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# Relative chronology of Palaeolithic drawings of the Great Ceiling, Rouffignac cave, by chemical, stylistic and superimposition studies

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## Keywords

Rouffignac cave, *Great Ceiling*, Palaeolithic black drawings, *in situ* non-invasive analysis, X-ray fluorescence, barium-containing manganese oxides, stylistic and superimposition study.

## Abstract

This paper presents the first systematic chemical study of the black pigments used on the *Great Ceiling* panel at Rouffignac Cave in combination with stylistic and superimposition studies. A refined quantification strategy was designed to address the large number of drawings and the considerable amount of data required for the effective study of the specific black compositions on the *Great Ceiling*. We show that at least three different types of barium-bearing manganese oxides (that could be assimilated to different black raw coloring materials) were used. The new chemical data obtained are interpreted in light of stylistic and superimposition analyses of the figures. The grouping of the figures in small sub-sets according to chemical composition and consideration of superimpositions allowed for a complete re-reading of this complex panel and suggests that a small group of artists, each using different crayons, may have drawn the figures in a short period of time.

## 1. Introduction

Palaeolithic rock art is one of the oldest testimonies of representational artistic expression and of the beliefs and ritual practices of our ancestors. Renowned examples of decorated caves such as Altamira in Spain and Lascaux in France display an abundance of panels with overlapping images, painted in various colors, drawn, or engraved (Breuil, 1952; Leroi-Gourhan, 1965; Lorblanchet, 2010). Since their discovery and recognition as art, many studies have been conducted to reveal the meaning of their complex arrangements and to understand how, when and why they were created. Understanding the organization of the figures that compose the impressive panels of Palaeolithic cave art is a central archaeological and art-historical issue. Detailed studies of rock art have sought to identify various distinct steps in the decoration of cave walls and ceilings in order to provide a relative chronology of the creation of the figures when direct dating of the prehistoric pigments is not possible. The combination of several analytical approaches (stylistic, physicochemical, and superimposition of the drawings) is essential to such studies. This is the basis of our present work. An in-depth, systematic study of a major panel of Rouffignac cave was performed to determine the use of different pigments in its creation as a promising line of research. Thus, a systematic chemical study of the *Great Ceiling* drawings complements stylistic and superimposition studies already carried out. It provides essential new insight into the creation and interpretation of the panel, structuring its organization in the absence of absolute dating possibilities.

Cave art can be directly dated using radiocarbon dating if carbon-based pigments were used in its creation (Lorblanchet, 1994; Quiles et al., 2016). However, preserved prehistoric pigments are most commonly minerals with no organic components that would allow for C-14 dating. The stylistic study of the technical and thematic evolution of rock art over time (Breuil, 1952) and the study of the superimposition of images (Aujoulat, 2004; Lorblanchet, 2010) have been used to clarify the chronology of images and distinguish between different phases of creation. For example, N. Aujoulat (2004) studied the famous “Salle des Taureaux” at Lascaux (Dordogne, France) and found that it was apparently created in three phases. During each phase, monothematic groups of animal representations were drawn (first the horses, then the aurochs and finally the deer) (Aujoulat, 2004). The study by M. Lorblanchet (2010) of the 25 overlapping black drawings (horse, bison, mammoth, and aurochs) of the “Frise noire” of Pech-Merle cave (Lot) revealed five distinct phases of creation. Others examples can be cited, coming from Australia (Chippindale and Taçon, 1998), where the relative chronology of the creation of complex decorative panels takes into account the figures style (nature of the figure, size and color used,) the creation technique of the figures, the superimposition of the representations and the conservation state both of the pictorial matter and of the bedrock, and is combined with absolute dating. Stylistic and superimposition studies, however, have certain limitations. More detailed studies, including physicochemical analyses, are required to further enrich our understanding of Palaeolithic cave art.

The first such physicochemical analyses, mostly focused on pigments, were carried out on samples from important prehistoric caves such as Altamira, Niaux, Lascaux and La Garma using electron microscopy (SEM, SEM-EDX and TEM) and X-ray absorption spectroscopy (XAFS) to access the artists' techniques and to understand the operational sequence of prehistoric painting activities (Cabrera-Garrido, 1978; Ballet, 1979; Vandiver, 1983; Clottes et al., 1990; Menu and Walter, 1992; Chalmin et al., 2003; Vignaud et al., 2006; Arias et al., 2011). During the last decade, important progress made in the

48 development of portable systems for non-destructive analysis has facilitated greater access to valuable works of art like  
49 Palaeolithic rock art, and thus enabled an increasing number of studies to be conducted while preserving the integrity of the  
50 artwork (de Sanoit et al., 2005; Roldán et al., 2010; Lahlil et al., 2012; Nuevo et al., 2012; Beck et al., 2012 and 2014;  
51 Olivares et al., 2013; Gay et al., 2015 and 2016; Sepulveda et al., 2015; Wallis et al., 2016; Mauran et al., 2019).  
52 Portable XRF spectrometry (pXRF) is a particularly suitable method for the *in situ* study of rock art, allowing for the quick  
53 and non-destructive identification of the elemental composition of pigments. Quantification is however challenging because  
54 of the heterogeneity of the analyzed material, in terms of the composition of the pigments and underlying rock surfaces,  
55 variable thickness of pigments applied. The karst environment also contributes to the complexity that can be encountered  
56 when applying pXRF to rock art: accessibility of art deposited on walls and ceiling, and uneven surfaces (geometric  
57 constraints) will depend directly on it, and with it, the use of a spectrometer device the handler (Huntley 2012). All of these  
58 difficulties, as well as the scope of work and time required (for work in the field and the evaluation of the data), have to be  
59 taken considered in the development of quantitative procedures specific to each individual cave (Gay et al., 2016).

60

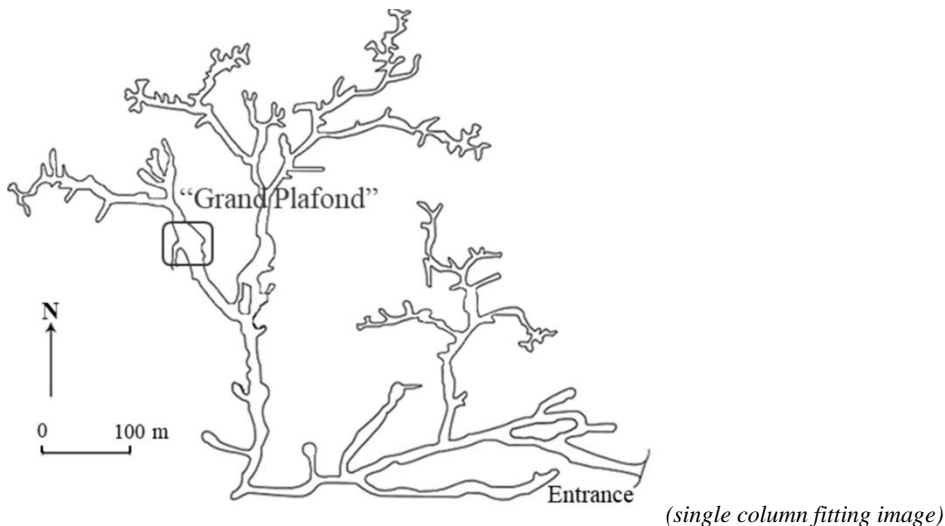
## 61 2. The Rouffignac cave

62

63 The cave of Rouffignac, also referred to as “the cave of a hundred mammoths” is famous for the striking number of  
64 mammoth representations (160), which account for about 30% of all mammoth representations inventoried in prehistoric  
65 rock art to date (Barrière, 1982; Plassard, 2005). Rouffignac Cave is one of the most prominent caves of the Vézère Valley, a  
66 major locus of Palaeolithic rock art classified as a UNESCO World Heritage Site in 1979. Its entrance has remained open  
67 since the Pleistocene and the cave has long been visited in more recent centuries, as indicated by many graffiti. However, the  
68 Palaeolithic rock art was only discovered in 1956 by Louis-René Nougier and Romain Robert (Nougier and Robert, 1957).  
69 The vast cave network comprised of nearly ten kilometers of galleries houses 250 engravings and black drawings, sometimes  
70 arranged in impressive compositions as in the example of the *Great Ceiling* panel. The art has been stylistically assigned to  
71 Leroi Gourhan style IV (Leroi Gourhan, 1965). However, new discoveries at the end of the twentieth century, in particular  
72 the discovery of the Chauvet Cave, overturned the current system based on a linear stylistic evolution of the Palaeolithic rock  
73 art (Valladas and Clottes, 2003; Fritz, 2017). Yet, it is still possible to accept the Rouffignac art attribution to the recent  
74 phase of the emblematic Middle Magdalenian period, whose is well-documented in other caves and rock-shelters in the  
75 region, such as Font-de-Gaume, Les Combarelles, Bernifal or Cap-Blanc, or in other occupied shelters, such as Laugerie-  
76 basse or la Madeleine (Tosello, 2003; Plassard, 2005; Bourdier, 2010).

77 The *Great Ceiling* is a major and singular ornamented panel in Rouffignac, on which a fourth of the total number of  
78 compositions are concentrated on a surface of less than 40 m<sup>2</sup> (Fig. 1).

79



80

81

82 Fig. 1: Map of Rouffignac Cave (according to Plassard, 2005) with the Great Ceiling (“Grand Plafond”) indicated.

83

84 The five principal animal species in the cave (mammoth, bison, ibex, horse, and woolly rhinoceros) are represented on it in  
85 the form of black figures that overlap each other and vary in size and orientation (Barrière, 1982). Even the largest animal on  
86 the Great Ceiling, a horse 2.70 m long was executed without distortion even though the floor of the cave was less than one  
87 meter below the ceiling in the Palaeolithic era. This evidences the remarkable artistic performance of its creators.

88 The creation technique of all the figures is the same and consists in drawing with dry matter. Indeed, the black matter from  
89 the strokes is agglomerated or adhered on one side of contact surfaces, but not on the other side (Fig. 2). These contact  
90 surfaces can be asperities of the substrate or relief. If it was moist painting matter, it would have covered entirely these  
91 irregularities. Moreover, these observations allow attesting the direction of the linear stroke.

92

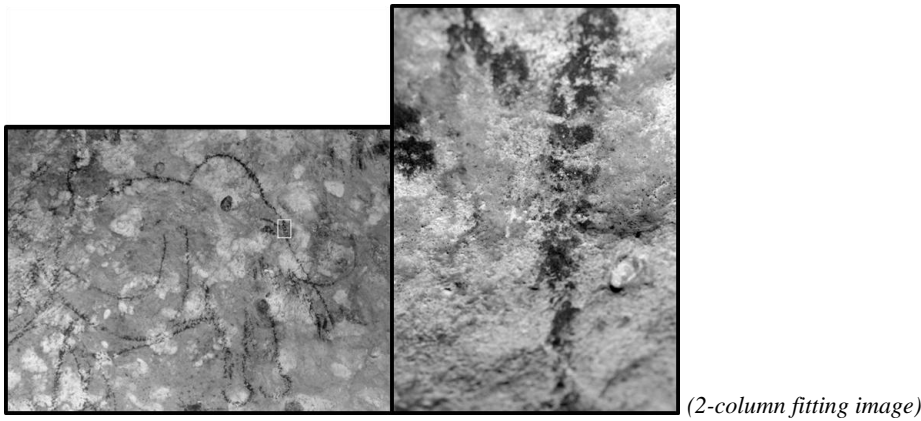


Fig. 2: Left: Macrophotography of the mammoth 121. Right: The side of the agglomerated matter on the wall substrate indicates the direction of the stroke.

No testimony of the use of crayons, commonly defined as faceted blocks showing signs of wear (Salomon, 2009; Dayet 2013, Chalmin and Huntley, 2017) has been found at Rouffignac Cave. Thus, the notion of dry raw coloring material seems more suitable in this studied case. The term of moist painting matter appears incorrect, given the macroscopic observation of the deposit of the black matter on the bedrock.

In the midst of this turmoil of images, with homogeneous style and application technique, no master plan of its realization brings out, become unreadable as a result of an accumulation of too many images. Despite a lack of coherence, the desire to comprehend the organization of these drawings and the will to identify various distinct steps in the decoration in order to access to a relative chronology of the creation of the figures lead to different detailed studies including stylistic and superimposition observations as well as physico-chemical analyses.

### 3. Previous studies and new issues

The stylistic study of art at Rouffignac Cave has been conducted since the earliest research in the cave in 1959 (Nougier and Robert, 1959). The authors of these studies (Leroi-Gourhan, 1965; Barrière, 1982; Plassard, 1999 and 2005) are in agreement that all of the figures share stylistic similarities with each other and with the rock art from other caves such as Font-de-Gaume and Combarelles.

The organization of the drawings on the *Great Ceiling* was first interpreted as animal groups distributed in a semi-circle around the entryway to the gallery, entering or leaving it (Barrière, 1980). More recently, F. and J. Plassard (2016) re-interpreted the composition based on superimposition studies and concluded that the *Great Ceiling* is a juxtaposition of monothematic or more complex ensembles, associated with single representations. However, the understanding of the overall organization of the *Great Ceiling* panel is still incomplete.

Preliminary chemical studies on samples from Rouffignac as well as isolated *in situ* analyses of a few drawings not only confirm the exclusive use of manganese (Mn) oxide-based pigments in the Palaeolithic figures in the cave, but additionally demonstrate that it is possible to distinguish between different kinds of Mn oxide (Grazioso, 1956; de Sanoit et al., 2005; Lahlil et al., 2012; Beck et al., 2012 and 2014). Anyhow, any relationship could not be established between the use of one kind of Mn oxide and the species, the size or orientation of the animals represented yet.

Based on these early results and the lack of chemical information for most of the drawings, a systematic chemical study of the *Great Ceiling* drawings is engaged to provide essential new insight into the creation and interpretation of the panel.

Specific research questions addressed in this study include:

1. Is it possible to define characteristic chemical fingerprints for the specific pigments used in the creation of each prehistoric figure based on *in situ* chemical analyses by pXRF?
2. Can groups of figures be defined according to the chemical signatures of specific pigments?
3. Is there a relationship between specific chemical signatures and the types of animals depicted?
4. Do chemical data support stylistic classifications of the figures or do they suggest a new interpretation of the drawings (or panel composition)?
5. Do the chemical data support the chronological order of the execution of the figures proposed by the superimposition study?

Furthermore, does the combination of chemical, stylistic and superimposition data provide new insights into the process of creation of the panel? Can conclusions be drawn regarding:

6. The identification of different hands (individual artists or groups of artists) and their assignment to distinct figures or figure groups?
7. The relative chronological sequence of the figures?
8. The identification of distinct phases of creation in different time periods (intervals of one generation to several or even many generations) or, in contrast, the quasi contemporaneity of the figures?

Interpretation of the data is based on several assumptions. One, that the chemical composition of a pigment is homogeneous within the strokes that form an individual figure and could imply that one figure was made in one session with one type of particular raw coloring material. Second, the applied analytical method is sensitive enough to distinguish between different raw coloring materials used by means of the analysis of the strokes on the wall. Thirdly, a specific chemical signature could be then attributed to a particular dry fragment of homogeneous raw material supposed homogeneous, with a common geological origin, assuming that the geological source of the raw material is also homogenous. Though various scenarios can be hypothesized for the creation process of the panel, we consider it most likely that one artist or a small group of artists used a specific type of raw coloring material at any given time. Furthermore, if two figures created with pigments of different chemical compositions overlap, the figures must necessarily have been created in succession, one before the other. However, the time interval between the creations of these figures could vary widely and will be discussed on the basis of our new results. Therefore, combining chemical data with the other forms of information available is crucial to the development of new interpretations.

#### 4. Materials and Methods

##### 4.1. Challenges of *in situ* chemical analysis in karst contexts

A total of 44 drawings from the *Great Ceiling* with three to five measurements per figure was analyzed at representative locations depending on accessibility (Fig. 3b). Several measurements had to be performed on the lines of the figures and on the bedrock next to them so that statistically relevant data could be obtained for each figure or parts of a figure. In all, 212 analyzed points on the pigment and 82 analyzed points on the bedrock have been performed (Appendix Table A and Table B). The *Great Ceiling* as a whole presents a remarkable state of conservation. Any taphonomic phenomenon, as coating, crust or other mineral accretion, doesn't affect both the pigment layer and the substrate, which simplifies the choice of the location of the analyzed point. The applied pXRF spectrometer was well adapted to the particular working conditions of a cave environment, given its high flexibility, minimal weight, and ease of handling. This was even more important as the animal drawings are not easily accessible at the cave's ceiling, currently 2.40 meters above the cave floor (Fig. 3a). The primary spectrometer is a non-commercial device composed of a 40 kV MOXTEK X-ray tube with a palladium anode and a beam-spot size of approximately 1 mm<sup>2</sup> on the sample achieved with a collimator. A 7 mm<sup>2</sup> Silicon Drift Detector with an energy resolution of 140 eV (FWHM at 5.9 keV) was used to collect the XRF signal. The X-ray tube and detector are fixed in 45°/90° geometry on a positioning system that allows micrometric movements. The head of the detector is at a distance of 5 mm of the analyzed surface. The spectrometer is mounted on a stable tripod. The measurement conditions were 30 kV, 300 μA, and 300 s / measurement.

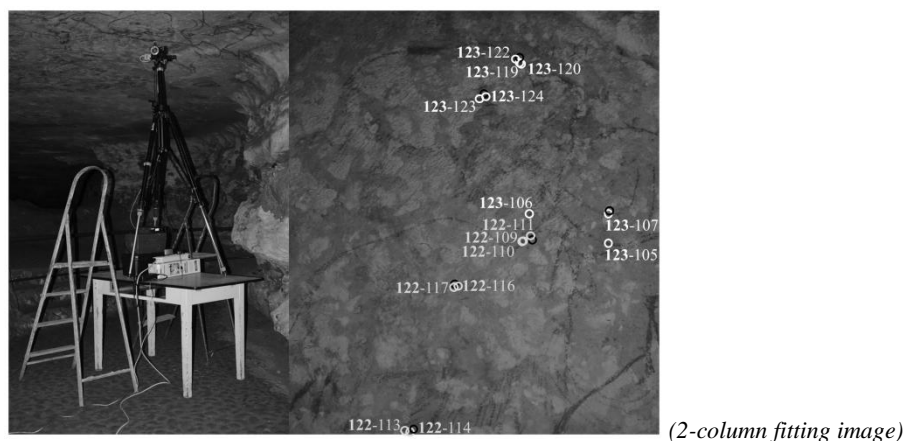


Fig. 3: **a)** Portable XRF spectrometer used in the analysis of the black drawings on the *Great Ceiling*. **b)** Multiple measurements at different locations on a single figure (bright points) and on the wall substrate (black points) were performed to acquire statistically relevant XRF data and to evaluate the heterogeneity of the cave wall: the example of Mammoths 122 and 123.

Additionally, two slightly different spectrometer set-ups were used in the analysis of Rouffignac's rock art during a total of seven three-day measurement campaigns between 2009 and 2016 (de Sanoit et al., 2005; Beck et al., 2012 and 2014; Gay et al., 2016). They differ in terms of the type of components (X-ray tube and/or detector) and in the geometry of the device (position of the X-ray tube and the detector with regard to each other and to the analyzed surface). The performance of the

three set-ups can be considered equal (see de Sanoit et al., 2005 and Beck et al., 2012 and 2014 to a detailed description of the two devices) and the results obtained were tested as equivalent (*Appendix Table C*).

#### 4.2. Quantification of analytical data

Quantification of the results poses an analytical challenge due to a suite of complicated conditions:

- the small quantity of pigment
- the heterogeneity of both the thickness of the pigment layers and the composition of the underlying cave wall. It must be taken into consideration that the signals captured from the pigments and the cave wall cannot be easily separated. The influence of the composition of the wall on the spectra obtained from the pigments varies as a function of the thickness and density of the applied pigment (Gay et al., 2016).
- the X-ray incidence and detection angles are difficult to control due to the uneven surface of the cave wall at macro- and micro-scales, which may introduce some inter-measurement variations that are difficult to control for.

Nevertheless, the composition of the black pigments can be (semi)-quantified in terms of the three main constituents of the pigments: manganese oxide ( $\text{MnO}_2$ ), barium oxide ( $\text{BaO}$ ) and iron oxide ( $\text{Fe}_2\text{O}_3$ ) (for the complete data set see *Appendix Table A*). The  $\text{MnO}_2$  and  $\text{BaO}$  contents of the cave wall are negligible and  $\text{Fe}_2\text{O}_3$  is found in the cave wall to a much lesser extent than in the pigment. Therefore, these oxides can be considered chemical markers for the pigment (Fig. 4).

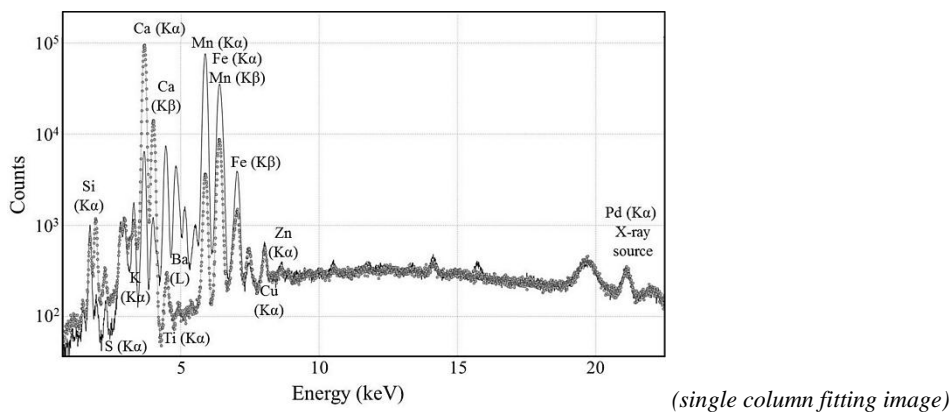


Fig. 4: XRF spectra obtained for the black pigment of the horse 84 measurement point 201 referenced 84-201 (continuous line) and the wall substrate right next referenced 84-203 (dotted line). These two XRF spectra are representative of the whole spectra of this study.

Two different procedures to semi-quantify the three oxide concentrations have been implemented through the previous studies, the fundamental parameter method (Beck et al., 2012 and 2014; Gay et al., 2016) and Monte Carlo simulations (de Sanoit et al., 2005). Monte Carlo simulations are very time-consuming, and the fundamental parameter method turned out to be more efficient and better suited to our large XRF datasets and was therefore used exclusively in this analysis. The XRF spectra were fitted using PyMca software (Solé et al., 2007). The sum of the three main constituents ( $\text{MnO}_2$ ,  $\text{BaO}$  and  $\text{Fe}_2\text{O}_3$ ) was normalized to 100 %, rendering measurement data comparable and independent from flux variations between different analyses. Although the wall contains a trace amount of  $\text{Fe}_2\text{O}_3$  it is considered negligible with respect to its content in the pigment. Therefore, the subtraction of the substrate signal from that of the pigment layer was not necessary (Gay et al., 2016).

#### 4.3. Stylistic study

The stylistic study of the *Great Ceiling* is based upon observations of the contexts and graphic conventions of the figures: the determination of the number of animals belonging to a species, and location of these animals in relation to others and the identification of associated themes, the description of the technique(s) used for drawing, the description of the animal profile and its degree of completion, the indication of anatomical elements related to a pictorial theme (sexes, legs, bellies, tails, eyes, tusks, horns, humps, etc.). Some of these details are highly relevant to the chronological attribution of the style. Thus, the presence of tectiforms and the anatomical precision of some of the figurative details such as the horses' heads and hooves, or the mammoths' bilobed trunks and anal flaps are considered typical of Leroi-Gourhan's Style IV.

#### 4.4. Superimposition study derived from F. and J. Plassard (2016)

The superimposition study was based on the compared observations of four different observers, made with the naked eye or with the help of a magnifying glass, of all overlapping or intersecting figures. In addition, the graphic elements and stylistic conventions chosen by the Palaeolithic artists have been considered in this study. These stylistic conventions included, for example, the orientation of the animals relative to one another (facing each other or not) and the fact that in some cases the artist obviously avoided drawing over an already existing figure when creating a new one. Each series of superimposed

241 motifs, when they are observable, can be assimilated to an archaeological stratigraphy and can therefore be presented in a  
 242 form of a Harris matrix (Harris and Gunn, 2017). A Harris matrix was thus constructed to evidence the superimposition  
 243 relationships between figures. The construction of the matrix is based on the relations that the stratigraphic units maintain  
 244 between themselves represented with lines and boxes: the figure of one ensemble is indicated in a box and connected by a  
 245 line to another figure. A vertical line represents the chronological relationship of one unit being later than another (the  
 246 younger event on the top) and a double horizontal line the relationship of equivalence or contemporaneity. Therefore, each  
 247 Harris matrix should be read from the base to the top.  
 248

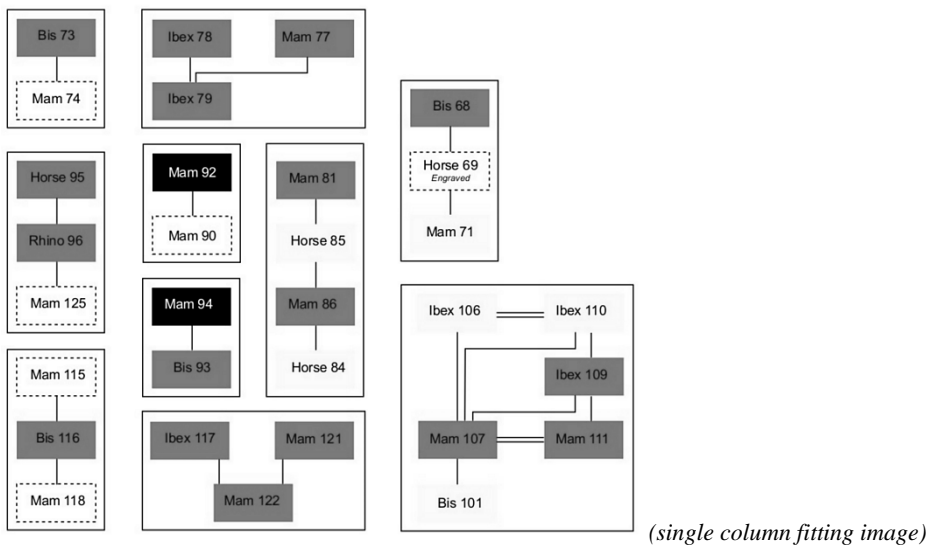
## 249 5. Results

### 250 5.1. Summary of the stylistic study

251  
 252 At the scale of the *Great Ceiling*, the stylistic similarity of the figures is underlined by their technical homogeneity, even in  
 253 the absence of a structured organizational layout. The small stylistic differences between pictures are not correlated with the  
 254 compositional groups, such as groups of facing animals. Therefore, these small differences are not enough to define distinct  
 255 stylistic groups. Nevertheless, some anatomical details depicted on the animals, such as the fat hump on bison or the horns of  
 256 ibex, allow us to identify groups of pictures with stylistic similarities even if it is not possible to decide if these reflect  
 257 stylistic choices or natural anatomical diversity.  
 258

### 259 5.2. Summary of the superimposition study derived from F. and J. Plassard (2016)

260  
 261 Approximately half of the 65 drawings showed intersections between the drawings of distinct figures in the superimposition  
 262 study. Only 27 superimpositions of strokes were observed in a total of 33 drawings (*Appendix Table B*). The compilation of  
 263 superimposition data, combined with graphical and stylistic elements of the figures that were deemed significant, allowed the  
 264 images to be classified into several subgroups. The construction of a Harris Matrix enables to highlight these subgroups (Fig.  
 265 5). This approach has already proved its efficiency to precise the stratigraphic sequence of an art panel in different contexts  
 266 (Harris and Gunn, 2017).  
 267  
 268



269  
 270  
 271 Fig. 5: Harris Matrix of the *Great Ceiling, Rouffignac cave, Dordogne*. Each series of superimposed motifs, when they are observable, can  
 272 be assimilated to an archaeological stratigraphy and can therefore be presented in the form of a Harris matrix (Harris and Gunn, 2017).  
 273 Each Harris matrix must be read from the base to the top ; the vertical line indicate the stratigraphic relationship (the younger event on the  
 274 top) and the double horizontal lines indicate a chronological equivalence.  
 275

276 As illustrated by the Harris matrix depicted in Fig. 5, the subgroup with Mammoths 120 to 123 creates a small frieze with  
 277 superimpositions and reserve between the pictures.

278 Mammoth 125, Rhinoceros 96 and Horse 95 can be considered as a second subgroup. The horse was drawn after the  
 279 mammoth and the rhinoceros, although it is the largest picture on the ceiling. This observation contradicts the classical  
 280 hypothesis that the biggest drawings are central to the organization of the panel and were drawn first (Barrière, 1980).

281 A last example of a subgroup is a complex set of drawings located around Mammoth 107. The sequence of execution  
 282 established by the patterns of superimpositions and several graphic elements, such as certain lines that deviate to respect  
 283 lines already present, is as follows: first the forehead of Bison 126, followed by Bison 101, whose backline respects the  
 284 outline of the forehead of Bison 126; Mammoths 107 and 111 came after and, finally, Ibexes 106, 109 and 110.  
 285

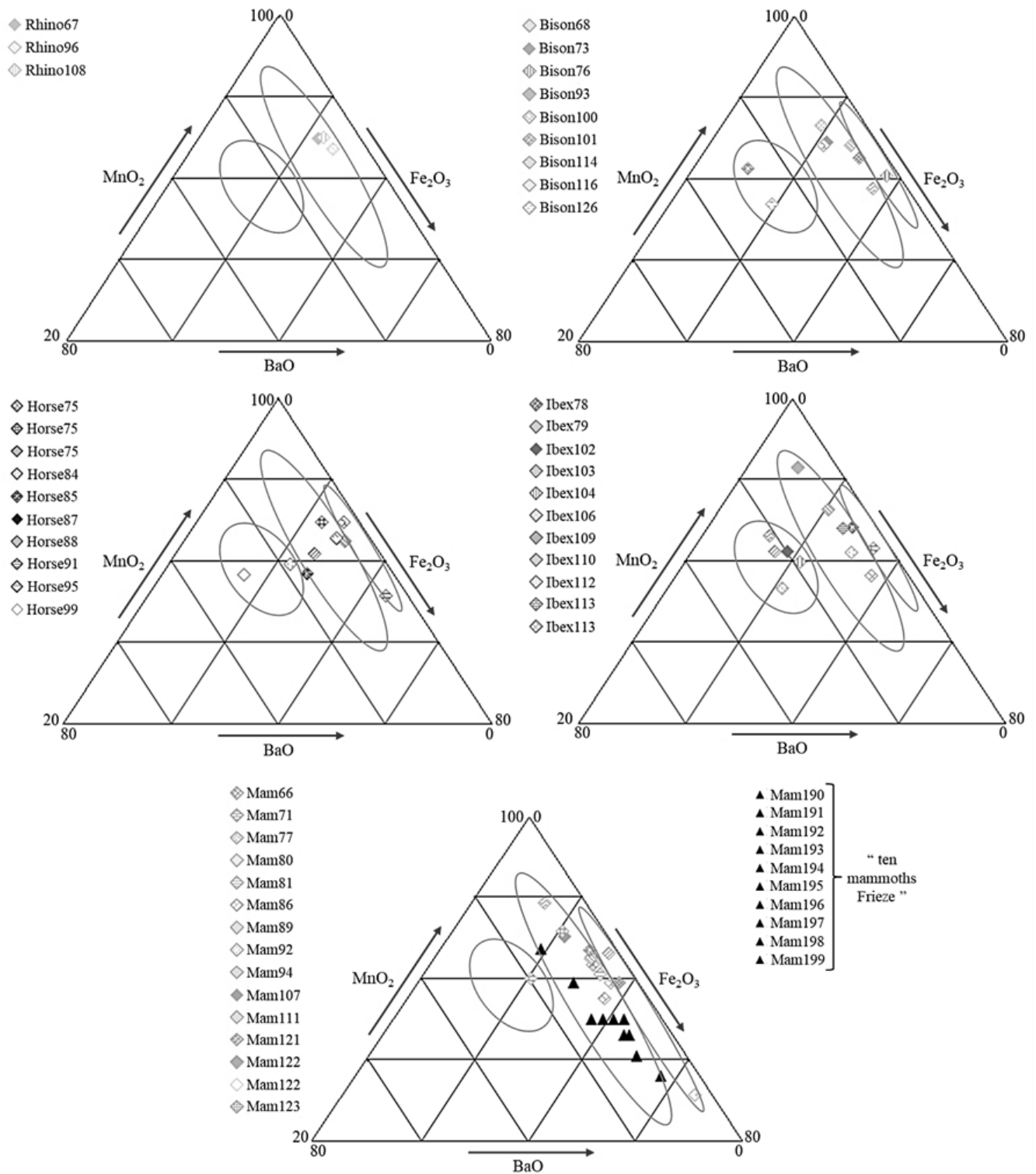
286 5.3. Chemical study. The composition of the strokes on the ceiling as chemical signature of specific raw coloring material

287  
288 As no crayons have been found at Rouffignac Cave, the chemical compositions of the pigments on the wall are the only  
289 sources of data on the coloring material used. The composition of pigments obtained from several pXRF measurements on a  
290 given drawing has proven to be generally homogeneous, and average values are used as representative compositional values  
291 of the figures (*Appendix Table A*). This supports our hypothesis that a characteristic chemical composition would be  
292 indicative of a specific coloring material; be it in the form of a raw material fragment or as transformed matter. Thus,  
293 chemical fingerprints were defined for various pigments used in the creation of individual figures. These chemical  
294 characteristics can be correlated to the use of raw material of a specific origin. In a first approximation, we will suppose that  
295 material from the same geological source might have similar compositions. In this case, though, it could still indicate  
296 (especially with the combined superimposition data) the same supply of raw material, e.g. a group arrives together having  
297 collected their raw coloring material at the same location on the way to the cave. At the same time, if different artists from  
298 different places met in the cave to create the drawings, each artist might have raw material from a different source.

#### 300 5.4. Distinguishing different groups of figures according to the chemical composition of the pigment strokes

301  
302 Our comprehensive dataset suggests the existence of three chemical groups of figures predominantly based on differences in  
303 the BaO contents contained in their outlines (Fig. 6). The first group is characterized by very low BaO concentrations,  
304 ranging from 2 % and 3 % (very-low BaO-group). A second group with low to intermediate BaO contents of up to 10 % has  
305 been defined (intermediate BaO-group). The pigments belonging to the third group (high BaO-group) contain significantly  
306 higher BaO concentrations ranging from 16 % to 27 %. The ternary diagram also shows great dispersion of Fe<sub>2</sub>O<sub>3</sub> values.  
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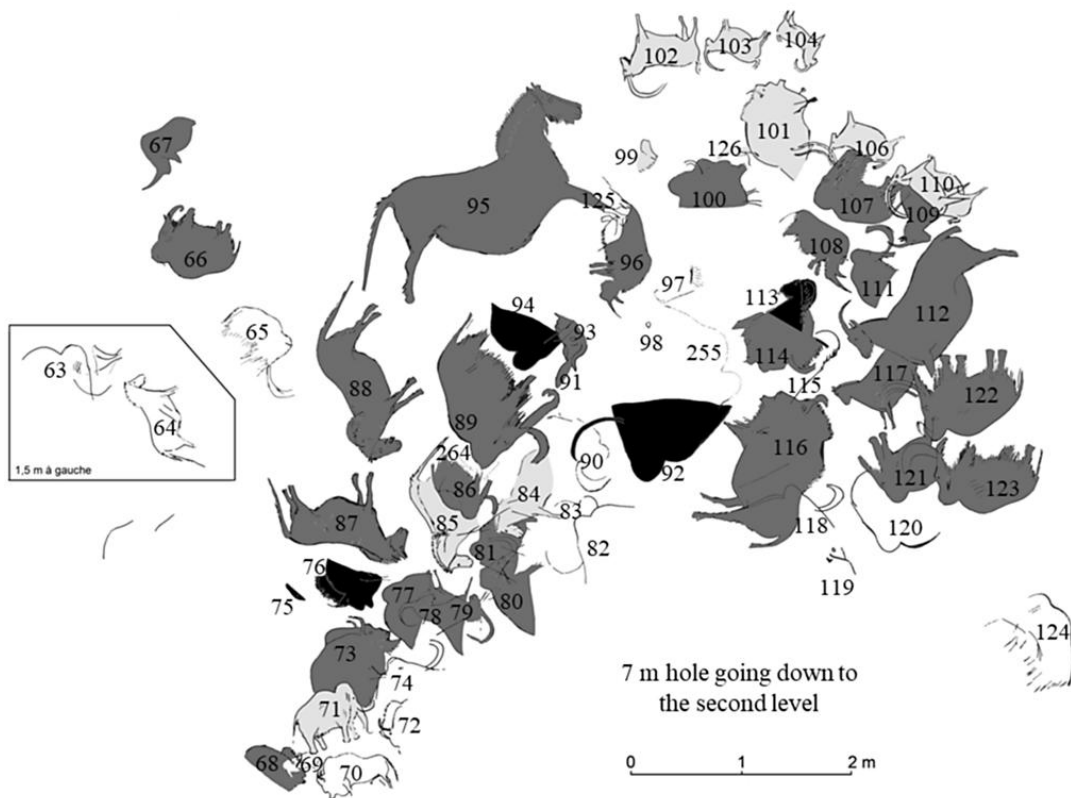




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(2-column fitting image)  
 Fig. 6: Ternary diagrams of the three main oxide concentrations ( $MnO_2$ ,  $Fe_2O_3$  and  $BaO$ ) characterizing the black pigments of the Great Ceiling and of the “Ten Mammoths Frieze” at Rouffignac Cave, including data from this study and the available literature (de Sanoit et al., 2005; Beck et al., 2012 and 2014; Gay et al., 2016). The data are grouped according to the animal species represented to enhance clarity of the plots. The three groups are indicated by ellipses.

A clear interpretation of the  $Fe_2O_3$  concentration values is not possible due to a small and varying Fe content in the cave wall substrate, which affects the pigment signal more or less as a function of the thickness of the pigment layer. Figure 7 provides an overview of the analytical data obtained from the Great Ceiling and illustrates the chemical classification of the drawings in three different groups, indicating the use of a minimum of three different crayons.



(2-column fitting image)

Fig. 7: Visualization of all analytical data in the general survey of drawings on the *Great Ceiling*. The different chemical groups are indicated: very-low BaO group (black), intermediate BaO group (dark-gray), high BaO group (light-gray) and inaccessible figures, not studied (not colored).

The three chemical groups identified in this study are consistent with the use of at least three different variants of black pigment (or mixtures of pigment) consisting of different amounts of the minerals pyrolusite ( $MnO_2$ ), corresponding at the low end of the range to the very-low BaO group, and of romanechite ( $Ba_2Mn_5O_{10}$ ), corresponding to the high end of the range to the high BaO-group. These Mn oxides have been identified in a preliminary study using portable X-ray diffraction (Lahlil et al., 2012; Beck et al., 2012 and 2014). The majority of the studied animal representations belongs to the very-low and intermediate BaO-groups, thus to pyrolusite and pyrolusite mixed with another Ba-containing Mn oxide like romanechite. No relationship was observed between the chemical groups and the animal species represented (Table 1). The chemical groups are apparently partly contradictory to those defined by stylistic/superimposition studies.

Table 1: The classification of the black animal drawings of the *Great Ceiling* in three chemical groups according to concentrations of the main constituents of the pigments.

Drawing	Very-low BaO-group (2-3 % BaO)	Intermediate BaO-group (~10 % BaO)	High BaO-group (16-27 % BaO)
Woolly rhinoceros		67, 96, 108	
Bison	76	68, 73, 93, 100, 114, 116	101, 126
Mammoth	92, 94	66, 77, 80, 81, 86, 89, 107, 111, 121, 122, 123	71
Horse	75	87, 88, 91, 95	84, 85, 99
Ibex	113	78, 79, 109, 112, 117	102, 103, 104, 106, 110

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## 6. Discussion

### 6.1. Re-reading of the Great Ceiling crossing different studies

The overall comparison of the data from superimposition, stylistic, and chemical studies does support a hypothesis of thematic structuring with one species drawn before another, as observed in other cases of prehistoric rock art, such as the “Frise noire” at Pech-Merle, the “Salle des taureaux” at Lascaux (Aujoulat, 2004; Lorblanchet, 2010).

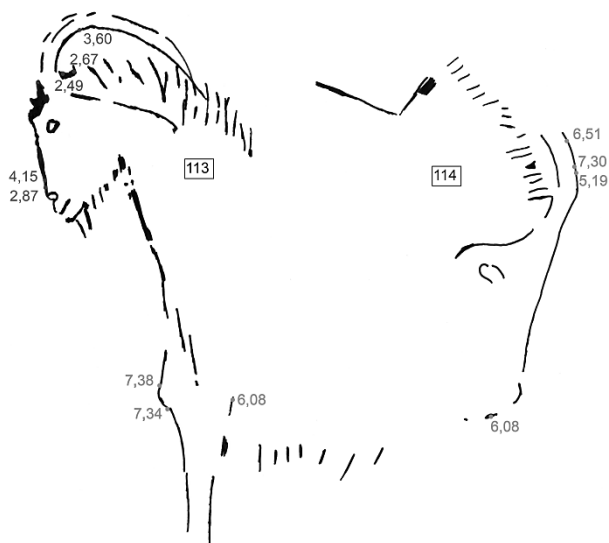
As discussed in F. and J. Plassard (2016), the *Great Ceiling* cannot be interpreted as a unified composition. It is more appropriate to interpret the panel as an arrangement of several subgroups of images highlighted first by the stylistic and the superimposition studies, on which we have chosen to focus our discussion given the new chemical results.

### 6.2. Insights on selected subgroups from the *Great Ceiling*

#### 6.2.1. *Re-reading of single-panel figures and evidence of a third chemical group*

The two cases discussed below illustrate particularly how chemical analyses can contribute to a new reading of the figures. The comprehensive chemical data obtained from the figures on the *Great Ceiling* allowed the definition of a third chemical group corresponding to the very-low BaO-group (~ 3 wt.% BaO), which has not been identified before in the preliminary studies (Beck et al., 2012 and 2014). The identification of this new chemical group allows additional insights into the unity of specific figures.

First, the neighboring drawings of Ibex 113 and Bison 114 are considered (Fig. 8). The pigment compositions found for the front legs of Ibex 113 (~ 7 wt.% BaO, intermediate BaO-group) are not in agreement with the ones obtained for the ibex muzzle, the back of its head and its horn (~ 3 wt.% BaO respectively, very-low BaO-group). Surprisingly, the chemical signature of the ibex legs corresponds perfectly to the one of the head of neighboring Bison 114. The chemical characteristics observed for these two figures may indicate that the lines initially interpreted as front legs of the Ibex 113 actually represent the front leg of Bison 114.



(single column fitting image)

Fig. 8: Chemical analysis of two neighboring drawings, Ibex 113 and Bison 114, calling into question the attribution of the front legs of Ibex 113. The BaO concentrations (wt.%) obtained are indicated for all XRF analysis locations.

Second, different chemical signatures were found for several drawing strokes attributes to Mammoth 122. The BaO concentrations obtained for the lines inside the outline of this mammoth and the lines illustrating the fur on its legs differ from those of the line that forms the outline (6 and 3 wt.%, respectively). This difference can be explained by a stylistic addition from the original artist itself or by another hand possibly at a later date, which we consider the more likely scenario.

#### 6.2.2. *Evidence of the creation of sub-ensembles of the Great Ceiling*

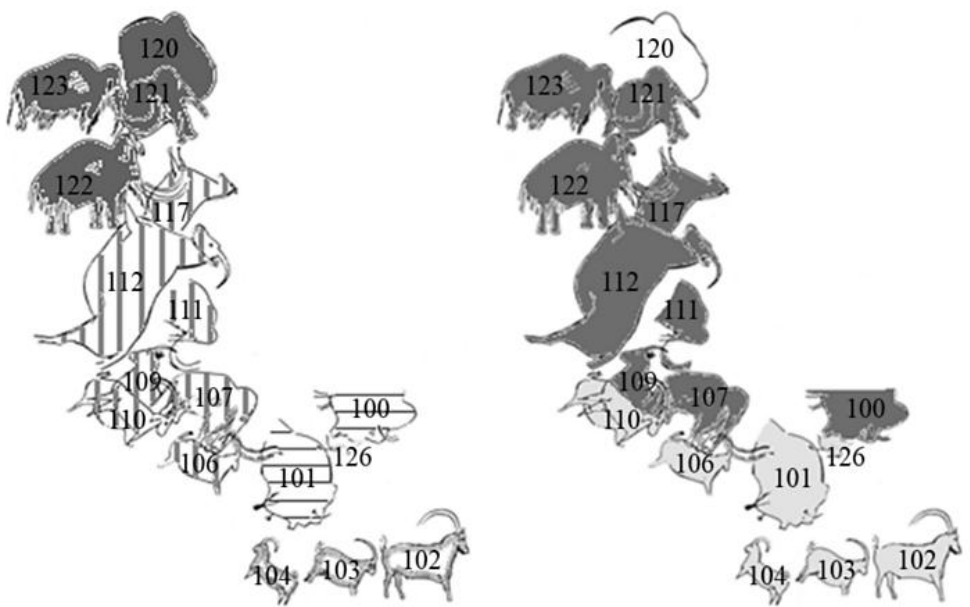
The following example illustrates how combining the results stylistic, superimposition, and chemical studies provides new information on the sequence of actions that created the panel. The example is a complex ensemble of four distinct but

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overlapping stylistic subgroups (Fig. 9). Group 1 is a homogeneous set of mammoths (Mammoths 120-123), group 2 is a coherent structure of mammoths and ibexes (Mammoths 107 and 111, Ibexes 106, 109, 110, 112 and 117), group 3 is a monothematic set of ibexes (Ibexes 102-104) and group 4 is a triptych of bison (Bison 100, 101 and 126) (Fig. 9a).

The chemical analyses reveal the same chemical fingerprint for all of the mammoths in the homogenous group 1. The results fall into the intermediate BaO group (Fig. 9b) and corroborate the conclusions of the stylistic and superimposition studies.



(single column fitting image)

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Fig. 9: a) Superimposition interpretation: four distinct units consisting of a homogeneous set of mammoths (gray fill), a coherent layered group of mammoth and ibex (vertical stripes), a triptych of bison (horizontal stripes) and a set of ibexes (gray outline). b) The different chemical groups are indicated: intermediate BaO group (dark-gray), high BaO group (light-gray) and inaccessible figures, not studied (not colored).

An identical chemical fingerprint (intermediate BaO-group) was found for all figures of the complex composition of mammoths and ibexes (group 2), except two of the ibexes (Ibexes 110 and 106) that show high BaO concentrations, placing them into the high BaO group. This suggests that these two ibexes could belong to the neighboring monothematic set of ibexes (group 3), which all belong to the high BaO group. In this case, the chemical data partly confirm the stylistic and superimposition studies and, moreover, provide evidence for the re-assignment of certain figures and thus a more accurate reading of the sub-panel composition.

Further, the chemical grouping determined for the triptych of bison (group 4) apparently contradicts the grouping of the three bison into a single unit, because Bison 100 belongs to the intermediate BaO group whereas the other two fall into the high BaO group. The superimposition of Bison 101 and 126 reveals that the backline of Bison 101 was broken to respect the forehead of Bison 126. These observations coincide with the chemical information and support the separation of the set of bison into two groups, with Bison 100 on one side and Bison 101 and 126 on the other side.

6.2.3. Evidence for the sequential creation of subgroups on the Great Ceiling

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Considering the overlap patterns of these four groups, a new reading of the organization of this complex ensemble can be provided, even if the chronology of creation appears to be very complex. There are two groups of figures with superimpositions within them, and no superimpositions between the groups.

Table 2 summarizes the sequential information that can be deduced from the new observations and analyses. A part of this ensemble (group 1 and part of group 2) was started with Mammoths 120 to 122 then Ibexes 112 and 117. The other part (part of groups 2 and 4, and group 3), with no superimposition with the previous, was started with Bison 126 then Bison 101. It was continued with Mammoths 107 and 111 and Ibex 109, looking to the left side, and then finished with Ibexes 110 and 106, looking to the right side and associated with Ibexes 102 and 103. Ibex 104 chemically belongs to the set of ibexes looking to the right side and was apparently drawn at last. Consequently, it seems that the two different black pigments were used in alternation: high BaO group for Bison 101, then intermediate BaO group for Mammoth 107 and 111 as well as for Ibex 109 and once again high BaO group for Ibexes 110, 106 and 102 to 104.

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Table 2. The interpreted relative chronological sequence of the panel of four superimposed groups.

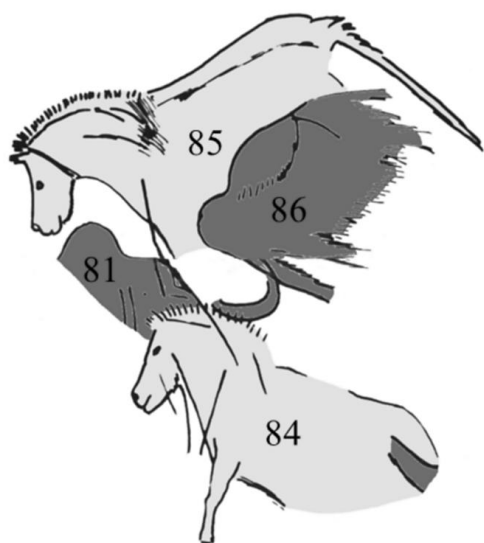
Creation phase	1	2	3
Group 1 and part of group 2	Mammoths 120, 121, 122	Ibexes 112, 117	
Chemical results	Intermediate BaO-group	Intermediate BaO-group	
Group 3 and part of groups 2 and 4	Bison 126 then Bison 101	Mammoths 107, 111, Ibex 109	Ibexes 110, 106 Ibexes 102, 103, 104
Chemical results	High BaO-group	Intermediate BaO-group	High BaO-group

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The application of several types of pigments on a complex, interlinked panel indicates that the panel was created in a relatively short period of time by a small group of people, using each a different type of black crayon.

#### 6.2.4. Evidence of the contemporaneous work of at least two artists on one ensemble

The case of two overlapping monothematic groups, Horses 84 and 85 and Mammoths 81 and 86, was chosen to demonstrate the complexity of the interpretation of the combined results of data from stylistic, superimposition, and chemical studies (Fig. 10). The graphic superimposition of the figures suggests the following timeline for their creation: Horse 84 was drawn first, followed by Mammoth 86, Horse 85 and finally Mammoth 81. According to these observations, the two monothematic pairs do not in fact belong together and seem to have been drawn independently. By contrast, the chemical analysis showed chemical differences between the pairs of mammoths and the pairs of horses, but the same chemical composition within the pairs: the two horses belong to the high BaO group whereas the two mammoths fall into the intermediate BaO group. Two different raw materials seem to have been used in alternation to draw two different pairs of animals.



(single column fitting image)

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Fig. 10: A detail of the *Great Ceiling* with two monothematic pairs, Horses 84 and 85, belonging to the high BaO group (light-gray) and Mammoths 81 and 86, belonging to the intermediate BaO group (dark-gray).

## 7. Conclusion

In addition to the superimposition and stylistic studies conducted on all figures of the *Great Ceiling* non-invasive, *in situ* chemical analyses were conducted on 44 out of the 65 animal representations to determine the chemical compositions of the pigments used. This study makes the *Great Ceiling* the most extensively-studied Palaeolithic panel of artwork in the world in terms of chemical analysis. Three different pigment groups were identified, indicating the use of at least three different types of crayon in the creation of this complex panel. The holistic approach combining results from stylistic, superimposition, and chemical studies of the figures revealed an absence of overall unity in the *Great Ceiling* decoration but allowed for the definition of several coherent subgroups in the panel. The new chemical classifications support a rereading of several drawings and sets of figures, providing crucial new insights to refine the organization of subsets or sequences of action in the creation of the drawings. Furthermore, the alternating use of different crayons in the creation of more or less complex compositions was revealed. As the style of the figures is relatively homogeneous, this reinforces the idea that a very small

465 team of artists probably created the *Great Ceiling* in a relatively short time, despite the apparent absence of a general  
466 structure. This conclusion is in line with observations made on the panels of drawings in other parts of the cave. The Ten  
467 Mammoths Frieze, in particular, forms a very structured and symmetric panel that can be regarded as a subset of the *Great*  
468 *Ceiling*. It is a significant example of a coherent panel drawn with one type of BaO-bearing Mn oxide pigment (intermediate  
469 BaO group) that was also found on the *Great Ceiling*. The question of whether the Ten Mammoths Frieze is contemporary  
470 with the figures on the *Great Ceiling* remains unresolved, although the two are consistent in terms of style and the pigments  
471 used. Still, our study demonstrates the value combined approaches to the non-invasive *in situ* analysis of prehistoric rock art  
472 in providing new organizational and chronological insights into sequences of creation in the absence of possibilities for  
473 absolute dating.

474  
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#### 482 483 **Appendix A. Supplementary data**

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485 Supplementary data to this article can be found online at <http://doi.org/10.1016/j.jasrep.2019.102006>.

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