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Analysis of sulphur isotopes to identify the origin of cinnabar in the Roman wall paintings from Badalona (Spain)



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ABSTRACT

Mural paintings bear witness to the lifestyle, wealth and prestige of a specific society and depict its ideas, its beliefs, and its religion. The characterization of the pigments used in mural paintings can shed light on a site's economic status or on the importance of a particular area within the site, since the price of different pigments varied widely. Isotope analysis can be used to identify the origin of pigments and to establish whether the raw materials are local/regional or imported. This, in turn, provides us with valuable information about the socioeconomic relations that developed in a specific society. The present study explores the sulphur isotope ratio of the cinnabar used in the Roman wall paintings sampled from Baetulo (modern-day Badalona, Spain) and compares the ratios found to the geological ores sampled at the various Spanish mercury mines in use in Antiquity. The general conclusion is that the red pigments used in the wall paintings seem to come either from Almadén (Castile la Mancha, Spain, known as Sisopo or Sisapu in Roman times), or from Tarna (Asturias, Spain).

1. Introduction

Mural paintings are faithful testimonies of the lifestyle, wealth, and prestige of a specific society and depict its ideas, its beliefs, and its religion. Many wall paintings reflect the daily activities of a society and the socioeconomic models that developed there. In the classical world, murals were used to decorate both public and private spaces. Roman paintings were influenced by traditional Etruscan painters and by Greek painters from the school of Apelles. The Etruscans used paintings in funerary buildings as magical evocations of spirits and goods, while the Greeks combined these evocations with the practical aim of decorating buildings and walls; finally, the Romans transformed the art of painting into a way of representing the most important features of their lives, thus turning it into a public expression (Vitruvio Polion, 1992). The characterization of the pigments used in mural paintings, in a particular building, at a specific location, may shed light on the economic status of the site and on the importance of the building inside it. As the price of pigments varied widely (Pliny Nat. hist.: 35, 31) their study (among others: Abad-Casal, 1992; Augusti, 1967; Barbet, 1990; Barbet et al., 1990, 1999; Colombo, 1995; Delamare et al., 1990; Giovanoli, 1969) can help to establish whether the raw materials used at a particular site were local/regional or imported, and thus give an idea of the socioeconomic relations that developed inside the society.

Cinnabar, a mercury sulphide (HgS), is a highly toxic, naturally occurring form of the mineral mercury which was used in Antiquity to obtain a bright orange (vermilion) pigment used in ceramics, murals and tattoos, and also in religious ceremonies. The difficult, time-consuming and dangerous process through which the pigment was produced made it a particularly valued substance. In the western Mediterranean the earliest known site of cinnabar production was the Neolithic site of Çatalhöyük in Turkey (7000–8000 BCE) (Çamurcuoğlu-Duygu, 2015; Martín-Gil et al., 1995; Schmitt et al., 2014). Recent investigations in the Iberian Peninsula at the Casa Montero flint mine and burial sites at La Pijotilla and Montelirio (Huarto-Pérez, 1988) suggest that cinnabar began to be used as a pigment approximately 5300 BCE; it has been identified in Neolithic funerary sites in various localities in

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Extremadura in southern Spain (Pavón-Soldevila et al., 2009). Strangely, Plini Secundi (1998) states that cinnabar was not used in Dynastic Egypt. Theophrastus, the fourth-century BCE Greek author of "De Lapidius", is the first writer to refer to this pigment, although it is believed that the pigment was already known to the Greeks in the sixth century BCE (Colombo, 1995) and to the inhabitants of Asia Minor long before that. During the Roman period cinnabar was well known and widely used. Vitruvio Polion (1992) mentions that this red pigment was prepared by breaking, grinding and homogenizing the natural mineral in iron bowls, heating it continuously until all the impurities disappeared, the toxicity was eliminated, and a strong and stable red colour was obtained. He also notes that the pigment remained stable (and thus retained its pure red colour) only if it was not exposed to the heat of the sun; therefore, it could be used only for interior decoration. In fact, when cinnabar is heated it turns grey-black, because its crystalline structure changes and it is transformed into meta-cinnabar (a'-HgS) which has this dark colour. Some called cinnabar the "Pompeian red", although in fact only one of the red pigments sampled from the mural paintings from that site was identified as cinnabar.

Cinnabar is essentially found in all mineral extraction sites that yield mercury, notably Puerto Princesa (Philippines); Almadén (Spain); New Almadén (California); Hastings Mine and St. John's Mine, Vallejo, California; Idrija (Slovenia); New Idria (California); Giza, Egypt; Moschellandsberg near Obermoschel in the Palatinate, Germany; Ripa, at the foot of the Apuan Alps and in Mount Amiata (Tuscany); Mount Avala (Serbia); Huancavelica (Peru); Murfreesboro, Arkansas and Terlingua, Texas and the province of Guizhou (Kweichow and Huanan) in China. It also occurs near Red Devil, Alaska on the middle Kuskokwim River (which was in fact named after the red colour of the cinnabar mine), and in Dominica, Lesser Antilles has its sulphur springs at the southern end of the island along the west coast. In Spain, several cinnabar mines are known today: Almadén (Castile la Mancha), Tarna (Asturias), Sta. Cecilia (Navarra), Riaño (León), Baza and Cástaras (Granada), Orihuela (Alicante), Afrondeguilla, Xóvar and Betxi (Castelló), Minas de Mariquita-Usagre-Badajoz and Torrejoncillo-Cáceres (Extremadura) (Fig. 1; see also Supplementary Fig. 1/Google

Cinnabar normally occurs in veins and impregnations near recent volcanic rocks and hypothermally altered deposits. It crystallises under different mineral forms such as metacinnabar, mercury (metal) or mercury chlorine and with different compositions. In some of the gangues in California and in Almadén in Spain it is contained in quartz, and in others in the same areas it seems to be hosted within carbonate host rocks, as it is in Idria, Slovenia. In Nevada (Humboldt County, Esmeralda County, Goldbanks deposit, Steamboat Springs) and Oregon (Opalite mine) the cinnabar is spread within hydrothermal silica (Foord and Berensen, 1974; Saupé and Arnold, 1992; McCormack and Dickson, 1998; Kim et al., 2000; Lavrič and Spangenberg, 2003; Esbri et al., 2010). In the ancient literature on the exploitation of this red ore in the Mediterranean, Plini Secundi (1998) mentions that during Roman times most of the supplies came from Sisopo, which plausibly corresponds to the mine of Almadén in Spain (Hernández et al., 1999).

The recent advances in isotope analysis technology have helped to expand its use for the identification of the origin of archaeological materials, and specifically of pigments. Several studies have been published on Roman wall paintings, where cinnabar was identified (among others: Edreira et al., 2001, 2003; Franquelo et al., 2009; Martín-Gil et al., 1995; Mazzocchin et al., 2003), very few of them dealt with the identification of sulphur isotopes in western Mediterranean (Saupé, 1990; Saupé and Arnold, 1992; Spangenberg et al., 2010). Using Pb isotope analysis (Mazzocchin et al., 2007), all these studies have indicated Almadén as a possible source. The most extended data is presented in, Spangenberg et al. (2010) who reported for Almadén values between -0.5 and 8.9. Nonetheless, these studies have generally dealt with the plausible sources of cinnabar in Europe, and none of them considered any Spanish mines other than Almadén, nor have any similar studies has been done on Roman wall paintings, in Spain, in spite of the fact that ancient sources mention the presence of many cinnabar mines in the Iberian Peninsula.

The aim of the present study is to compare, for the first time, the sulphur isotope values detected in Roman mural paintings at the site of Badalona with the values of the cinnabar ores sampled at several Spanish mines. We will then use the results to establish whether the pigments are local, and then identify the mines they probably came from. This is the first time this kind of study is carried out for the Spanish territory, is no other works available in isotope measuring of cinnabar in wall paintings in Spain, and the data available for sulphur isotopes in the raw minerals is, equally, limited for Almadén (Castilla-



Fig. 1. Map of Spain with the most important cinnabar ore mines' locations.

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