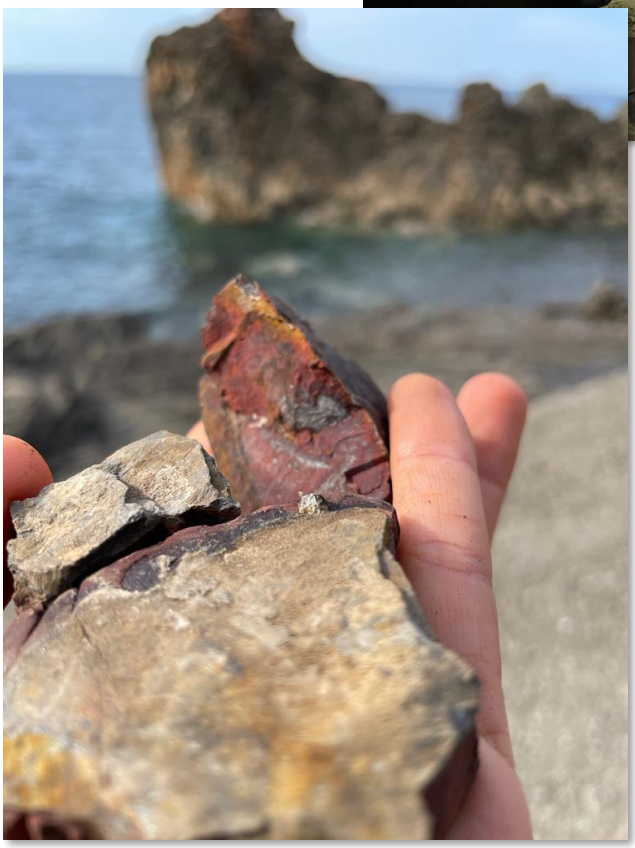
“ Taste of Colors ” in the Cantabrian mountain range during prehistory (Gravettian - Magdalenian)

Claire Chanteraud, Diego Garate, David Stalla, Alexandra Kuo, Brandi L. Macdonald

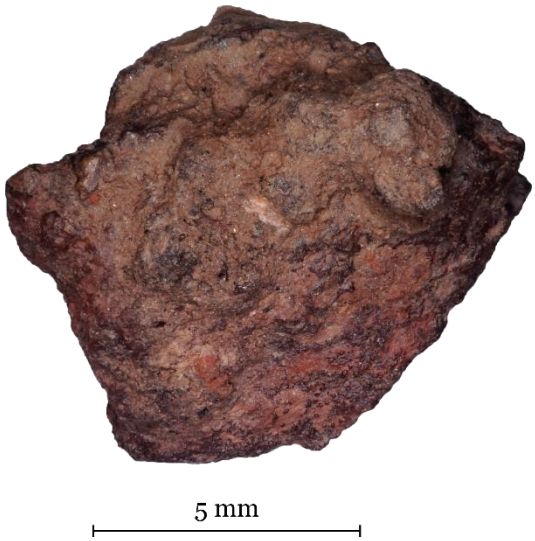
« Taste of Colors » in the Cantabrian mountain range during prehistory (Gravettian - Magdalenian)

Claire Chanteraud, *Diego Garate, David Stalla, Alexa Kuo, Brandi Lee MacDonald*

Excavation & in situ analysis at Altxerri Cave
(Photo : Iñaki Intxaurbe)



Geological iron rich rocks from Artmintza
(Photo : Claire Chanteraud)



Colouring remains from Morgotà Cave
(Photo: Claire Chanteraud)

Report on the analysis and observations of the colouring remains in the caves of Morgota, Ondaro, Lumentxa and Armiña

Claire Chanteraud, Diego Garate, Alexa Kuo, David Stalla, Brandi L. MacDonald,

In the first half of 2022 (January-June) a collection of colouring remains was provided by the Arkaeologi Museoa de Bilabo under the supervision of the Basque Government. Half of the collections of colouring remains from the caves of Morgota, Lumentxa, Ondaro and Armiña were sent by UPS, covered by a transport insurance of 10k€ and an import permit issued by the US Department of Agriculture, to the Archaeometry Laboratory of the Missouri University research reactor (MURR), the sponsor of Dr. Claire Chanteraud's post-doctorate.

The laboratory and the University of Missouri offer a wide range of analytical tools to characterise archaeological colouring materials from macroscopic to microscopic and geochemical scales in a non-invasive and non-destructive (SEM-EDS, Raman, pRaman, XRF, DRX) or µ-invasive way (LA-ICP-MS).

4.3.1 Macroscopic observations

Firstly, the objects were weighed (Table 01) with a high-precision balance and then observed under a stereomiscrope in order to determine their surface petrographic characteristics and to classify them into lithological groups. The observation of the blocks of material from the different sites allowed us to identify nine lithological types (Table 02)

Table 01: *Weighing table of colouring remains for the four sites: Morgota (AGT), Ondaro (OND), Armiéna (Ar) et Lumentxa (L)*

Object name	Weight (mg)	Object name	Weight (mg)	Object name	Weight (mg)
AGT.5A.ALM.3.1.19	236	**OND.22J.SLEC.9.5.458a**	62,985	**AR.W15.1.9.18-33b**	285,48
AGT.5A.0ALM.6.2.11	5024,1	**OND.22J.SLEC.9.5.458b**	62,985	**AR.W15.1.9.18-33c**	285,48
AGT.5A.0ALM.3.2.164a	315	**OND.22J.SLEC.9.5.458c**	279,87	**AR.W15.1.9.18-33d**	285,48
AGT.5A.0ALM.3.2.164b	4,125	**OND.22J.SLEC.9.5.458d**	20,0175	**AR.W15.1.9.18-33e**	285,48
AGT.5A.0ALM.3.2.164c	4,125	**OND.22J.SLEC.9.5.458e**	20,0175	**AR.W15.1.9.18-33f**	285,48
AGT.5A.0ALM.3.2.164d	4,125	**OND.22J.SLEC.9.5.458f**	20,0175	**AR.W15.1.9.18-33g**	285,48
AGT.5A.0ALM.3.2.164e	4,125	**OND.22J.SLEC.9.5.458g**	20,0175	**AR.W15.1.9.18-33h**	285,48
AGT.5A.0ALM.6.2.173a	66,61	**OND.22J.SLEC.9.5.413**	9660	**AR.W15.1.9.18-33i**	285,48
AGT.5A.0ALM.6.2.173b	23,42	**OND.22K.SLEC.7.4.395**	386,65	**AR.W15.1.9.18-33j**	285,48
AGT.5A.0ALM.6.2.173c	30,962			**AR.W15.1.9.18-33k**	285,48
AGT.5A.0ALM.6.2.173d	30,962			**AR.W15.1.9.18-33l**	285,48
AGT.5A.0ALM.6.2.173e	30,962	**AR.W15.3.10.1**	120,86	**AR.W15.1.9.18-33m**	285,48
AGT.5A.0ALM.6.2.173f	30,962	**AR.V15.2.4.3.8a**	303,19	**AR.W15.1.9.18-33n**	285,48
AGT.5A.0ALM.6.2.173g	30,962	**AR.V15.2.4.3.8b**	129,2375	**AR.W15.1.9.18-33o**	205,33
AGT.5A.0ALM.6.2.173h/l/m	227,960	**AR.V15.2.4.3.8c**	129,2375	**AR.W15.1.7.1-5a**	421,44
AGT.5A.0ALM.6.2.173i	20,71	**AR.V15.2.4.3.8d**	129,2375	**AR.W15.1.7.1-5b**	421,44
AGT.5A.0ALM.6.2.173j	87,63	**AR.V15.2.4.3.8e**	129,2375	**AR.W15.1.7.1-5c**	421,44

AGT.5A.0ALM.6.2.173k	48,57	**AR.V15.2.4.3.8f**	574,44	**AR.V15.1.7.1-2a**	530,4
AGT.5A.0ALM.6.2.173l	113,98	**AR.V15.2.4.3.8g**	633,93	**AR.V15.1.7.1-2b**	120,53
AGT.5A.0ALM.6.2.173m	113,98	**AR.W15.1.15.1**	307,11	**AR.W15.1.13.3**	1204,19
AGT.5B.AM0.8.4.359a	94,5666667	**AR.V15.4.6.1**	131,32		
AGT.5B.AM0.8.4.359b	94,5666667	**AR.W15.1.5.3**	896,17		
AGT.5B.AM0.8.4.359c	94,5666667	**AR.W15.1.9.5-17a**	598,06	**L.6B.145.01**	2810
AGT.5B.AM0.8.4.359d	16,28	**AR.W15.1.9.5-17b**	209,687	**L.8F.170.01**	24870
AGT.5B.AM0.8.4.359e	20,5675	**AR.W15.1.9.5-17c**	209,687	**L.6B.145.03**	4900
AGT.5B.AM0.8.4.359f	20,5675	**AR.W15.1.9.5-17d**	209,687	**L.6B.145.06**	2060
AGT.5B.AM0.8.4.359g	20,5675	**AR.W15.1.9.5-17e**	209,687	**L.6B.145.04**	10390
AGT.5B.AM0.8.4.359h	20,5675	**AR.W15.1.9.5-17f**	209,687	**L.8F.170.03a**	3000
AGT.5B.AM0.8.3.315a	536,54	**AR.W15.1.9.5-17g**	209,687	**L.8F.170.03b**	720
AGT.5B.AM0.8.3.315b	71,01	**AR.W15.1.9.5-17h**	209,687	**L.6B.145.07**	49140
AGT.5B.AM0.8.3.315c	7,54	**AR.W15.1.9.5-17i**	209,687	**L.5E.90.01**	73140
AGT.5B.0ALM.2.2.226a	12,49	**AR.W15.1.9.5-17j**	209,687	**L.6B.145.02**	590
AGT.5B.0ALM.2.2.226b	191,805	**AR.W15.1.9.5-17k**	209,687	**L.12E.11.01**	12980
AGT.5B.0ALM.2.2.226c	191,805	**AR.W15.1.9.5-17l**	267,01	**L.11G.230.01**	16590
AGT.6A.ALM.3.1.5	654	**AR.W15.1.9.5-17m**	151,04	**L.12B.120.02**	2180
AGT.6B.0ALM.1.2.10	273,2	**AR.W15.4.11.1.3a**	239,926667	**L.8F.170.02**	12210
AGT.5A.0ALM.9.2.192a	34,19	**AR.W15.4.11.1.3b**	239,926667		
AGT.5A.0ALM.9.2.192b	60,51	**AR.W15.4.11.1.3c**	239,926667		
AGT.5A.0ALM.9.2.192c	34,23	**AR.W15.3.9.1-4a**	472,05		
AGT.5A.0ALM.9.2.192d	30,09	**AR.W15.3.9.1-4b**	762,38		
AGT.5B.0ALM.4.2.27a	1675,74	**AR.V15.4.4.1-2a**	2367,4		
AGT.5B.0ALM.4.2.27b	81,07	**AR.V15.4.4.1-2b**	285,55		
AGT.5B.0ALM.2.2.10	4963	**AR.W15.3.7.1-12a**	398,305		
AGT.5B.AM0.8.6.429a	113,34	**AR.W15.3.7.1-12b**	398,305		
AGT.5B.AM0.8.6.429b	113,34	**AR.W15.3.7.1-12c**	966,165		
AGT.5B.AM0.8.5.407a	23,42	**AR.W15.3.7.1-12d**	966,165		
AGT.5B.AM0.8.5.407b	23,42	**AR.W15.3.7.1-12e**	966,165		
AGT.5B.AM0.8.5.407c	23,42	**AR.W15.3.7.1-12e**	966,165		
AGT.5B.AM0.8.5.407d	23,42	**AR.W15.1.8.10-18a**	222,262		
AGT.5B.AM0.8.5.407e	23,42	**AR.W15.1.8.10-18b**	222,262		
AGT.5B.AM0.8.5.407f	23,42	**AR.W15.1.8.10-18c**	222,262		
AGT.5B.AM0.8.5.407g	23,42	**AR.W15.1.8.10-18d**	222,262		
AGT.5B.AM0.8.5.407h	23,42	**AR.W15.1.8.10-18e**	222,262		
AGT.5B.AM0.8.5.407i	23,42	**AR.W15.1.8.10-18f**	683,02		
AGT.5B.AM0.8.5.407j	23,42	**AR.W15.1.8.10-18g**	209,04		
AGT.5B.AM0.8.5.407k	23,42	**AR.W15.1.8.10-18h**	581,95		
AGT.5B.AM0.8.5.407l	23,42	**AR.W15.1.9.18-33a**	285,48		

Table 02: *Surface petrographic characteristics of the lithologies identified (macroscopic scale) for the colouring remains of the Lumentxa, Morgota, Ondaro and Armiña caves. Some images are missing because they are still being acquired.*

LITHOLOGIE	DESCRIPTION	ILLUSTRATION
A	Cohesive purple iron-clay material with an oily to metallic sheen, hard	

B	Cohesive orange-red clay-ferruginous material, soft mat	
C	Soft cohesive clay-iron material with a greasy to metallic sheen showing desiccation cracks and botrioidal minerals	
D	Pure purple hematite with a metallic sheen, hard	
E	Red to orange siltstone/sandstone	*No objects with these characteristics in the first part of the collection*
F	Red to orange iron-clay bedded concretion hematite + goethite cohesive hard	

G	Soft orange-pink porous silt clay	
H	Soft, powdery, purplish-red clay-ferruginous material with a matt, dry appearance	*Image in acquisition*
I	Yellow consolidated soft matte sand	*Image in acquisition*

By cross-referencing the weighing, i.e. the quantity of material, with the lithological classification it was possible to draw graphs representing the quantity of material (mass in grams) of these classes in the archaeological levels. This makes it possible to assess the changes in the selection of raw material over time at the four sites studied (Figure 01 to 04).

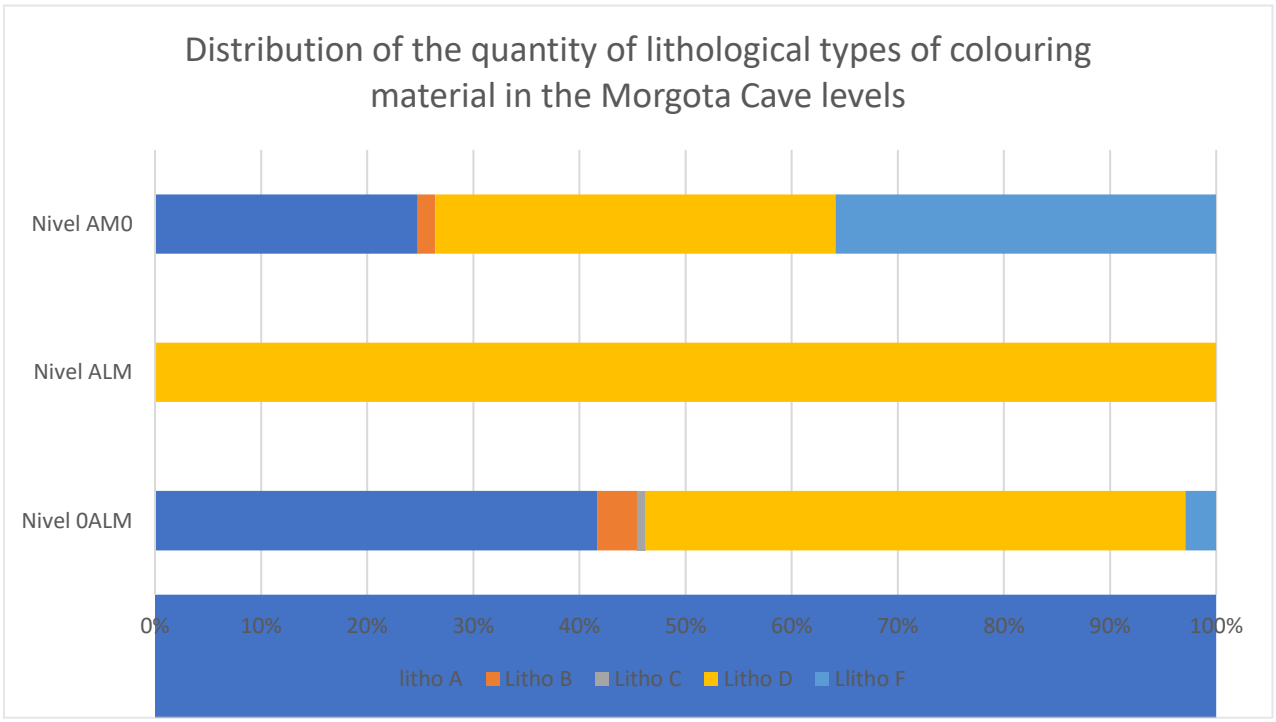


***Figure 01** : lithological determination of the remains of the Morgota cave and representation of the quantity of material per archaeological level*

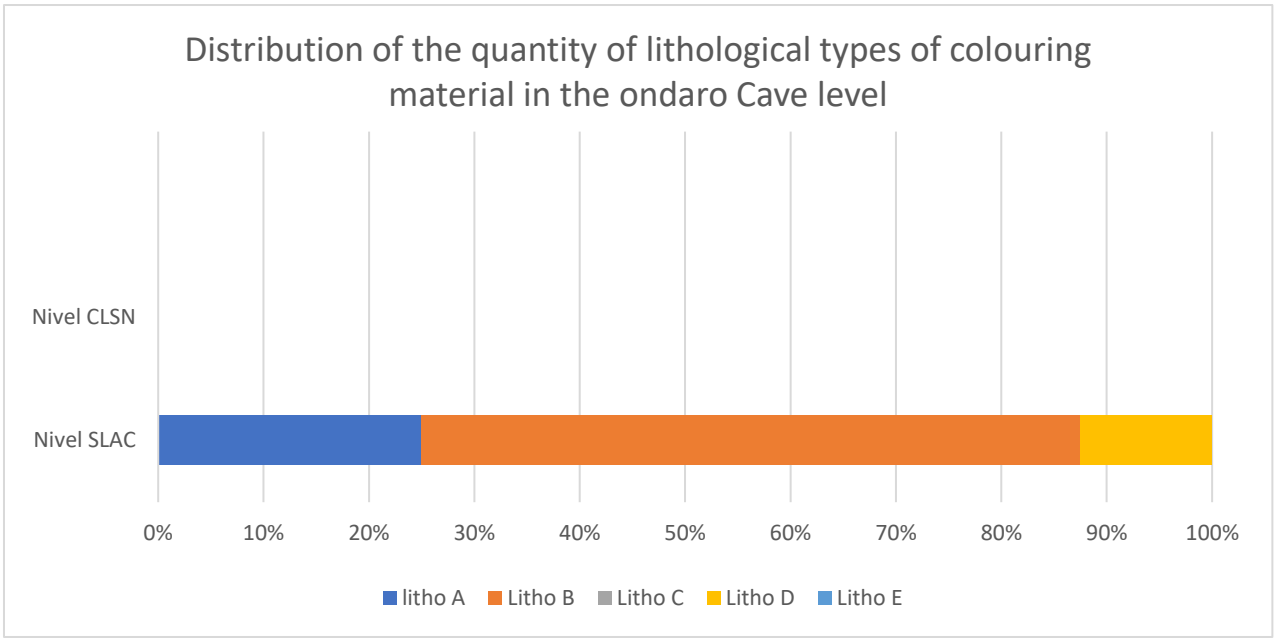


***Figure 02** : lithological determination of the remains of the Ondaro cave and representation of the quantity of material per archaeological level*

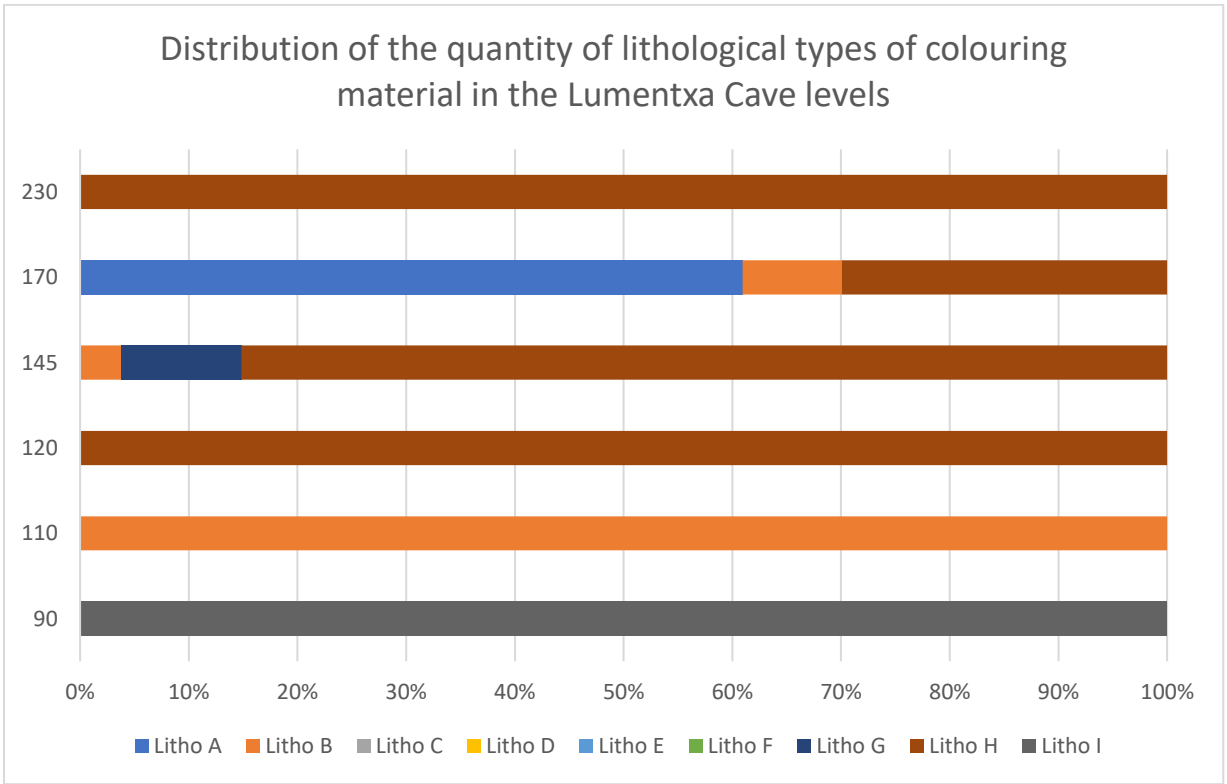


***Figure 03** : lithological determination of the remains in the Lumentxa cave and representation of the quantity of material per archaeological level*

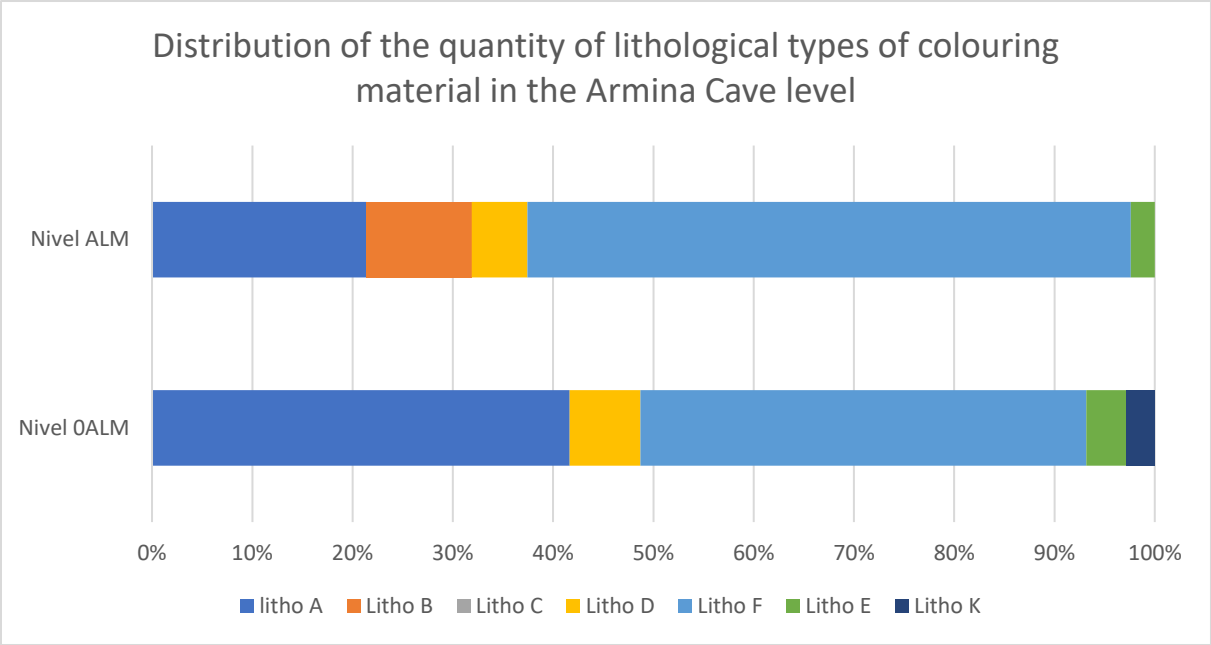


***Figure 04** : lithological determination of the remains of the Armiña cave and representation of the quantity of material per archaeological level*

The first macroscopic observations show a diversity of materials selected and exploited in the four ornate sites. Several lithological types, such as lithology A and B, are present in all the sites, which may reflect a selection of the same raw materials: similar taste? Furthermore, the study of the quantity of material per stratigraphic unit in the different sites does not show any evolution in the selection strategy over time or comparable between sites. This is due to 1/ an insufficient global quantity of objects for certain sites (Ondaro, Lumentxa, Armiña), 2/ a study based on half of the collections, 3/ insufficient stratigraphic data (Lumentxa). The study of the rest of the collection will enable us to clarify these observations by occupation level for all the sites.

The lithological determination has enabled us to classify the colouring remains into groups with petrographic characteristics that can be appreciated by the eye (obvious and appreciable characteristics in prehistory). Observation on a microscopic scale provides a precision of these characteristics in order to pursue the scientific objective of researching the origin of the materials.

4.3.2 Observations en microscopie électronique

Preparation

In order to link the raw materials mined at the archaeological sites with potential sources of supply, the micromorphologies of iron oxide and associated minerals in the dyes were characterised. This allows the modes of formation of the exploited rocks to be determined and compatible geological formations to be selected for comparison. Energy dispersive electron microscopy (EDSEM) observations were made at the University of Missouri Microscopy Laboratory (EMCore).

For these observations, the residue from the bottom of the bag was collected and placed on carbon tape, thus allowing the remains to be completely preserved. When there was no residue available in the bottom of the bags, the objects were introduced into the microscope chamber without preparation (simply placed on the analysis stage) (Figure 05).

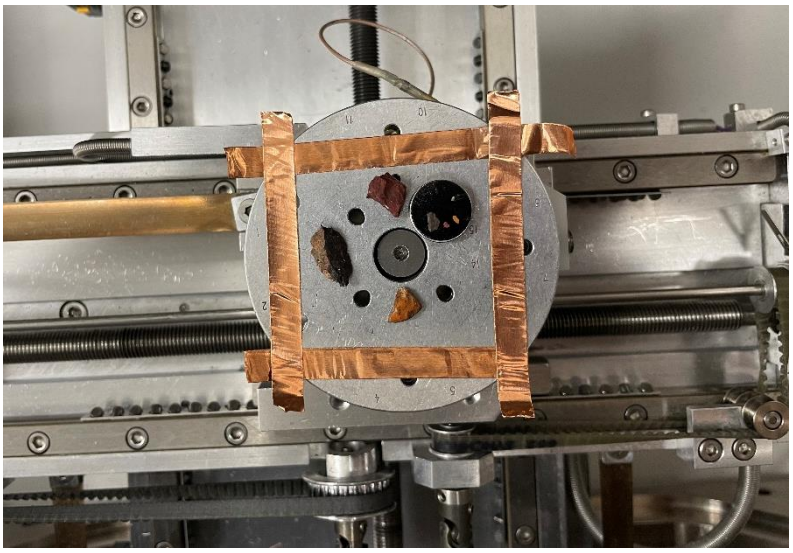
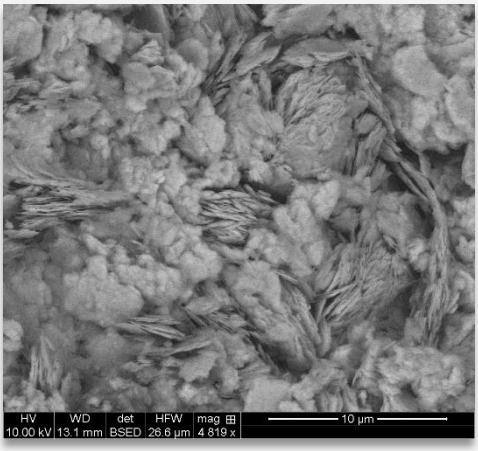
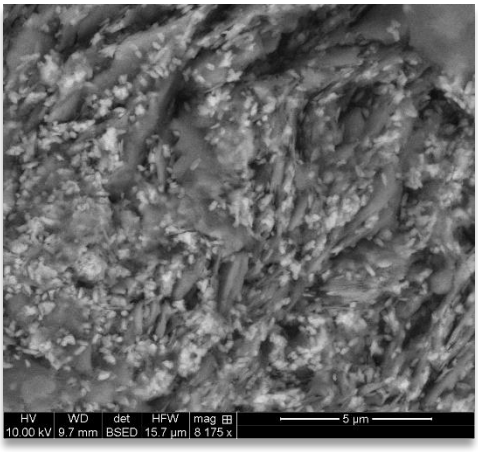
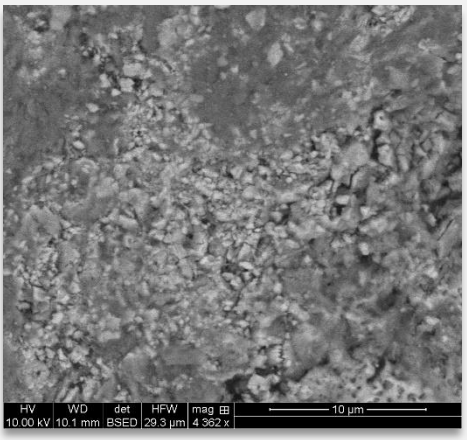
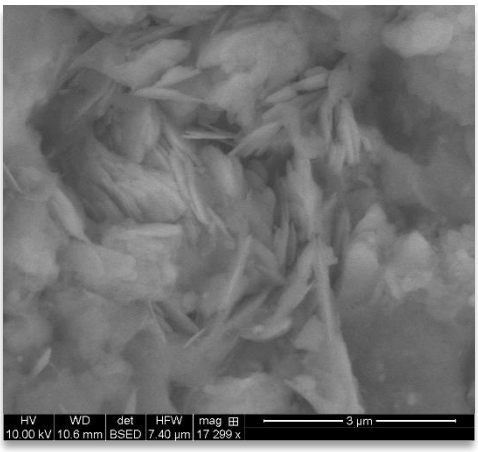


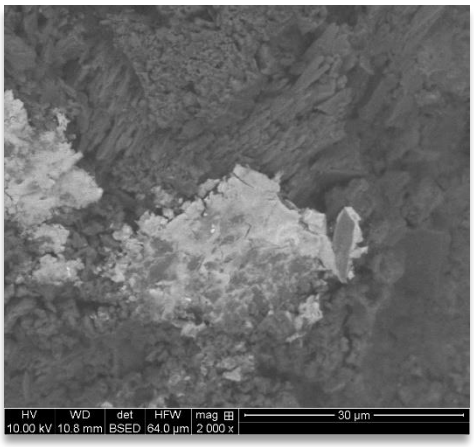
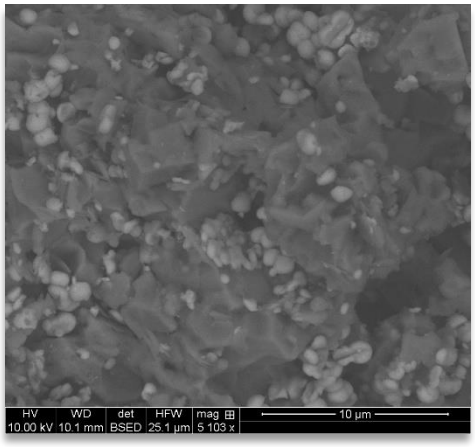
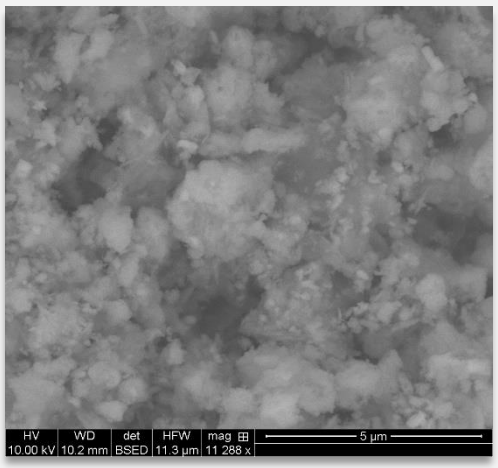
Figure 05: *Vue d'échantillons de références géologique (Pigmentothèque), de vestiges archéologiques non préparés simplement déposer sur le carrousel d'analyse et de résidus de fond de sachet fixés sur un plot métallique par scotch carbone avant introduction dans la chambre de microscope.*

Observation results

Table 03: *Surface characteristics of the lithologies identified on a microscopic scale for the colouring remains of the Lumentxa, Morgota, Ondaro and Armiña caves. Some images are missing because they are still beinin acquisition. BSE: Backscatering Electron = surface chemical contrast image, SE: Scanning Electron = surface topography image.*

LITHOLOGIE	DESCRIPTION	ILLUSTRATION
A	Macro-scale: Cohesive purple iron-clay material with an oily to metallic sheen, hard Micro-scale: micrometric sized zoned hematite plates in an Al/si matrix	*Image BSE sur AR.W15.1.8.10-18a* 

B	Macro-scale: Cohesive orange-red clay-ferruginous material, soft mat Micro-scale: iron oxide particles (hematite or goethite) smaller than one micrometer in an Al/Si clay matrix	*Image BSE sur AGT.5A.0ALM.3.2.164c* 
C	Macro-scale: Soft cohesive clay-iron material with a greasy to metallic sheen showing desiccation cracks and botrioidal minerals Micro-scale: iron oxide present in zonation angular morphology micrometre-sized particles	*Image BSE sur AGT.5A.0ALM.9.2.192a* 
D	Macro-scale: Pure purple hematite with a metallic sheen, hard Micro-scale: Hematite in 1µm thick plates	*Image BSE sur AGT.5B.0ALM.4.2.27a* 
E	Red to orange siltstone/sandstone	-

F	Macro-scale: Red to orange iron-clay bedded concretion hematite + goethite cohesive hard Micro-scale: morphology of the iron oxides undetermined, presence of crystals enriched in Uranium.	*Image BSE sur AGT.5B.AM0.8.4.359a* 
G	Soft orange-pink porous silt clay	*In acquisition*
H	Macro-scale: Soft, powdery, purplish-red clay-ferruginous material with a matt, dry appearance Micro-scale: Iron oxide as a spherical concretion deposited on quartz crystal	*Image BSE sur L.6B.145.07* 
I	Macro-scale: Yellow consolidated soft matte sand Micro-scqle: iron oxides of undetermined morphology in association with silicate minerals and aluminosilicates	*Image BSE sur L.5E.90.01* 

Scanning electron microscopy (SEM) observation allows us to observe the shapes of the minerals and their association in the material. These characteristics are directly conditioned by the mode of formation of the rocks (genesis) and give indications on the type of rock from which the colouring remains are derived. Thus hematites in the form of fine platelets (size < µm) of lithology A and D are often characteristic of a hydrothermal formation (very rich context in the Bilbao syncline). The spherical and botrioidal morphology in association with clay and silicate minerals can indicate a formation in a context of continental alteration of a parent rock, lithology B, H and I.

4.3.3 Physico-chemical analysis (LA-ICP-MS)

Finally, after an observation phase, the objects were selected for geochemical analysis by LA-ICP-MS (Laser Induced Mass Spectrometry).

This method provides very high precision data on the content of trace elements that indicate the origin of the material. The laser ablates a 40µm area on the surface of the object, the material is transformed into plasma which is then transported to the mass spectrometer which counts the atoms present. The method is µ-destructive leaving a crater of a few micrometres on the surface of the object invisible to the naked eye.

However, because of this micro-destructive factor only a limited selection of objects was analysed. The selection was aimed at objects representative of the petrographic lithologies, and objects bearing potential traces of use were discarded in favour of small fragments with no apparent traceological significance.

Table 04: *liste des objets analyser en LA-ICP-MS (SP=Spain, MURR project code)*

Analyse N°	Object	Analyse N°	Object
SP001	AGT.5A.0ALM.3.2.164d Litho A	**SP020**	AGT.5B.BAM0.8.6.429a Litho F
SP002	AGT.5A.0ALM.3.2.164e Litho A	**SP021**	AGT.5B.0ALM.2.2.226a Litho B
SP003	AGT.5A.0ALM.3.2.164a Litho B	**SP022**	AGT.5B.AM0.8.5.407a litho A
SP004	AGT.5A.0ALM.3.2.164b Litho B	**SP023**	OND.22K.SLEC.7.4.395 litho D
SP005	AGT.5A.0ALM.3.2.164c Litho B	**SP024**	OND.22J.SLEC.9.5.458a Litho A
SP006	AGT.5A.0ALM.9.2.192a Litho D	**SP025**	OND.22J.SLEC.9.5.458c litho B
SP007	AGT.5A.0ALM.9.2.192b Litho A	**SP026**	AR.W15.3.10.1 Litho B
SP008	AGT.5A.0ALM.9.2.192c Litho A	**SP027**	AR.V15.2.4.3-8a Litho D
SP009	AGT.5A.0ALM.9.2.192d Litho C	**SP028**	AR.W15.1.15.1 litho A
SP010	AGT.5B.0ALM.4.2.27b Litho A	**SP029**	AR.V15.4.6.1 litho E
SP011	AGT.5B.AM0.8.3.315a Litho D	**SP030**	AR.V15.2.4.3.8g Litho F
SP012	AGT.5B.AM0.8.3.315b Litho A	**SP031**	AR.W15.1.5.3 Litho F

SP013	AGT.5B.AM0.8.3.315c Litho B	**SP032**	AR.W15.3.9.1-4a litho D
SP014	AGT.5B.AM0.8.4.359a Litho D	**SP033**	AR.W15.1.9.5-17a Litho F
SP015	AGT.6A.ALM.3.1.5 litho D	**SP034**	AR.W15.1.9.5-17l Litho D
SP016	AGT.5A.ALM.3.1.19 Litho B	**SP035**	AR.W15.1.9.5-17a Litho F
SP017	AGT.6B.0ALM.1.2.10 litho C	**SP036**	AR.W15.4.11.1.3a Litho F
SP018	AGT.5A.0ALM.6.2.173a Litho A		
SP019	AGT.5A.0ALM.6.2.173b Litho D		

This report does not present the study of geochemical data. Indeed, in order to be exploited, the data obtained on the archaeological objects needs to be integrated into a geological reference system. The data on the local and extra-regional geological references is in progress (scheduled for autumn 2022). The exploitation of the data is plan for the beginning of 2023. The addition of the second half of the collection would be an undeniable asset to ensure the robustness of the approach.

Acknowledgements

This project is financially supported by the University of Missouri Nuclear Research Center (MURR) and the Before Art Research Program. The study of colouring materials is carried out under the direction of and in collaboration with the Government of Bizkaia and the Archaeological Museum of Bilbao (Arkaeologi Museoa).