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Main Manuscript for

Painting the place: insights from Cueva de Ardales into the symbolic role of caves in the Middle Paleolithic.

Africa Pitarch Martí, 1*, João Zilhão, 1,2,3*, Francesco d'Errico, 4, 5, Pedro Cantalejo-Duarte, 6, Salvador Domínguez-Bella, 7, Josep M. Fullola, 1, Gerd C. Weniger, 8,9, José Ramos-Muñoz 10.

1 Seminari d'Estudis i Recerques Prehistòriques (SERP), Facultat de Geografia i Història, Departament d'Història i Arqueologia, Universitat de Barcelona, Montalegre 6, 08001, Barcelona, Spain.

2 Institució Catalana de Recerca i Estudis Avançats (ICREA), Passeig Lluís Companys 23, 08010 Barcelona, Spain.

3 Centro de Arqueologia da Universidade de Lisboa (UNIARQ), Faculdade de Letras, Campo Grande, 1600-214 Lisboa, Portugal.

4 Univ. Bordeaux, UMR 5199 CNRS, De la Préhistoire à l'Actuel : Culture, Environnement, et Anthropologie (PACEA), Bâtiment B18, Allée Geoffroy St Hilaire, 33615 Pessac, France.

5 SSF Centre for Early Sapiens Behaviour (SapienCe), University of Bergen, Øysteinsgate 3, Postboks 7805, 5020, Bergen, Norway.

6 Centro de la Prehistoria/Cueva de Ardales, Avenida de Málaga, 1, 29550 Ardales (Málaga), Spain.

7 Unidad de Geoarqueología y Arqueometría aplicada al Patrimonio Histórico-Artístico y Monumental (UGEA-PHAM). Departamento de Ciencias de la Tierra, Facultad de Ciencias. Universidad de Cádiz. Campus Río San Pedro. 11510. Puerto Real, Cádiz, Spain.

8 Department of Prehistoric Archaeology, University of Cologne, Weyertal 125, 50931 Cologne, Germany.

9 Neanderthal Museum, Talstraße 300, 40822 Mettmann, Germany.

10 Departamento de Historia, Geografía y Filosofía, Universidad de Cádiz, Avenida Gómez Ulla s/n, Cádiz, Spain.

* Corresponding author

Email: africa.pitarch@ub.edu; joao.zilhao@ub.edu

Africa Pitarch Martí 0000-0002-8396-9487

João Zilhão 0000-0001-5937-3061

Francesco d'Errico 0000-0002-2422-3079

Pedro Cantalejo 0000-0001-6408-7385

Salvador Domínguez-Bella 0000-0003-3892-763X

Gerd-Christian Weniger 0000-0002-5270-657X

José Ramos-Muñoz 0000-0002-6042-2446

Josep M. Fullola 0000-0001-7089-1425

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APM, JZ, FD, PCD, SDB, G-CW, JRM designed research

APM, JZ, FD, PCD, SDB, G-CW, JRM performed research

APM, JZ, FD, PCD, SDB, G-CW, JRM analyzed data

JZ, PCD, G-CW, JRM provided and reviewed archaeological data

APM, JZ, FD interpreted data

APM, JZ, FD, PCD, SDB, JMF, G-CW, JRM wrote the original paper

APM, JZ, FD, PCD, SDB, JMF, G-CW, JRM reviewed the original paper

This PDF file includes:

Main Text
Figures 1, 2, 3
Table 1

Abstract

Cueva de Ardales (Málaga, Spain) is one of the richest and best-preserved Paleolithic painted caves of southwestern Europe, containing over a thousand graphic representations. Here, we study the red pigment in panel II.A.3 of “Sala de las Estrellas”, dated by U-Th to the Middle Paleolithic, to determine its composition, verify its anthropogenic nature, infer the associated behaviors and discuss their implications. Using optical microscopy, scanning electron microscopy coupled with energy dispersive X-ray spectroscopy, micro-Raman spectroscopy, and X-ray diffraction we analyzed a set of samples from the panel and compared them to natural coloring materials collected from the floor and walls of the cave. The conspicuously different texture and composition of the geological samples indicate that the pigments used in the paintings do not come from the outcrops sampled in the cave. We confirm that the paintings are not the result of natural processes and show that the composition of the paint is consistent with the artistic activity being recurrent. Our results strengthen the hypothesis that Neanderthals symbolically used these paintings and the large stalagmitic dome harboring them over an extended time span.

Significance Statement

The emergence of symbolic behavior in our genus is a controversial issue. The dating of paintings in three caves from the Iberian Peninsula supports the view that Neanderthals developed a form of cave art more than 20,000 years before the emergence of anatomical modernity in Europe. In this study, we confirm that the paintings on a large speleothem from one of these sites, Cueva de Ardales, were human made, and we show that the pigments were not collected at the outcrops sampled inside the cave. Variations in the composition of the paint correspond to differences in the age of the paintings, supporting the hypothesis that Neanderthals used the speleothems symbolically over an extended time span.

Main Text

Introduction

The production of art is considered a big leap forward in the cultural evolution of humankind. It represents a means of recording and transmitting complex symbolic representations in a durable way (1, 2). However, despite the work of generations of researchers, many questions concerning the origin, chronology, technology, function and meaning of Paleolithic art remain open. Research conducted over the last two decades has focused on the earliest instances of graphic representation (3, 4), the interdisciplinary analyses of key cave sites (5-8), the study of open air sites (9-11), the presentation of new discoveries (12-14), and the dating of the earliest instances of cave painting (15-21).

Of particular relevance is that the application of U-series dating to stratigraphically associated calcite accretions has shown these artistic manifestations to be of much greater antiquity than hitherto thought. At El Castillo cave (Spain), a minimum age of 40.8 ka was obtained for a red disk (18), consistent with Neanderthal authorship of Europe’s earliest cave art, as eventually corroborated by the non-figurative paintings and hand stencils from three Iberian sites dated to >64.8 ka (20). Hand-stencil art from Borneo and a naturalistic painting from Sulawesi have yielded minimum ages of 39.9 ka and 43.9 ka (15-16), convincingly demonstrating broad contemporaneity with the earliest European manifestations of this practice, as predicted (22). The Iberian evidence has been challenged (23-27), but all the criticisms have been exhaustively responded to (28-31).

One of the early Iberian sites is Cueva de Ardales (Fig. 1, *SI Appendix, Archaeological context*), which has a long but intermittent history of research, beginning with Breuil more than a century ago (32) and continuing with the recent investigations, carried out by an international research team led by two of us (G-CW and JRM). Until now, however, the pigments composing the paintings in the cave, including those dated by U-series, remained unanalyzed. As part of a broader project to study

the origin and evolution of southwestern Europe's Paleolithic painting technologies, this paper focuses on the microscopic and chemical analysis of panel II.A.3 (Fig. 2).

Based on the U-series dating of calcite samples, the age constraints for the red stains in panel II.A.3 are as follows: >45.9 ka in Curtain 5; >45.3 ka and <48.7 ka in Curtain 6; >65.5 ka in Curtain 8 (20). These results place the artistic activity in the regional, Neanderthal-associated Middle Paleolithic, and there is nothing to suggest that the decoration of the panel's undated curtains might be of a different, later age.

Our aims here are twofold. Firstly, we intend to characterize the composition of the red pigment of panel II.A.3 (*SI Appendix, Materials and Methods*, Fig. S1). It has been suggested (23) suggested that the pigment could represent natural staining, which, macroscopic observation does not support (30) rejected as unsubstantiated. Secondly, we investigate whether patterns in pigment composition and technology can provide additional detail on the different phases of Middle Paleolithic artistic activity demonstrated by the dating. We also analyzed natural Fe-rich coloring materials collected from the floor and walls of the cave (*SI Appendix, Materials and Methods*, Fig. S2) to see if the chemical fingerprint of these geological materials was consistent with their being the source of the pigments used in the paintings.

Background to the site

Cueva de Ardales is situated close to the eponymous village, in Málaga, southern Spain (Fig. 1). The cave is 1577 m long and features two superimposed levels: the Lower and the Upper Galleries. The site was discovered in 1821 after an earthquake re-opened a cave entrance previously sealed by colluvial sediments, but it was not until 1918 that the Paleolithic rock art was found by Henri Breuil (32). Over a thousand graphic representations, mostly attributed to the Upper Paleolithic, have been described. They include both figurative and non-figurative engravings and paintings grouped into two hundred and fifty-two panels (33). Most abstract red paintings are on speleothems and located near the entrance rather than at the back, i.e., in those parts of the cave in which archaeological excavation has corroborated broadly coeval use of the space by Middle Paleolithic Neanderthals (34). Panel II.A.3, which is the focus of the present study, is located in an impressive stalagmitic dome of *Sala de las Estrellas* (Fig. 2), in the Lower Gallery. More detailed information on the cave setting, research history, art and human use is provided in (*SI Appendix, Archaeological context*)

Results

Micro-samples from panel II.A.3. The samples are composed of hematite, aluminosilicates (clay minerals and micas), calcite and, in some cases, quartz and amorphous carbon (Table 1, *SI Appendix, Results*). Analyses also detected traces of P and S that may come from small amounts of sulphates and phosphates. SEM observation of these micro-samples indicates a mineral origin, since none of them features the particle morphologies typically found in biomineralizations (e.g., filaments, coccoid forms, beads-on-a-string, rods arranged in rows, biofilms; 35-37). The shape and size of crystals in the micro-samples are also consistent with a mineral habit. Closer analysis reveals interesting textural and compositional differences (*SI Appendix, Results*, Fig. 3, Fig. S3 to S7). Samples from Curtains 5 (P-ARD-06) and 8 (P-ARD-03 and P-ARD-04) are composed of tightly bound agglomerates of sub-micrometric to micrometric platy Fe-rich minerals and clays while in Curtain 6 (P-ARD-05) the hematite and aluminosilicate particles do not appear as agglomerates but are recorded in the form of individual particles. The red stains of Curtain 9 (P-ARD-02) differ from those of Curtains 5 and 8 for the presence of coarse isolated mica platelets (15-30 μm) and the lack of hydrated clay minerals. Unlike Curtain 6, and similarly to Curtain 5 and 8, hematite and clay particles occur in Curtain 9 in the form of agglomerates. In addition, Curtain 6 (P-ARD-05) reveals the presence of amorphous carbon (*SI Appendix, Results*, Fig. S8), not detected in the other samples.

Geological samples. Six types of Fe-rich deposits that could have potentially been used as pigments were identified in the cave. They are composed of heterogeneous materials ranging from loose ochraceous sediment to compact violaceous rocks (*SI Appendix, Materials and Methods*, Fig. S2, Table S2, *Results*, Fig. S11). SEM analysis makes it clear that these materials share no similarities with the samples from panel II.A.3 (*SI Appendix, Results*, Fig. S12, Table S3). The XRD analysis (*SI Appendix, Results*, Table S4) shows that only two of the geological samples (G-ARD-01 and G-ARD-11) include hematite, the mineral responsible for the color of the red staining on the stalagmites, but none of them is comparable to those from the panel: sample G-ARD-01 is composed of micrometric to submicrometric granular, massive and acicular Fe/Mn-rich crystals (Mn reaching concentrations ranging from 0.5 to 2 %), Fe-rich sulphate spherules, and K-rich micas (Si, K, Al, Ca, Ti, Mg), while sample G-ARD-11 consists of clusters of 2 μm disc-shaped Fe-rich crystals in a foliated matrix of clays (Si, K, Al, Mg, Ti, Ca, Mn).

Speleothem sample. Microscopic and Elemental analyses performed on the speleothem layer covering one stain (*SI Appendix, Results*, Fig. S13) show that it is mainly composed of low-magnesium calcite. Small amounts of aluminum, probably from Al-rich hydroxide, were also detected. No iron or clay minerals were detected.

Discussion

The analysis of the micro-samples collected on the panel indicates, as suggested on the basis of close observation of the panel by naked eye (30), that the staining is mineral in origin and cannot be interpreted as the result of microbial activity. The staining cannot be interpreted either as the result of natural geological processes typically occurring in caves such as fluvial flows, infiltration from soils, percolating waters, or weathering of the walls (38). Although flooding may coat the walls and even the roof of a cave, most accumulation occurs on the floors and is, in general, widespread. In Cueva de Ardales, traces of a deposit formed by flooding are visible neither on the floor nor on the walls of the chamber in which panel II.A.3 is located. In addition, clay platelets transported by flooding generally show, under SEM, broken or rounded edges (39), which is not the case with our samples. Deposition of iron oxides by dripping water would produce a diffuse red staining of the calcite while the deposit interpreted as paint occurs in the form of a distinct layer on top of and/or covered by calcite. Inclusion within the calcite of iron-rich particles present in drip water would not lead to the formation of loose hematite and clay platelets, as observed in panel II.A.3. Weathering is the only process that could produce thin layers of well-preserved iron oxides and clay platelets but is inconsistent with the exclusive affection of a small area in the middle of a stalagmite located in a large room on the walls of which no similar deposits are observed. Microscopic and chemical analysis of the calcite layer overlying the pigment do not detect these compounds, which discard the hypothesis that iron and clay rich mineral could originate from the speleothem. Furthermore, in terms of morphology, the paintings are markings characterized by a central area with high color density surrounded by an aureole that features a gradual reduction in the concentration of red matter (30). This pattern suggests an application of the paint by splattering as experimentally reproduced (40). Our results reject speculations that the panel might be the result of natural processes (23).

It has been proposed that some red stains found in Paleolithic decorated caves may be the consequence of accidental contact rather than due to a deliberate will of marking the caves' walls (41). In a narrow passage it is indeed possible that visitors wearing ochered clothes or body painting might inadvertently touch the walls, but, in the case of panel II.A.3, accidental staining can be excluded because the painted dome is in the middle of a very large chamber. In addition, the traces of color are found in both salient and recessed areas of the stalagmitic drapery. Indeed, some of the folds of this drapery where color can be seen are very deep and even beyond arm's reach; the only way pigment accretions could have reached some of the places where they can be observed is as drops and droplets blown via experimentally reproduced techniques (40).

The striking differences between the geological and the archaeological samples indicate that none of the cave deposits sampled in this study were used as sources for the pigments used to paint panel II.A.3. Furthermore, we observed neither substantial changes in the intensity and width of the hematite bands in the archaeological samples' Raman spectra nor features suggesting that goethite-rich raw materials could have been heat-treated. No support therefore exists for a hypothesis whereby goethite-rich material naturally present in the cave was heated to produce the paint on panel II.A.3. Our results strongly support that the Paleolithic artist(s) used Fe-rich lumps collected in geological formations from an as yet unknown source likely to be found outside the cave. Future research will need to survey local Fe-rich formations to establish whether the ochre used to paint the stalagmites is found nearby or comes from more distant sources.

The differences observed in the composition of the micro-samples from panel II.A.3 may be attributed to different causes. Assuming a single episode of artistic activity, conducted by single or multiple artists, slight differences in composition might be due to incomplete homogenization of the mixture applied to the draperies, or to different persons using pigment powders of diverse geological origin and produced with distinct techniques (e.g., because those persons belonged to different cultural traditions or came from different regions). Alternatively, such variations might be due to the fact that different draperies were painted at different times with a slightly different paint recipe or with a different source being used each time. These alternatives can be assessed against available dating evidence. The markings in Curtains 5 and 8 date to, respectively, >45.9 and >65.5 ka and, therefore, we cannot exclude that they represent a single painting episode taking place at some point in time before 65.5 ka ago. Such a hypothesis would be consistent with our finding that the markings were made in both curtains with a quite similar paint, composed of agglomerates of fine-grained platy Fe-rich minerals and clays. Curtain 6 was painted between 45.3 and 48.7 ka ago and therefore must represent a different incursion. Our finding that the Curtain 6 pigment differs from that applied to Curtain 8—it is also composed of clay-sized platy particles of hematite and aluminosilicates, but such particles are in the case of Curtain 6 scattered rather than forming agglomerates—suggests variation through time in the nature of the colorants used. The pigment in Curtain 9—composed of agglomerates of fine-grained Fe-rich minerals and clays associated with coarser K-rich micas—differs from the paint used in Curtains 5, 6 and 8 for the presence of isolated mica platelets and the lack of hydrated clay minerals.

To summarize, the dating evidence implies a minimum of two incursions. Based on Occam's Razor, similarity in composition is more likely to reflect appurtenance to the same painting event, and it is the opposite for dissimilarity. This is the more so since, in addition to their different compositions, the Ardales samples also feature different grain sizes, which is consistent, although in itself not demonstrative, of the use of a different paint. Thus, combining both lines of evidence (dating and composition), we can be certain that our samples represent a minimum of two painting events, and we can additionally suggest that the real number probably is of at least three, maybe even four. Additional precision on the number of painting episodes is currently not possible; it depends on the acquisition of additional dating evidence.

As the paintings are the result of recurrent addition, questions arise about it being a piece of art, subject to a sort of rejuvenation or restoration. Rejuvenation of motifs has been shown to occur in rock art, and ethnographic research has demonstrated that repainting is a common practice amongst traditional communities (42-49). Deliberate modification/restoration has also been proposed for Paleolithic cave art (e.g., the Pech Merle horses; (1)), and would seem to be ubiquitous in sites of the Spanish Levantine rock art tradition, which features panels that are thought to have been restored, altered, or expanded, either for ritual purposes or for the restoration of degraded figures (50-51). Examples include Cova Remigia, in the Valltorta-Gassulla area of Castellón, where total or partial repainting, addition of new elements or of another colour have been described, suggesting graphical and narrative re-appropriation (52). In Coves de la Saltadora (Castellón), the detection of a chemically distinct pigment led to interpret one figure as repainted (53). A clear example of two-color combination has been recently documented in Barranco de

Segovia (Letur), where the use of red over black was interpreted as enhancement of the original's value (54). Other examples of intentional overpainting that modified the nature and identity of the imagery are found at Ceja de Piezarrodilla (Albarracín), Cueva del Chopo (Obón), Canto Blanco (Jumilla), Prado de las Olivanas (Tormón), or Cueva de la Vieja (Albacete), amongst others (55-57).

In the case of panel II.A.3 of Cueva de Ardales, assessing whether image renovation occurred could be evaluated via the identification of distinct pigment layers separated by Ca/Mg-rich accretions. However, such a micro-stratigraphic study would entail significant damage to both painting and canvas, which is precluded by one of the key premises of our study — not to damage the paintings during our sampling work. It is unlikely, however, that such is the behavior reflected by our results. Ethnographic examples of rock art restoration show that this practice is often applied to abstract or figurative representations presenting characteristics (shape, details, color association) that fade out over time and that need to be rejuvenated in order to guarantee, on the one hand, the visual recognition of the diagnostic features identifying them as discrete, recognizable symbols by the concerned members of the group and, in some cases, to renew the symbolic link between the place and the representations. By restoring the painting, the group cultivates its ancestral link with a place charged with meaning and renews the art that binds the group to the place. Commitment to the land, however, is generally not the only reason for making rock art and one cannot directly use ethnographic examples to infer ancient cultural behavior. The techniques used to apply the paint in panel II.A.3 of Cueva de Ardales and the resulting markings do not allow the recognition of discrete features. This suggests that cultivating the link with the place, rather than associating it with a particular representation, must have been the main reason for marking the stalagmites. Restoration of an image makes sense when the image itself carries symbolic information and is the focus of the artistic activity but what we see at Cueva de Ardales is distinct: the carrier of the symbolic information is, in this case, the large stalagmitic dome harboring the panel, not the panel itself. Put another way, treating the dome as the canvas is useful shorthand but should not be taken to imply that this large formation is no more than a convenient surface used to appose markings and that these markings are in and of themselves the repositories of symbolic information irrespective of where they were made. Instead, the dome is the symbol and the paintings are there to mark it as such, not the other way around. In this context, recurrent marking is not akin to the restoration or modification of a pre-existing motif to maintain, enhance or modify its meaning. Rather it must stand instead for the renewed assertion of the symbolic value of the dome itself.

Based on the results of this study, we hypothesize that panel II.A.3 is not “art” in the narrow sense of the word —“the making of objects, images, music, etc. that are beautiful or that express feelings, or “the activity of painting, drawing, and making sculpture” (<https://dictionary.cambridge.org/dictionary/english/art>)— but rather the result of graphic behaviors intent on perpetuating the symbolic significance of a space. The evidence from Bruniquel cave, in France, shows that Middle Paleolithic Neanderthals were involved in symbolic activities taking place deep inside the karst that involved the intentional modification of speleothems and their use in the construction of complex arrangements (8). Evidence from Cueva de Ardales confirms the view that speleothems played a fundamental role in the symbolic systems of some Neanderthal communities. Paint-marking using splattered red pigment on such large, imposing domes as that which panel II.A.3 decorates can thus be seen as a development deeply rooted in a long-standing tradition, of which other examples exist at Ardales. Rock art may therefore have begun in Europe as a form of place marking, with the Middle Paleolithic hand stencils and geometric signs seen at other Iberian cave sites (Maltravieso, La Pasiega, El Castillo and Gorham's Cave; (18, 20, 40, 58)) representing much the same type of symbolic behavior. We predict that more markings bearing similarities with those from Cueva de Ardales will be identified in the future in the Iberian Peninsula and dated to the Middle Paleolithic. Although Upper Paleolithic cave art is technically and thematically more complex, signs and hand stencils play a prominent role in it (59). It is possible that markings such as those identified at Cueva de Ardales and other Iberian sites represent the

prolegomena of a long process during which new needs linked to social complexification have triggered the development of novel symbolic traditions supported by the development of more varied and innovative technical practices.

Materials and Methods

Permission to sample the paintings was granted by the Department of Culture of the Regional Government of Andalusia. Subsequent to visual assessment of the pictorial representations, five samples were collected. Micro-agglomerates of pigments were recovered by scratching the outer surface at a unique spot. We paid special attention not to leave visible traces of the sampling. Photographs were taken before and after the extraction procedure to precisely document the location of the sampled area. To assess whether Fe-rich deposits present in the cave could have provided the pigmentary material sampled in panel II.A.3, eight geological samples of natural coloring materials were collected from different Fe-rich deposits. The pigment and geological samples were submitted to a variety of microscopic, elemental, and mineralogical analyses. More information on methods is given in (*SI Appendix, Materials and Methods*, Fig. S1-S2; Table S1-S2).

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Figure captions

Figure 1. Cueva de Ardales, Spain. Geographic location and plan of the cave. The position of recently excavated areas (zones 2, 3, 5), panel II.A.3 where samples P-ARD-02 to 06 were

extracted, and the areas where the geological samples were collected (stars) are indicated. Europe map modified from https://d-maps.com/carte.php?num_car=2233&lang=es

Figure 2. Panel II.A.3 in *Sala de las Estrellas*. A) General view of the speleothem with the location of one sampled area (P-ARD-06). The square indicates the area enlarged in B; B) close-up view of the drapery hosting most of the red stains with arrows indicating the sampled areas. Scale bars = 50 cm.

Figure 3. Close-up view of red stains, microphotographs and selected SEM images in BSE mode of red micro-samples extracted from panel II.A.3. A: curtain 9; B, C: curtain 8; D: curtain 6; E: curtain 5; F, K: P-ARD-02; G, L: P-ARD-03; H, M: P-ARD-04; I, N: P-ARD-05; J, O: P-ARD-06. Arrows in A to E indicate the extraction spot. Scale bars in A to E = 1 cm.