The symbolic role of the underground world among Middle Paleolithic Neanderthals

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Cueva de Ardales in 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 indicates that the pigments used in the paintings do not come from the outcrops of colorant material known 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.

cave art | symbolism | pigment | spectroscopic analyses | Iberian Peninsula

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 in 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 nonfigurative 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 in refs. 28–31.

One of the early Iberian sites is Cueva de Ardales (Fig. 1 and *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 G.C.W. and J.R.-M. 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, and >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* and Fig. S1). It has been suggested (23) that the pigment could represent natural staining, which macroscopic observation does not support (30). Secondly, we investigate whether

ANTHROPOLOGY

Significance

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 do not come from the outcrops of colorant material known 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.

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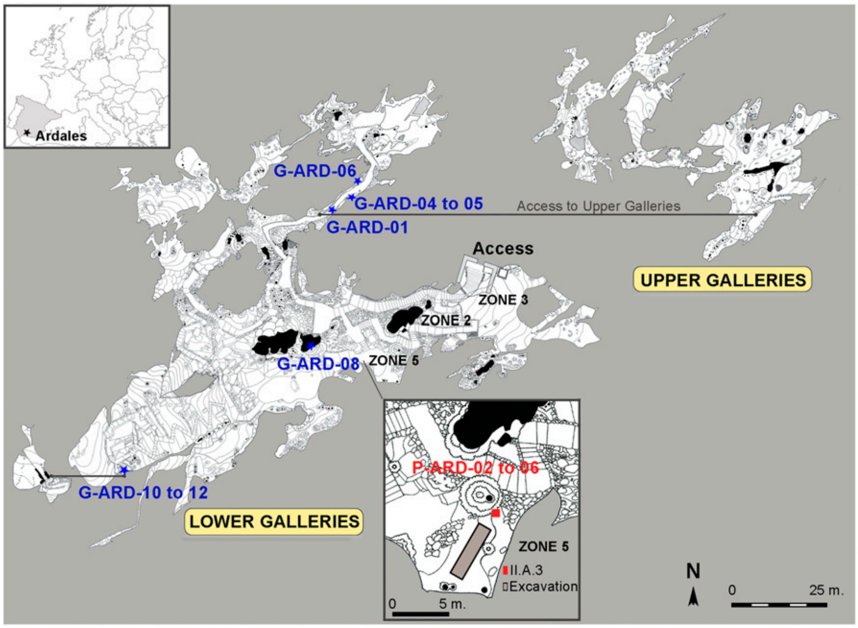

Fig. 1. Cueva de Ardales, Spain. Geographic location and plan of the cave. The position of recently excavated areas (zones 2, 3, and 5), panel II.A.3 where samples P-ARD-02 through 06 were extracted, and the areas where the geological samples were collected (stars) are indicated. Europe map is modified from https://d-maps.com/carte.php?num_car=2233&lang=es.

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* and Fig. S2) to see whether 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 1,577 m long and features two superimposed levels: the Lower and the Upper Galleries. The site was discovered in 1821 after an earthquake reopened 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 nonfigurative engravings and paintings grouped into 252 panels (33). Most abstract red paintings are on speleothems and located near the entrance rather than at the back (i.e., they are found in those parts of the cave in which archaeological excavation has corroborated broadly coeval use of the space by Middle Paleolithic Neanderthals; ref. 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 cave setting, research history, art and human use is provided in *SI Appendix*, *Archaeological Context*.

Results

Microsamples 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 and *SI Appendix*, *Results*). Analyses also detected traces of phosphor and sulfur that may come from small amounts of sulfates and phosphates. SEM observation of these microsamples indicates a mineral origin, since none of them feature 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 microsamples are also consistent with a mineral habit. Closer analysis reveals interesting textural and compositional differences (*SI Appendix*, *Results*, Fig. 3, and *SI Appendix*, Figs. S3–S7). Samples from Curtains 5 (P-ARD-06) and 8 (P-ARD-03 and P-ARD-04) are composed of tightly bound agglomerates of submicrometric to micrometric platy Fe-rich minerals and clays, while in Curtain 6 (P-ARD-05), the hematite and aluminosilicate particles appear not as agglomerates but 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 to 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

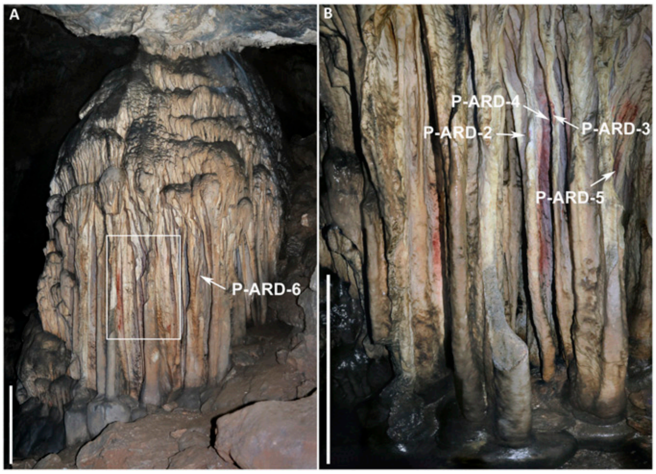

Fig. 2. Panel II.A.3 in Sala de las Estrellas. (*A*) General view of the speleothem with the location of one sampled area (P-ARD-06). The square indicates the area enlarged in *B*. (*B*) Close-up view of the drapery hosting most of the red stains with arrows indicating the sampled areas. Scale bars,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

Table 1. Results of elemental and mineralogical analyses on red microsamples from panel II.A.3, Cueva de Ardales, Spain

			SEM observations and EDS analyses						
						Elemental composition			
Sample	Curtain	Age constrains (ka) (20)	*N*	Size (μm)	Shape	Major elements	Minor elements	XRD and RS	Unresolved RS bands (cm^{-1})
P-ARD-02	9	–	7	1–3	Agglom. of platy and nonfacetted cr.	Fe, Ca, Si	Al	hem, cal, arg, qz	
			4	15–30	Platy	Si, Fe, C, Al, K, Ca, Mg, Ti, V, Ni			
			4	1–3	Agglom. of platy and nonfacetted cr.	Fe, Ca, C, Si	Al		
P-ARD-03	8	>65.5	5	1–2.5	Agglom. of platy and nonfacetted cr.	Fe, C, Ca, Si	Al	hem, cal, qz, arg	3,272 (br), 3,348 (br)
			6	1–2.5	Agglom. of platy and nonfacetted cr.	Fe, Ca, Si	Al, Ti		
P-ARD-04	8	>65.5	5	2.5	platy	Fe, Ca, Si	K, Al, Mg	hem, cal, arg	453, 1,092 (w), 1,144, 1,576 (br), 1,717 (w), 3,272 (br), 3,348 (br)
			7	1–1.5	Agglom. of platy and nonfacetted cr.	Fe, Ca, P, Si, C	Al, K, S, Mg		
P-ARD-05	6	Between 45.3 and 48.7	7	2–3.5	platy	Fe, Ca, C, Si, P	S, Al, K, Mg	hem, cal, C, arg	911 (w), 927 (w)
			2	1–1.5	Agglom. of platy and nonfacetted cr.	Fe, Ca, C	Si, K, Al		
P-ARD-06	5	>45.9	13	0.9	Agglom. of platy and nonfacetted cr.	Fe, Ca, Si, C	Al, P, K, Mg	hem, cal	3,272 (br), 3,348 (br)
			2	0.9	Agglom. of platy and nonfacetted cr.	Ca, Fe, C	K, P		458 (w)

"*N*" records the number of times each feature was observed; cr: crystals; hem: hematite; cal: calcite; arg: aragonite; qz: quartz; C: amorphous carbon. Unresolved RS bands around 900 could be explained by the presence of sulfates. RS bands in the 1,000 to 1,300 cm^{-1} region may be caused by rare earth elements luminescence. Broad RS bands in the range 3,200 to 3,500 cm^{-1} may be due to the stretching modes of water molecules in hydrous minerals. RS bands: br, broad; w, weak; s, strong. SEM, scanning electron microscopy; EDS, energy dispersive spectroscopy; XRD, X-ray diffraction; RS, Raman spectroscopy.

in the form of agglomerates. In addition, Curtain 6 (P-ARD-05) reveals the presence of amorphous carbon (*SI Appendix*, *Results* and 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*, Figs. S2 and S11, Table S2, and *SI Appendix*, *Results*). SEM analysis of representative samples makes it clear that these materials share no similarities with the samples from panel II.A.3 (*SI Appendix*, *Results*, Fig. S12, and Table S3). The X-ray diffraction analysis (*SI Appendix*, *Results* and 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 stalagmites' red staining, but neither is comparable to the panel's samples; 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 sulfate spherules, and K-rich micas (Si, K, Al, Ca, Ti, and Mg), while sample G-ARD-11 consists of clusters of 2-μm disk-shaped Fe-rich crystals in a foliated matrix of clays (Si, K, Al, Mg, Ti, Ca, and Mn).

Speleothem Sample. Microscopic end elemental analyses performed on the speleothem layer covering a stain from Curtain 9 (*SI Appendix*, *Results* and Fig. S13) show that it is mainly composed of low-magnesium calcite. Small amounts of aluminum, probably from an Al-rich hydroxide, are also present. No iron or clay minerals were detected.

Discussion

The analysis of the microsamples 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 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), either. 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. Ruling out the hypothesis that the presence of iron- and clay-rich minerals could be related to speleothem formation, the analyzed calcite sample includes neither, and any iron-rich particles present in drip water would in any case not lead to the formation of the loose hematite and clay platelets seen in the pigments of panel II.A.3. Weathering of the bedrock 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. Furthermore, in terms of morphology, the markings are 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

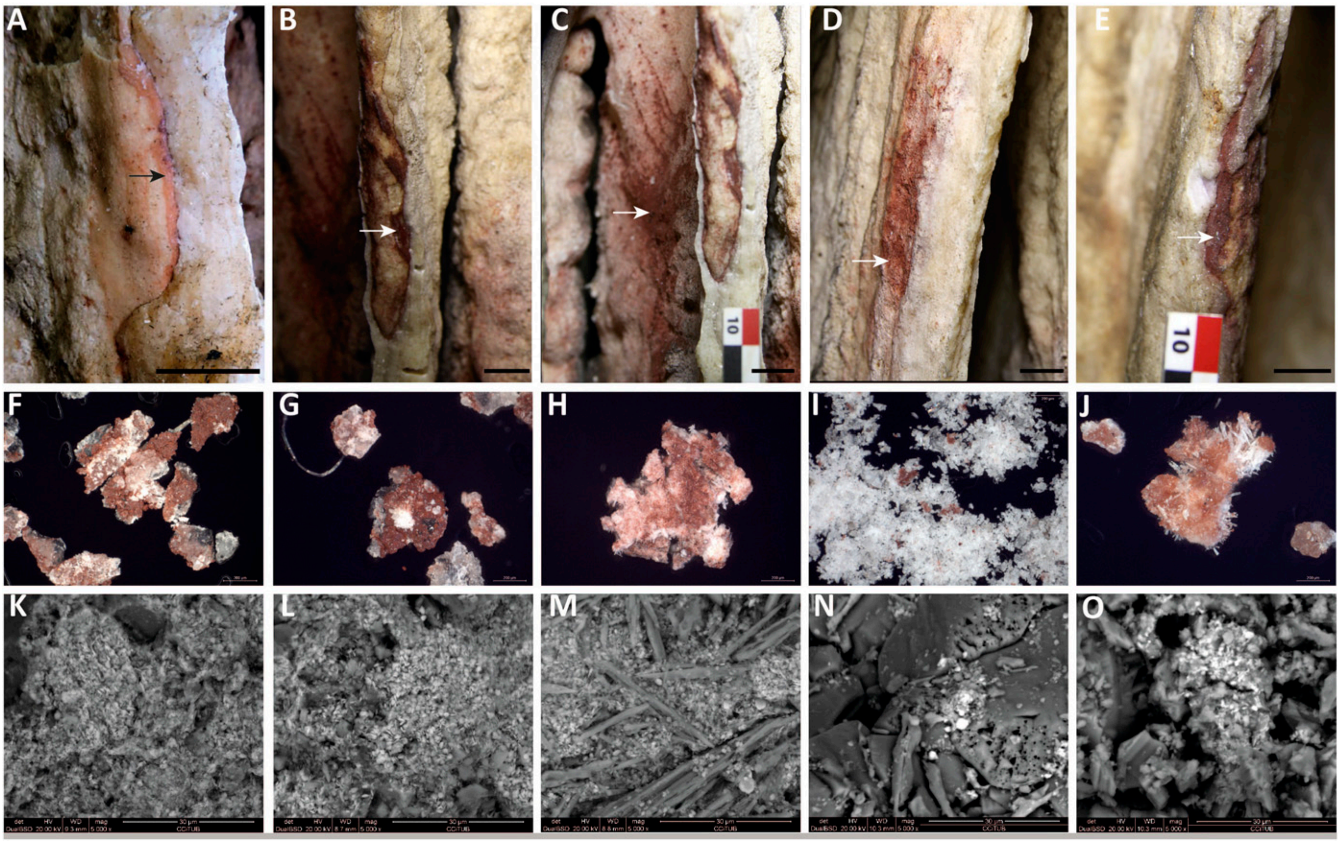

Fig. 3. Close-up view of red stains (*A*–*E*), microphotographs (*F*–*J*), and selected SEM images in backscattered electron mode (*K*–*O*) of red microsamples extracted from panel II.A.3. (*A*) Curtain 9, (*B* and *C*) Curtain 8, (*D*) Curtain 6, (*E*) Curtain 5, (*F* and *K*) P-ARD-02, (*G* and *L*) P-ARD-03, (*H* and *M*) P-ARD-04, (*I* and *N*) P-ARD-05, and (*J* and *O*) P-ARD-06. Arrows in *A*–*E* indicate the extraction spot. Scale bars in *A*–*E*, 1 cm.

by splattering as experimentally reproduced (40). Our results are inconsistent with speculations that panel II.A.3 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 intention 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 Raman spectra of the archaeological samples 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 would have been 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 microsamples 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 travelled to the site 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 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 is probably at least three, or maybe even four. Precising the number of painting episodes further must await the acquisition of more 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 among traditional communities (42–49). Deliberate modification and/or restoration has also been proposed for Paleolithic cave art (e.g., the Pech Merle horses; ref. 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 and addition of new elements or of another color have been described, suggesting graphical and narrative reappropriation (52). In Coves de la Saltadora in Castellón, the detection of a chemically distinct pigment led to interpreting one figure as repainted (53). A clear example of two-color combination has been recently documented in Barranco de Segovia in 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 in Albarracín, Cueva del Chopo in Obón, Canto Blanco in Jumilla, Prado de las Olivanas in Tormón, and Cueva de la Vieja in Albacete, among 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, but such a microstratigraphic 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.

Even though ethnographic analogy cannot be relied upon to make direct inferences about ancient behaviors, it is nonetheless useful in illustrating the range of possibilities and helping with the interpretation of the archaeological evidence. Ethnographic examples of rock art restoration (42–49) show that this practice is often applied to abstract or figurative representations presenting characteristics (shape, details, color association) that fade out over time. The rejuvenation guarantees the visual recognition by the concerned members of the group of the diagnostic features identifying the representations as discrete, recognizable symbols and, in some cases, are also intent on renewing the symbolic link between the place and the people (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).

The techniques used to apply the paint in panel II.A.3 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: It would seem to us that 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 made. Instead, we believe that 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 preexisting motif to maintain, enhance, or modify its meaning. Rather, it must stand instead for the renewed assertion of the symbolic value of the place or of the "canvas" 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"—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 included the intentional modification of speleothems and their use in the construction of complex arrangements (8). The evidence from Cueva de Ardales supports the notion 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; ref. 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 emergence 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. Microagglomerates 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 colorant material sampled in panel II.A.3, eight representative 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*, Figs. S1 and S2, and Tables S1 and S2).

Data Availability. All study data are included in the article and/or *SI Appendix*.

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