine source. However, they also add another possible path involving calcite sulphation, resembling the process at the origin of black crust formation.

Regarding Pompeian frescoes, the famous eruption of the Vesuvius and the corresponding temperature increase deserve further mention.

Ballirano et al. (2013) observed that the conversion temperature of the Almadén samples they investigated (> 673K = 399.85° C) was higher than that fixed as the formation temperature of the pyroclastic deposits of Pompeii (653K = 379.85° C) and thus claimed that the blackening of Pompeian cinnabar also had to be due to impurities contained in the pigment.

What unites the work of Cotte et al. (2006) and that of Ballirano et al. (2013) is that while thinking of two different processes, both recognise the blackening of cinnabar as a process chiefly due to impurities, either naturally contained within it or brought from the outside.

On the other hand, the applied and experimental research carried out by Radepont et al. (2011, 2015) confirmed that both light and chlorine presence are the key factors in the discolouration of pictorial surfaces. Their main results may be summarised as follows:

(a) evaluation of the colour changes over time;
(b) identification of degradation products such as calomel, corderoite, kenhsuite, terlinguaite and sulphates;
(c) iInvestigation of the reaction sequence leading to calomel and corderoite formation (in the various layers).

In 2013, Nöller proposed a review on cinnabar where the darkening by radiation, the influence of substrates and the reactions with halogens, other pigments and binding media are summarised. The author reports numerous discolouration examples in artworks and highlights how natural cinnabar alters differently from the artificial vermilion, depending on structural impurities (Nöller 2013). While confirming the importance of the role played by light and chlorine, the experimental work carried out by Kegelman Neiman et al. (2015) also demonstrated that high relative humidity must be added to the factors responsible for colour variations. Furthermore, the formation of calomel in tests exposed to saline solution, with or without light exposure, combined with the absence of corderoite led to a revision of the previous assumptions on reaction mechanisms and, especially, on the catalyst role of chlorine ions (see Keune and Boon 2005). Lastly, Kegelman Neiman and coworkers observed that the presence of white calomel and the absence of black corderoite made it challenging to explain the colour variations observed during alteration and cautiously put forward some hypotheses, including that of the presence of metallic mercury.

Other works have been conducted on this topic but they left the big picture unchanged; if anything, more details were presented on specific issues such as pigment-binder interactions upon weathering (*e.g.*, Elert and Cardell 2019). Lastly, the work done by Hogan and Da Pieve (2015) on the mechanisms involved in the darkening of cinnabar is worth mentioning because it clarifies the role of chlorine. For the weathering of a sample exposed to light and humidity, the authors proposed a process in 4 steps:

(1) uptake of Cl on α-HgS surface;
(2) consequent formation of γ-$Hg_3S_2Cl_2$ first and of the more stable cubic phase α-$Hg_3S_2Cl_2$ after;
(3) structural instabilities cause the release of elemental Hg (0);
(4) chloride phases form (over the degradation products previously formed) and undergo further degradation processes.

In a nutshell, (a) chlorine, light and humidity or, possibly, a considerable temperature rise induce the blackening of cinnabar, (b) none of the researches presented here found metacinnabar.

The bad news is that blackening appears to be a relatively simple process to happen and, for now being irreversible, we do not currently have the means to counter it.

Calomel

The presence of calomel in an artwork is generally referred to as a degradation process. Still, on this point, we need to clarify.

The Art of limning by Nicholas Hilliard (ca. 1537/1547-1619) is often quoted in this regard (Burgio et al. 2018; Crippa et al. 2020) because the author mentions a white mercury pigment recommended for limning: "*ther is also an excelent whit to be made of quicksilver/ which draweth a very fine line / this whit the women painters vsse*"[13].

These few lines testify to the use of a mercury-based white pigment and are enough to open a little discussion. However, before going into the discussion, it must be assumed that there is no confusion with cerussite/hydrocerussite because Hilliard talks about lead white production a few lines above. The fact that Hilliard delimits the use of this pigment to "women painters" is perplexing. Thornton and Cain (*i.e.* the editors of his treatise) suggest that Hilliard was referring to the painter Levina Teerlinc, "*appointed paintrix to Henry VIII around 1546*", but one may also think that he refers to the cosmetic use of the contemporary sublimates.

This last reading is perhaps the least likely. Still, the doubt comes when it is found that Edward Norgate does not mention white quicksilver in the course of dealing with whites and that his 17th-century manuscript, *Miniatura or the Art of Limning*, is believed to be largely derived from Hilliard work. On the order hand, Norgate mentions "mercury Sublimata" while discussing a method to produce gold in a fine, fair and cheap way.

The nature of the "sublimate" is clearly described in the third book of "*A tracte containing the artes of curious paintinge, carvinge and buildinge*, written first in Italian by Paul Lomatius painter of Milan and englished by Richard Haydocke" in 1598:

> "*Of sublimate and the bad effects thereof.*
>
> *Diverse women use Sublimate diversly prepared for increase of their beauty. Some bray it with quicksilver in a marble morter, with a wodden pestle; and this they call argentatum. Others boile it in water, & therwith wash their face. Some grinde it with Pomatum, and fundry other waies. But this is sure, that which way soever it be used, it is very offensive to mans flesh, and that not only to the face; but unto all the other parts of the body besides, where it is applied. For proofe whereof Sublimate is called deadfier; because of his malignant, and biting nature. The composition whereof is of salte, quicksilver, and vitrioll, distilled together in a glassen vessell.*
>
> *This the Chirurgions call a corrosive. Because if it bee put upon mans flesh it burneth it in a short space, mortifying the place, not without great paine to the patient.*

[13] There is also an excellent white to be made of quicksilver which draweth a very fine line; this white the women painters use.

Wherfore such women as use it about their face, haue alwaies black teeth, standing far out of their gums like a Spanish mule; an offensive breath, with a face halfe scorched, and an uncleane complexion. All which proceede from the nature of Sublimate. So that simple women thinking to grow more beautifull, become disfigured, haftening olde age before the time, and giving occasion to their husbandes to seeke strangers insteede of their wives; with divers other inconveniences."

Further information on the cosmetic use of these sublimates is provided by Karim-Cooper (2006), where another recipe handed down by Hugh Plat[14] in 1600 is reported, and in the chapter "Making Calomel" of the Swiderski (2008) book.

In any case, the production of these sublimates and calomel was widespread since earlier times. For example, cosmetic use is documented in China (Needham et al. 1976) and Japan (Takamatsu 1878). Divers (1894) provides a very detailed review of Chinese and Japanese terminology and technology, further supplemented by sketches of the tools and furnaces. One of the recipes mentioned by Divers is reported more succinctly, albeit detailed, by Takamatsu (1878):

"*According to old history this substance was very early known in the year 714 (the 6th year of Wado) when it was presented to the empress Gemmiyo from the province of Ise. Up to the present time it has been manufactured only in that province, hence it is also called Ise Oshiroi, which possessing important medicinal properties is mostly used for that purpose rather than as a pigment. (...) The mode of preparation is the following: —A mixture of 2 parts of alum, 1 part of mercury, and 1 part common salt is well pulverized in a mortar until no globules of mercury can be seen. The mass is then placed in a iron pot covered with an earthen ware cover, which is carefully luted up by means of a mixture made from wood ashes and salt water. 36 of such pots are placed in a rectangular furnace, and gradually heated by a charcoal fire for some time, meanwhile the covers are moistened with water. In this way about 2 parts of the calomel can be obtained from 1 part of mercury, subliming on the inner surface of the cover as a white crystalline powder like snow.*"

Recipes vary in relative quantities and procedures on both a geographical and temporal scale, but it is interesting to note the morphological similarity between the *Hozukigama*, the pot used for sublimation (Fig. 11A), and those used for the production of lead white (see Gliozzo and Ionescu 2021 in this TC).

Based on archaeometric evidence available to date, the deliberate use of calomel as a white pigment has been claimed for a few artworks:

- four *mopa mopa* objects — A 17th century richly decorated *barniz de Pasto* table cabinet "*probably made during the seventeenth century in the northern zone of the Vice-royalty of Peru*", two *barniz de Pasto* gourd flasks and a *barniz brilliante* casket, all held by the Victoria and Albert Museum in London, UK (Burgio et al. 2018; Melchar et al. 2021). Please note that the authors refer to calomel as "mercury white", whenever it is clear that this material has been used intentionally as a pigment in its own right;
- a viceregal Spanish American *barniz de Pasto* gourd (*mopa mopa*) produced in the 17th century (pre-1650) at Pasto in Colombia and acquired in 2014 by The Hispanic Society Museum and Library at New York in the USA (Pozzi et al. 2020);
- The "Fitzwilliam Missal" (MS 34), that is a 15th century English illuminated manuscript "*probably made in York around 1470 for Sir Richard Fitzwilliam and his wife, Elizabeth Clarell*" and preserved in the Fitzwilliam Museum in Cambridge, UK (Crippa et al. 2020);
- a late 16th-century English portrait miniature "*of a fashionably attired unknown lady*" by Isaac Oliver's in the Fitzwilliam Museum collections (FM 3868; Crippa et al. 2020).

Conversely, calomel as an alteration product has been identified in:

- the vaults of the Hall of the Kings (13th–15th centuries) within the Alhambra complex at Granada in Spain (Dominguez-Vidal et al. 2012);
- the *Virgin and child enthroned* (1461–1462) by Benozzo Gozzoli's (National Gallery, London, UK; Spring and Grout 2002);
- the oil on panel *The Crucifixion of St Julia* (Saint Wilgefortis Triptych, ~1497) by Hieronymus Bosch (1450–1516), kept in the Gallerie dell'Accademia di Venezia (Sodo et al. 2019);
- the oil on canvas *Adoration of the Magi* (1624) by Peter Paul Rubens (Royal Museum of Fine Arts, Antwerp, Belgium; Radepont et al. 2011);
- the decorative panels of the Japanese tower (~1905) in Laeken, Belgium (Vermeulen et al. 2015);
- probably, the *Portrait of a Lady* (~ 1625) by Peter Paul Rubens, kept at the Mauritshuis (The Hague, The Netherlands) and the *Minerva and Hercules*

[14] From Platt (1600) Delights for Ladies. Sig. G12v. "*incorporate with a wooden pestle, and in a wooden mortar with great labour, four ounces of sublimate, and one ounce of crude Mercurie, at the least 6 or 8 houres*".

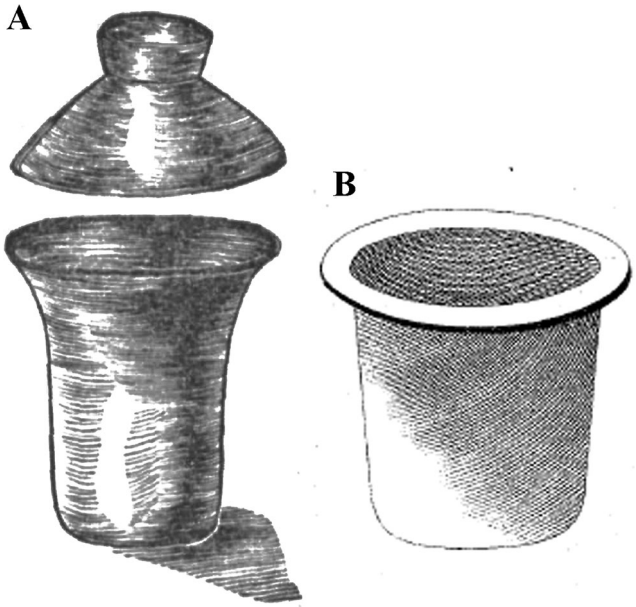

Fig. 11 **A** The *Hozukigama* (pot) and the *Hozuki* (lid) described by Takamatsu (1878). **B** The iron-furnace pot sketched by Divers (1894). For the former, the reported dimensions are 5 *sun* in diameter and in height, corresponding to about 15 cm. The dimensions of the pot B are not indicated.

Opening the Doors for Victory (1651) by Christiaan van Couwenberg, kept at the Oranjezaal Huis ten Bosch Palace (The Hague, The Netherlands; Keune and Boon 2005);

– probably, a Pompeian wall painting (before 79 AD) excavated in 1988 (Cotte et al. 2006).

In conclusion, as far as we know to date, calomel may have been used as a pigment or may have formed as an alteration product. Hence, its occurrence in artworks must be discussed on a case-by-case basis.

Analytical methods for a good practice

Before going into detail, it should be noted that the choice of the methodology to adopt for investigation supports and follows the formulation of archaeological questions and the selection of samples and not vice versa. In many articles, we often read that a technique was chosen because not time-consuming and not expensive but these factors have no value in quality research. The survey methodology must be suitable to answer the research questions and respectful of the artefact's conservation. Other criteria are not to be considered a priority.

Identification and characterisation of the pigment

Identifying cinnabar would be relatively simple if we were *a priori* sure of the absence of other Hg-based phases. However, we have seen that:

(1) other phases such as calomel may be present. Therefore, it is necessary to employ a technique capable of distinguishing the various phases with due accuracy. A simple assumption that determines cinnabar's presence by combining a red pigment's observation with mercury individuation no longer appears sufficient.
(2) the presence of impurities and the morphology of the grains may indicate the nature of cinnabar, *i.e.* natural or synthetic. This determination requires both microscopic observation and a technique capable of providing the chemical composition of compounds present in small amounts;
(3) the surface of the frescoes tends to form thick calcite crusts that can prevent a poorly penetrating beam from reaching the pigment. Therefore, invasive sampling is required unless the find is already detached and lends itself to being transported.

In more than 250 research studies in which cinnabar was reported, the most used techniques were Raman spectroscopy (24%), X-ray fluorescence (XRF, 16%), scanning electron microscopy (SEM, 17%), Fourier transform infrared spectroscopy (FT-IR, 16 %) and optical microscopy (OM, 14%) followed by other less common techniques such as fibre optic reflectance spectroscopy (FORS), electron microprobe analysis (EMPA), time-of-flight secondary ion mass spectrometry (ToF-SIMS), inductively coupled plasma mass spectrometry combined with laser ablation (LA-ICP-MS), particle-induced X-ray emission (PIXE), electron paramagnetic resonance (EPR), laser-induced breakdown spectroscopy (LIBS), Mössbauer spectroscopy and the whole range of imaging techniques.

While using a single technique is inadvisable based on the limited result achievable, even the exclusive use of portable techniques may lead to a partially accurate search.

The optimum is therefore achieved with a combination of techniques capable to:

– observe the pigment (OM, SEM);
– determine the chemical composition even of elements present in small minimal quantities (SEM, EMPA, LA-ICP-MS or portable XRF when there is really no possibility of transferring the sample/product to the laboratory);
– provide an unquestionable phase/pigment assessment (XRD, Raman, FTIR).

Undoubtedly, the problems related to the conservation of the artefact are a priority. The help comes from the vast range of portable instruments existing to date and, above all, from large facilities such as synchrotrons. These latter offer different techniques with the advantage of optimised analytical conditions (such as penetration depth).

The provenance of the pigment

Several authors have repeatedly attempted to determine the provenance of cinnabar through Raman spectroscopy. For example, Villar and Edwards et al. (2005) related the coexistence of cinnabar with calcium carbonates or quartz with a provenance from Tarna or Almaden, respectively. The basic assumption was that the Spanish mines exploited in Roman times were essentially two: Tarna (León) to the north, associated with sedimentary carbonates and Almaden (Ciudad Real) to the south, associated with quartz.

Recently, Botticelli et al. (2020) have made a further attempt in this direction. They obtained structural data for 31 cinnabar samples from China (Tsa Tien Mine or modern Chatian Mine, Hunan; War Shan-Chang Nmer, Guizhou, Kweichow; unspecified locality in the Hunan territory), Czech Republic (Hořovice in Bohemia), Germany (Moschellandsberg), Italy (Serravezza, Lucca; Val di Castello, Lucca; Grosseto; Miniera San Filippo, Mount Amiata; Abbadia San Salvatore, Mount Amiata; unspecified locality at Mount Amiata; Loibel Valley, Carnia, Udine), Serbia (Mount Avala), Slovakia (Rosenau), Slovenia (Idrija), Spain (Almadén) and Ukraine (Saizewka, Nikitovka, Charkov train station, Asow; Nikitovka Donetz). The results achieved were processed through statistical analysis. Some distinctions were observed between Almadén and Idrija and especially Chinese samples, further characterised by selenium's presence.

Very few authors have tried the same type of approach using XRD. For example, Maras et al. (2013) investigated several mineralogical samples from Austria (Carnia, 1 sample), China (Tsar Lien Mine, Honan, 1), Germany (Moschellandsberg, 1), Italy (Cerreto Piano, Grosseto, 2; Val di Castello, Lucca, 1; Sele Mine, Castell'Azzara, Monte Amiata, 1), Romania (Rosenau, 1), Russia (Nikitowska, Charkov railway station, 1), Serbia (Avala, 1), Slovenia (Idrija, 1) and Spain (Almadén, 3). Since the results provided measurable differences in unit cell parameters and volume (due to Hg: S variable ratios) between the various samples, the authors suggested that the method could also work in provenance studies.

These two methods could provide effective developments in the future, but it seems that both suffer from two main problems: (a) representativeness, due to the investigation of a few areas only and (b) nature of the pigment (natural or artificial). Indeed, one may wonder (a) what these methods' discriminating power could be when the items used for comparison become numerically consistent and (b) are there possible overlaps in spectra and/or structural parameters between natural cinnabar from a given location and artificial cinnabar?

Another issue that does not seem to have been addressed so far (except in part for Almaden) concerns the intra-site variability. This could represent another critical factor in the comparison.

At present, therefore, the most promising techniques seem to be isotopic ones, although much work is still needed before having sufficiently comprehensive and representative data for comparison. In this case, the archaeometric literature may also use geological studies and take advantage of a pre-existing and developing database.

As for sulphur isotopes, data on natural cinnabar occurrences are limited but particularly focused on ancient mining districts (Table 7). This method has been used for provenance investigation by Damiani et al. (2003), Minami et al. (2005), Domínguez-Bella (2010), Spangenberg et al. (2010), Kawano et al. (2014), Tsantini et al. (2018) and Minami et al. (2019). Further data on natural ores are provided by Lavrič and Spangenberg (2003) for the Idrija mine in Slovenia and Jébrak et al. (2002) for the Nuevo Entredicho deposit in Spain. The authors report different methods for sample preparation. Depending on the materials analysed, some considered an initial grinding, followed by analytical techniques such as optical microscopy and XRD, to assess the possible presence of contaminants. Others adopted more laborious procedures including the following steps: 5h heating at 95° C, dilution with a HNO_3/HCl solution (3:1), washing with ultrapure water, addition of reverse aqua regia to the insoluble part followed by 5h heating at 95° C; cooling, bromine addition, 2h heating at 95° C, washing with ultrapure water, addition of 1-ml $BaCl_2$, overnight heating at 65°C, repeated washing with ultrapure water of the resulting barium sulphate, drying and addition of 1 mg of vanadium pentoxide (see Tsantini et al. 2018 for further details). The analyses are typically carried out using isotope ratio mass spectrometers (IRMS), using pure SO_2 gas as a reference. Data are conventionally reported as $\delta^{34}S$ ‰ (*i.e.* $^{34}S/^{32}S$) and standardized to the Vienna Cañón Diablo Troilite. The standards mostly used are the IAEA-S-1 and IAEA-S-2 silver sulphides (−0.3% and + 22.7 ± 0.2%, respectively) and, when reported, reproducibility values are better than 3%.

Lead isotopes (Table 8) are consistently used for metal provenancing, first for ancient lead (Brill and Wampler 1967; Grögler et al. 1966) and later also for copper-based alloys (Gale and Stos-Gale 1982; Pernicka et al. 1990, 1993; Niederschlag et al. 2003). Their use for cinnabar investigation requires suitable sample preparation to eliminate mercury and purify lead. As a matter of fact, ^{204}Hg interferes with ^{204}Pb; moreover, the lead isotope ratios of the leachates and residues are different mainly due to lead contamination from associated phases such as pyrite (Higueras et al. 2005). For this reason, sample preparation is as crucial as the analytical technique used for the measurements. Archaeometric issues have been approached through this method by Mazzocchin et al. (2008), Hunt et al. (2011), Minami et al. (2013, 2021)

Table 7 Sulphur isotope composition of cinnabar from several mining districts ($\delta^{34}S$ values provided as ‰). [In the first column: Pf stands for profile. Among data from Lavrič and Spangenberg 2003, the age of host rock is indicated. For several Chinese and Japanese ores, the average value is provided, followed by the standard deviation (after ±)]

Locality	$\delta^{34}S$	Reference	Locality	$\delta^{34}S$	Reference
Almaden District (Spain)			**Idrija (Slovenia)**		
M. Las Cuevas (n=9)	+13.6(1)[(1)]	Rytuba et al. 1988	MGL51355	+8.2	Spangenberg et al. 2010
El Entredicho (n=3)	+8.4(0.8) [(2)]	Rytuba et al. 1988	MGL51357	+5.9	Spangenberg et al. 2010
Almadén (n=8)	+4.3(2.8) [(3)]	Rytuba et al. 1988	MGL51357	+5.1	Spangenberg et al. 2010
San Pedro (Pf 1) AL1740/4.1	+7.3 (0.07)	Saupé and Arnold 1992	MGL51361	+8.5	Spangenberg et al. 2010
San Pedro (Pf 1) AL17-40/4.III	+6.5	Saupé and Arnold 1992	MGL51361	+9.1	Spangenberg et al. 2010
San Pedro (Pf 1) AL17-40/6.I	+5.2 (0.03)	Saupé and Arnold 1992	MGL51366	+3.1	Spangenberg et al. 2010
San Pedro (Pf 1) AL17-40/7.1	+6.4 (0.04)	Saupé and Arnold 1992	MGL30392	+6.0	Spangenberg et al. 2010
San Pedro (Pf 4) IV-1	+7.2 (0.05)	Saupé and Arnold 1992	MGL30392	+7.7	Spangenberg et al. 2010
San Pedro (Pf 4) IV-2	+6.9 (0.05)	Saupé and Arnold 1992	MGL51478	+1.2	Spangenberg et al. 2010
San Pedro (Pf 4) IV-3	+6.8 (0.06)	Saupé and Arnold 1992	MGL51478	+1.5	Spangenberg et al. 2010
San Pedro (Pf 4) IV-5	+5.9 (0.05)	Saupé and Arnold 1992	MGL51478	+6.1	Spangenberg et al. 2010
San Pedro (Pf 4) IV-6	+4.9 (0.06)	Saupé and Arnold 1992	MGL51478	+7.0	Spangenberg et al. 2010
San Pedro (Pf 4) IV-8	+4.5 (0.04)	Saupé and Arnold 1992	MGL51639	+0.0	Spangenberg et al. 2010
San Pedro (Pf 6) VI-1	+5.8 (0.06)	Saupé and Arnold 1992	MGL51639	-0.9	Spangenberg et al. 2010
San Pedro (Pf 6) VI-2	+4.5 (0.05)	Saupé and Arnold 1992	MGL52647	+8.9	Spangenberg et al. 2010
San Pedro (Pf 6) VI-3	+5.3 (0.05)	Saupé and Arnold 1992	MGL34995	+1.3	Spangenberg et al. 2010
San Nicolas II-11	+0.1 (0.08)	Saupé and Arnold 1992	MGL34995	+0.5	Spangenberg et al. 2010
San Nicolas II-7	-0.3 (0.08)	Saupé and Arnold 1992	MGL34995	+4.1	Spangenberg et al. 2010
San Nicolas II-5	+1.2 (0.06)	Saupé and Arnold 1992	MGL34989	+0.0	Spangenberg et al. 2010
San Nicolas II-4	+0.8 (0.07)	Saupé and Arnold 1992	MGL34989	-0.5	Spangenberg et al. 2010
San Nicolas II-3	+1.2 (0.05)	Saupé and Arnold 1992	MGL34976	+7.1	Spangenberg et al. 2010
San Nicolas II-2	-1.6 (0.07)	Saupé and Arnold 1992	MGL34996	+7.6	Spangenberg et al. 2010
San Francisco III-1	+7.4 (0.05)	Saupé and Arnold 1992	MGL34981	+1.0	Spangenberg et al. 2010
San Francisco III-2/1	+7.3 (0.04)	Saupé and Arnold 1992	JSID26	+3.8	Spangenberg et al. 2010
San Francisco III-2/3	+7.3 (0.04)	Saupé and Arnold 1992	Upper Ladinian (n=40)	+6.7[(5)]	Lavrič and Spangenberg 2003
San Francisco III-3	+8.2 (0.04)	Saupé and Arnold 1992	Ladinian, Karoli oreb. (n=14)	+0.8[(6)]	Lavrič and Spangenberg 2003
San Francisco III-4	+8.1 (0.05)	Saupé and Arnold 1992	Anisian (n=10)	+4.9[(7)]	Lavrič and Spangenberg 2003
San Francisco III-5	+9 (0.03)	Saupé and Arnold 1992	Upper Scythian (n=3)	+5.6[(8)]	Lavrič and Spangenberg 2003
San Francisco III-7 top	+8.4 (0.04)	Saupé and Arnold 1992	Lower Scythian (n=39)	+0.6[(9)]	Lavrič and Spangenberg 2003
San Francisco III-7 bottom	+8.9 (0.02)	Saupé and Arnold 1992	Upper Permian (n=34)	-6.4[(10)]	Lavrič and Spangenberg 2003
San Francisco III-8	+8.1 (0.05)	Saupé and Arnold 1992	Middle Permian (n=3)	+2.6[(11)]	Lavrič and Spangenberg 2003
San Francisco III-9	+8.9 (0.03)	Saupé and Arnold 1992	Permocarboniferous	-4.9	Lavrič and Spangenberg 2003
San Francisco III-10	+7 (0.06)	Saupé and Arnold 1992	Range	From -6.4 to 9.1	
Las Cuevas – LC-PL3-36	+13	Higueras et al. 1999	Average (n=24)	3.5	
Las Cuevas – LC-PL3-37	+13	Higueras et al. 1999			
Las Cuevas – LC56	+12.2	Higueras et al. 1999	**Monte Amiata (Italy)**		
Las Cuevas – 2-pow	+12.8	Higueras et al. 1999	MGL-Bickel	-1.6	Spangenberg et al. 2010
Nuevo Entredicho 14	+10.3	Jébrak et al. 2002	MGL-Bickel	-1.7	Spangenberg et al. 2010
Nuevo Entredicho 18	+10.8	Jébrak et al. 2002	MGL-SGAM1	+2.3	Spangenberg et al. 2010
Nuevo Entredicho 19	+10.7	Jébrak et al. 2002	MGL-SGAM2	+2.4	Spangenberg et al. 2010
Nuevo Entredicho 22b	+10.6	Jébrak et al. 2002	MGL-NM-1	-5.0	Spangenberg et al. 2010
Nuevo Entredicho 30	+10.7	Jébrak et al. 2002	MGL-ICMA	-0.8	Spangenberg et al. 2010
MGL25234	+6.6	Spangenberg et al. 2010	MGL-IMP	+0.1	Spangenberg et al. 2010
MGL40128-1	+6.2	Spangenberg et al. 2010	MGL-NM-2	-7.6	Spangenberg et al. 2010
MGL40128-2	+5.4	Spangenberg et al. 2010	MGL-SGAM3	+0.9	Spangenberg et al. 2010
MGL25229	+6.3	Spangenberg et al. 2010	Range	From -7.6 to 2.4	
MGL14308-1	+8.8	Spangenberg et al. 2010	Average (n=24)	-1.2	
MGL14308-2	+8.0	Spangenberg et al. 2010			
MGL51356	+0.6	Spangenberg et al. 2010	**Genepy (France)**		
MGL51349	+4.6	Spangenberg et al. 2010	MGL58789	-2.2	Spangenberg et al. 2010
MGL34986-1	+4.4	Spangenberg et al. 2010	1 Aupt	-3.5	Spangenberg et al. 2010
MGL34986-2	+5.1	Spangenberg et al. 2010			
MGL34986-3	+4.6	Spangenberg et al. 2010	**Moschellandsberg (Germany)**		
MGL34986-4	+5.4	Spangenberg et al. 2010	MGL3499-1	-19.6	Spangenberg et al. 2010
MGL34986-5	+7.0	Spangenberg et al. 2010	MGL3499-2	-15.6	Spangenberg et al. 2010
M. de Almadén	+9.6	Domínguez-Bella 2010			
Castilla - La Mancha	+7.51	Tsantini et al. 2018	**Turkey**		
Castilla - La Mancha	+9.61	Tsantini et al. 2018	Haliköy (n=3)	-6.7±0.1	Akçay et al. 2006
Castilla - La Mancha	+6.855	Tsantini et al. 2018			
Castilla - La Mancha	+8.365	Tsantini et al. 2018	**China**		
Castilla - La Mancha	+6.8	Tsantini et al. 2018	Qingtongguo mine (n=3)	+10.54±0.14[(12)]	Minami et al. 2021
Range	From -1.6 to +13.6		Wanshan mine (n=15)	+22.6±3.6	Kawano et al. 2014

Table 7 (continued)

Locality	$\delta^{34}S$	Reference	Locality	$\delta^{34}S$	Reference
Average (n=43)	+5.8		Wanshan mine (n=9)	+24.91± 2.56[13]	Minami et al. 2021
			XunYang mine (n=4)	+10.5±0.1	Kawano et al. 2014
Other localities in Spain					
Escarlati deposit, León	+9.75[4]	Martín-Izard et al. 2009	**Japan**		
M. Sultana-Mariquita, Usagre	+18.3	Domínguez-Bella 2010	Itomuka (n=22)	-4.6±1.8[14]	Minami et al. 2019
M. B.&F. Albuñol, Granada	+4.3	Domínguez-Bella 2010	Ryushoden (n=53)	-2.0±5.5[15]	Minami et al. 2019
M. Sierra de Espadán Chóvar, Castellón	-6.6	Domínguez-Bella 2010	Meiji (n=6)	+8.4±9.5[16]	Minami et al. 2019
M. Oriental Chóvar Castellón	-10.7	Domínguez-Bella 2010	Horokanai (n=1)	+9.5	Minami et al. 2019
M. Encarnación, Caunedo, Asturias	+10.1	Domínguez-Bella 2010	Niu mine (n=34)	-7.3±1.9	Kawano et al. 2014
M. La Uña, León	+10.2	Domínguez-Bella 2010	Niu (n=58)	-8.0±4.7[17]	Minami et al. 2019
M. Pantano de Riaño, León	+9.1	Domínguez-Bella 2010	Niu (n=28)	-8.96±1.84[18]	Minami et al. 2021
M. Riosol Tarna, León	+17.2	Domínguez-Bella 2010	Yamato (n=107)	-2.4±4.4[19]	Minami et al. 2019
Tarna, Asturias	+18.10	Tsantini et al. 2018	Yamato (n=61)	-3.34±1.76[20]	Minami et al. 2021
Caravia, Asturias	+7.29	Tsantini et al. 2018	Yamato-Suigin mine (n=66)	-2.1±1.6	Kawano et al. 2014
Riaño, León	+21.84	Tsantini et al. 2018	Sui mine (n=17)	-2.4±1.4	Kawano et al. 2014
Guipúzcoa, Bask Country	-23.27	Tsantini et al. 2018	Sui (n=16)	-4.8±3.3[21]	Minami et al. 2019
Xóvar, Castelló	-7.82	Tsantini et al. 2018	Suii Mine (n=13)	-3.01±2.70[22]	Minami et al. 2021
Unknown, Castelló	-14.65	Tsantini et al. 2018	Ohita (n=3)	-4.5±1.1[23]	Minami et al. 2019

[1] Range +12.2/+16.6; [2] range +7.3/+9.1; [3] range −0.1/+8.6; [4] range +9.1/+10.7; [5] range +3.6/+22.8; [6] range +0.1/+3.2; [7] range +3.1/+8.0; [8] range +4.9/+7.0; [9] range +3.6/+5.8; [10] range +19.1/+7.7; [11] range +0.1/+6.2; [12] range +10.39/+10.66; [13] +21.30/+28.10; [14] range −7.6/+0.7; [15] range −20.9/+12.7; [16] range +3.9/+27.7; [17] range −15.4/+15.6; [18] range −12.28/−5.85; [19] range −11.2/+20.2; [20] range −6.90/+2.30; [21] range −1.3/+3.3; [22] range −6.21/+3.30; [23] range −5.7/−3.1

and Rodríguez et al. (2020). Other data on cinnabar ores have been further provided by Jébrak et al. (2002), on recrystallised pyrite and Higueras et al. (2005) on cinnabar. These authors used different preparation procedures and analytical techniques; therefore, their results bear different levels of accuracy.

Higueras et al. (2005) prepared the sample through the following procedures (in order): grinding, sieving, electromagnetic separation, ultrasonic bath, dissolution via HCl and HNO_3, heating on a hotplate 100° C for 24 hours and ion exchange in hydrobromic acid. Measurements were performed using a micromass multicollector thermal ionisation mass spectrometer (TIMS). The reference standard was the SRM981 and the results bear an estimated error of 0.15–0.48% for $^{206}Pb/^{204}Pb$, 0.13–1.07% for $^{207}Pb/^{204}Pb$ and 0.10–0.45% for $^{208}Pb/^{204}Pb$ and an overall confidence level of 95%.

Mazzocchin et al. (2008) prepared the sample through the following procedures (in order): dissolution in hydrochloric and nitric acids, heating on a hotplate, cooling and addition of ultra-pure water. The analysis of the samples was performed by quadrupole inductively coupled plasma mass spectrometry (QICP-MS) to study ^{206}Pb, ^{207}Pb and ^{208}Pb.

Hunt et al. (2011) did not provide data regarding sample preparation and the accuracy of the proposed results. Their own measurements were achieved using TIMS.

Minami et al. (2013, 2021) prepared the sample through the following procedures (in order): decomposition with reverse aqua regia, dissolution in hydrochloric acid, ion exchange, dissolution by hydrochloric acid solution and nitric acid and evaporation. Measurements were made by TIMS and MC-ICP-MS. In this second case, the measurements were corrected using the reference NIST981 and NIST99 standards. The isotopes analysed were ^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb. In Minami et al. (2013), the accuracy of the isotopic ratios was of 0.1–0.3% and 0.3–0.6% with ^{206}Pb and ^{204}Pb as the denominator, respectively[15].

Rodríguez et al. (2020) prepared the sample through the following procedures (in order): preliminary digestion by HNO_3 (1st day), removal of supernatant and rinsing of the residue with H_2O, digestion with HF (2nd day), removal of supernatant, rinsing of the residue with H_2O and XRD, digestion with aqua regia (3rd day), evaporation to dryness, ion exchange in hydrobromic acid, lead elution by HCl. Measurements were made using a multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS). Correction was obtained by the addition of thallium with NBS997 as isotopic reference material ($^{205}Tl/^{203}Tl$ ratio of 2.3889) and for lead the reference material NBS981 was used. Further tests allowed these authors to measure a ^{204}Hg contribution lower than 3.5% of the total intensity at mass 204 and established that "accurate Pb ratios are obtained for solutions containing up to 3.6% of Hg and for total signals above 40×10^{-14} A on mass 204".

Lastly, Jébrak et al. (2002) analysed recrystallised pyrite instead of cinnabar. After assessing the absence of mercury contamination through the measurement of ^{202}Hg, they analysed the samples using an ICP-MS (reproducibility of 0.12%, 0.16% and 0.22% for $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios, respectively).

[15] I apologise for any translation errors of the original text in Japanese.

Table 8 The lead isotopic analyses performed on cinnabar ores (G) and archaeological finds (A). All measurements were performed on cinnabar samples, except for the first five by Jébrak et al. (2002) on recrystallised pyrite from the Nuevo Entredicho deposit. The number of digits in the ratios and 2σ values are reported as it appears in the consulted texts but they are considered realistic when they remain within 5 decimal digits for the $^{207}Pb/^{206}Pb$ and $^{204}Pb/^{206}Pb$ ratio; 4 decimals for $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$, $^{208}Pb/^{207}Pb$ and $^{206}Pb/^{207}Pb$ ratios; three decimals for $^{208}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ ratios and two decimals at most for analytical uncertainty. For example, $^{206}Pb/^{204}Pb$ values with four decimal digits are unlikely and uncertainties of almost 1% (*e.g.*, sample Shaanxi 3) make data unreliable [A.Tech. stands for analytical technique. Some measurements taken from Minami et al. 2013 and 2021 are likely the same]

G/A		Locality	Sample	Pb^{208}/Pb^{207}	Pb^{208}/Pb^{206}	Pb^{208}/Pb^{204}	Pb^{207}/Pb^{206}	Pb^{207}/Pb^{204}	Pb^{206}/Pb^{207}	Pb^{206}/Pb^{204}	A.Tech.	Reference
G	China	China*	O	2.442±0.005	2.045±0.002	-	0.837±0.002	-	1.194±0.002	-	ICP-MS	Mazzocchin et al. 2008
G	China	Guizhou	-	-	2.0919±0.0013	38.49±0.20	0.85688±0.00088	15.767±0.041	-	18.40±0.12	TIMS/MC-ICP-MS?	Minami et al. 2013
G	China	Hunan	-	-	2.0830±0.0009	38.58±0.30	0.85608±0.00052	15.840±0.12	-	18.41±0.15	TIMS/MC-ICP-MS?	Minami et al. 2013
G	China	Qingtongguo Mine	n=7	-	2.06424±0.01818	-	0.85112±0.01463	15.14219±0.70113	-	17.73752±1.211	TIMS	Minami et al. 2021
G	China	Shaanxi	1	-	2.0491±0.0011	38.62±0.27	0.83921±0.00005	15.812±0.011	-	18.843±0.013	TIMS/MC-ICP-MS?	Minami et al. 2013
G	China	Shaanxi	2	-	2.0466±0.0003	38.34±0.03	0.83658±0.00014	15.670±0.014	-	18.732±0.014	TIMS/MC-ICP-MS?	Minami et al. 2013
G	China	Shaanxi	3	-	2.0458±0.0002	38.76±0.30	0.83589±0.00051	15.840±0.12	-	18.95±0.15	TIMS/MC-ICP-MS?	Minami et al. 2013
G	China	Wanshan Mine	n=4	-	2.08514±0.00994	-	0.85557±0.00205	15.67945±0.18343	-	18.30175±0.20051	TIMS	Minami et al. 2021
G	Italy	Monte Amiata*	M	2.444±0.006	2.086±0.004	-	0.853±0.002	-	1.172±0.003	-	ICP-MS	Mazzocchin et al. 2008
G	Japan	Mie (Nyu)	1	-	2.0925±0.0002	38.63±0.08	0.84554±0.00010	15.555±0.032	-	18.382±0.038	TIMS/MC-ICP-MS?	Minami et al. 2013
G	Japan	Mie (Nyu)	2	-	2.0910±0.0002	38.56±0.04	0.84350±0.00009	15.559±0.015	-	18.443±0.019	TIMS/MC-ICP-MS?	Minami et al. 2013
G	Japan	Nara (Yamato)	1	-	2.0902±0.0004	38.50±0.07	0.84428±0.00016	15.554±0.030	-	18.421±0.035	TIMS/MC-ICP-MS?	Minami et al. 2013
G	Japan	Nara (Yamato)	2	-	2.1011±0.0002	38.64±0.04	0.84515±0.00010	15.540±0.015	-	18.389±0.017	TIMS/MC-ICP-MS?	Minami et al. 2013
G	Japan	Niu Mine	n=6	-	2.09463±0.00679	-	0.84355±0.00212	15.57194±0.17065	-	18.38939±0.3359	TIMS	Minami et al. 2021
G	Japan	Suii Mine	n=3	-	1.94903±0.00206	-	0.79427±0.00308	15.85788±0.14858	-	19.96679±0.0812	TIMS	Minami et al. 2021
G	Japan	Tokushima	1	-	1.9511±0.0001	39.07±0.05	0.79735±0.00009	15.963±0.021	-	20.024±0.028	TIMS/MC-ICP-MS?	Minami et al. 2013
G	Japan	Tokushima	2	-	1.9470±0.0002	38.77±0.07	0.79119±0.00017	15.753±0.028	-	19.909±0.035	TIMS/MC-ICP-MS?	Minami et al. 2013
G	Japan	Yamato Mine	n=6	-	2.0968±0.0247	-	0.84615±0.00722	15.51989±0.13885	-	18.34016±0.07696	TIMS	Minami et al. 2021
G	Slovenia	Idria Hg-Mine	N	2.455±0.005	2.104±0.003	-	0.857±0.002	-	1.167±0.002	-	ICP-MS	Mazzocchin et al. 2008
G	Spain	Almadén	ALMD-3	-	-	38.826±0.017	-	15.681±0.016	-	18.460±0.016	TIMS	Higueras et al. 2005
G	Spain	Almaden 1*	I	2.442±0.006	2.091±0.003	-	0.856±0.001	-	1.168±0.001	-	ICP-MS	Mazzocchin et al. 2008
G	Spain	Almaden 2*	L	2.447±0.008	2.093±0.002	-	0.855±0.002	-	1.169±0.003	-	ICP-MS	Mazzocchin et al. 2008
G	Spain	El Entredicho	ETD-1	-	-	38.577±0.083	-	15.663±0.081	-	18.357±0.092	TIMS	Higueras et al. 2005
G	Spain	El Entredicho	ETD-2	-	-	38.531±0.062	-	15.641±0.060	-	18.266±0.061	TIMS	Higueras et al. 2005
G	Spain	El Hembrar	Hembrar		2.0973±0.0001	38.105±0.006	0.85984±0.0004	15.622±0.002		18.169±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Escarlatti-Riaño	PA20129		2.0220±0.0001	38.630±0.005	0.82416±0.0003	15.746±0.002		19.105±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Las Cuevas	LC-10	-	-	38.643±0.096	-	15.663±0.086	-	18.112±0.096	TIMS	Higueras et al. 2005
G	Spain	Lois-Riaño	PA20125		2.0780±0.0001	38.638±0.005	0.84457±0.0003	15.704±0.002		18.593±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Miñera de Luna	PA20133		2.1022±0.0001	38.643±0.005	0.85392±0.0003	15.697±0.002		18.383±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Nuevo Entredicho	14 (ME124–126.3 m)	-	-	38.604	-	15.753	-	18.352	ICP-MS	Jébrak et al. 2002
G	Spain	Nuevo Entredicho	15 (ME124–128.8 m)	-	-	38.781	-	15.755	-	18.550	ICP-MS	Jébrak et al. 2002
G	Spain	Nuevo Entredicho	18 (ME124–149.8 m)	-	-	38.594	-	15.748	-	18.386	ICP-MS	Jébrak et al. 2002
G	Spain	Nuevo Entredicho	22 (ME142–127.8 m)	-	-	38.492	-	15.702	-	18.381	ICP-MS	Jébrak et al. 2002
G	Spain	Nuevo Entredicho	23 (ME142–175.5 m)	-	-	38.591	-	15.726	-	18.458	ICP-MS	Jébrak et al. 2002
G	Spain	Pedrosa Rey-Riaño	PA20130		1.8361±0.0001	38.507±0.005	0.75461±0.0003	15.826±0.002		20.972±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Pedrosa Rey-Riaño	PA20131		1.9258±0.0001	38.655±0.005	0.78597±0.0003	15.776±0.002		20.072±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Riaño	PA20132		2.0690±0.0001	38.623±0.005	0.84119±0.0003	15.703±0.002		18.668±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Riosol-Riaño	PA20126		1.9591±0.0001	38.676±0.005	0.79947±0.0003	15.783±0.002		19.742±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Timar, Granada	Complejo El Cruce	-	2.10801	-	0.85758	-	-	18.2065	TIMS	Hunt et al. 2011
G	Spain	Usagre	Rampa		2.1348±0.0001	37.741±0.006	0.87841±0.0004	15.530±0.002		17.679±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Usagre	Pozo Sultana		2.1413±0.0001	37.665±0.006	0.88265±0.0004	15.526±0.002		17.590±0.002	MC-ICP-MS	Rodríguez et al. 2020
G	Spain	Usagre, Badajoz	Mina Rampa	-	2.13803	-	0.88020	-	-	17.6452	TIMS	Hunt et al. 2011
G	Spain	Usagre, Badajoz	Pozo Sultana	-	2.14354	-	0. 88381	-	-	17.5664	TIMS	Hunt et al. 2011
G	Spain	Usagre, Badajoz	US-1	-	2.11676	-	0.86666	-	-	17.9823	TIMS	Hunt et al. 2011
A	Italy	Montegrotto Terme (PD)	D	2.478±0.004	2.085±0.003	-	0.841±0.001	-	1.189±0.002	-	ICP-MS	Mazzocchin et al. 2008
A	Italy	Pompeii (Casa Bracciale d'oro)	H	2.455±0.003	2.082±0.003	-	0.848±0.003	-	1.179±0.006	-	ICP-MS	Mazzocchin et al. 2008
A	Italy	Pompeii (I. Centenario)	G	2.458±0.007	2.079±0.003	-	0.846±0.003	-	1.182±0.005	-	ICP-MS	Mazzocchin et al. 2008

Table 8 (continued)

G/A	Locality		Sample	Pb^{208}/Pb^{207}	Pb^{208}/Pb^{206}	Pb^{208}/Pb^{204}	Pb^{207}/Pb^{206}	Pb^{207}/Pb^{204}	Pb^{206}/Pb^{207}	Pb^{206}/Pb^{204}	A.Tech.	Reference
A	Italy	Pordenone (Torre)	E	2.450±0.006	2.073±0.005	-	0.846±0.001	-	1.182±0.005	-	ICP-MS	Mazzocchin et al. 2008
A	Italy	Trieste (Crosada)	F	2.472±0.006	2.081±0.004	-	0.842±0.003	-	1.188±0.006	-	ICP-MS	Mazzocchin et al. 2008
A	Italy	Verona (San Cosimo)	A	2.446±0.004	2.093±0.005	-	0.856±0.001	-	1.168±0.001	-	ICP-MS	Mazzocchin et al. 2008
A	Italy	Verona (Vicolo Agnello)	B	2.475±0.005	2.059±0.004	-	0.832±0.002	-	1.202±0.001	-	ICP-MS	Mazzocchin et al. 2008
A	Italy	Vicenza (S. Biagio)	C	2.484±0.003	2.071±0.003	-	0.834±0.001	-	1.199±0.002	-	ICP-MS	Mazzocchin et al. 2008
A	Japan	Kurozuka	-	-	2.0921±0.0003	39.32±0.02	0.83791±0.00012	15.745±0.011	-	18.791±0.011	TIMS/MC-ICP-MS?	Minami et al. 2013
A	Japan	Kurozuka tumulus	n=1	-	2.09206	-	0.83791	15.74523	-	18.79072	TIMS	Minami et al. 2021
A	Japan	Nishidani	-	-	2.1683±0.0071	37.75±0.36	0.89014±0.00431	15.500±0.13	-	17.41±0.16	TIMS/MC-ICP-MS?	Minami et al. 2013
A	Japan	Ohburo-minami	-	-	2.1026±0.0008	38.61±0.02	0.85693±0.00017	15.709±0.007	-	18.330±0.006	TIMS/MC-ICP-MS?	Minami et al. 2013
A	Japan	Sakurai-Chausuyama	-	-	2.0941±0.0012	38.51±0.20	0.84422±0.00056	15.574±0.080	-	18.437±0.094	TIMS/MC-ICP-MS?	Minami et al. 2013
A	Japan	Sakurai-Chausuyama	n=1	-	2.09414	-	0.84422	15.57379	-	18.43720	TIMS	Minami et al. 2021
A	Japan	Tatetsuki	-	-	2.1798±0.0002	38.37±0.05	0.88613±0.00010	15.601±0.018	-	17.610±0.021	TIMS/MC-ICP-MS?	Minami et al. 2013
A	Japan	Tenjinyama tumulus	n=3	-	2.14032±0.04307	-	0.8661±0.01778	15.58407±0.02967	-	17.9993±0.36253	TIMS	Minami et al. 2021
A	Japan	Yamato-Tenjinyama	-	-	2.0906±0.0002	38.50±0.24	0.84562±0.00010	15.567±0.097	-	18.42±0.12	TIMS/MC-ICP-MS?	Minami et al. 2013
A	Spain	Casa Montero	7985.3G-2	-	2.09672	-	0.85284	-	-	18.3613	TIMS	Hunt et al. 2011
A	Spain	Humanejos	PA20123		2.0886±0.0002	39.088±0.008	0.83992±0.0004	15.719±0.003		18.715±0.004	MC-ICP-MS	Rodriguez et al. 2020
A	Spain	La Pijotilla	T-1.11.157	-	2.07707	-	0.84703	-	-	18.4791	TIMS	Hunt et al. 2011
A	Spain	La Velilla	PA20120		2.1049±0.0001	38.758±0.006	0.85296±0.0004	15.705±0.002		18.413±0.002	MC-ICP-MS	Rodriguez et al. 2020
A	Spain	Montelirio	DJ07-32,C46	-	2.08951	-	0.84730	-	-	18.4778	TIMS	Hunt et al. 2011
A	Spain	Montelirio	DJ07-32,C46		2.0763±0.0002	38.728±0.007	0.83948±0.0004	15.659±0.002		18.653±0.002	MC-ICP-MS	Rodriguez et al. 2020

*Kremer pigmente GmbHαCo, Airchstetten

From these indications, it is possible to assign different values of precision to the proposed results. The reader must consider that, while the typical precision of TIMS is ca. 0.1% or better (Begemann et al. 1995) and the precision of MC-ICP-MS can reach 0.03% and better (Nørgaard et al. 2019), the quadrupole ICP-MS is not able to provide equally precise results. The latter remains at substantially lower precision (0.1%), often ranging between 0.2 and 0.5% (Gulson et al. 2018) and sometimes reaching even the percent range (Mišta-Jakubowska et al. 2019).

Lastly, mercury isotopes (^{199}Hg, ^{200}Hg, ^{201}Hg, ^{202}Hg) are perhaps the least used in archaeometry. Hintelmann and Lu (2003) demonstrated that different ratios of Hg isotopes might possibly discriminate cinnabar sources, although with some limits due to contamination. To the best of my knowledge, only Cooke et al. (2013), Prieto et al. (2016)[16] and Minami et al. (2021) used mercury isotopes to investigate the use and trade of "archaeological" cinnabar. Conversely, data on natural cinnabar ores (and Hg^0_L) are also available for other areas such as Almaden (δ^{202}Hg ranging between −0.92 and 0.15‰, mean of −0.56‰, σ = 0.35‰, *n* = 7; Gray et al. 2013), Monte Amiata (δ^{202}Hg ranging between −0.96 and −2.25‰, mean of −1.34‰, σ = 0.5‰, *n* = 7; Pribil et al. 2020) and a few Chinese and Japanese cinnabar ores (Minami et al. 2021).

Overall archaeometric investigations have shown that (a) it is possible to obtain significant results through isotopic analyses and (b) the investigated cinnabar ores are still few. Conversely, the database used for comparison should be large enough not to incur "false positives" or those cases in which provenance is assessed based on a non-representative ground for comparison.

Concluding summary of key concepts

Was cinnabar that rare and expensive?

The question may come to mind after such a long and geographically large list of occurrences.

Actually, this should be true only for antiquity, that is, when the use of cinnabar is attested in contexts related to kings, queens and wealthy families. Starting from about the 8th–9th centuries (before/after depending on the geographical area), the possibility of synthesising vermilion must have gradually lowered its cost.

The rarity of the raw material may undoubtedly have favoured its use in neighbouring regions or regions linked by political and commercial relations with the supply areas, such as Spain and Italy in Roman times. Still, it is too early to trace a diachronic and comprehensive picture of its distribution. On the cinnabar trade, we still have little data available, and, at the same time,

[16] Raw data not shown.

the comparison database for provenance assessment is yet small whatever technique is to be used. The isotopic data are still few and cover only some deposits; therefore, much work is needed to reach a larger geographical area. If more effective techniques are not discovered, it would be advantageous to follow already traced paths, such as measuring sulphur and/or lead isotopes. There is already a database available that only needs to be expanded.

What is certain today is that cinnabar has been used to paint almost everything, from human bones to furnishings, from architectural surfaces to manuscripts, from lacquers to lustre. The problems related to *fresco* painting have been typically solved with the *secco* technique. The association with other organic and inorganic pigments such as madder lake or minium seems variously motivated. It may reflect a technical skill, aimed at conferring stability to the pigment and preventing the blackening, or it may represent a counterfeit aimed at increasing the weight of the powder to sell it at an even higher price than the declared one. With the use of cinnabar in lustre production, perhaps a sort of technological milestone is reached because cinnabar is no longer used for its beautiful red colour but as a functional additive to reduce copper.

The discoveries relating to the use of calomel as a pigment have opened a new field of investigation, closely related to the study of the degradation products into which cinnabar can transform. However, recent findings have also highlighted that an approximate, albeit quick and inexpensive analytical approach is no longer sufficient or acceptable.

Pending desirable future developments, we may still agree on what to call it. The distinction between natural cinnabar and synthetic vermilion has worked well for a long time, and it would therefore be helpful to keep it.

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Code availability Not applicable

Declarations

Competing interests The author declares no competing interests.

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