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Research Article

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Inferring Upper Palaeolithic Human Mobility Strategies in the Pyrenees

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Abstract

The use of geochemistry to characterize chert tools recovered at hunter-gatherer sites is particularly interesting for inferring the territorial behavior of past societies and their mobility routes. In western Europe, the Pyrenean mountain range is one of these areas where analysing past human mobility is especially challenging. This mountain chain was believed to have been a barrier for Palaeolithic communities; however, recent studies have demonstrated that it was frequented and crossed by human groups almost throughout the last glaciation. To infer Upper Palaeolithic human mobility strategies in the Pyrenees, lithic artefacts recovered at Cova del Parco and Montlleó open-air site (Lleida, Spain) and Caune de Belvis (Aude, France) have been analysed using geochemical tools: energy dispersive X-ray fluorescence (ED-XRF), and laser-ablation inductively coupled plasma mass spectrometry (LA-ICP-MS). The results have shown that geochemistry is a useful tool for establishing differences between sources and for connecting archaeological cherts with specific formations.

Keywords: Chert; geochemistry; Palaeolithic mobility; Pyrenees

Introduction

Lithic artefacts are among the best preserved archaeological remains in Palaeolithic sequences from Western Europe and chert was one of the most exploited rocks during the period. The use of geochemistry to characterize chert outcrops and to relate chert tools recovered at hunter-gatherer sites with a specific geological formation has increased all over the world during the past decade [1-5]. Studies of this kind can help to infer the territorial behaviour of past societies and their mobility routes, especially regarding their

strategies for acquiring and moving lithic raw materials. The analysis of the territories frequented by Palaeolithic hunter-gatherer groups is particularly interesting in areas with specific environmental and orographic conditions such as mountain regions. In Western Europe, the Pyrenean mountain range is one of these areas where analysing past human mobility is especially interesting. This mountain chain naturally divides the Iberian Peninsula from the rest of continental Europe along the S-N axis, extending for almost 500 km from the Bay of Biscay to the Mediterranean Sea. During the 19th century,

the Pyrenees were defined as a border area separating France and Spain, and mobility during the Upper Palaeolithic was thought to have been restricted to the extremes of the mountain chain. However, environmental studies have demonstrated that ice retreat in this area occurred earlier than previously thought [6], making human occupation feasible throughout the Upper Palaeolithic and even during the Last Glacial Maximum [7]. Moreover, archaeological work has demonstrated that this mountain chain was frequented by different hominin groups from the Lower Palaeolithic onwards, at least in the eastern and western margins, and even Neanderthal populations crossed the Pyrenees at least at the western end, following what is known as the Basque corridor [8]. However, it is not until the arrival of the first modern humans that a regular frequentation of the Pyrenean mountain chain is clearly attested. Thus, studies focusing on the homogeneity of rock art [9], the

techno-typology of lithic tools [10] and the origin of the exploited rocks [11-13] prove that the Pyrenean chain was crossed regularly, and that contact between the two slopes was common. To infer the mobility strategies followed by human groups that frequented the Pyrenees during the Upper Palaeolithic, lithic artefacts recovered at three Magdalenian Pyrenean sequences have been analysed using geochemical tools. Three sites with archaeological sequences from the late Upper Palaeolithic located in the Eastern Pyrenees were selected for study: an archaeological site in northern Eastern Pyrenees (Caune de Belvis, Aude, France), another archaeological site in the south-eastern Pyrenees (Cova del Parco, Lleida, Spain) and an open-air site at the natural crossroads in the eastern part of the Pyrenees towards the coastal range (Montlleó open-air site, Lleida, Spain) (Figure 1).

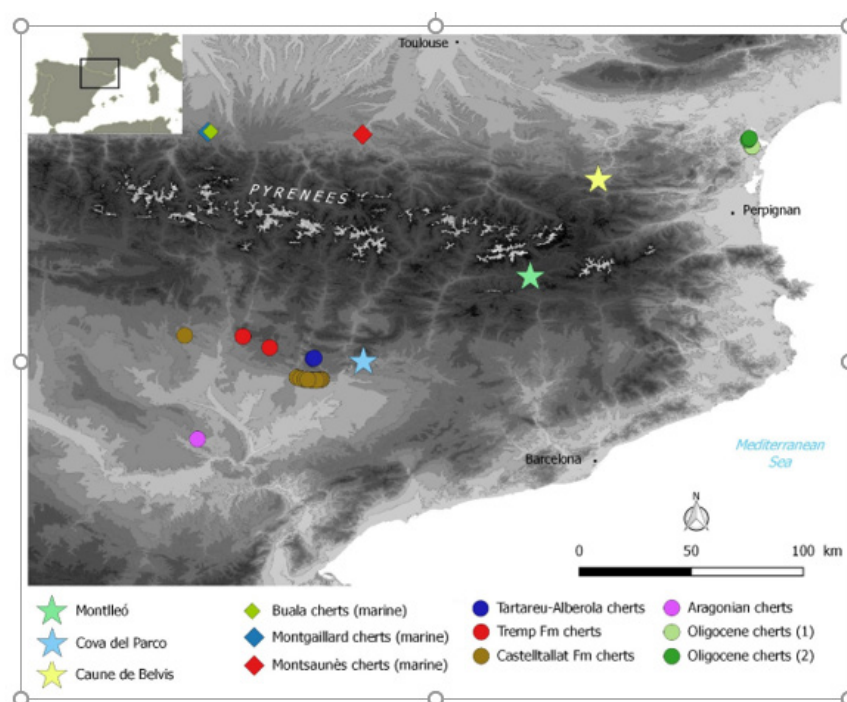


Figure 1: Location of the archaeological sites of Montlleó, Cova del Parco and Caune de Belvis and the studied outcrops containing lacustrine and marine chert varieties.

Materials and Methods

The open-air site of Montlleó is located at 1144 m asl in the Cerdanya valley, one of the largest high-altitude valleys in the Pyrenees. It is a crossroads for human circulation in the Eastern Pyrenees following the axes of the River Segre to the south and the River Têt to the north. The site was discovered in 1998 and archaeological work has been carried out systematically since 2000. Two different cultural human occupations have been identified up to now. The typology of the stone and bone materials as well as several radiocarbon dates indicate that the more recent occupation occurred during the Lower Magdalenian (22500 - 18500 cal BP). The earliest frequentation of the site dates from the Badegoulian period (23000 - 22500 cal BP) (Figure 2) and was characterized

by the abundance of retouched flakes, among them a few raclettes, and the presence in the same level of some Solutrean shouldered points that may indicate an even earlier phase of activity [14]. The stone materials from both levels vary widely, with exploitation of several types of chert, rhyolites and lydites, among others. Cova del Parco is in the south-eastern Pyrenees, in the Segre river valley, at 420 m asl, in a sheltered area. The site was discovered and first excavated in the 1970s and is still under excavation. It presents a long sequence of human occupation from, at least, the Middle Magdalenian (currently under excavation) to the Bronze Age [11,15]. The Magdalenian period is well represented, with several archaeological layers revealing occupations from the Late Upper Magdalenian (dated 15200 - 14600 cal BP), the Upper Magdalenian

(dated 16300 - 15500 cal BP) and the Middle Magdalenian, and already dated to 16800 - 15700 cal BP (Figure 2) [11]. Chert was

practically the only rock exploited, and different types have been identified under the stereoscopic and the petrographic microscopes.

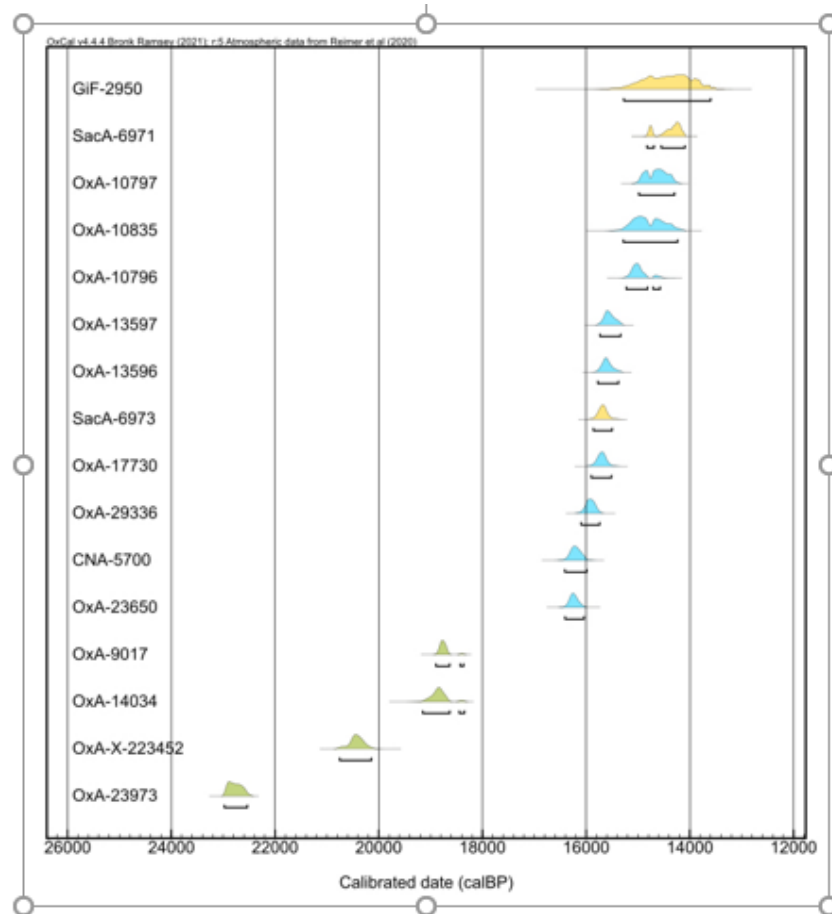


Figure 2: Radiocarbon dates with calibration after OxCal v.4.4.4 (Bronk Ramsey 2021) with IntCal20 calibration curve (Reimer et al., 2020).

Caune de Belvis is located in the north-eastern Pyrenees, in the department of Aude, in southeastern France, at 960 m asl. The cave opens to the south in a limestone massif from the Lower Cretaceous, the entire deposit covering an area of 65 sq m. Archaeological work carried out during the 1970s and 1980s identified the existence of two human occupations with a long hiatus between them. In this study we have analysed archaeological materials from the last human occupation of the cave, ascribed to the Upper Magdalenian (layers 1 to 4), dated to 16000-14000 cal BP by three radiocarbon dates (Figure 2) [16]. Chert was the most exploited rock, but rhyolites, lydites and jaspers were also used.

The first step in the analytical protocol was the observation of the archaeological collections through macroscopic characterization, to infer visual and micropalaeontological descriptions. This characterization was performed using a Leica M125 binocular microscope (from 6.7 to 100 x magnification) and a coupled Leica DFC420C model camera for the analysis of Caune de Belvis materials and a Olympus SZ61 (from 6.7 to 45 x magnification) and a coupled Olympus SC30 model camera for the analysis of the Montlleó and Cova del Parco sets.

Geological formations with cherts with similar visual and micropalaeontological characteristics were also analysed using the same methods with the aim of comparing and connecting archaeological samples with specific geological units. Several formations located on both slopes of the Central and Eastern Pyrenees as well as in the Bages-Sigean basin (in the southeast of France) and the Ebro Basin (in the north-east Iberia) were thus surveyed and the samples collected were studied under the same analytical conditions.

As the aim of the study was to analyse both geological and archaeological samples under the same analytical conditions, non-destructive and micro-destructive techniques were prioritized. Thus, after the macroscopic study, geochemical analyses were undertaken to quantify major, minor and trace components so as to be able to compare the raw materials used to make the archaeological tools with those from known geological outcrops.

To analyse major and minor elements, Energy Dispersive X-Ray Fluorescence (ED-XRF) was employed. One hundred and thirty-eight archaeological tools and 294 geological samples were studied at the Archeosciences Bordeaux laboratory in France. Nine element

concentrations were quantified as oxides (Na, Mg, Al, Si, p, K, Ca, Ti, Fe) using a SEIKO SEA 6000 VXX-ray fluorescence spectrometer, with fundamental parameters corrected by the granodiorite GSP2 from the US Geological Survey (USGS) international standard (Wilson, 1998). A 3 x 3 mm collimator was prioritized, and analysis time was set to 400 s for each measurement condition (three conditions with air or He environment and Cr or Pb filter were established). To check instrument calibration and accuracy, JCh-1 chert standard from the Geological Survey of Japan (GSJ) was used [17]. To prove and validate the formula used and to check instrument accuracy, a measurement with the JCh-1 chert standard was established. The standard deviation obtained was always below 0.08w%, thus validating the accuracy of the formula used [18].

Trace elements were analysed using Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) at the Ernest Babelon laboratory, IRAMAT, Orléans, France. Thirty-seven archaeological tools and 425 geological samples were studied. Elements were quantified using a Thermo Fisher Scientific Element XR mass spectrometer associated with a Resonetics Resolution M50e ablation device. This spectrometer has the advantage of being equipped with a dual mode (counting and analogue modes) secondary electron multiplier with a linear dynamic range of over nine orders of magnitude, associated with a single Faraday collector which allows an increase in the linear dynamic range by an additional three orders of magnitude. This feature is particularly important for laser ablation analysis of lithic samples, as it allows analysis of major, minor and trace elements in a single run regardless of their concentrations and their isotopic abundance. The ablation device was an excimer laser (ArF, 193 nm), which was operated at 7-8 mJ and 20 Hz; only if saturation was observed, conditions were reduced to 10 Hz. A dual gas system with helium (0.65 l/min) released at the base of the chamber, and argon at the head of the chamber (1.1 l/min) carried the ablated material to the plasma torch. Ablation time was set to 40 s, with 10 s pre-ablation to let the ablated material reach the spectrometer and 30-s collection time. Laser spot size was set to 100 µm and only reduced to 80 or 50 µm if saturation was detected. Line mode acquisition was chosen to enhance sensitivity. Background measurements were run every 10-20 samples. Fresh fractures were analysed on geological samples to reduce potential contamination. Priority was given to characterizing large numbers of samples; thus, only one ablation

line was carried out per specimen. However, if heterogeneities in element spikes were observed during analysis due to the presence of inclusions, the results were discarded and a new ablation location was selected.

Calibration was performed using standard reference glass NIST610 which was run periodically (every 10-20 samples) to correct for drift. NIST610 was used to calculate the response coefficient (k) of each element (Gratuze, 1999, Gratuze, 2014) and the measured values of each element were normalized against ²⁰Si, the internal standard, to produce a final percentage. Glass Standard NIST612 was analysed independently of calibration to provide comparative data. A total of 29 elements (Li, Be, B, Mg, Al, Si, Ca, Ti, V, Cr, Fe, Ga, As, Rb, Sr, Y, Zr, Nb, Cs, Ba, La, Ce, Pr, Nd, Sm, W, Bi, Th, U) were quantified.

Results

Chert was the most exploited rock in the three studied archaeological sets. Several varieties were identified during the first visual description but only two of them were selected for the application of geochemical methods; they form the basis of this study. The first were cherts originated in a lacustrine environment and characterized by a micropalaeontological content composed of sections of cells and oogonium of charophyte algae and some lacustrine gastropods as well as micritic mud residues, metal oxides and grains of detrital quartz as inclusions. The second variety were marine cherts with sponge spicules and, in some cases, macro foraminifera as the main micropalaeontological content, as well as abundant metal oxides and detrital quartz crystals as inclusions.

The lacustrine cherts

Several geological formations outcropping in northeast Iberia and south-east France contained cherts macroscopically similar to the archaeological cherts originating in a lacustrine environment. Outcrops were located in the Bages-Sigean basin in south-east France (Oligocene cherts from Étang du Doull outcrops and Port Mahon outcrops), in the Ebro basin (Aragonian cherts from Puente Candanos outcrop) and on the southern slope of the central Eastern Pyrenees (Tresp formation cherts from Mentirosa and Zurita outcrops, Castellatallat formation cherts from Castelló de Farfanya, Peraltila and Alfarràs outcrops and Oligocene cherts from Alberola outcrops) (Figure 1). A complete description and characterization of these formations can be found in [19,20].

Table 1: Median value of the lacustrine chert outcrops with the 9 elements quantified by ED-XRF.

Variable	Na2O	MgO	Al2O3	SiO2	P2O5	K2O	CaO	TiO2	Fe2O3
Alberola 1	<LOD	0,094	0,971	95,440	<LOD	0,163	3,015	0,043	0,274
Alberola 2	<LOD	0,082	0,750	95,924	<LOD	0,124	2,879	0,034	0,209
Alfarràs	<LOD	0,148	0,558	98,689	<LOD	0,050	0,360	0,011	0,183
Castelló de Farfanya	<LOD	0,018	0,605	97,068	<LOD	0,033	2,165	0,013	0,099
Mentirosa	<LOD	0,000	0,312	99,483	<LOD	0,010	0,178	<LOD	0,013
Peraltila	<LOD	0,000	0,416	96,681	<LOD	0,047	2,753	0,016	0,088
Port Mahon 1	<LOD	0,415	0,041	96,289	<LOD	0,035	3,166	0,001	0,051
Port Mahon 2A	<LOD	0,436	0,036	97,369	<LOD	0,025	2,090	<LOD	0,041
Port Mahon 2B	<LOD	0,107	0,215	97,986	0,013	0,026	1,613	0,001	0,037
Puente Candanos	<LOD	0,007	0,377	99,294	<LOD	0,023	0,282	0,003	0,015

Zurita	<LOD	0,000	0,430	99,253	<LOD	0,017	0,282	<LOD	0,017
Étang du Dou1	<LOD	0,010	0,300	99,519	<LOD	0,011	0,152	<LOD	0,009
Étang du Dou2	<LOD	0,012	0,477	98,759	<LOD	0,021	0,699	0,003	0,029
Étang du Dou3	<LOD	0,005	0,351	99,425	<LOD	0,013	0,182	0,001	0,019

Geochemistry was used to distinguish between formations and to link the archaeological samples with a specific outcrop, as since visual and micropalaeontological characteristics of the several units and outcrops were insufficient for this purpose. ED-XRF was applied to determine the major and minor chemical composition. Nine elements were quantified (Na₂O, MgO, Al₂O₃, SiO₂, P₂O₅, K₂O, CaO, TiO₂ and Fe₂O₃) but data relating to Na₂O and P₂O₅ were limited, as their values were below the instrument's detection limits. Median values were calculated for each geological outcrop (Table 1) and a Principal Component Analysis (PCA) was calculated using XLSTAT software [21] to see whether the quantified distinctions were visible in this approach.

PCA of the seven elements given results with the median of each geological outcrop and all the archaeological samples indicates certain differences between formations (Figure 3). In all, 74.05% of the total variance-F1 (57.60%) and F2 (16.46%) is represented

in the PCA. The large amount of archaeological samples is close to the median values of Oligocene cherts from Étang du Dou1, the Aragonian cherts from Puente Candanos outcrop, the Tremp Fm cherts from Zurita and Mentirosa outcrops and some cherts from the Castellatallat formation (Alfarràs and Castelló de Farfanya outcrops). However, the Oligocene cherts from the Alberola and Port Mahon outcrops do not conform entirely to the main dispersion of the archaeological samples. Similarly, the scatterplot for Log Al₂O₃/SiO₂ vs Log CaO/SiO₂ shows that the main dispersion of the archaeological samples mostly coincide with the median values of some Oligocene cherts from Étang du Dou1, the Aragonian cherts from Puente Candanos and the Castellatallat formation cherts from Castelló de Farfanya. In this regard, in addition to the discarded cherts from the Alberola and Port Mahon outcrops, the scatterplot revealed that the Zurita and Mentirosa outcrops (Tremp formation) were also far from conforming to the main dispersion of archaeological cherts (Figure 3).

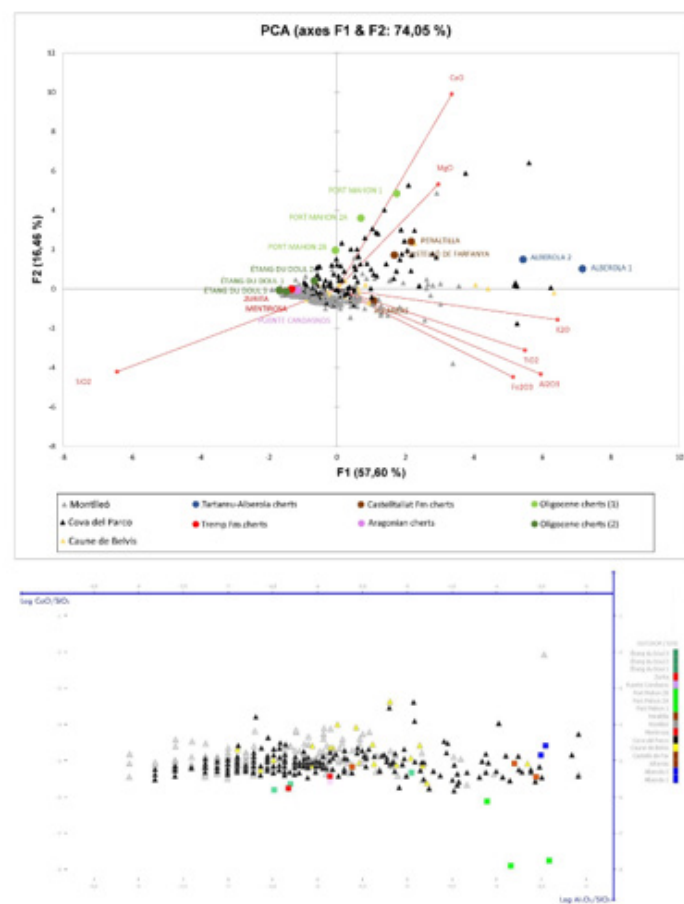


Figure 3: Top: Results of Principal Component Analysis (PCA) of the seven quantified elements by ED-XRF with the median value of each outcrop and all the archaeological samples accounting for the 74.05% of the total variance. Bottom: Scatterplot concerning Log Al₂O₃/SiO₂ vs Log CaO/SiO₂ and representing all the geological samples (squares in colors) and the archaeological samples from the three sites (triangles in colors).

As ED-XRF was not conclusive regarding the lacustrine cherts and in order to refine the connection between archaeological and geological cherts, LA-ICP-MS was conducted to quantify the trace elements. Twenty-nine elements were quantified, and some of them were able to distinguish between the different geological formations and thus make it possible to connect the archaeological chert samples directly. For example, the scatterplot for Log As/U vs Log W/U identified differences between the three cherts probably used (Aragonian cherts from Puente de Candanos, Oligocene cherts from Étang du Doul and Castellallat formation cherts from Castelló de Farfanya). The archaeological samples from Montlleó

seem to connect mainly with the main dispersion of Étang du Doul and Castelló de Farfanya, with only one sample being connected to the Puente de Candanos outcrop. Cherts from Cova del Parco are mostly linked to the Castelló de Farfanya main dispersion, but a large number of samples do not fit with any geological group studied. This may mean that other as yet unstudied outcrops or formations may have been frequented by past groups settled at Cova del Parco site. Finally, the archaeological cherts from Caune de Belvis fit entirely with the main dispersion of Étang du Doul group (Figure 4).

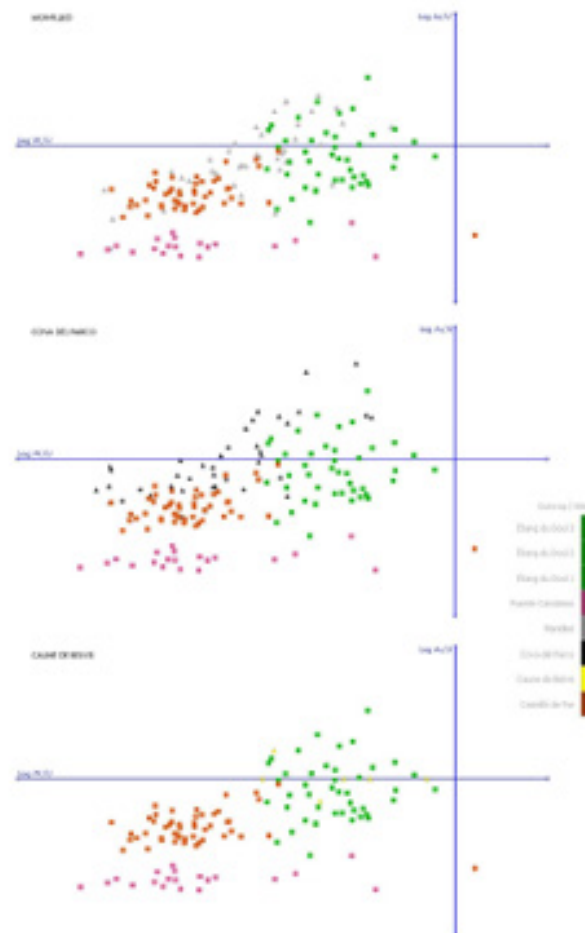


Figure 4: Scatterplot for Log W/U vs Log As/U quantified by LA-ICP-MS and representing the geological samples from Castelló de Farfanya, Puente Candanos and Étang du Doul outcrops (squares in colors) and the archaeological samples from each site (triangles in colors): on the top Montlleó, on the middle Cova del Parco and on the bottom Caune de Belvis.

The marine cherts

The archaeological cherts identified at the three archaeological sites and macroscopically defined as originating in a marine environment, characterized by the abundance of metal oxides, grains of quartz crystals and abundant sponge spicules were similar to three different chert sources outcropping in the northern slope of the Central Pyrenees: the Campanian-Maastrichtian flysch cherts from the Buala outcrop, the Maastrichtian cherts from

the Nankin Formation (Montsaunès outcrop) and the Turonian-Santonian cherts from the Montgaillard outcrop. A complete description of these cherts can be found in [5]. First, ED-XRF analyses were conducted to determine the major and minor components of geological and archaeological samples, but they did not yield conclusive results, as the geological sources possessed similar values. Therefore, LA-ICP-MS was run to identify and quantify trace elements and try to establish differences between

sources. The results showed that some trace elements (Ti, Sr and Th) could help to discriminate between outcrops. The scatterplot concerning Log Ti/Sr vs Log Th/Sr clearly separates the three sources and archaeological samples can be directly associated to a specific source. For example, the archaeological tools from Montlleó

fit with both the Montgaillard and Montsaunès main dispersions. In the case of Cova del Parco, it seems that all the samples fit with the Montgaillard group. Finally, as at Montlleó, the chert tools from Caune de Belvis can be connected with both Montgaillard and Montsaunès main dispersion (Figure 5).

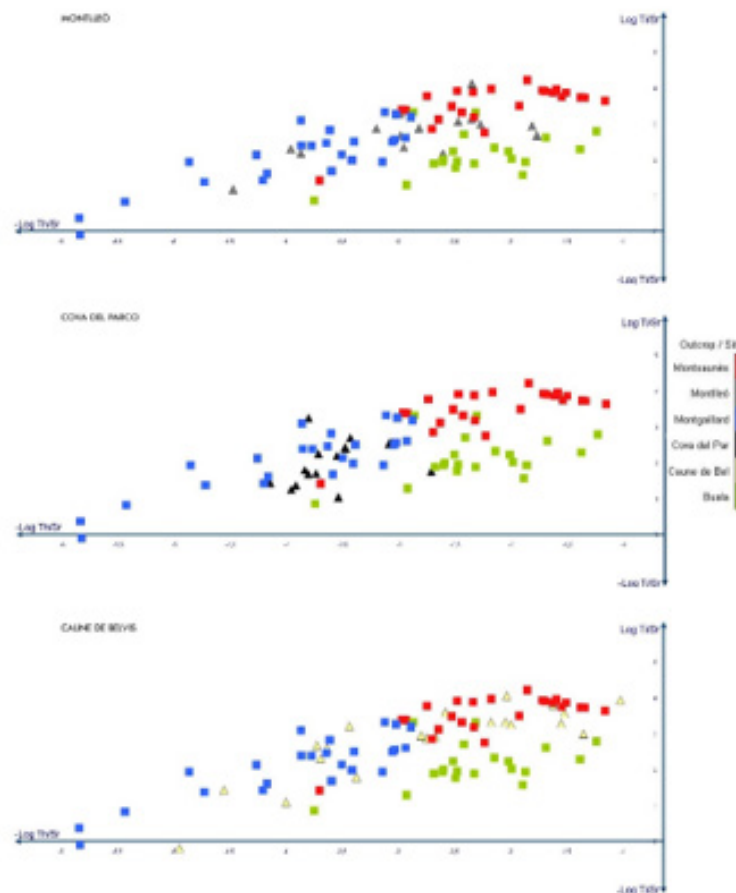


Figure 5: Scatterplot concerning Log Ti/Sr vs Log Th/Sr quantified by LA-ICP-MS with the three geological marine outcrops (squares in colors) and the archaeological samples from each site (triangles in colors): on the top Montlleó, on the middle Cova del Parco and on the bottom Caune de Belvis.

Discussion

The geochemical analysis of the main chert types exploited at the Magdalenian occupations of Montlleó, Cova del Parco and Caune de Belvis indicates a large territory of lithic raw material acquisition involving both slopes of the Pyrenean mountain chain. The application of geochemical methods to refine the first visual and micropalaeontological description has made it possible to directly connect the archaeological samples studied with specific geological units from cherts outcropping in NE Iberia and the southeast of France (Figure 6). The lacustrine cherts were the most exploited type in the three collections studied, and were mostly collected from regional outcrops at Caune de Belvis (with frequentation of Étang du Doul outcrops in the Bages-Sigean basin) and Cova del Parco (with frequentation of Castelló de Farfanya outcrops, in the contact between the Ebro basin and the first Pre-Pyrenean

mountain ranges). However, some samples from Cova del Parco do not directly fit with any of the geological cherts studied. Thus, either post-depositional processes may have altered the chemical composition of some samples, or other as yet unstudied sources may have been exploited. At Montlleó, in an area without good-quality chert sources, cherts located both north of the Pyrenees (Étang du Doul cherts) and south of the mountain range (Castelló de Farfanya cherts) were frequented. This duality in the acquisition of the most exploited chert type proves that the Magdalenian groups that settled the site regularly crossed the Pyrenean mountain chain.

The marine cherts studied in this paper had their origin in the northern slope of the Central Pyrenees. Two main sources seem to have been exploited by Magdalenian groups. While in Caune de Belvis and Montlleó open-air site both Montgaillard and Montsaunès outcrops seem to have been frequented, groups settled

at Cova del Parco exploited only the Montgaillard cherts. This difference between sites may be linked to the specific preferences of these groups or to the existence of cultural barriers.

Thus, the groups that settled Montlleó regularly frequented both Pyrenean slopes, acquiring cherts both to the north and to the south of the Pyrenean mountain range. In Cova del Parco it seems that the most exploited chert type was obtained from regional sources, but the northern Pyrenees were also frequented in order to acquire the marine cherts that regularly arrived at the site. In

Caune de Belvis, the regional lacustrine cherts were the most exploited, but a connection with the central northern Pyrenees is attested with the exploitation of the marine sources. Our studies of other chert types exploited at this site have also shown that the southern Pyrenees were also frequented by groups settled at Caune de Belvis, thus demonstrating that the Pyrenean mountain chain was not a barrier for these groups. In this sense, it seems that the Têt and Segre rivers could have functioned as main axes for Palaeolithic human mobility in the Eastern Pyrenees [22].

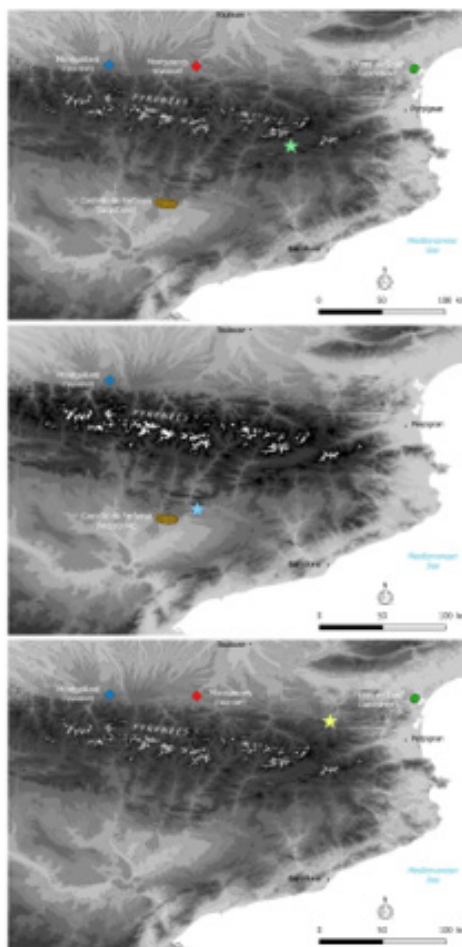


Figure 6: Location of each archaeological site (on the top Montlleó, on the middle Cova del Parco, on the bottom Caune de Belvis) and the connected outcrops for each site after the geochemical study of the lacustrine and marine varieties.

Conclusion

This study shows that geochemistry is a useful tool for establishing differences between geological sources and thus for connecting archaeological samples with specific formations [23]. The data obtained have allowed us 1) to define the mobility strategies followed by human groups that settled the Pyrenees during the last glaciation, and 2) to demonstrate that the mountain chain was not just sporadically crossed by Magdalenian groups; similar strategies for acquiring the lithic raw materials were conducted by past groups inhabiting the Central Eastern Pyrenees

[24]. However, specific strategies were applied at each site, mostly involving exploitation of the closest areas to obtain most of the material, but also procuring cherts whose origin is in some cases more than 100 km from the site [25]. Future studies incorporating other archaeological sites and chert types are planned to obtain further insights into the human societies inhabiting Western Europe during the Upper Palaeolithic [26].

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Conflict of Interest

No conflict of interest.

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