# A MULTI-ANALYTICAL STUDY OF ROCK PAINTINGS FROM LEANDRO 5 MEGALITHIC BARROW, NORTH-WESTERN PORTUGAL

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**Abstract.** The colourant composition of a northern Portugal megalithic barrow decorated with 'solar' motifs was studied using a multi-analytical approach, allowing the characterisation of the painting techniques, pigments and binders. The red pigment was prepared with iron oxide minerals, using vegetal oils as organic additives, while the black pigment was charcoal without any organic additives or binders. The solar motif was first drawn with charcoal and subsequently painted with a red pigment.

## 1. Introduction

Megalithic monuments are probably the most striking remains of the Neolithic period of western Europe. Each building project was unique, sometimes differing considerably from one structure to another, even between contemporary monuments within a single region. The detailed study of individual sites exposes how barrows were built as architectural, social and symbolic undertakings (Bueno-Ramírez et al. 2016; Laporte and Scarre 2016). Alongside the manner in which the materials were used, the presence of painted, carved or sculpted decorations was a common practice.

Some western Europe megaliths are well known for their paintings. Despite their diffusion throughout Atlantic Europe (Shee 1974; Bueno-Ramírez and Balbín-Behrmann 2002; Bueno-Ramírez et al. 2012; 2016), they are most common in north-west Iberia, predominantly in Galicia (Spain) and the centre-north and north of Portugal (Vasconcelos 1907; Correia 1924; Coelho 1931; Shee 1974, 1981; Jorge 1994, 1997; Silva 1997a, 1997b; Carrera 1997, 2005, 2011; Cruz 1998, 2001; Santos et al. 2017).

The rock painting motifs have been interpreted as an essential part of funeral rites (Bueno-Ramírez et al. 2016). Pigment for painting is produced using an array of mined or quarried organic and inorganic substances (Burgio and Clark 2001; Gomes et al. 2013; 2015). However, archaeologists are often unaware of both the colourant's composition and the technical solutions used in their production, which could also have symbolic importance. In that context, the colourant composition of two northern Portugal megalithic barrows decorated with rock art motifs (Eireira, in Viana do Castelo, and Leira das Mamas, in Braga) were already studied using a multi-analytical approach (XRD, SEM-EDS, FTIR and GC-MS) and the results published, allowing the characterisation, for the first time in Portugal, of the painting techniques, pigments and the organic compounds used as binders (Oliveira et al. 2017). Following a similar analytical approach, the colourant composition of a new megalithic monument was also studied — Leandro 5 barrow, located in Maia municipality, north of Portugal — and the results are presented here.

## 2. Archaeological context

The barrow of Leandro 5 is placed in a megalithic necropolis located at a low-altitude plateau in the parish of Silva Escura, municipality of Maia (Oporto district) in the north-west of Portugal (Fig. 1). It was partially excavated in 2008 and 2009 (Ribeiro and Loureiro 2010, 2011; Loureiro 2018) evidencing a partially destroyed monument, still preserving a sub-circular mound of about 20 m in diameter and a height of 1.80 m, composed of sediments and covered by a lithic protection of angular pebbles, cobbles and granitic blocks (Fig. 2).

While the funerary chamber presented a high degree of destruction, with only an orthostat in situ, a medium-sized east-oriented corridor was well preserved. Two well-imbricated granitic slabs enclosed this corridor. In front of the corridor, there was an atrium, which was partially excavated, containing most of the located depositions, about one thousand
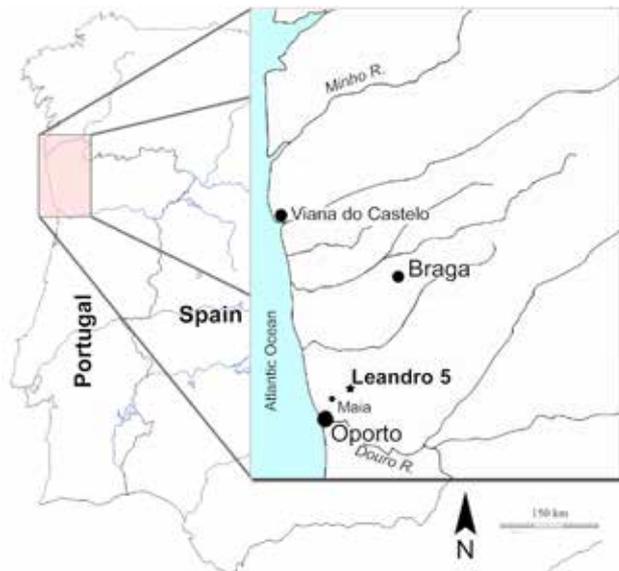
*Figure 1. Location of Leandro 5 mound and other barrows with paintings cited in this text in the Iberian Peninsula and in the north of Portugal.*
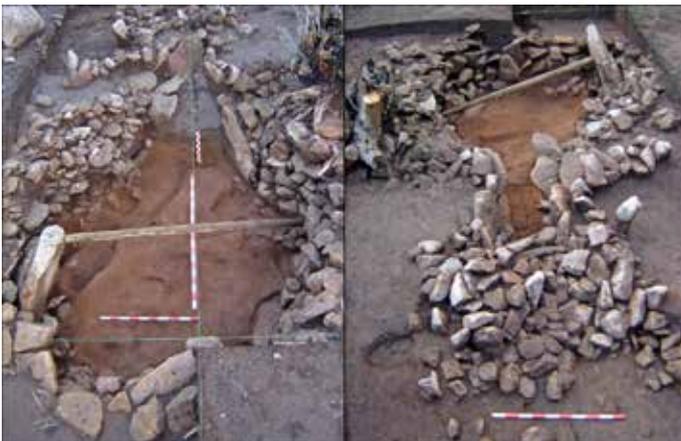


*Figure 3. Drawing of a decorated hemispherical container with some incised motifs (drawing of Amélia Marques).*



*Figure 2. General view of Leandro 5 mound (Ribeiro and Loureiro 2010).*



*Figure 4. Some of the recovered ceramic and lithic artefacts (photo by Manuel Santos from D. Diogo de Sousa Museum).*

pottery sherds corresponding to various hemispherical containers, some of them decorated with motifs performed through incisions (Fig. 3).

Due to the poor preservation of the funerary chamber, very few archaeological materials were recovered there. It was inside the intact corridor that most of the lithic collection was found, composed of more than a hundred artefacts, including arrowheads, blades, lamellae, splinters, microliths, cores, polished axes, gouges, necklace beads etc. (Fig. 4). These objects were manufactured with local and exogenous raw materials. Reflecting the geological substratum of the area (Costa and Teixeira 1957; Pereira et al. 1992), the local objects were made of slate, schist, quartz, quartzite and amphibolite. The others were constructed with flint, revealing supra-regional contacts. It is important to note that the nearest flint deposits are in the centre of Portugal, in Cantanhede, near Coimbra (Barbosa et al. 2008; Aubry et al. 2014), about 100 km to the south.
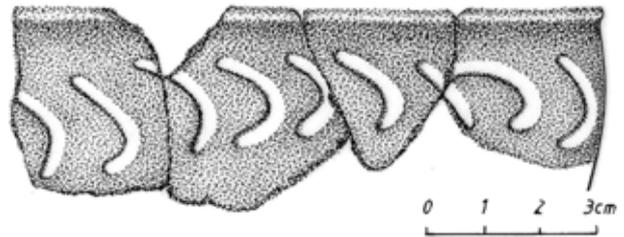
Although there is no carbon dating ($^{14}$C) data available, the monument is thought to be from the end of the fifth or the fourth millennia BCE. This estimation was achieved by correlating with the $^{14}$C dates for this phenomenon in north-western Portugal and Spain (Bettencourt 1991–1992; Jorge 1986, 1992; Cruz 2001; Bettencourt et al. 2008; Bueno-Ramírez et al. 2016; Carrera 2016).

At the funerary chamber in the head orthostat, though broken and displaced from its original position (Ribeiro and Loureiro 2010, 2011), the remains of a painted red sun-shaped motif were identified. It is composed of a central 'solar' motif between two straight vertical segments (Ribeiro and Loureiro 2010, 2011). Already in the scope of this work, we detected the additional presence of black lines also composing the sun-shaped motif, which appear to have been drawn before the paintings. This fact was particularly well visualised by analysis of digitally processed pho-
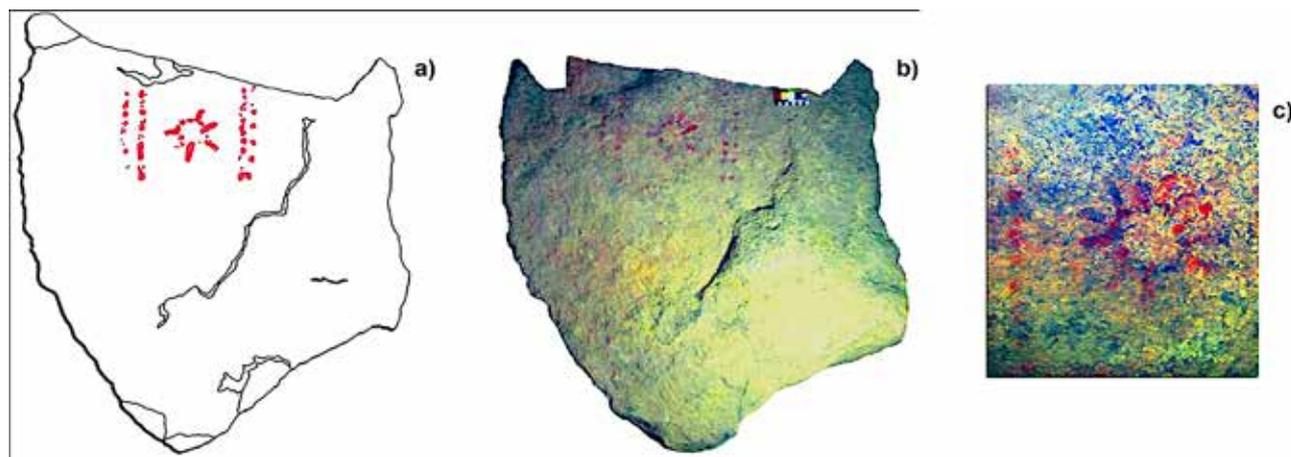
*Figure 5. Chamber's head orthostat with the painting motifs: (i) the first recorded painting motifs (Ribeiro and Loureiro 2011); (b) photographic survey of the chamber's head orthostat with a 'solar' motif; (c) magnified picture of the 'solar' motif showing two different pigments (digitally processed photos with Image J software, plugin D-Stretch).*

tos with Image J software, plugin D-Stretch (Fig. 5).

## 3. Material and methods

The analytical methodologies have already been described in detail elsewhere (Oliveira et al. 2017). Briefly, the pigments were scraped with clean scalpels gathering small aliquots of about 0.5 g that were stored in Eppendorf tubes until analysis. Different aliquots of the stone were also collected and analysed to study the possible contamination of the pigment samples with minerals from the stone. All the procedures were performed following the best laboratory practices, e.g. using gloves, clean glassware and sterilised instruments to prevent sample contamination (Oliveira et al. 2017).

The collected materials (black and red pigments and stone material) were analysed with three different analytical techniques: x-ray diffraction (XRD); scanning electron microscopy - energy dispersive spectrometry (SEM-EDS) and gas chromatography coupled to mass spectrometry (GC-MS).

*XRD:* The crystalline phases characterisation and identification were performed in a Philips PW 1710 diffractometer with a graphite monochromator, Cu-K$\alpha$ radiation and operating at 40 kV and 30 mA. The powder XRD patterns were examined in the region of 2° to 65° with step size 0.02° for 2θ and scan speed of 2 s per step. The identification of the mineral phases was achieved by comparison with a database based on JCPDS-ICDD patterns. All the studied samples were manually ground in an agate mortar

*SEM-EDS*: The SEM-EDS analyses were carried out using a JSM-610 LV scanning electron microscope from Jeol (Japan), equipped with the low vacuum mode as a standard feature. This SEM is equipped with an energy dispersive spectroscope (EDS - INCAx-Act, PentaFET Precision, Oxford Instruments). EDS resolution: 5.9 keV–129 eV.

*GC-MS*: The chromatographic analyses were performed with a quadrupole analyser Thermo Scientific™ IS device that was operated in full scan mode with the following experimental conditions: (a) column

HP-5MS, 60 m × 0.25 mm × 0.25 μm using helium as carrier gas with a constant flux of 1 mL min$^{-1}$; (b) injection volume of 1 μL; (c) injector temperature of 250°C; (d) heating program: initial temperature, 60°C; hold time 1 min; 60°C to 150°C (10 min$^{-1}$); 150 to 290°C (5°C min$^{-1}$); 290°C (27 min); (e) ionisation mode, electronic impact at 70 eV; (f) interface and ionic source at 290°C; (g) scanned masses from m/z 50 to 600. Compound identification was based on the GC/MS spectra libraries Wiley6 and NIST08, co-injection with authentic standards and analysis of fragmentation patterns.

## 4. Results
*XRD*

The study by XRD provided diffractograms with information about the crystalline phases from pigments and rock samples (Figs 6.a to 6.c). It is necessary to emphasise that small amounts of pigment were analysed and that the crystalline phases can only be identified by conventional XRD if the amount in the sample is enough to give a good signal-to-noise ratio (Duran et al. 2010; Rogerio-Candelera et al. 2013). Consequently, minority compounds can be masked either by the background noise or by the existence of strong reflexions from the substratum.

Quartz ($SiO_2$), feldspars (K-feldspars - $KAlSi_3O_8$ and plagioclases - $NaAlSi_3O_8$–$CaAl_2Si_2O_8$), micas (biotite - $K(Mg,Fe)_3(AlSi_3O_{10})(F,OH)_2$ and muscovite - $KAl_2(AlSi_3O_{10})(F,OH)_2$) were the main mineral phases identified in all the samples. This composition is a reflection of the rock mineralogy, as the samples from undecorated areas (Fig. 6.b) confirmed the composition of the granitic substratum (quartz, feldspars and micas), and are a strong signature in the XRD data from the pigment samples.

In spite of the low signal-to-noise ratio and the strong signature of the substratum minerals, it was possible to detect the presence of haematite ($\alpha$-$Fe_2O_3$) in the red pigment samples (Fig. 6.a), characterised by some very small and broad reflections with d-spacing of 2.70, 2.51 and 1.69 Å. This mineral is the main crys-

talline compound associated with the red colour of the pigment.

It was also possible to identify kaolinite ($Al_2Si_2O_5[OH]_4$) (peaks with d-spacing of 7.17, 3.58 and 1.49 Å) and clinochlore ($[Mg,Fe]_5Al[Si_3Al]O_{10}[OH]_8$) (peaks with d-spacing of 14.1, 7.16 and 2.54 Å) in both the red (Fig. 6.a) and black pigments (Fig. 6.c) samples. Kaolinite is a clay mineral while clinochlore is a phyllosilicate mineral from the chlorite group.

The black pigment (Fig. 6.c) was more difficult to characterise due to the strong signatures of the granitic substratum. In fact, no crystal phase that could be associated with the black colour of the pigment was identified. A slightly broad band, between about 20º and 26º (2θ), is probably due to an amorphous carbon black (probably charcoal).

*SEM-EDS*

Figure 7 presents SEM images and EDS spectra from the pigments and undecorated rock, while Figure 8 presents SEM images and EDS spectra from sectors of the pigment samples. It is necessary to emphasise that the EDS spectra shown in the Figures represent the composition of the areas marked with the 'P' in the SEM image, reflecting a punctual analysis from a complex inhomogeneous mixture. The EDS analysis of the red pigment samples (Figs 7.d and 8.c) confirmed the presence of iron, which is consistent with the presence of haematite in the pigment. Silicon, magnesium, aluminium, potassium and sodium were also detected. These results also reflect the orthostat granitic composition (quartz, micas and feldspars) and the presence of kaolinite.

The presence of Mg (Figs 8b and 8d) can be associated with biotites from the substratum or with the presence of clinochlore, identified in the XRD analysis. The SEM observations (Fig. 7.a) strengthen these conclusions, showing iron oxide flakes (of less than
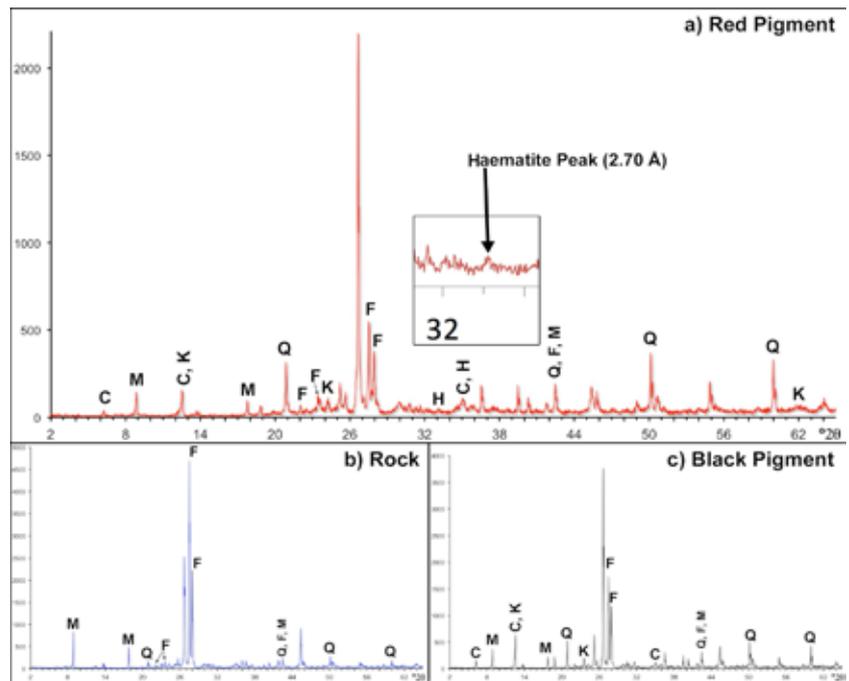
**Figure 6.** *XRD analysis of the Leandro 5 mound pigments: (a) red pigment, (b) rock from an undecorated area and (c) black pigment. The identified mineral phases were as follows: C, clinochlore; K, kaolinite; H, haematite; Q, quartz; M, micas; F, feldspars.*
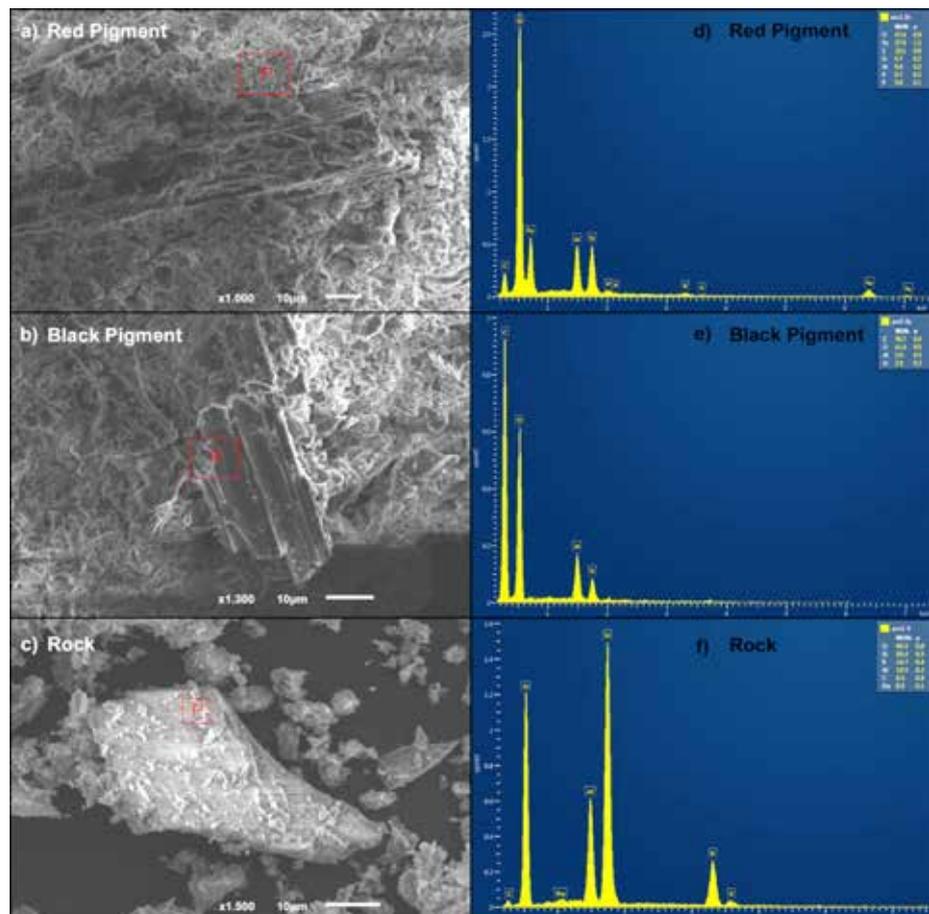
**Figure 7.** *SEM images from the (a) red pigment, (b) black pigment and (c) undecorated rock. EDS spectra from the (d) red pigment, (e) black pigment and (f) undecorated rock.*
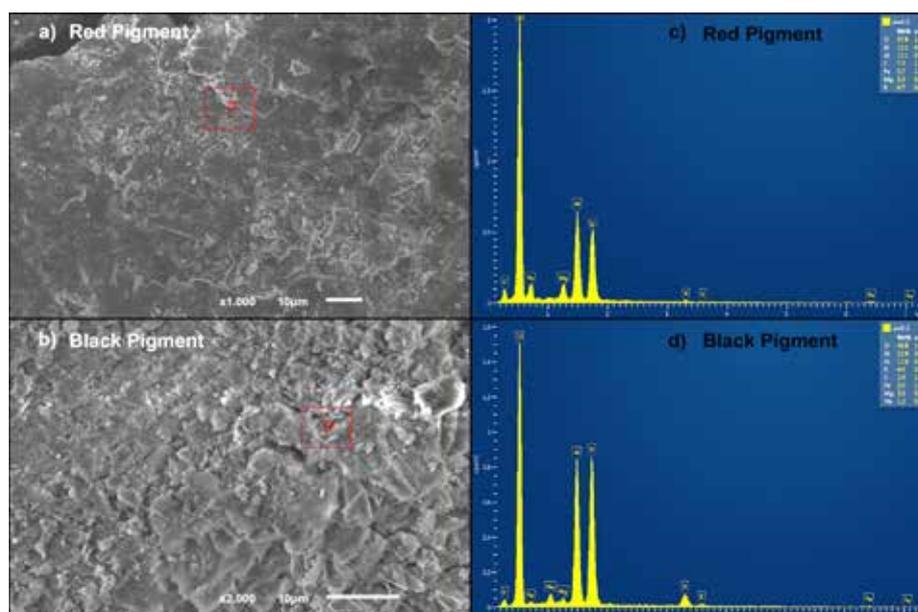
**Figure 8.** *Analysis of the Leandro 5 mound pigments. SEM images (a and b) and EDS spectra (c and d) from the black and red pigment. In the EDS spectra it is possible to observe the presence of Mg.*

5μm) and several crystalline grains with structural and textural characteristics from the minerals referred to above. The analysis of samples from undecorated areas indicated a residual presence of iron and magnesium, probably associated with the biotite mica. In this case, the SEM observations showed only the presence of the granite mineral phases. Small quantities of carbon were also detected, this element probably being associated with the organic material in the pigments and/or with biological or human contamination, or only being due to the SEM manipulation in a low vacuum.

The EDS analysis from the black pigment (Fig. 7.e) shows a strong signal for carbon. These results are consistent with the presence in the pigment of a material like charcoal, responsible for the black colour. The SEM observation (Fig. 7.c) showed a structure similar to charcoal (Winter 1983; Zicherman and Williamson 1981). The identification of silicon and aluminium (Figs 7.e) is probably due to the presence of kaolinite. The spectrum from Figure 8.d is from a sector of the black pigment sample with low carbon content, probably reflecting the composition of an extender and/or the orthostat. The presence of silicon, aluminium, magnesium, potassium, sodium and iron reflects the granite mineralogy (orthostat) and can also be related to the presence of clinochlore and kaolinite.

*GC-MS*

Figure 9.a presents a typical chromatogram of red pigment samples (several samples were analysed, resulting in very similar chromatographic data). Lipids from archaeological matrices are frequently highly degraded, particularly with the breakdown of triacylglycerols by hydrolysis, creating diacylglycerols (DAG) and monoacylglycerols (MAG) through the loss of one

or two fatty acyl moieties, free fatty acids and glycerol. The amount of free fatty acids in an archaeological sample increases with ageing, reflecting the extent of hydrolysis of the triacylglycerols.

The red pigment chromatogram was dominated by 1-monostearin (MAG $C_{18:0}$), also containing monopalmitin (MAG $C_{16:0}$), traces of palmitic ($C_{16:0}$) stearic ($C_{18:0}$) acids and glycerol. This profile is typical of degraded animal fats and plant oils.

The absence of cholesterol, an animal fat biomarker (Baeten et al. 2013; Kimpe et al. 2001, 2002) together with the detection of high-levels of oleamide and oleanitrile, suggests that a reaction of oleic acid in a basic environment could have taken place, which in turn points to the presence of fatty acids from vegetable oils (Pecci and Cau-Ontiveros 2010; Vaccaro et al. 2013). Despite the absence of additional tracers that would allow the identification of the oil composition, the high abundance of these two compounds suggests the use of plant oils extremely rich in oleic acids, such as those from *Olea europaea* var. *europaea L.* (olive) or *Olea europaea* subsp. *europaea* var. *sylvestris* (oleaster). The detection of docosenoic acid ($C_{22:1}$) could suggest the presence of *Brassicaceae*, e.g. plants of the mustard family (Colombini et al. 2005; Eerkens 2007; Romanus et al. 2008; Pollard and Heron 2015). However, other plants also produce this molecule in small amounts (Eerkens 2007).

Dehydroabietic acid is a diterpenic compound produced by the heat of abietic acid throughout a dehydrogenation process (Eerkens 2002; Jerković et al. 2011; Malainey 2011; Pollard and Heron 2015). It is one of the main components of *Pinaceae* resins, suggesting the use of resin as a pigment binder.

The chromatographic results suggest that the red pigment was prepared using vegetal oils as organic additives, in a mixture that was probably heated and stabilised using cellulosic material of *Pinaceae* origin.

Contrasting with the chromatograms obtained for the red pigment, the black pigment chromatograms evidence a restricted number of organic tracers (Fig. 9.b), as the only relevant detected compounds were monostearin (MAG $C_{18:0}$) and oleamide. The absence of additional traces, particularly of dehydrobietic acid and oleanitrile, suggests that the black pigment was applied without organic additives or binders. It also seems that the 'solar' motif was drawn first with charcoal and then painted red. No organic matter was detected by GC-MS analysis upon the rock surface.

## 5. Data discussion and final considerations

A range of techniques (XRD, SEM-EDS and chromatographic analysis) was applied on sample pigments collected on a chamber's head painted granitic orthostat from the Leandro 5 barrow.

The XRD and SEM-EDS analyses showed that the reddish colour of the pigment found in the orthostat is due to the presence of iron oxide minerals, particularly haematite, while the black colour from the pigment comes from charcoal. The detection of kaolinite and clinochlore (phyllosilicate mineral from the chlorite group) in the red pigment could be due to the use of these minerals as extenders in the paint composition. The chlorite group minerals are commonly used in the composition of pigments in art and archaeology in several continents and chronologies (Uda et al. 2005; Sotiropoulou et al. 2012; Gebremariam et al. 2016; Siddall 2018) but it is the first time it has been detected in pigments from the Neolithic of west Iberia. Usually, these minerals are used as extenders in different colours, from red to green, blue and black. Kaolinite is also used as white pigment or as an extender in other colours (Oliveira et al. 2017; Siddall 2018). It was probably the case in these pigments. Kaolinite and chlorites are common minerals in clay sediments, soils and weathered rocks from the region (Pereira et al. 1992). Their detection can be due to the deliberate use of these materials or to its presence in the material (clay/soil) used as an extender. The iron oxide mineral pigments were prepared in a mixture with vegetal oils, acting as organic additives, while the charcoal was used without any organic additives or binders.

Although few in quantity, the motifs painted in the chamber's head orthostat from Leandro 5 barrow are extremely interesting as sources of new knowledge about the Neolithic funerary scenarios.

They present a sun-shaped motif inside a frame of vertical lines of points, placed on the central area of the head orthostat, which, associated with the orientation of the corridor to the sunrise, allows to consider the
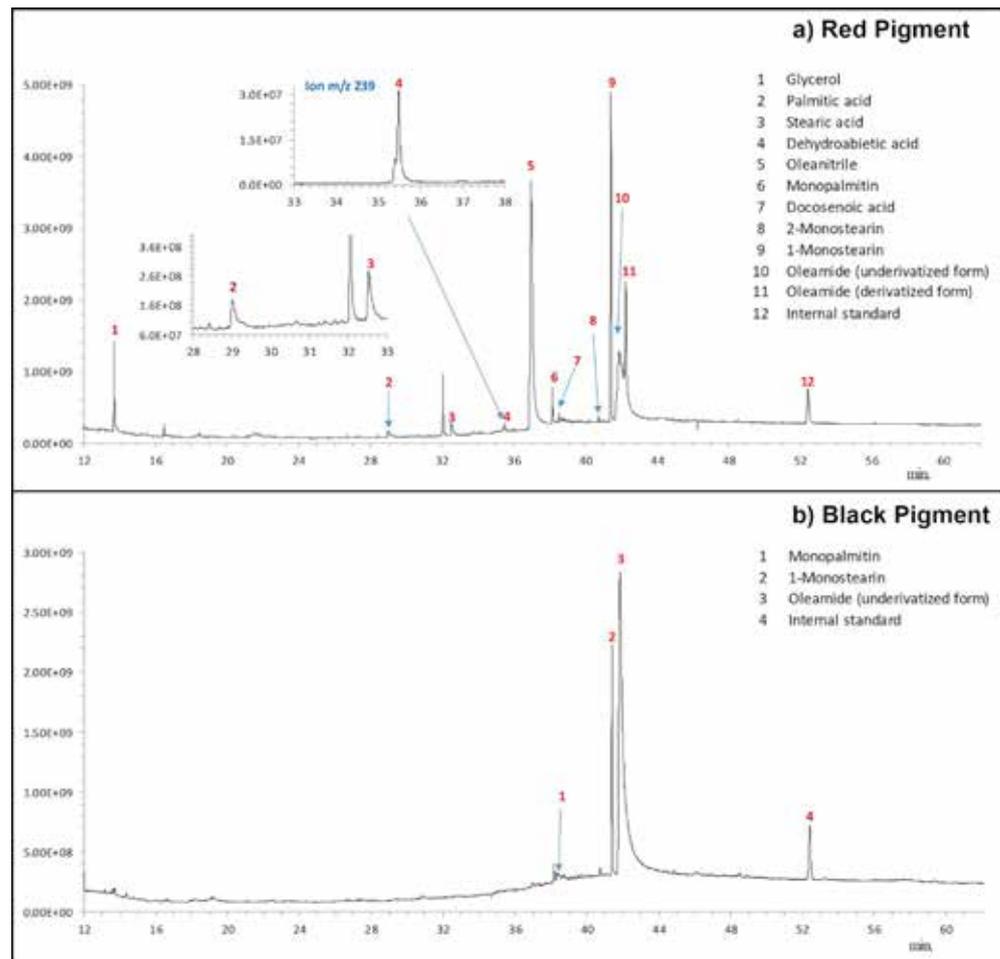


**Figure 9.** *Chromatograms of Leandro 5 extracts of the: (a) red pigment and (b) black pigment.*

hypothesis of a symbolic interaction between death and the solar cycle, as has already been proposed in relation to other cases (Bettencourt 2013).

Due to the partial preservation of these paintings on the head orthostat, the apparent absence of other paintings in the located chamber and corridor orthostats, it is possible to include this monument in the scope of the dolmens with simple paintings where the drawing or painting was applied directly over the orthostats, according the classification of Bueno-Ramirez and Balbín-Behrmann (1997), Bueno-Ramírez et al. (1999a) and Bueno-Ramírez et al. (1999b). They would have been built before the first half of the fourth millennium BCE, following Carrera (2011).

Another aspect to emphasise is the particularity of the sun-shaped motif being drawn at first with a black pigment, later painted in red. We are thus faced with the use of two different techniques (drawing and painting) and two different colours (black and red). The colours appear overlapped, attesting two possible pictorial moments in this monument. What is the significance? Is it a restoration or a new painting?

The absence of black pigment in other places from the orthostat, especially in the area of the lateral figures, allows us to consider that it was not a restoration but a

decision that changed the original colour of the central motif and the original composition, enhancing it. However, any architectural modifications that may have accompanied these actions are unknown, although the monument was reused during the third millennium BCE (Ribeiro and Loureiro 2010). Therefore, it is unknown if these changes correspond to their first occupation-reoccupation moment, during the fourth millennium BCE, or if they occurred at a stage during the third millennium BCE.

What seems obvious is that the sun-shaped motif remains significant within communities that were investing in time-consuming paint-making techniques for use in the megalith, where minerals, oil and resinous material are major contributing agents. It is a recipe that uses, in a balanced and lasting way, different elements of nature, like the haematite red pigment, added to kaolinite and clinochlore, as extenders, and vegetal oils as organic additives, in a mixture that was probably heated and stabilised using cellulosic material of *Pinaceae* origin, revealing a deep knowledge of the resources available.

It also seems relevant that the plant oils used are extremely rich in oleic acid, for example, those obtained from *Olea europaea* var. *europaea L.* (olive) or *Olea europaea* subsp. *europaea* var. *sylvestris* (oleaster). The former is treelike and the second tends to take a shrubby form (Carrión-Marco et al. 2010). The wild olive that grows in Iberia is associated with different plant communities present nowadays in the Thermomediterranean level (below c. 200 m) and in the lower Mesomediterranean level (c. 200–500 m) (Rivas-Martínez 1987; Costa et al. 1998). During the Holocene, charcoal sequences in Iberia document the presence of *Olea* in the Mesomediterranean level, particularly in its lower part where its presence seems to be strongly linked to favourable orographic conditions — sunny slopes and valley floors that are warm and protected from continentality (Rodríguez-Ariza and Moya 2005; Carrión-Marco et al. 2010, 2013). The presence of *Olea* charcoal has been reported in north-east Portugal in the Late Neolithic context of Dólmen de Arcã (Mirandela), in the Copper Age occupation of Barrocal Alto (Mogadouro) (Figueiral and Sanches 1998–1999; 2003) and in Early-Middle Bronze Age contexts of Foz do Medal and Terraço das Laranjeiras (Martín-Seijo et al. 2017).

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