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RESEARCH

Inferring the territoriality of Upper Palaeolithic hunter-gatherer's groups settled at Cueva del Gato 2 (Épila, Zaragoza)

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Abstract

In recent decades, the development of different analytical procedures applied to the study of archaeological lithic remains has allowed us to approach the territoriality of past societies. The application of geochemical tools has improved the study of lithic raw materials, allowing direct connections between archaeological samples and specific geological formations. In a similar way, the incorporation of GIS tools to the study of past mobility and territoriality has allowed to define which could have been the most probable routes used by past groups to stock up on rocks. In this paper, we present the results obtained after the geochemical study by Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) of lithic cherts found at Upper Palaeolithic human occupations at Cueva del Gato 2 (Épila, Zaragoza, Spain) as well as the least cost path routes obtained after GIS analyses. The Cueva del Gato 2 archaeological site is a large cave placed in the first foothills of the Iberian System, close to the Middle Ebro Basin, in Eastern Iberia. The archaeological work has revealed a long sequence of occupation with archaeological levels from the Bronze Age, the Ancient Neolithic and the Upper Palaeolithic. In this paper, we have focused on the study of level II, which has been dated in the range 21,000–23,000 cal BP. Results have demonstrated that past human groups from the Last Glacial Maximum knew and frequented the Sierra de Lanaja – Montes de Castejón lacustrine chert sources, outcropping in the Middle Ebro Basin.

Keywords Chert sourcing · Human mobility · Geochemistry · Upper Palaeolithic · Ebro Basin

Introduction

This study constitutes the first step towards defining the territoriality and the mobility of the hunter-gatherer human populations that settled the first foothills of the Iberian System near the Middle Ebro Basin. It is based on the analysis of the chert lithic remains recovered at the archaeological site of Cueva del Gato 2 (Épila, Zaragoza province, Spain). Chert was the rock preferred by Upper Palaeolithic communities who settled in this location. Thus, thanks to the study of this

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archaeological site, we can gain a deeper knowledge not only of the lithic raw materials acquisition strategies of these populations, but also of their relation with the geographical space.

The study of chert as a reliable marker to trace past human mobility and territoriality started several decades ago and has largely improved in recent years, with the implementation of non-destructive and accurate techniques. Thus, to characterize the lithic assemblage, we are nowadays moving beyond the classic approach mostly based on the use of destructive and sometimes limited techniques like petrographic studies. If it is true that petrography is one of the best ways to define the mineralogical phases in rock characterization, it involves the destruction of the artefact. Moreover, the macroscopic approach at the stereoscopic microscope often does not enable lithic artefacts to be directly connected with a specific geological origin in the case of facies convergence. To do so, and to generate reliable data, in the last decades, the use of quantitative approaches has been introduced with the use of spectroscopy and geochemistry to characterize lithic sources and archaeological remains (Finkel et al. 2022; Olausson et al. 2017; Parish 2018; Pereira et al. 2021; Sánchez de la Torre et al. 2019a, b; Sánchez de la Torre et al. 2020a, b, c; Senesi et al. 2023; Stewart et al. 2020).

In this paper, we apply multielemental analysis using Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) to quantify major, minor and trace elements and thus characterize chert tools. The study is based on the use of a non-destructive or micro-destructive technique that offers replicable and repeatable data, which can then be compared with other geological sources or archaeological materials analyzed under the same analytical procedure (Brandl et al. 2018; Gurova et al. 2021; Sánchez de la Torre et al. 2017; Speer 2014). Moreover, based on the results obtained with this multielemental analytical approach, we include spatial analysis to determine the mobility of these past societies. The tools offered by the Geographical Information Systems (GIS) make it possible to suggest the probable routes that could have been used by past populations for acquiring lithic raw materials and in what manner the orographic conditions could have affected these interactions (García-Rojas et al. 2017; Prieto et al. 2016; Sánchez et al. 2016).

Cueva del Gato 2 archaeological site

The archaeological site of Cueva del Gato 2 is located on the left bank of the Ebro River Basin, 10 km from the Jalón River, in the foothills of Sierra de la Virgen or Sierra de Rodanas, which is an extension of the Iberian System towards the Ebro Basin. The coordinates of the site are: ETRS89 / UTM 30 T X 631661 – Y: 4,608,069. The site is situated in the municipality of Épila, at 620 m asl, in a limestone dolomitic system from the Jurassic that structures the aforementioned Mountain Range. The cave, which is south facing, possesses two entrances, the main one of which has a maximum length of 11 m and a maximum heigh of 3 m. Inside, there is a large room (22 m × 15 m) well lit by sunlight. The presence of a masonry wall reveals the use of the cave until recent times. Archaeological works were developed in the main room in three archaeological seasons between 2002 and 2011 by a team from the University of Zaragoza, Spain. Four archaeological surveys were made. Here we focus our attention on one of them (Survey II), made in the central area of the large room. Up to three sedimentary levels were identified (levels I to III), of which level II attracted our attention. This level II was documented in 10 m^2 and several hundreds of archaeological materials were recovered, which include lithic tools, faunal remains, bone industry remains and perforated marine shells. With a sedimentary depth of 30 cm, this archaeological level has been dated by radiocarbon to 21,000–23,000 cal BP (Fig. 1) (Blasco and Rodanés, 2009; Rodanés et al. 2021; Utrilla et al. 2012, 2010). The lithic industry is composed of 1.381 chert samples, and is characterized by large blades and flakes, while burins and scrappers are the most abundant retouched tools, followed by troncatures. The bone industry is formed by 30 tools, also including a pendant (Rodanés et al. 2021), the javelins being the most abundant remain, followed by perforated marine shells. The set is completed by a needle and an awl.

The faunal remains, currently under study, are very abundant, with more than 4000 samples being recovered during the first archaeological season. The most documented specimen is the rabbit, representing more than the 90% of the faunal remains, followed by the deer, the horse, the ibex and the lynx. Occasionally, also birds were identified. Generally, the faunal bones belong to adult animals and all or almost all the skeletal parts are represented. Some of them possess burned evidences and, in general, they present typical fractures of the processing when consumed by humans (Blasco and Rodanés 2009).

Palaeoclimatic conditions have only been analyzed by anthracology, as pollens have not been recovered. Thus, charcoals, which present an excellent state of preservation, have been studied, the *Juniperus* being the only documented specimen. Nevertheless, these results are consistent with other regional studies, suggesting that, during the Last Glacial Maximum, conditions of extreme aridity and low temperatures prevailed (Badal et al. 2012).

The cultural ascription of this archaeological levels is complicated and not yet solved. The radiocarbon dates suggest that these occupations could be related to the Badegoulian period, well documented in the northern Pyrenees and the Cantabric region (Corchón et al. 2015; Ducasse et al. 2017). However, the lithic technocomplex does not entirely fit with the traditional Badegoulian horizon, *raclettes* being the most representative tool, which has not yet been identified at Cueva del

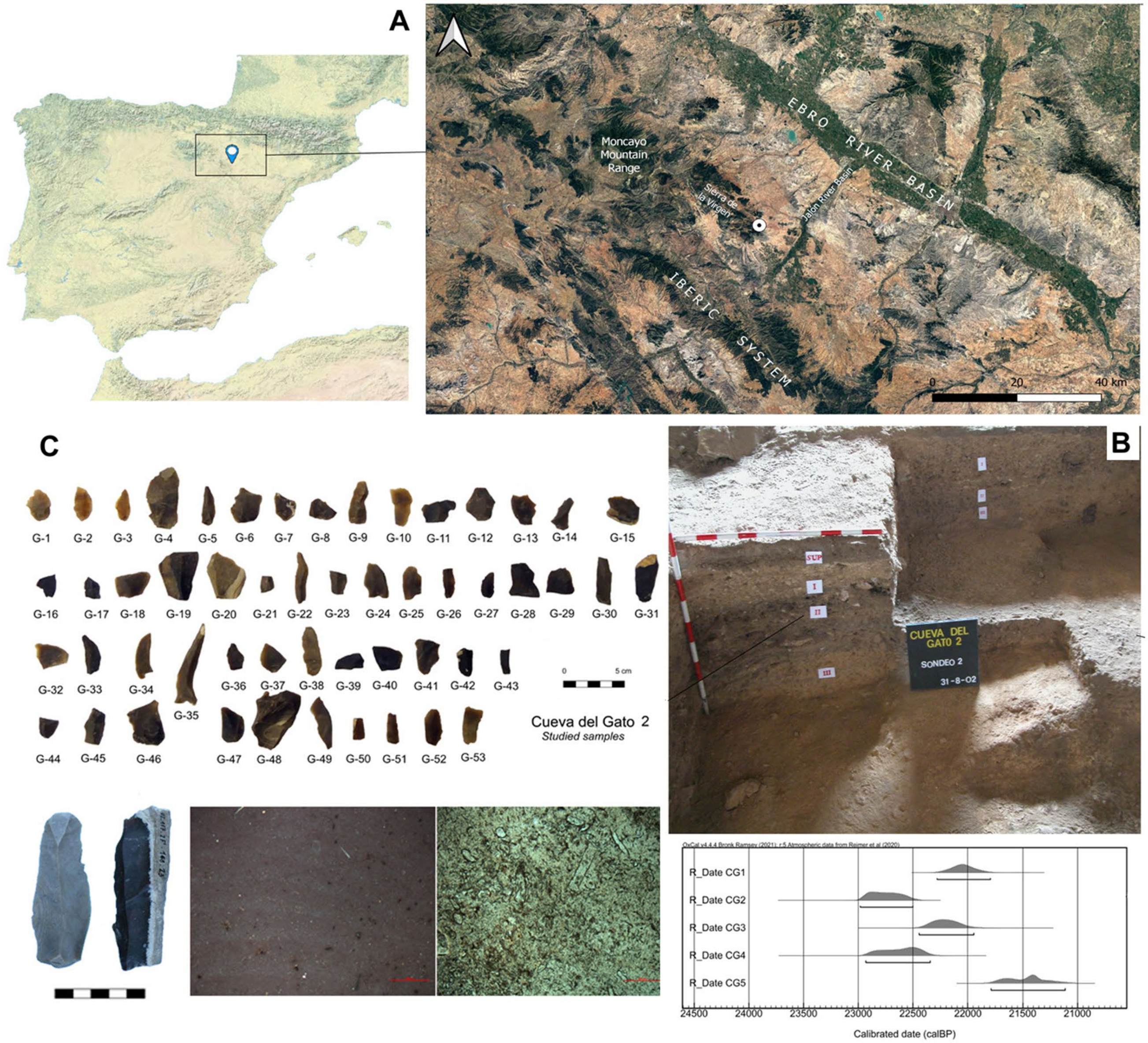

Fig. 1 (**A**) Location of Cueva del Gato 2 in the Middle Ebro Basin, in NE Iberia. (**B**) Archaeological grid and stratigraphic sequence of the survey II, with the calibration of the radiocarbon dates of level II (Ramsey 2017). (**C**) Archaeological chert remains selected for LA-ICP-MS analysis and main texture of archaeological samples observed at the petrographic scale (source of macroscopic and petrographic images: (Leorza et al. 2017)

Gato 2. Nevertheless, the characteristics of the lithic *chaîne opératoire* and the typology of the bone industry could fit well within this Badegoulian horizon, as regards a javelin typologically defined as Placard type (Séronie-Vivien 2005).

The lithic set ascribed to the Upper Palaeolithic human occupation from Cueva del Gato 2 is composed of 1381 (level II) lithic samples. Chert was almost the only rock for confectioning the lithic assemblage. The preliminary observation of almost the entire set (1099 samples), made by one of us (E.D.), indicated that 65% of the assemblage (715 samples) could be related to the same macroscopic texture. Macroscopically defined as *Monegros* type, this chert variety possesses brownish colors with an extremely fine grain and a good knapping aptitude given the absence of internal fractures. Their texture can be distincted as a wackestone type, with abundant mud carbonatic relicts and fragments of Charophyte algae and lacustrine gastropods. In some specimens, Liesegang rings are observed. The inclusions content indicates that this chert type originated in a continental lacustrine sedimentary environment. The anecdotic number of cores and the low averages of cortical surfaces (the 5% of this chert type) as well as some remounts suggest that lithic arrived preformed as supports and only the confection and repair of tools was done at the site. A petrographic characterization was done for a selection of archaeological samples. A microquartz mosaic fabric was the main silica texture observed, with some inclusions of subangular grains of detrital quartz. Metal oxides were also identified, as well as remains of carbonate residues, previous to the silicification, in the form of bioclastic remains in process of silicification and mud remains (Fig. 1D).

Another chert variety was identified during the macroscopic approach. They were cherts originated in a hypersaline sedimentary environment, without bioclastic remains and just scarce inclusions of metal oxides and lenticular gypsum pseudomorphs were identified in specific samples. This chert variety represented the 11% of the studied assemblage. The remaining 24% were cherts whose ascription was not possible mostly due to postdepositional alterations (Leorza et al. 2017).

The geological setting: lacustrine chert outcrops

Four geological units outcrop in the Monegros region of the Middle Ebro Basin: the Sierra de Pallaruelo – Monte de la Sora Unit, the Sierra de Lanaja – Montes de Castejón Unit, the Bujaraloz – Sariñena Unit and the Torrente de Cinca – Alcolea de Cinca Unit. The remaining three geological units outcrop in the southeastern Pyrenean foothills: being the Tremp formation, the Castelltallat formation and the Tartareu – Alberola Unit. We briefly describe each unit below, which has been fully described and macroscopically and petrographically characterized in our previous works (Sánchez de la Torre et al. 2019a, b; Sánchez de la Torre et al. 2017) (Fig. 2).

1. The **Sierra de Pallaruelo – Monte de la Sora Unit** (Lower Aragonian, Miocene) is composed at its base of pelitic sediments of alluvial origin outcropping in the Ebro Basin. The top of the unit is practically entirely made up of margo-carbonate levels of lacustrine origin that regularly contain lacustrine chert nodules (Quirantes 1978). Chert outcrops were identified where at the present time stands the hermitage of Santa Quiteria, near the municipality of La Almolda (Zaragoza province) (Santa Quiteria outcrop) (SQ) and near the villages of Muel and Mezalocha (Zaragoza province), in the mounts of San Borombón. (San Borombón 1 and San Borombón 2 outcrops) (SB1 and SB2).
2. The **Sierra de Lanaja – Montes de Castejón Unit** (Upper Aragonian, Miocene) outcrops in the Eastern area of the Ebro Basin and is mostly constituted by distant alluvial facies and fan border facies at the base, and margo-

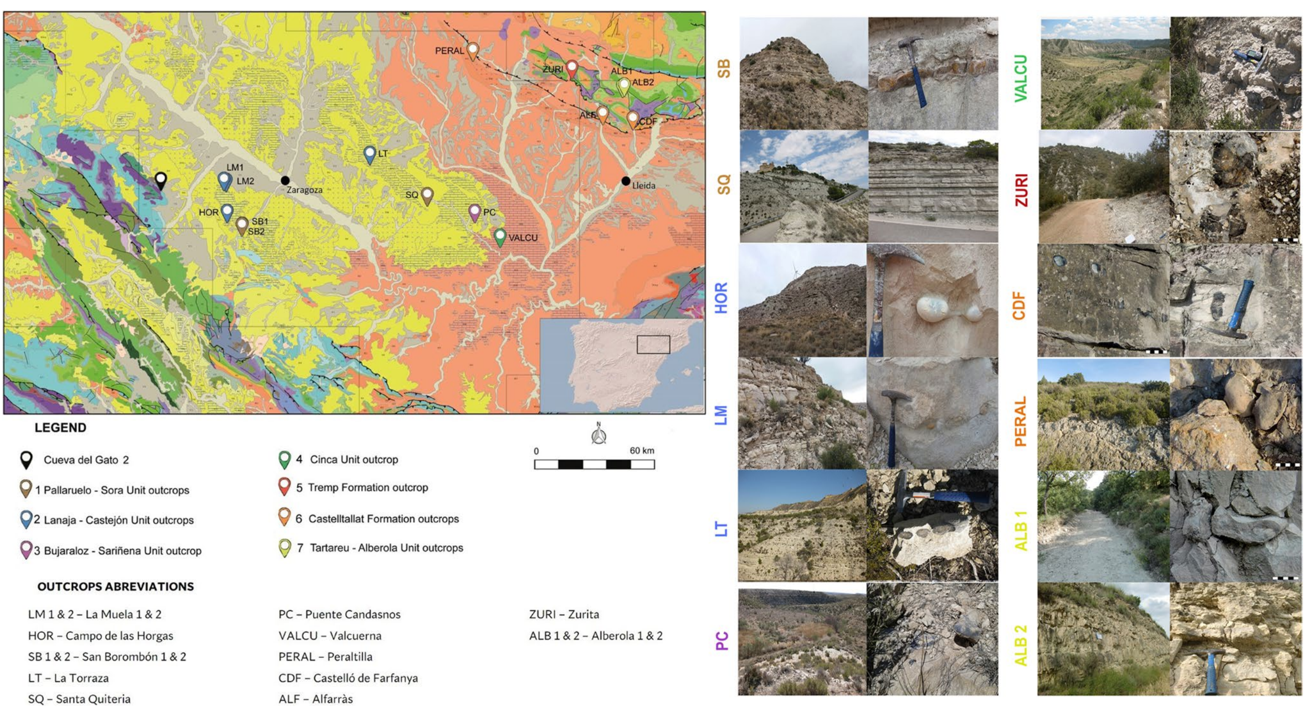

Fig. 2 Main location of the archaeological site and the outcrops analysed in the framework of this study in a geological map. Source: www.infoIGME.es/visor, modified

carbonate sediments created in a lacustrine environment at the top. Between their lithologies, chert is found, being one of the best-quality chert types from the Monegros region (Quirantes 1978). Different outcrops were identified near the town of Farlete (Zaragoza province), in the San Caprasio mount (La Torraza outcrop) (LT), the villages of Muel (Zaragoza province) (Campo de las Horgas outcrop) (HOR) and La Muela (Zaragoza province) (La Muela 1 and La Muela 2 outcrops) (LM1 and LM2). In the La Muela outcrops, sign of past knapping was found during the surveys, these being frequently related to the production of gunflints during the eighteenth and nineteenth centuries (Barandiarán 1974; Tarriño et al. 2016).

3. The **Bujaraloz – Sariñena Unit** (Aagenian – Aragonian, Miocene) is mainly composed of evaporitic facies present in a large area of the central sector of the Ebro Basin. In the south-eastern sector of the Ebro Basin, these materials are crowned by carbonate levels of lacustrine origin, where cherts appear implanted (Quirantes 1978). A chert outcrop was located near the municipalities of Peralba and Candasnos (Zaragoza province) (Puente Candasnos outcrop) (PC).
4. The **Torrente de Cinca – Alcolea de Cinca Unit** (Chattian – Agenian, Miocene – Oligocene) appears as detrital facies in the northern and southern margins of the Ebro Basin. In the central area of the basin it is largely constituted by carbonated sediments created in shallow lacustrine conditions. Chert appears embedded within these carbonate deposits, largely outcropping in the Valcuerna ravine, near the town of Peñalba (Huesca province) (Valcuerna outcrop) (VALCU).
5. The **Tremp Formation** (Maastrichtian, Upper Cretaceous) outcrops in the Carrodilla mountain range, a Pre-Pyrenean foothill placed between the Cinca and the Noguera Ribagorzana rivers. This formation holds a level of laminated micritic limestones with embedded nodular cherts, that originated in a lacustrine sedimentary environment (García Senz et al. 1990). Several outcrops were located in the Carrodilla mount, from which one outcropping near the municipality of Zurita (Huesca province) has been selected for geochemical analysis (Zurita outcrop) (ZURI).
6. The **Castelltallat Formation** (Rupelian, Oligocene) largely outcrops in the Serra Llarga mountain range, which is located between the towns of Castelló de Farfanya and Alcarràs (Lleida province), in the contact among the Pre-Pyrenees and the Central Depression (Anadón et al. 1989; Ortega et al. 2018). Nodular cherts appear within the stratified limestones in the Serra Llarga (Castelló de Farfanya and Alfarràs outcrops) (CDF and ALF) and also near the village of Peraltilla (Huesca province), 20 km to the west of Serra Llarga and close to the Cinca River (Peraltilla outcrop) (PERAL).

Table 1 Reference of the archaeological samples studied in the context of this work and their specific provenance. All of them were recovered in the Survey II and are ascribed to the Upper Paleolithic humans' occupations

Reference	Square	Depth (cm)	Reference	Square	Depth (cm)
G-1	6E'	-196	G-28	4D'	-185
G-2	6E'	-196	G-29	4D'	-185
G-3	6E'	-196	G-30	4D'	-185
G-4	4D'	-205	G-31	4D'	-185
G-5	4D'	-205	G-32	2D'	-190
G-6	4C'	-222	G-33	4F'	-170
G-7	4C'	-222	G-34	4B'	-103
G-8	6E'	-196	G-35	4D'	-188
G-9	4B'	-264	G-36	4C'	-215
G-10	4B'	-264	G-37	4C'	-208
G-11	4F'	-170	G-38	2D'	Cut
G-12	2D'	-186	G-39	2D'	-175
G-13	2D'	-186	G-40	4D'	-174
G-14	4C'	-196	G-41	2D'	-194
G-15	4C'	-196	G-42	2D'	-194
G-16	2D'	-190	G-43	2D'	-194
G-17	2D'	Cut	G-44	4C'	-222
G-18	2D'	-190	G-45	2D'	-190
G-19	4E'	-175	G-46	2B'	-249
G-20	2D'	-211	G-47	2D'	-190
G-21	4F'	-170	G-48	4D'	-205
G-22	4C'	-215	G-49	2D'	-182
G-23	4D'	-188	G-50	4D'	-188
G-24	4D'	-205	G-51	4F'	-170
G-25	4D'	-205	G-52	4D'	-205
G-26	6E'	-196	G-53	2D'	-194
G-27	6E'	-196			

7. The **Tartareu – Alberola Unit** (Rupelian, Oligocene) appears within the lacustrine stratified limestones outcropping in the Sant Miquel mountain range, a Pre-Pyrenean mountain chain located to the north of Serra Llarga and being limited by the Noguera Ribagorzana and the Farfanya rivers (IGC 2008). Two outcrops were identified near the town of Alberola (Lleida province) (Alberola 1 and Alberola 2 outcrops) (ALB1 and ALB2).

Materials and methods

Materials

The study presented here has focused on the analysis of a selection of 53 lithic fragments recovered at the archaeological survey II and mostly ascribed to level II, which is probably related to a human occupation of the cave during the Badegoulian period (Table 1). The preliminary visual

Table 2 Reference of the geological outcrops studied in the context of this work and the number of samples analysed by LA-ICP-MS

Reference	Outcrop name	Formation	Age	UTM zone	Coordinates	LA-ICP-MS
HOR	Campo de las Horgas	Lanaja – Castejón Unit	Miocene	30	659754, 4592686	20
LM1	La Muela 1	Lanaja – Castejón Unit	Miocene	30	656671, 4604842	10
LM2	La Muela 2	Lanaja – Castejón Unit	Miocene	30	656447, 4604961	20
LT	La Torraza	Lanaja – Castejón Unit	Miocene	30	707804, 4617567	13
SB1	San Borombón 1	Pallaruelo – Sora Unit	Miocene	30	660105, 4592066	11
SB2	San Borombón 2	Pallaruelo – Sora Unit	Miocene	30	660153, 4592276	20
SQ	Santa Quiteria	Pallaruelo – Sora Unit	Miocene	30	732866, 4603917	12
PC	Puente Candasnos	Bujaraloz – Sariñena Unit	Miocene	30	747366, 4598483	20
VALCU	Valcuerna	Cinca Unit	Miocene—Oligocene	31	749859, 4591833	20
CDF	Castelló de Farfanya	Castelltallat Formation	Oligocene	31	308619, 4631742	49
ALF	Alfarràs	Castelltallat Formation	Oligocene	31	298254, 4634033	6
PERAL	Peraltilla	Castelltallat Formation	Oligocene	30	746514, 4660207	20
ALB1	Aberola 1	Tartareu-Alberola Unit	Oligocene	31	288003, 4651622	20
ALB2	Alberola 2	Tartareu-Alberola Unit	Oligocene	31	307168, 4645549	17
ZURI	Zurita	Tremp Formation	Maastrichtian	31	307413, 4645217	13

and micropalaeontological description to define the main macroscopic textures indicated that the 53 selected lithic remains belonged to the same chert type, originated in a continental lacustrine sedimentary environment. Thus, once the type of chert was defined, geological samples that macroscopically fitted with the archaeological specimens were included in the study with the aim of finding the probable sources. These comprised samples from all the lacustrine chert sources outcropping in the Middle Ebro Basin and the southeastern Pyrenean foothills, being a total of seven geological formations studied, with 15 geological outcrops and 271 samples analyzed (Table 2).

Geochemical analytical approach

To improve the analysis time and to avoid surface alterations, the geological samples were prepared in squares of 5×5 mm, removing cortex surfaces. Archaeological samples with plat surfaces and without cortical or postdepositional alterations were preferred. Before the analysis, both geological and archaeological remains were cleaned with distilled water and ethanol. Then, geochemical analyses were undertaken to quantify major, minor and trace elements. Analyses were done at the Ernest Babelon laboratory, IRAMAT, Orléans, France. 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 is possible to analyse 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 and only if saturation was observed, were the conditions 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 25 s; 8 s pre-ablation to let the ablated material reach the spectrometer and 17 s collection time. The 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 characterising a large number of samples; thus, only one ablation line was carried out per specimen. However, if element spikes due to the presence of inclusions or heterogeneities were observed during analysis, 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, 2014), and the measured values of each element were normalised against 29Si, the internal standard, to produce a final percentage. Glass Standard NIST612 was analysed independently of the calibration to provide comparative data. A total of 29 elements were quantified (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). The results were statistically analyzed using the XLSTAT software (Addinsoft 2022).

GIS analytical approach

Regarding the GIS analytical method, we present in this paper a first approach to the study of the territorial connections that can be inferred from the archaeological site with the chert outcrops which, according to the geochemical analyses, possess a similar elemental chemical composition. To do this, we start with the methodology proposed by researchers from the Department of Geography, Prehistory and Archaeology of the University of the Basque Country (García-Rojas et al. 2017; Prieto et al. 2016; Sánchez et al. 2016). The methodology is based on the development of the "cost unit" as a way of measuring the physical effort on a human scale and its translation to an area's geographical space, in this case the Middle Ebro Basin. To develop this analysis, we have used the free access program QGIS in its version 3.22.

The spatial study starts with the Digital Elevation Model (DEM) from the STRM-Downloader plugin, which provides terrain models from NASA's EARTHDATA service. Once the rasters of the northeastern area of the Iberian Peninsula are combined, the *r.slope.aspect* function of the GRASS software, implemented in the aforementioned version of QGIS, is applied to establish a connection between the altitude of the relief and the slope degree of the terrain. With this, a raster is generated, on which a reclassification function is applied based on the slope-cost unit calibration curve to establish a correlation between the measured slope in degrees and the variable physical effort depending on the slope. The *r.cost* function, also present in the GRASS software, is applied to the raster obtained to generate an Accumulated Cost Map that starts from the geographical coordinates of the archaeological site and whose range of values is segmented into 52 lines of cost. Finally, the Least Cost Path function integrated in QGIS is applied to calculate the optimal paths from the archaeological site to the outcrops, based on the slope raster.

Results and discussion

Geochemistry and statistics

Geochemical analyses were developed to determine the elemental chemical composition of a selection of lithic tools recovered at Cueva del Gato 2 which were then compared with a series of geological samples with similar macroscopic features. Firstly, a descriptive analysis was run to identify which where the elements that presented the largest standard deviation (Table 3). Despite having quantified 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), values obtained for Ge were generally below the detection

Table 3 Descriptive analysis with the minimal value (Min. V.), the maximal value (Max. V.) and the standard deviation (Std Dev) for each of quantified element by LA-ICP-MS

	Li	Be	B	Mg	Al	Si	Ca	Ti	V	Cr	Fe	Ga	As
Min. V	0,02	0,01	9,29	22,32	11,9	264705,7	5,37	5,41	0,13	0,01	63,08	0,555	0,01
Max. V	43,97	6,54	146,26	6284,4	35849,5	467215,2	300919,4	2061,33	410,76	37,50	16269,96	279,72	42,51
Std Dev	8,80	0,39	18,26	1047,6	3708,5	30029,5	44826,7	245,18	24,01	4,81	1893,45	29,34	7,69

	Rb	Sr	Y	Zr	Nb	Cs	Ba	La	Ce	Pr	Nd	Sm	W	Bi	Th	U
Min. V	0,09	2,42	0,01	0,11	0,03	0,01	1,05	0,02	0,04	0,01	0,02	0,01	0,01	0,02	0,02	0,24
Max. V	58,53	1612,77	15,39	65,72	5,41	11,91	1200,51	23,89	48,08	4,33	15,14	2,33	35,26	10,94	5,94	496,46
Std Dev	6,43	202,65	1,31	7,07	0,67	0,78	139,28	2,23	4,21	0,40	1,49	0,25	3,29	1,17	0,62	54,52

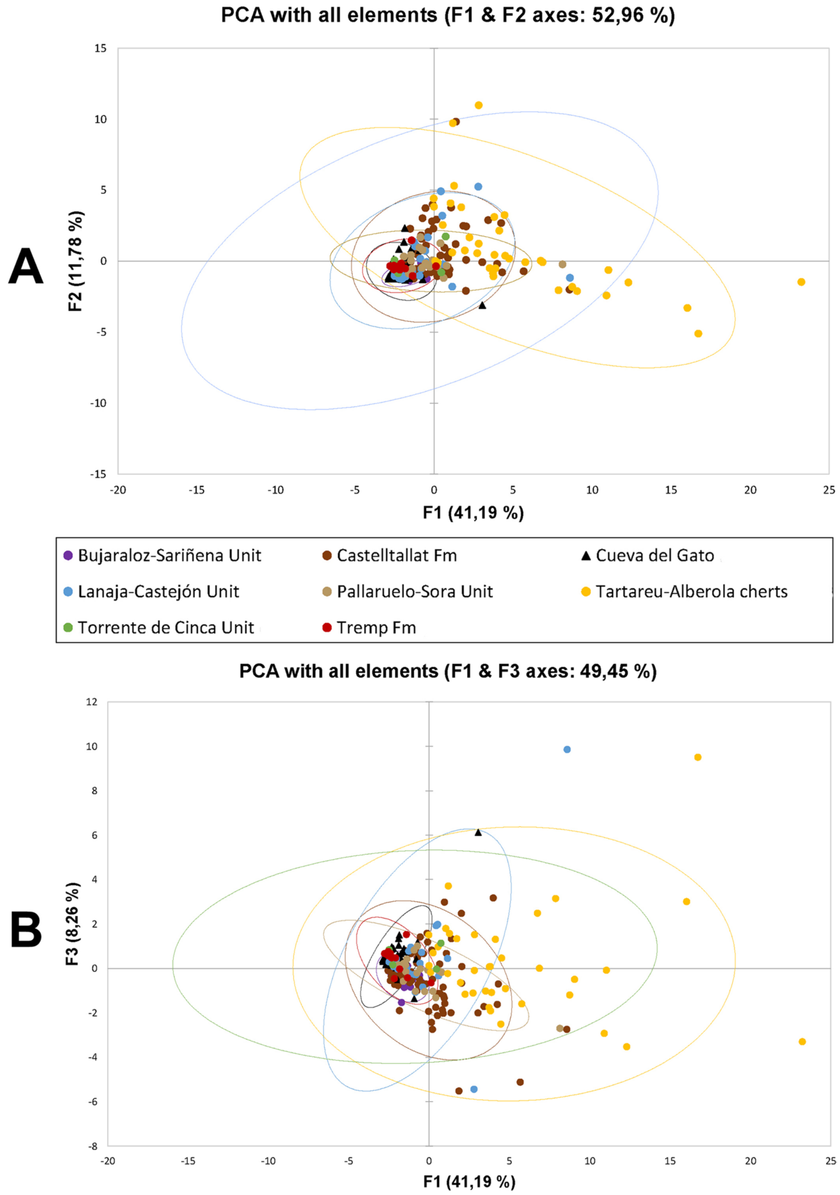

Fig. 3 Principal Component Analysis plots with all the elements (except Ge) and all the archaeological and geological samples for F1 + F2 (**A**) and F1 + F3 (**B**). Ellipses indicate the 95% of the probability

limits, so it was removed from the interpretation. The largest differences were observed in the main components (Mg, Al, Si, Ca and Fe). Specifically regarding the trace elements, the largest standard deviations were founded in B, Ti, V, Cr, Ga, As, Rb, Sr, Ba and U values.

After eliminating the data obtained for Ge, a Principal Component Analysis (PCA) with the dosed elements was run to see if quantified distinctions between geological units were observed (Fig. 3). Data was automatically normalized prior to PCA computation by Pearson correlation. After the analysis, 77 measures were eliminated because they possessed some values below the detection limits. The PCA was calculated with all the geological samples and the archaeological specimens being considered. In the plot obtained, a value of 55.19% of the total variance F1 (44.17%) and F2 (11.02%) is represented (Fig. 3A). All the geological samples are symbolized with a dot and the archaeological samples with a triangle. The ellipse with the dispersion area of the archaeological samples is presented. The plot does not give extremely useful values, as there is a significant overlapping area concerning the archaeological tools as well as the geological samples from different units. Only the geological cherts from Tartareu-Alberola (ALB1 and ALB2 outcrops) and partially some samples from the Castelltallat formation (CDF, PERAL and ALF outcrops) seem to appear far from the main archaeological dispersion. Then, to try to obtain a clearer plot, we repeated the PCA with F1 (41.19%) and F3 (8.26%) with similar results (Fig. 3B) (see descriptive data in the Supplementary information).

Linear Discriminant Analysis (LDA) was then run to see if differences between geological sources and thus, ascriptions for the archaeological samples could be established on the basis of trace elements. Samples with measures below the detection limits were discarded from the analysis. We firstly tried to run a LDA with the major and minor elements with highest standard deviations (Al, Si and Ca) but they did not allow the discrimination between samples. Thus, the large standard deviation observed within these elements must be due to a large internal variability mostly depending on the inclusions content. In this way, trace elements data were preferred to calculate the DA. Within the trace elements with largest standard deviations, Ti, Sr and U values allowed better distinguishing between the different geological units. The DA plot obtained of the three selected elements for all the geological and archaeological samples shows differences between the geological units. In the plot, 97.50% of the total variance is represented (F1: 59.95% and F2: 37.55%) (Fig. 4). To better observe the differentiation, the centroids for each group are symbolized. It can be observed that cherts from the Bujaraloz – Sariñena Unit, the Tartareu – Alberola Unit and the Castelltallat formation can easily be differentiated from the remaining formations, which appear overlapped with the archaeological cherts. Next, another DA was calculated, again considering the As, W and U values but groups were performed on the basis of each geological outcrop, and not by geological unit. The generated plot represents 82.82% of the total variance (F1: 52.60% and F2: 30.22%) (Fig. 5, top). The results indicate that cherts from the Tremp formation (Zurita outcrop) (ZURI) and the Cinca Unit (Valcuerna outcrop) (VALCU) can be distinguished from the other samples and do not fit with the main dispersion of the archaeological samples. Similarly, one of the studied outcrops from the Pallaruelo – Sora Unit (Santa Quiteria outcrop) (SQ) can be separated from the other outcrops of the same geological unit, and is located far from the archaeological samples, so it can also be discarded as a potential source. However, the remaining two outcrops from the Pallaruelo – Sora Unit (San Borombón 1 and San Borombón 2 outcrops) (SB1 and SB2) as well as all the outcrops from the Lanaja – Castejón Unit (Campo de las Horgas, La Muela 1, La Muela 2 and La Torraza outcrops) (HOR, LM1, LM2 and LT) are close to the Cueva del Gato 2 main dispersion.

Finally, to try to directly relate the studied archaeological cherts with a specific outcropping area, we made a new DA analysis, this time adding to the previously selected trace elements (As, W and U) the B data, as we observed that the values of this element helped to increase the differences between geological outcrops. In the generated DA plot, a value of 84.87% of the total variance F1 (63.19%) and F2 (21.68%) is represented (Fig. 5, bottom). Two geological formations were considered in this analysis: all the geological outcrops from the Lanaja – Castejón Unit, which previously have been shown to be closer to the main distribution of the archaeological samples (HOR, LM1, LM2 and LT), and the geological outcrops of San

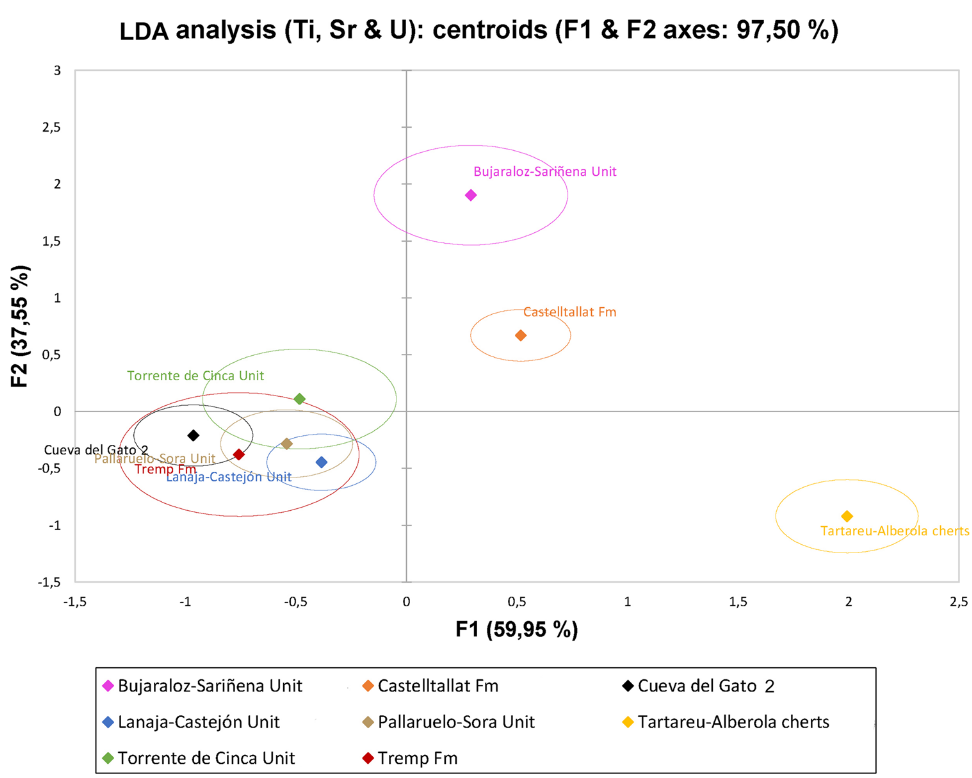

Fig. 4 Linear Discriminant Analysis plot with the centroids of all the geological units and archaeological samples for the Ti, Sr and U values

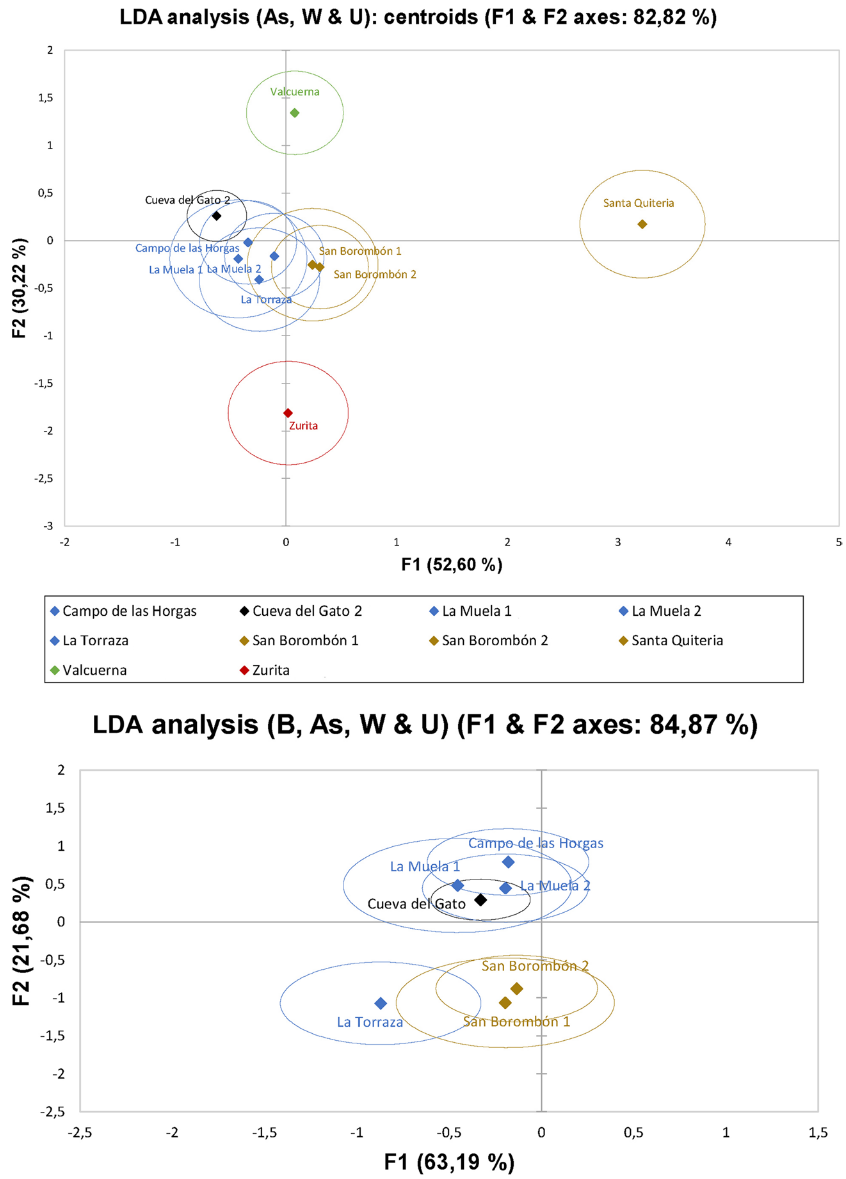

Fig. 5 Linear Discriminant Analysis plot with the centroids of the geological outcrops from Lanaja-Castejón Unit (in blue), Pallaruelo-Sora Unit (in brown), Cinca Unit (in green) and Tremp formation (in red) and the centroid of the archaeological samples for the As, W and U values (top) and for B, As, W and U values (bottom)

Borombón 1 and 2 (SB1 and SB2) from the Pallaruelo – Sora Unit. The resultant plot indicates that both outcrops from the Pallaruelo – Sora Unit (San Borombón 1 and San Borombón 2) (SB1 and SB2) can be distinguished from the other studied formation, and do not fit with the Cueva del Gato 2 dispersion. Similarly, one of the outcrops from the Lanaja – Castejón Unit (La Torraza outcrop) (LT) can also be discarded, as it is not placed in the same distribution area as the archaeological cherts. However, the remaining outcrops from the Lanaja – Castejón Unit (Campo de las Horgas, La Muela 1 and La Muela 2 outcrops) (HOR, LM1 and LM2) are located close to the archaeological samples and, in fact, the centroids from La Muela 1 (LM1) and La Muela 2 (LM2) are placed in the dispersion ellipse of the Cueva del Gato 2, thus showing a clear direct relation in terms of the elemental chemical composition.

GIS results

GIS tools were applied with the aim of determining the mobility of these hunter-gatherers' societies and trying to establish the probable routes the humans followed to find the rocks. Following the methodology described, we obtained a "Costs Map" (Fig. 6) that shows the relationship between the Cueva del Gato site and the outcrop locations described above. Although Campo de las Horgas (HOR), La Muela 1 (LM1) and La Muela 2 (LM2) were the outcrops confirmed by DA analysis, we thought that comparing the possible routes, distances, and cost units (Fig. 7, Table 4) obtained from the GIS analysis of these three outcrops with the rest of the considered non-confirmed outcrops, could be more interesting for the purposes of comparison.

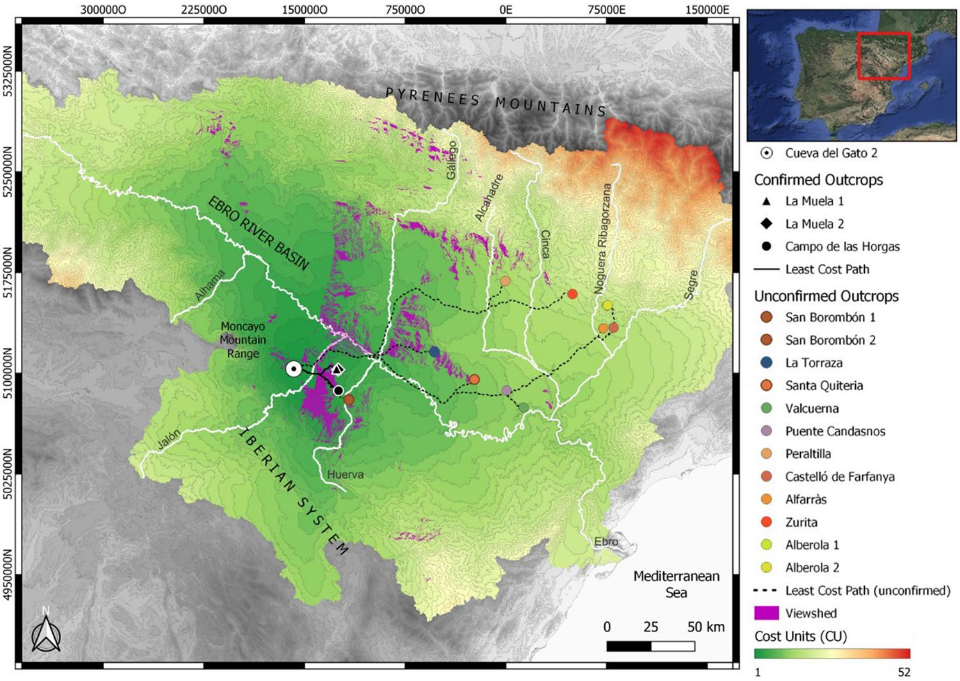

Fig. 6 Cost map of Cueva del Gato and the outcrops considered in this study

As we can observe, the position of Cueva del Gato in the Ebro basin geographical context, gives the site a privileged point of access to the surrounding environment. The Least Cost Path and the Euclidean distance differences for La Muela 1 (LM1),La Muela 2 (LM2) and Campo de las Horgas (HOR) (Table 4 in bold, Fig. 7 shaded) are quite small (34–35 km vs. 23–24 km respectively), while the cost unit value for these three outcrops is the lowest of all those considered: 4 CU. Access to these chert outcrops is made by an easy route that requires walking across the plains of the

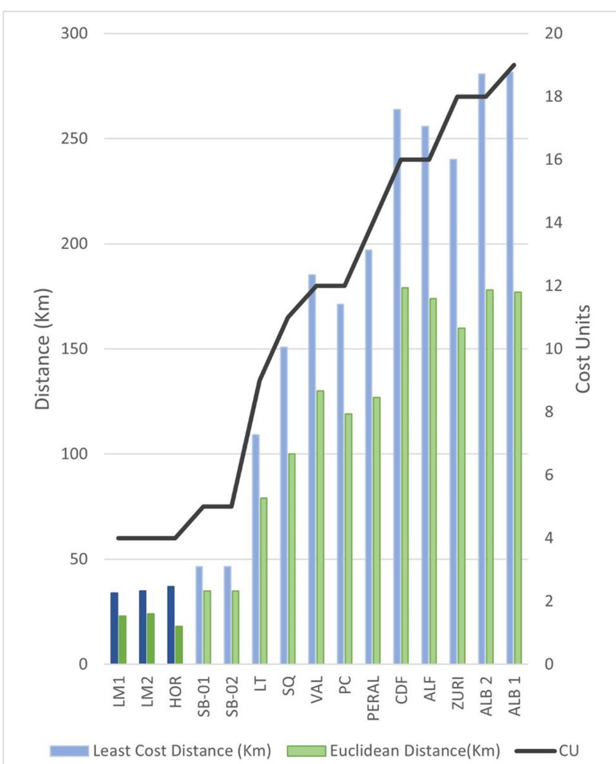

Fig. 7 Composite plot of Least Cost Path distance and Euclidean distance (bars) and Cost Units (CU) (line) of the studied outcrops

Table 4 Reference of Least Cost Path distances, Euclidean distances and Cost Units of the studied outcrops

Reference	Least cost distance (km)	Euclidean distance (km)	Cost units
LM1	**34**	**23**	**4**
LM2	**35**	**24**	**4**
HOR	**35**	**18**	**4**
SB-01	37	35	5
SB-02	47	35	5
LT	46	79	9
SQ	109	100	11
VAL	151	130	12
PC	185	119	12
PERAL	171	127	14
CDF	197	179	16
ALF	264	174	16
ZURI	256	160	18
ALB 2	240	178	18
ALB 1	281	177	19

The bold entries correspond to the sources that are similar to the archaeological cherts according to the geochemical approach

Jalón basin, and crossing the river halfway to reach the La Muela hills and the Campo de las Horgas outcrop (HOR), which is located in the south region of the la Muela Plateau. All this environment has a perfect view from the cave's hilltop. In this sense, the Jalón river and its valley must not have posed any significant barrier to cross this area, since the surrounding orography does not offer any other type of obstacle. All of this shows that the supply of resources in the Cueva del Gato is regional or immediately accessible, which is logical considering the ease of access to these two main chert outcrops. Similarly, this route would be followed for San Borombón 1 and 2 outcrops (SB1 and SB2), which are located in the nearby Huerva River basin. It is communicated with the Jalón River through a depression easy to traverse, which gives these outcrops also a low Cost Unit value (4–5 CU).

For the rest of the unconfirmed outcrops, we observe that the cost units gradually and proportionally grow with the distance separating them from the site. This is due to the fact that the main axis of communication of the archaeological site with these other outcrops occurs through the Ebro valley, after which, the routes diversify following the tributary rivers of the pre-Pyrenean area. Thus, we obtain that the access to the outcrops of La Torraza (LT) and Santa Quiteria (SQ) (both at the range of the viewshed from the cave) and Puente Candasnos (PC) and Valcuerna (VALCU), would follow the natural riverbed and the foothills of the Middle Ebro River, which still places us in an easy access context (9–12 CU). In contrast, to reach the Peraltilla outcrop (PERAL) (14 CU), it would be necessary to go up the courses of the Gállego and Alcanadre rivers. Finally, going into the pre-Pyrenean areas, it would be necessary to ascend the Cinca (Zurita) (ZURI) and Segre-Noguera Ribagorzana River courses (Alfarrás –ALF-, Castelló de Farfanya –CDF- and Alberola 1 & 2 –ALB1 and ALB2-), which significantly increases both the Least Cost Path distances (almost twice the Euclidean distances) and the cost units (tending to between 16 and 19 CU). Thus, the characteristics and position of these outcrops would require an investment of physical effort which, due to the characteristics of the terrain surrounding Cueva del Gato, would not make it feasible to collect siliceous resources at the site.

The data presented regarding the CU investment for achieving the geological outcrops needs to be compared with other criteria such as abundance, the size, the suitability for knapping or the type of extraction involved (if it is easy or not to extract the chert nodules from the embedded rock), as suggested by other researchers (Soto et al. 2017; Wilson 2007). For the specific case of the confirmed outcrops by geochemical methods, the chert outcrops from La Muela 1, La Muela 2 and Campo de las Horgas present similar results. La Muela 1 and La Muela 2 outcrops are placed in the La Muela plateau, a very rich area in chert nodules, with sizes ranging between the 10 and 30–40 cm. Cherts are embedded in soft limestones and marls, being not difficult the extraction. Moreover, the chert nodules have naturally been eroded from the embedded rock and they are abundant in the hillside. Campo de las Horgas is a subprimary outcrop where cherts have been already removed from the embedded rock. In that case, the abundance of chert nodules is not as abundant as observed in La Muela 1 or La Muela 2 outcrops, but they are more accessible. In the three concerned outcrops, the quality of these cherts is exceptional, with extremely fine surfaces without fissures. Thus, it seems clear that in addition to the proximity of these outcrops to the archaeological sites, other criteria such as the abundance of high-quality and easy to extract chert nodules was key for the selection of these cherts.

Archaeological inferences

The first macroscopic characterization of the archaeological tools recovered at the Upper Palaeolithic human occupations of Cueva del Gato 2 revealed that one main siliceous type was exploited. The textural and micropalaeontological content observed at the stereoscopic microscope showed similarities with the badly-name chert type of *Monegros* cherts. This geographical name has by tradition been used by prehistorians from Western Europe to describe a very high-quality, brownish chert originated in a carbonate lacustrine environment that frequently contains Liesegang rings and charophyte algae and gastropods sections. Their supposed outcrops have frequently been located loosely in the Middle Ebro Basin. However, in a previous study the term *Monegros* was redefined (García-Simón and Domingo 2016; Sánchez de la Torre et al. 2019a, b). The results indicated that different geological formations outcropping in the Middle Ebro Basin contained chert macroscopically similar to that traditionally defined as *Monegros* chert. Moreover, in aforementioned study, a first geochemical approach to quantify the major and minor elements was carried out using Energy Dispersive X-Ray Fluorescence (ED-XRF). The results revealed differences between geological units at the chemical elemental composition and confirmed the value of geochemical methods and statistical procedures to establish differences among cherts that were macroscopically similar.

Thus, in this paper we have tried to go further and try to directly connect the archaeological lacustrine cherts of Cueva del Gato 2 with a specific geological formation based on the quantification of trace elements, made by LA-ICP-MS. We have only included the strictly geological formations outcropping in the Middle Ebro Basin, but we have also considered these geological units containing lacustrine cherts and outcropping in the first Pre-Pyrenean foothills of the Central-Eastern Pyrenees. The aim was to be able to establish clear differences on the basis of the elemental chemical composition between all the geological formations containing macroscopically similar lacustrine cherts in an area largely frequented by human populations during the Upper Palaeolithic.

This study has proved that it is possible to establish clear differences between macroscopically similar geological units based on the specific analysis of the trace elemental composition and the use of statistical procedures. Thus, in this particular case, we have been able to directly connect the archaeological lacustrine cherts of Cueva del Gato 2 with the lacustrine cherts of the Lanaja – Castejón Unit, precisely with the geological outcrops of La Muela 1 (LM1), La Muela 2 (LM2) and Campo de las Horgas (HOR). These significant results encourage us to apply this analytical approach to other archaeological sets. In this way, the use of LA-ICP-MS to quantify major, minor and trace elements has revealed to be a useful technique for the characterization of chert tools aiming at define the procurement of these abiotic resources. While ED-XRF had already been applied to the analysis of geological cherts defined as *Monegros*, it is for the first time that LA-ICP-MS has been applied to identify differences between geological units in the base of the quantification of trace elements. This technique, that despite being micro-destructive does not involve the destruction of the archaeological artefact, has become a very useful tool for the characterization of geological chert sources and their relation with archaeological artefacts. The LA-ICP-MS offers the possibility to quickly and precisely quantify a large number of elements, with extremely low detection limits. It is at the moment one of the most used techniques for the complete quantification of the elemental chemical composition for chert and other rocks, being the only inconvenient that the samples need to be moved to the laboratory for the analysis.

The geochemical characterization by LA-ICP-MS combined to the application of GIS analysis has given valuable data regarding the territoriality of the hunter-gatherer populations inhabiting Cueva del Gato. The obtained radiocarbon dates for level II place this human occupation during the transition moment between the Solutrean to the Magdalenian. The absence of *raclettes* would place this occupation during the Magdalenian 0 or the Lower Badegoulian, a period not yet well known in this region of the Ebro Basin. Thus, the data obtained after the geochemical and GIS analyses are key to define the territoriality of these groups. It seems that we are facing an occupation of groups with regional mobility strategies, at least regarding the procurement of lithic raw materials. Thus, the lithic industry does not reveal the existence of long distances for acquiring this resource, probably due to the good quality and high quantity of available rocks in the Middle Ebro Basin. Nevertheless, this large mobility (by direct procurement or exchange with groups) seems to have existed, according to the presence of perforated marine shells in the site. However, and probably linked to the great availability of high quality raw materials in the nearby territory, chert did not move long distances.

The absence of exogenous cherts in this human occupation at Cueva del Gato contrasts with the data obtained for other Upper Palaeolithic sites from NE Iberia. Thus, we already know that human groups that settled the first Pre-Pyrenean foothills during the Upper Palaeolithic largely used lacustrine cherts. A first geochemical characterization based on the quantification of major and minor components at Chaves Cave (Bastarás, Huesca province, Spain), Fuente del Trucho (Asque-Colungo, Huesca province, Spain), Cova Alonsé (Estadilla, Huesca province, Spain) and Forcas I Shelter (Graus, Huesca province, Spain) indicated that some of the recovered lithic tools could have had their origin in cherts outcropping in the Middle Ebro Basin, establishing a direct relation between the first Pre-Pyrenean foothills and the Middle Ebro Basin (Sánchez de la Torre et al. 2017; Sánchez de la Torre et al. 2020a, b, c; Sánchez de la Torre et al. 2020a, b, c). However, in most of these sites (being Cova Alonsé an exception), exogenous cherts were also documented, in some cases involving the circulation of chert from the northern to the southern Pyrenees. Thus, future studies that include the quantification of trace elements by the use of LA-ICP-MS in other archaeological sites with Upper Palaeolithic human occupations would be key in order to understand human mobility in this area of Eastern Iberia and the relationship these humans had with the geographical space. during and after the Last Glacial Maximum.

Conclusions

In this paper, we have presented the results obtained after the geochemical analysis of chert tools recovered from the Upper Palaeolithic human occupations of Cueva del Gato 2, a site located in the first foothills of the Iberian System, near the Middle Ebro Basin. The quantitative data obtained after the elemental chemical characterization by LA-ICP-MS of a series of archaeological tools and their comparison with a collection of macroscopically similar geological cherts has suggested that the past human groups who settled at Cueva del Gato 2 preferred cherts from the Lanaja – Castejón Unit for the confection of their lithic assemblages. Moreover, the statistical approach to precisely define their origin through a comparison of the elemental chemical compositions has shown that the geological samples from La Muela 1 (LM1) andLa Muela 2 (LM2) outcrops and probably also Campo de las Horgas (HOR)outcrop better fit with the studied archaeological tools. Then, the application of GIS resources to investigate the mobility and territoriality of these hunter-gatherer populations has suggested that the Middle Ebro Basin was the least-cost route connecting the outcrops of La Muela Plateau with the archaeological site. The application of these analytical procedures to other archaeological collections will be essential in order to offer a wider territoriality analysis of these past hunter-gatherers' populations that chose this area of the Iberian Peninsula to develop their subsistence strategies.

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Author contribution All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by all authors. The first draft of the manuscript was written by Marta Sánchez de la Torre and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript. The authors declare they have no relevant financial or non-financial interests to disclose.

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Data availability The authors confirm that the data supporting the findings of this study are available within the article and its supplementary materials.

Declarations

Competing interests The authors declare no competing interests.

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