



Chert procurement for inferring Neanderthal mobility in central western Iberia

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Abstract

Lithic raw material sourcing is one of the most effective approaches for inferring exploitation territories during the Palaeolithic, including for Neanderthals. Certain raw materials, particularly chert, are especially well suited to this type of investigation because the distinctive characteristics of each geological source can be reliably identified through a combination of macroscopic observation, low-magnification analysis, and geochemical methods. With that in mind we questioned if chert sourcing would allow identification of territorial patterns for the Neanderthal populations of central Portugal, in western-most Iberia. To answer that question, we conducted an exploratory study comparing archaeological cherts from Gruta Nova da Columbeira, Praia Rei Cortiço and Mira Nascente with geological samples from nearby sources using macroscopic, low-magnification PIXE and pXRF analysis. Our results showed similarities between archaeological and geological cherts proving that the sites, sources and analytical protocols used were viable for such investigation and, therefore, can be developed and refined in the future. They also showed that correspondences between geological and archaeological cherts are limited to the area between the coast and the western façade of the first mountain ridge (Serra d'Áire e Candeeiros) to the east, and between the Cesaredas Plateau to the south and the River Lis to the north. This territory is adjacent to the area already delimited by chert sourcing and isotopic analysis for the Neanderthals that occupied Oliveira Cave, located at the southeastern façade of the same mountain range. This contributes to a better understanding of Neanderthal territorial patterns, which is particularly important in regions where research is underdeveloped and where identifying other patterns of variability, particularly across time, has proven difficult.

Keywords Western Iberia · Neanderthals · Mousterian · Lithic assemblages · Chert

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Introduction

Raw material sourcing provides one of the most reliable sources of information to access Neanderthal selection preferences (Pereira et al. 2017; Abrunhosa et al. 2019; de Lombera-Hermida et al. 2020), transport and curation (Baumler 1996; Burke 2006), and overall exploitation territory, even if the exact position of some sources is not yet recognized (Matias 2016; Doronicheva et al. 2017; Gómez De Soler et al. 2019, 2020; Abrunhosa et al. 2020; Eixea et al. 2022). Overall, western European Neanderthals seem to have had complex strategies of raw material sourcing, particularly focusing on chert (Turq et al. 2017; Delpiano et al. 2018; Romagnoli et al. 2022). Research on lithic raw material procurement during the Middle Palaeolithic in the Iberian Peninsula has shown that technological organisation and mobility strategies were closely conditioned by local geological contexts, while remaining flexible and adaptive. Studies across eastern Iberia (Eixea et al. 2014, 2016; Rios-Garaizar and García Moreno 2015; Gómez De Soler et al. 2020, 2023; Mayor et al. 2022; del Pozo et al. 2023) and central Iberia (Prieto et al. 2016; Vallejo Rodríguez et al. 2017; Abrunhosa et al. 2020; Sánchez De La Torre et al. 2023; Ramírez-Amador et al. 2025; Herrero-Alonso et al. 2025) demonstrate that Neanderthal groups predominantly exploited locally available raw materials, often within short distances from sites, while selectively incorporating exogenous resources when these offered specific functional or technological advantages.

Even in chert-rich regions, other raw materials are consistently present in assemblages, indicating diversified procurement strategies, emphasizing that raw-material selection cannot be explained solely by availability or distance to sources (Gómez De Soler et al. 2020; Romagnoli et al. 2022; Herrero-Alonso et al. 2025). Despite significant advances, research remains uneven across Iberia.

In Portugal, chert is only available in the Algarve and Estremadura Limestone Massifs, which make up approximately 12% of the total area, and in some adjacent Quaternary deposits downstream (Pereira 2013; Pereira and Benedetti 2013). The systematic study of lithic raw material sourcing by Neanderthals in Portugal remains limited (Matias 2012, 2016; Pereira et al. 2015), but the available inventories show abundant chert at sites located in chert-rich territories, and its marked decline at sites in different geological settings, even in regions adjacent to chert-rich areas (Pereira et al. 2012). Within the scope of developing and encouraging further investigation on raw material sourcing in Portugal (Pereira et al. 2016), we conducted an exploratory research using a multi-method analytical approach. This included macroscopic and low-magnification characterization, and elemental analysis by pXRF

and PIXE to determine the variability of the raw materials from Gruta Nova da Columbeira, Mira Nascente and Praia do Rei Cortiço, and to identify the probable origin of the chert. Noting that results from Oliveira Cave based on raw material sourcing (Matias 2012, 2016) and isotope analysis (Linscott et al. 2023) converge towards a relatively well-delimited Neanderthal territory, we examined whether our data could similarly shed light on the area exploited by the Neanderthal groups that occupied the coastal area.

The geomorphological background of the sites

Portuguese Estremadura is a coastal region located in central western Iberia (Fig. 1). Of particular interest for this study are the Mesozoic and Cenozoic sedimentary rocks of the Lusitanian Basin (Kullberg 2000; Kullberg et al. 2006) (Table 1). The landscape is mostly based on uplifted Lower to Upper Jurassic limestone and dolomite units that underlie the ca. 800 km² Estremadura Limestone Massif (ELM) (Martins 1949; Manuppella et al. 1985) and spread to the nearby submerged continental platform (Ribeiro et al. 1979; Almeida et al. 2000; Ferreira 2000).

The ELM is marked by bounding tectonic faults that enclose wide karst depressions and valleys (Martins 1949; Reis et al. 2023). In the major river valleys (e.g. Tejo, Lis,



Fig. 1 Orography and hydrology of the Iberian Peninsula with the location of the study area (white rectangle) and archaeological sites: 1- Gruta Nova da Columbeira; 2- Praia Rei Cortiço; 3- Mira Nascente

Table 1 Phases, ages and sediments of the Lusitanian basin

Phase	Age	Sedimentary sequence
Pre-rift	Late Triassic and the Early Jurassic	Sandstones, siltstones, conglomerates, halite, gypsum, and lacustrine limestones and shales
Syn-rift	Middle Jurassic and Early Cretaceous	Marine limestones, sandstones, siltstones, and mudstones organic-rich shales, reef and carbonate platform deposits, and some volcanoclastic layers
Post-rift	Late Cretaceous and the Cenozoic	Marine limestones and marls, sandstones and shales, flysch-type deposits, and alluvial and deltaic sands and gravels

and Mondego) the Upper Cretaceous beds and Miocene to Quaternary deposits and terraces have abundant silicic gravels mostly composed of quartzite and quartz (Cunha 2019).

In the karstic areas, chert lenses and nodules are well known within the Middle and Upper Jurassic limestone beds, (Matias 2012; Jordão 2023). The incised drainage network exposes at several locations bedrock layers displaying chert nodules that can be found detached in the foot slopes, in terraces and in the shoreface (Matias 2012; Aubry et al. 2012; Pereira et al. 2016; Jordão 2023).

The main primary chert areas are near the municipalities of Leiria, Nazaré, Torres Vedras, and Lisbon as well as the Serra d'Aire and Candeeiros mountains and Cesaredas Plateau. Rio Maior is the largest chert source with nodules in sub primary and secondary position, while Montes has a similar situation, but on a smaller scale (Fig. 2J). Secondary chert deposits are found at fluvial gravels as in Zêzere, Alenquer, Leiria, and along the coast (Fig. 2).

The region was strongly influenced by sea level oscillations throughout the Pleistocene, causing the coastline to be far west of its present position for most of the time (Daveau

1980; Quézel 1985; Boski et al. 2000), probably exposing extensive gravel deposits on the continental shelf. Many locations in the study area have outcrops of Upper Cretaceous conglomerates rich in quartzite and other hard silicic rocks from the Iberian Massif, and devoid of sedimentary limestone-related cherts. The Mesozoic rocks are covered by Pleistocene and Holocene dune fields along most of the coastal margin (Daveau 1980; André et al. 2009; Cunha 2019; Haws et al. 2020).

The sites

Gruta Nova da Columbeira (39°17'53.42"N | 9°12'2.60"W) (Figs. 1 and 2 - #1, Fig. 3a and b) is a cave with seven Mousterian layers, rich in fauna and lithics, located in a canyon that runs across the Cesaredas Plateau. At its mouth, the canyon meets the Óbidos valley close to the seashore where several streams coming from the western façade of the Aire e Candeeiros mountain flow.

The site was found during the exploitation of a limestone quarry, and the excavation was done relatively

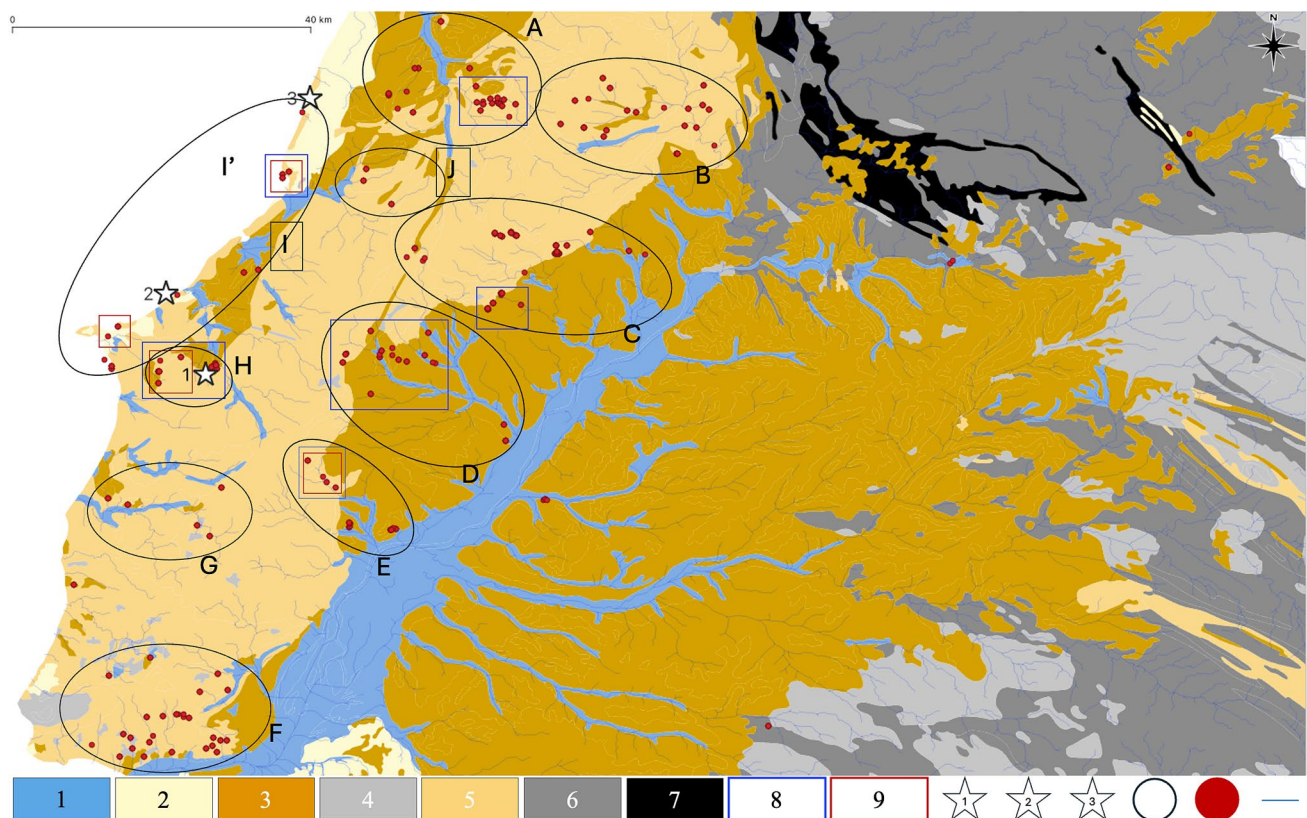


Fig. 2 Lithological background of the study area. Colors: 1- Alluvial; 2- Dunes, aeolian sands; 3- Quartz and quartzite conglomerates, sandstone, soft sandstone, gravels, sand, clay, marls, limestone; 4- Diorite, gabbro, granite, porphyry, quartzodiorite, syenite, basalt, dolerite, other eruptive rocks; 5- Limestone (chert in primary position), marls, sandstone, conglomerates (chert in secondary position); 6- Schist, mica schist, greywacke, sandstone, amphibolite, carbonate rocks, gnaiss, corneal, quartzite; 7- Quartzite. Colored rectangles: 8- Chert sources identified with pXRF; 9- Chert sources identified with PIXE. Stars: 1- Gruta Nova da Columbeira; 2- Praia Rei Cortiço; 3- Mira Nascente. Black ovals: chert sources: A- Leiria; B- Zêzere; C- Serra d'Aire e Candeeiros; D- Rio Maior; E- Alenquer; F- Lisbon; G- Torres Vedras; H- Cesaredas; I- Coast gravels and Nazaré outcrop (I'), J- Montes. Red dots: chert sources. Dark blue lines: rivers and streams

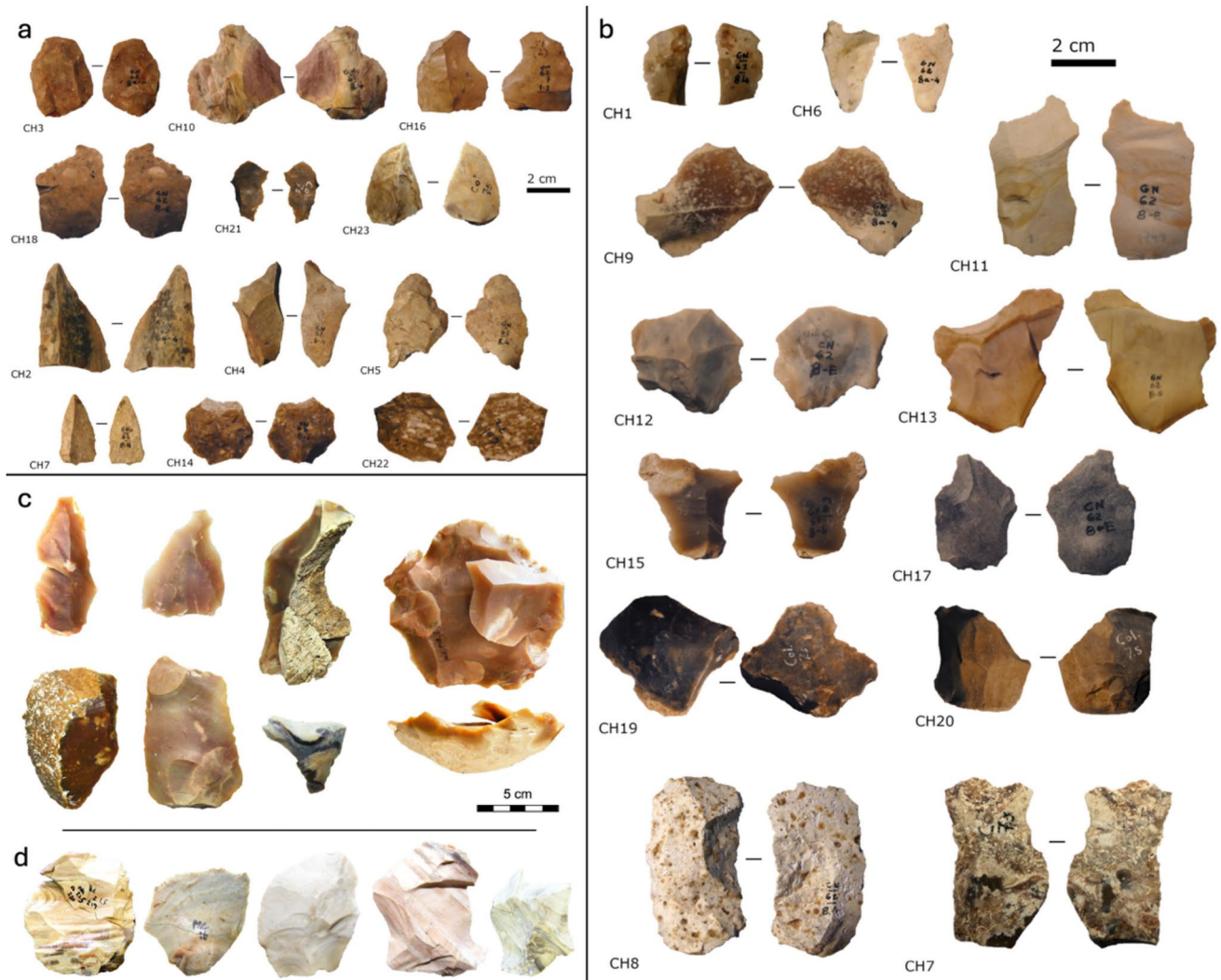


Fig. 3 Chert variability at **a**) and **b**) Gruta Nova da Columbeira, **c**) Mira Nascente and **d**) Praia Rei Cortiço

quickly (Raposo and Cardoso 1997, 1998; Cardoso et al. 2002; Fernández-Laso et al. 2015; Carvalho et al. 2018; Figueiredo et al. 2018). Over the years, the site has been dated through different methods, but its chronology is still far from clearly established (Zilhão et al. 2011). Nevertheless, it is still considered one of the most important Mousterian sites in Portugal. The Mousterian sequence is capped by a thick stalagmitic crust dated to >21 ka (Carvalho et al. 2018). The layers below have U-series dates that vary between 35 to 101 ka, but all with a standard deviation of dozens of thousands of years. So far, the most precise date seems to be 85 ± 8 ka for layer 8, which is near the base of the sequence. At the back of the room, the Mousterian sequence was cut by an infill during the Upper Paleolithic, dated by 14C between 29 and 15 ka (Zilhão et al. 2011).

Praia Rei Cortiço ($39^{\circ}25'22.38''N$ | $9^{\circ}14'35.69''W$) (Figs. 1 and 2 - #2, Fig. 3d) is a single layer, open-air site, with hundreds of lithics, several refits, charcoal, and without

fauna. The site is exposed at the foot of an eroding coastal cliff in sediments dated by OSL to about 100 ka (Benedetti et al. 2009; Haws et al. 2010, 2020). The cliff exposes Late Pleistocene and Holocene valley fill deposits incised into the Cretaceous conglomerate bedrock, which is exposed adjacent to the site and contains abundant quartz, quartzite, and chert pebbles. The presence of this chert near the site results from the combination of cliff erosion and transport by colluvial and fluvial processes.

Mira Nascente ($39^{\circ}42'21.56''N$ | $9^{\circ}2'58.74''W$) (Figs. 1 and 2 - #3, Fig. 3c) is a single layer, open-air site with hundreds of lithics, several of them refitting, and charcoal but no fauna. The archaeological stratum is located at the middle of an eroding coastal cliff, with C14 and OSL ages of about 40 ka (Benedetti et al. 2009; Haws et al. 2010, 2020). At this site, 30 m of late Quaternary sand and pebbles overlie the limestone bedrock, which outcrops on the beach. Chert nodules such as those found at the site are not visible

in the immediate area, but may have been available in the shore face or in bedrock outcrops that have since been covered or eroded.

The lithic assemblage of Gruta Nova da Columbeira has three main features: 1) a wide diversity of raw materials with balanced amounts of quartz, quartzite and chert, along with other rocks that appear in very low quantities; 2) few cortex pieces that, when present, are waterworn, and 3) high curation rates (Raposo and Cardoso 1997, 1998; Cardoso et al. 2002). These features (particularly the first two) are considered as markers of distance decay (Renfrew 1969, 1977; Wilson 2007; Clarkson and Bellas 2014). Here, their combined occurrence reinforces the idea of chert acquisition from areas beyond the immediate vicinities of the cave, which is consistent with the geochemical analysis.

Interestingly, a similar pattern is observed with quartz and quartzite, raw materials that are abundant and ubiquitous across the landscape. At Gruta Nova da Columbeira, these also show low quantities of waterworn cortex (that do not require cortex removal) within each layer. The absence of the configuration phase, and low amounts of cortex in the cores (particularly the pre-determinate ones) and on the blanks across the stratigraphy, indicate that these materials were shaped and produced elsewhere and brought as so into the cave. This is the opposite of what is seen for the lithics from Mira Nascente and Praia Rei Cortiço, where cortex is present and some flakes, including Levallois, refit into the cores (Benedetti et al. 2009; Haws et al. 2020).

Methods and materials

The geological samples for comparison were obtained from the LusoLit reference collection (Pereira et al. 2016) (red dots in Fig. 2). Archaeological samples were selected based on the macroscopic and low-magnification variability of the cherts present at the three sites and covering such recognized variability. Given the exploratory scope of this study and its aim of establishing chronologies and initial models for chert sourcing at these sites, the analysis focused specifically on raw-material characterisation, providing a foundation for the future integration of techno-typological data. Similarly, the relative proportions of each chert type were not addressed, as the primary objective was to identify the sourcing areas associated with each site rather than to explore the economic specificities of individual occupations. This aspect represents a clear avenue for future research.

Macroscopic and low-magnification analysis of chert has some known limitations, but is priceless for exploratory investigations and for large assemblages (such as Columbeira) where a great variability of raw materials can

be identified and grouped by the naked eye. Macroscopic and low-magnification observations followed established descriptive criteria (Dunham 1962; Embry and Klován 1971; Hurlbut and Klein 1977; James 1984; Vernon 2004; Abrunhosa et al. 2020). A 30× magnification loupe was used, focusing on diaphaneity, fracture, color variation, porosity, surface texture, homogeneity, and the presence of microfossils. Raw materials were then grouped and sub-grouped based on shared characteristics. Color, including its variability and homogeneity, was described using up to three Munsell rock color chart codes (Munsell Color 2009) per specimen: two for internal color variations and a third for the cortex, if distinguishable (Ferguson 2014) (Table 2).

Geochemical elemental analysis was performed in 2017 solely on the Gruta Nova da Columbeira assemblage. A portable Bruker™ S1 Titan® energy dispersive X-ray fluorescence spectrometer, equipped with a rhodium X-ray tube, FAST® SDD detector, 5 mm collimator, S1RemoteCtrl filter, and S1Sync software, was used. The spectrometer was operated in a closed desktop configuration. Elemental analysis, covering elements from Magnesium (^{12}Mg) to Uranium (^{92}U), was conducted on cleaned, flat, homogeneous surfaces. Each analysis consisted of a 240-s measurement, split equally between heavy and light elements (120 s each). Multiple analyses were performed per specimen. The default "Application Geochem General Method Dual Mining" calibration was used, consistent with previous successful applications (Carvalho and Pereira 2017; Pereira et al. 2017, 2021; Paixão et al. 2019). PIXE analyses were carried out on all three assemblages at the IST laboratory in 2014, following the established protocol used by the laboratory (Alves et al. 2000; Corregidor et al. 2011). Profiting from the external beam setup, the samples could be examined in ambient air, rendering sample size irrelevant. Each specimen was exposed to a 2 MeV and 500 pA proton beam for 15 min, targeting an area of $0.9 \times 0.9 \text{ mm}^2$. The resulting X-rays were collected using a 30 mm^2 SDD detector, and the spectra were processed and quantitatively analysed using the GUPIXWIN software (Campbell et al. 2010). With the chosen experimental conditions, only elements heavier than Si can be detected and quantified. All detected elements were considered to be present in its most common oxide form. While most specimens received a single measurement, some refitted nodules and specimens with multiple-colored zones underwent multiple measurements to verify the consistency of their elemental composition and proportions. In total, 46 specimens yielded 60 readings (Supplementary information Tables 1 and 2). With both pXRF and PIXE, several of the common elements are measured as their oxides readings (Supplementary information Tables 1 and 2).

Table 2 Results from the macroscopic analysis of the Gruta Nova da Columbeira samples

	Sample Name	Porosity	Transparency	Luster	Singular Components	Colour Variability	Munsell 1	Munsell 2	Munsell 3 (Cortex)
Mudstone	CH1	Compact	Semi-Opaque	Dull/Waxy	Chalcedony	Variable	10YR 6/2	5YR 8/1	-
	CH6	Semi-Porous	Semi-Opaque	Waxy	Geodes	Homogeneous	N9	10YR 7/4	-
	CH9	Compact	Translucent	Dull	-	Variable	5Y 8/1	5YR 4/1	5YR 5/6
	CH11	Semi-Porous	Semi-Opaque	Dull/Waxy	Liesegang Rings, Iron Oxides (5%), Bioclasts (Gastropods)	Homogeneous	5Y 8/1	10YR 8/6	-
	CH12	Compact	Semi-Opaque	Dull	-	Variable	5Y 8/1	5B 7/9	-
Mudstone to Wackestone	CH13	Compact	Semi-Opaque	Waxy	Liesegang Rings, Iron Oxides (1%)	Homogeneous	10YR 8/2	10YR 6/6	-
	CH15	Compact	Translucent	Waxy/Dull	-	Homogeneous	5YR 5/2	-	10YR 8/2
	CH17	Compact	Opaque	Waxy	-	Homogeneous	N5	N4	-
	CH19	Compact	Semi-Opaque	Waxy	Iron Oxides (1%)	Homogeneous	5YR 2/1	5YR 5/2	-
	CH20	Compact	Opaque	Waxy	-	Homogeneous	N3	-	10YR 6/2
Wackestone	CH3	Compact	Semi-Opaque	Waxy	Bioclasts	Variable	5R 4/6	10R 6/2	-
	CH10	Compact	Semi-Opaque	Dull/Waxy	Liesegang Rings, Iron Oxides (30%), Bioclasts	Variable	5RP 8/2	10YR 8/6	-
	CH16	Compact	Semi-Opaque	Waxy	-	Homogeneous	5YR 6/4	-	5Y 5/2
	CH18	Compact	Opaque	Dull	Bioclasts	Variable	5R 4/2	10R 7/4	-
	CH21	Compact	Semi-Opaque	Waxy	Iron Oxides (20%)	Variable	5R 3/4	10R 2/2	-
Wackestone	CH23	Compact	Translucent	Dull	Bioclasts	Homogeneous	10YR 6/2	-	-
	CH2	Compact	Semi-Opaque	Vitreous/Waxy	Geodes	Variable	10R 5/4	5YR 2/1	-
	CH4	Compact	Semi-Opaque	Dull	Bioclasts	Variable	5YR 8/1	5YR 6/1	-
	CH5	Compact	Semi-Opaque	Dull/Waxy	Bioclasts, Chalcedony	Variable	10YR 68/2	10YR 8/6	-
	CH7	Semi-Porous	Semi-Opaque	Dull	Bioclasts	Variable	10YR 8/2	10R 2/2	-
Packstone	CH14	Compact	Semi-Opaque	Dull/Waxy	Geodes, Iron Oxides (1%)	Variable	5Y 5/2	10YR 8/6	10YR 8/2
	CH22	Compact	Semi-Opaque	Dull	-	Variable	10R 4/2	5R 8/2	-
	CH8	Semi-Porous	Opaque	Waxy	Geodes	Variable	N9	5YR 5/6	-
	CH24	Compact	Semi-Opaque	Dull	-	Variable	5YR 5/2	10YR 8/2	-
	Summary of Chert Type Classification								
Chert Type	Texture	Porosity			Main Features	Examples			
Mudstone-Dominated Cherts	Compact	Low			Liesegang rings, iron oxides, bioclasts	CH1, CH9, CH10, CH11, CH12, CH13, CH15, CH17, CH19, CH20			
Wacke-Dominated Cherts	Compact	Low			Bioclasts, geodes, chalcedony	CH2, CH3, CH4, CH5, CH16, CH18, CH23			
Packstone Cherts	Semi-porous to compact	Moderate			Bioclasts, iron oxides, geodes	CH7, CH14, CH21, CH22			
Boundstone-Associated Cherts	Compact	Low to moderate			Geodes, silicified boundstone	CH8, CH24			

Results

Praia Rei Cortiço, Gruta Nova da Columbeira and Mira Nascente contain quartzite, quartz, chert, and other raw materials in trace amounts. However, Mira Nascente is dominated by chert, Praia Rei Cortiço by quartzite, and at all layers of Gruta Nova da Columbeira the three main raw materials are balanced (Table 3).

Quartzite and quartz are highly abundant in Cenozoic deposits widespread across the Lusitanian Basin region. They arrive by fluvial transport from the Iberian Massif Zone to the east, or by remobilization from the nearby outcropping Cretaceous conglomerates (Praia Rei Cortiço). Because of their various sources, and mixing during transport, elemental concentrations are highly variable within a given gravel deposit. Quartzite and quartz in the lithic assemblages were derived from local gravels, given that virtually all were knapped from rounded pebbles/cobbles. As a result, matching artifacts to a specific source is nearly impossible and these materials do not provide useful information about human mobility.

Chert, in contrast, is only available at specific locations with different geochemical compositions. This allows a relatively good understanding of their distribution across the landscape as primary and secondary sources and, consequently, they can be used to infer human mobility and territoriality. While the most prominent chert sources in the

region are well-known and sampled for this study, we cannot rule out the possibility of other unknown primary and secondary sources that may currently be submerged, buried, or lost to erosion.

Macroscopic and low-magnification analysis

The macroscopic and low-magnification analysis of chert from Gruta Nova da Columbeira, following the methodology described above, identified five primary groups (Table 2).

The characteristics of this chert assemblage shows diversity in texture, mineralogy, and color, suggesting different depositional and diagenetic conditions. Most samples are fine-grained, ranging from mud-supported (Mudstone, Wackestone) to grain-supported (Packstone, Boundstone).

Mud-supported cherts indicate low-energy sedimentation of the host limestone layers, while grain-supported types point to higher-energy environments or biogenic frameworks. Cherts are generally compact, indicating low primary porosity, or high diagenetic silicification though some semi-porous varieties (e.g., CH6, CH7, CH8, CH11) may show secondary porosity from diagenesis. Iron oxides, Liesegang rings, and geodes indicate post-depositional alteration, including fluid-rock interactions and oxidation, while chalcedony and bioclasts contribute to variability in luster.

Mud-supported cherts, such as CH1, CH6, and CH9, are compact to semi-porous with predominantly semi-opaque to translucent transparency. These samples exhibit dull to waxy luster, with some showing notable chalcedony content (e.g., CH1). CH6 is distinguished by the presence of geodes, suggesting a diagenetic overprint involving silica-filled void spaces. CH9, on the other hand, stands out for its high translucency, possibly indicating a high degree of silica purity and a lack of detrital impurities. The Munsell color values range from light grayish brown (10YR 6/2) to darker gray tones (5YR 4/1), suggesting generally low iron content with subtle variation, and potential organic matter influences.

Wacke-supported cherts (e.g., CH2, CH4, and CH5) are compact with low porosity, exhibiting semi-opaque transparency and a mix of dull, vitreous, and waxy lusters. CH2 is notable for the presence of geodes, which suggest cavity-filling silica precipitation during late-stage diagenesis. CH4 and CH5 contain bioclasts, indicating biological input and potential silica replacement of carbonate components. CH5 further distinguishes itself with the presence of chalcedony, reinforcing the interpretation of a mixed biogenic and chemically precipitated silica origin. Color variability within this group is significant, with hues ranging from reddish brown (10R 5/4) to light gray (5YR 8/1), likely reflecting iron variations and diagenetic oxidation processes.

Table 3 Raw material distribution from layers 4 to 9 of Columbeira Cave, Mira Nascente and Praia Rei Cortiço

Context	Flint	Quartz	Quartzite	Others	Reference
Columbeira Cave Layer 4	37,4	35,5	27,1	0	Raposo et al., 2002
Columbeira Cave Layer 5	36,5	36,5	26,9	0,1	Raposo et al., 2002
Columbeira Cave Layer 6	30,8	43,8	25,4	0	Raposo et al., 2002
Columbeira Cave Layer 6a	34,1	36,2	29,7	0	Raposo et al., 2002
Columbeira Cave Layer 7	30,4	37,7	31,8	0,1	Raposo et al., 2002
Columbeira Cave Layer 8	26,9	39	34	0,1	Raposo et al., 2002
Columbeira Cave Layer 9	30,6	47,9	21,5	0	Raposo et al., 2002
Mira Nascente	86,6	12	1,4	0	Haws et al. 2020
Praia Rei Cortiço	3,1	29,5	67,0	0,4	Haws et al. 2020

A subset of samples, including CH3, CH10, and CH21, exhibit characteristics indicative of mixed mud-wacke compositions. These cherts maintain compact textures and semi-opaque transparency but display notable diagenetic features such as Liesegang rings (CH10, CH21) and iron concentrations. The Liesegang banding, particularly in CH10, suggests rhythmic silica precipitation influenced by fluctuating redox conditions. Iron-rich samples, such as CH21, exhibit darker hues (5R 3/4, 10R 2/2), consistent with ferruginous silica cementation.

Pack-supported cherts (CH7, CH14, and CH22) exhibit semi-porous to compact textures, with dull to waxy luster and moderate variability in bioclastic content. CH7, for instance, contains bioclasts and shows significant color heterogeneity (10YR 8/2 to 10R 2/2), potentially indicative of depositional variations in oxygenation levels. CH14 contains minor iron, which contribute to subtle reddish-brown tones (5Y 5/2, 10YR 8/6). These cherts may represent higher-energy depositional settings, where silica replacement of pre-existing carbonate frameworks was influenced by more dynamic diagenetic conditions.

Boundstone-associated cherts (e.g., CH8, CH24) exhibit a more massive texture, typically with opaque to semi-opaque transparency and waxy or dull luster. These samples occasionally contain geodes (CH8), suggesting syngenetic or early diagenetic silicification of carbonate-rich sediments. Their color profiles, ranging from white (N9) to reddish-brown (5YR 5/6), indicate a combination of primary mineral composition and later diagenetic alteration.

Overall, the chert assemblage reflects a spectrum of silica diagenetic processes, from primary biogenic sources to secondary chemical precipitation and silica replacement of pre-existing carbonate substrates prior to raw material selection and knapping. Variability in iron content, Liesegang banding, and porosity characteristics further suggest differential diagenetic histories, likely controlled by localized geochemical conditions and fluid-rock interactions.

It was possible to detect secondary patinas reflected mostly on the lustre of the assemblage. The great majority of the pieces analysed present a waxy to dull lustre. Thermal alterations after knapping caused thermal domes, cracking and a waxy sheen. Likewise, these elements may have alterations in their colouring and fracture pattern which partly limit the correct correlation of these pieces with macroscopic groups. When thermic alterations were evident, these elements were excluded from the raw material identification analysis. However, their presence is important to note because it confirms the occurrence of fires inside the cave, as suggested by the ash detected during the excavation (Cardoso et al. 2002).

pXRF results

The pXRF analysis (Supplementary information Table 1) was performed on the assemblage from Gruta Nova da Columbeira and a wider number of samples from LusoLit. Results show the predominance of silica often with values close to 100% but in some cases as low as 81,5%. Phosphorus, sulfur, and chlorine are present in all samples. Calcium is present in all archaeological samples from Gruta Nova da Columbeira and in several geological samples from primary outcrops and nearby gravels, reflecting the environmental inputs from the cave and their original association with limestone or dolostone.

Nickel is present in very few specimens, and it only appears in the samples from further north in the eastern façade of the Serra d'Aire and Candeeiros (Arneiro da Milharia and Espinheiro). Gold appears in several archaeological and geological samples, the latter from the eastern drainage of Serra d'Aire and Candeeiros. Samples from farther west are associated with secondary quartzite, quartz, and chert cobbles in the gravels of Leiria and Nazaré. Tungsten, barium, and silver appear in geological samples from this same region on either side of the Serra d'Aire e Candeeiros. Some of the geological samples from this area may have moved North–South due to the river dynamics, but they seem to be relatively consistent in their position. The samples with less silica are those that tend to have higher magnesium. Aluminum is present in most but not all specimens, which is a common feature of several sources in western Iberia.

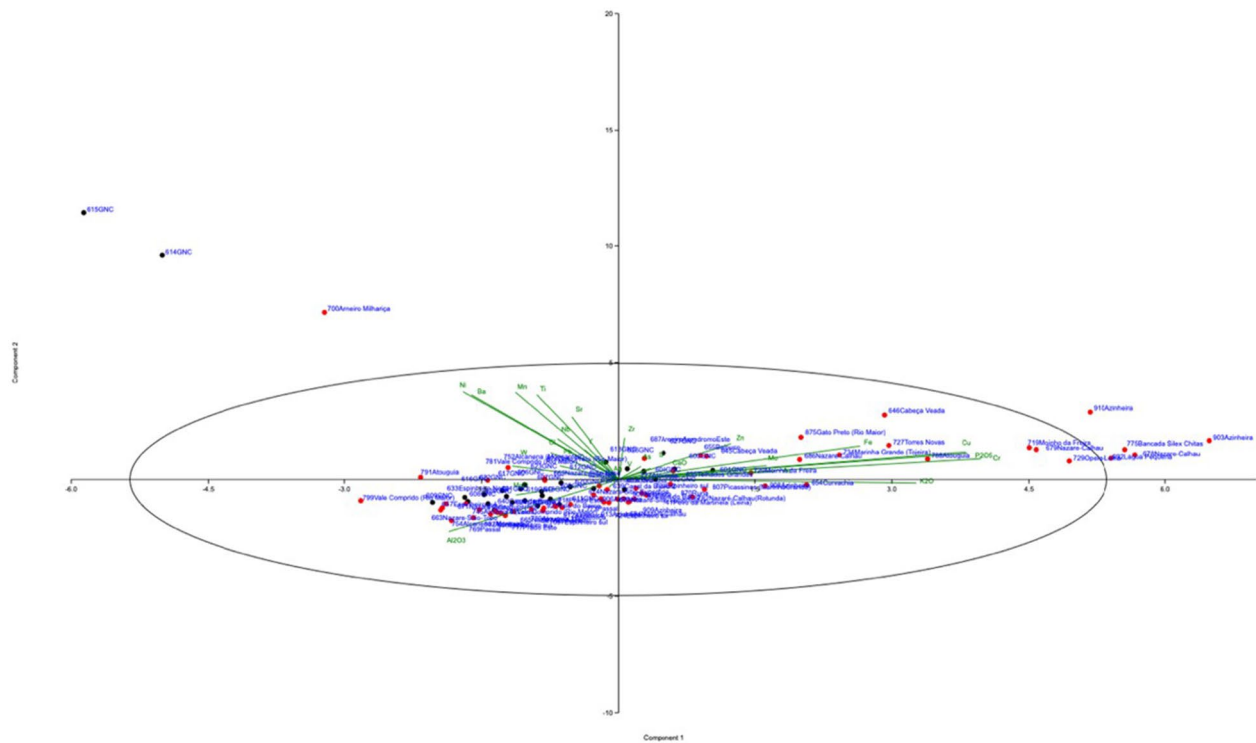
PCA shows that chert artefacts are mainly distributed within the composition bounds of the geological sources, indicating a strong geochemical compatibility between artefacts and sampled sources, without marked outliers. This suggests a systematic use of raw materials represented in the reference collection (Fig. 4a).

The cluster analysis has a cophenetic correlation coefficient of 0.7679, and shows two major branches. One links Gruta Nova da Columbeira with Rio Maior, Nazaré, and Leiria, but also with loose nodules from Serra d'Aire and Candeeiros. The other links with samples from Serra d'Aire and Candeeiros, Leiria, Nazaré and with gravels from the western parts of Rio Maior and Alenquer. This branch identifies several geological samples that do not cluster with archaeological specimens and vice-versa (Fig. 5a).

PIXE results

Silica is dominant throughout the samples, but its frequency shifts between 99,9% and 61,4% (Supplementary information Table 2). Phosphorus, sulfur, chlorine, potassium, calcium, titanium and manganese are the most common

a



b

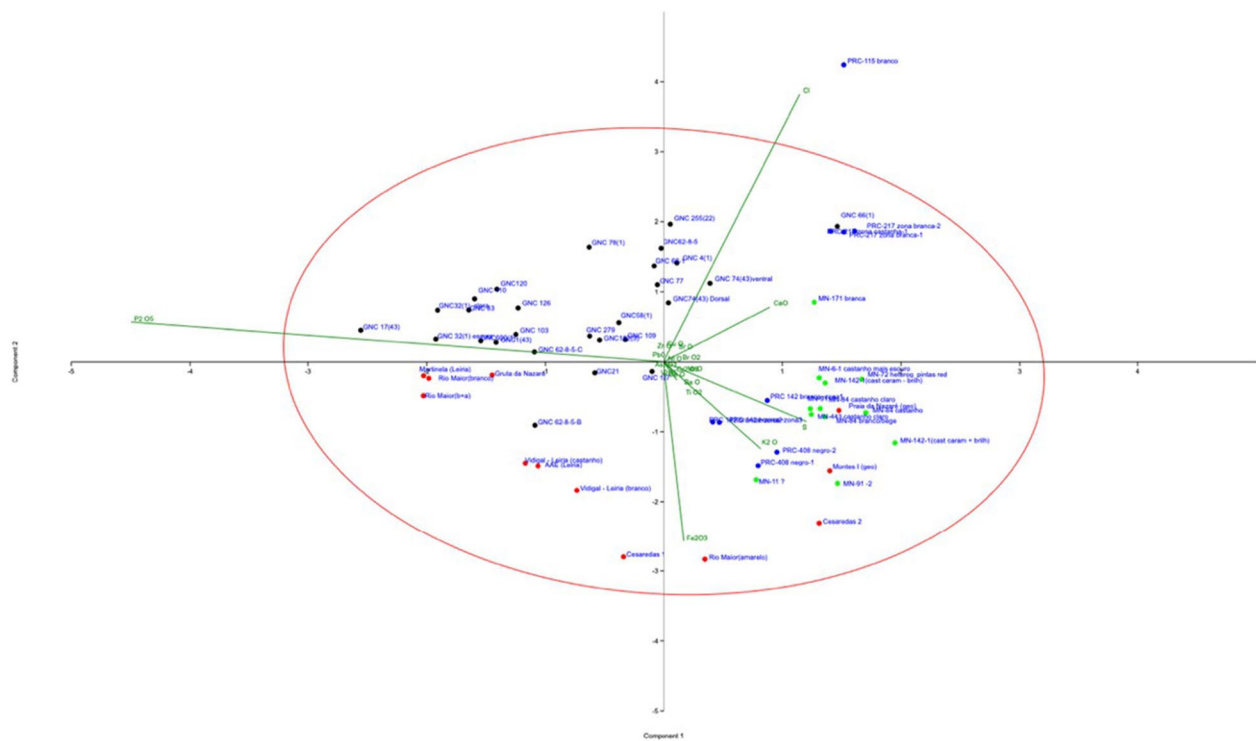


Fig. 4 a) PCA analysis with the pXRF results of Gruta Nova da Columbeira. b) PCA analysis with the PIXE results of Gruta Nova da Columbeira, Praia Rei Cortiço and Mira Nascente

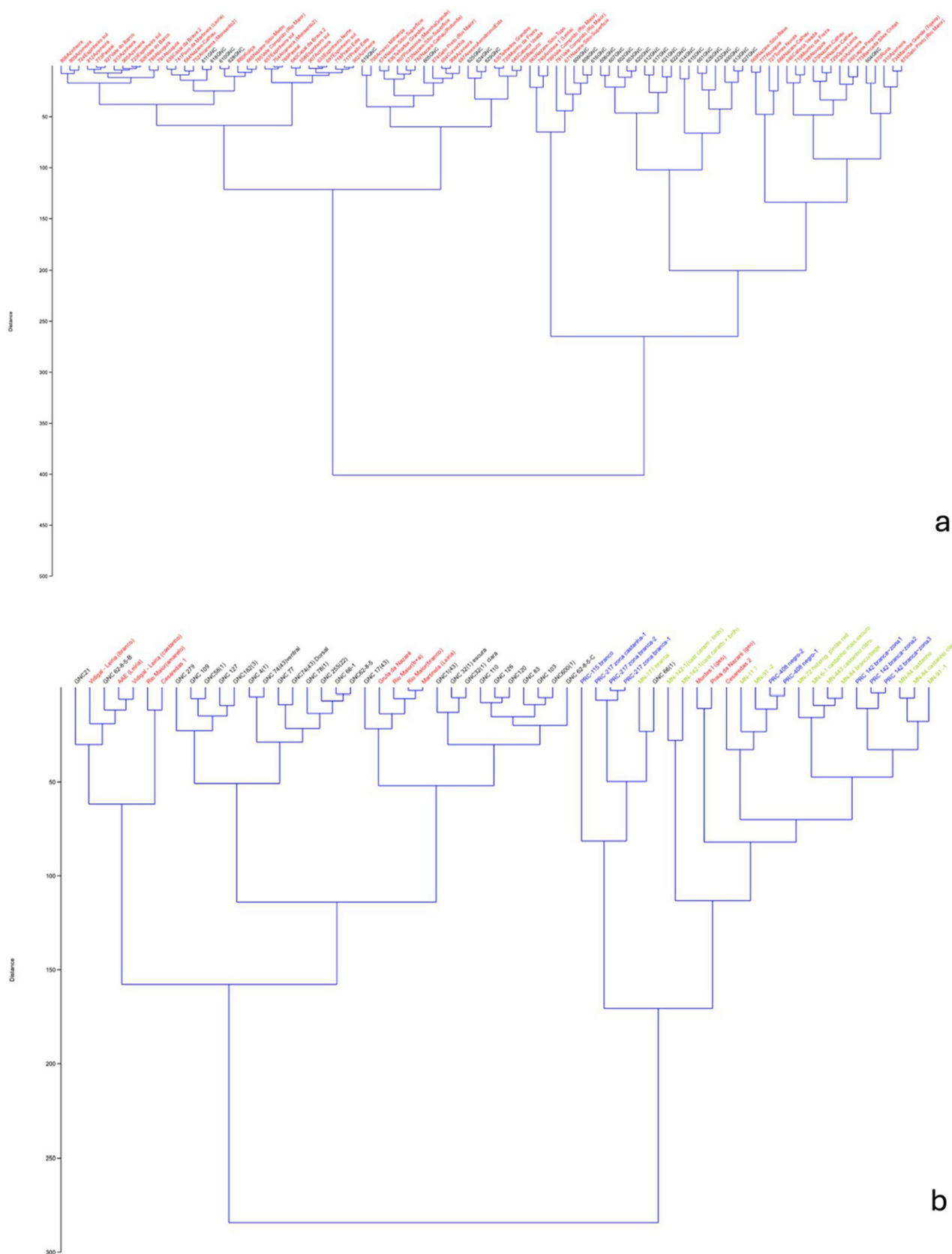


Fig. 5 Cluster analysis using Ward's Method Algorithm Euclidean Similarity index. **a)** pXRF results of Gruta Nova da Columbeira retrieving a cophenetic correlation coefficient of 0,7679; **b)** PIXE of Gruta Nova da Columbeira, Praia Rei Cortiço and Mira Nascente; retrieving a cophenetic correlation coefficient of 0,6111

elements while cobalt, nickel, copper, zinc, bromine and strontium are present in lesser amounts. Bromine (usually associated with evaporitic deposits) and strontium are only present in archaeological specimens from Praia de Rei Cortiço and Gruta Nova da Columbeira, which might indicate a local chemical signal that is not from the western face of Cesaredas, where two of the geological samples were collected. Nickel is only present in Nazaré and cobalt is only present in Nazaré and Montes (12 km inland from Nazaré), both areas more than 30 km from the cave.

The PCA on PIXE results is aligned with those from pXRF. In this case, the artefacts from all three sites show substantial overlap, and are mainly distributed within the 95% confidence bounds of the geological samples. This indicates overall geochemical compatibility between the geological samples and the archaeological assemblages, with only minor differences in the relative contribution of specific chemical elements (Fig. 4b).

The cluster analysis grouping, with a cophenetic correlation coefficient of 0.6111, shows two major branches. One includes archaeological specimens from Gruta Nova da Columbeira associated with geological samples from Leiria, Rio Maior, Cesaredas Plateau, and Nazaré. The other includes archaeological specimens from Praia Rei Cortiço, Mira Nascente and one from Gruta Nova da Columbeira associated with Montes, Nazaré, and Cesaredas Plateau (Fig. 5b).

Discussion

Silica of course dominates in all siliceous raw materials, and links between geological sources and archaeological materials are therefore established through variations in minor and trace elements. In the case of the study area, multi-proxy analyses give consistent results about the potential raw material sourcing for the three sites.

The combination of results seems to reveal the distance between the sites and the sampled sources (Fig. 6a-f and Table 4). The PIXE results show that the coastal sites of Praia Rei Cortiço and Mira Nascente seem to be related to the coastal sources of Nazaré, Montes and Cesaredas Plateau, at the western façade of the Serra d'Aire and Candeeiros mountains, but not with other sources located more inland that feed part of Gruta Nova da Columbeira. Since nickel is only present in Nazaré and cobalt is only present in Nazaré and Montes (12 km inland from Nazaré), these trace elements are common and unique to some geological strata of this coastal region. Unexpectedly, Mira Nascente, although closer to Leiria, does not cluster with the known sources from here, but has close similarities with Praia Rei Cortiço. Also surprising is the fact that some samples

from Mira Nascente and Praia Rei Cortiço are more similar between them than with any other geological specimen. This suggests acquisition of chert with similar properties from sources presently buried or submerged, despite the 35 km distance between the two sites.

In turn, Gruta Nova da Columbeira shows a diversified and dispersed range of acquisitions that includes outcrops from the Cesaredas Plateau (ca. 9.5 km to the west), gravels from Rio Maior (ca. 25 km to the east), the Nazaré outcrop (ca. 40 km to the north), and gravels from Leiria (more than 70 km to the north). This seems to suggest a wider territorial range of chert procurement compared with the other two sites.

The pXRF data are consistent with the PIXE results and further enrich them. It confirms that the chert assemblage from Gruta Nova da Columbeira clusters with samples from different areas, at different distances and directions. These include geological samples collected from gravels at the immediate vicinity of the cave (in a range of ca. 2.5 km), gravels from Rio Maior, outcrops and gravels from Leiria (above 70 km to the north), and even gravels at more than 90 km (also to the north). Not surprisingly, it also clusters with multiple samples collected from the Serra d'Aire and Candeeiros outcrops. Among them are samples collected from Telhados Grandes, Moinho da Freira and Bajouco, outcrops that are close to each other. Presently, these outcrops are only known at the SE face of the Serra d'Aire and Candeeiros. However, since the geological strata continue across the mountain, tracing about 1 km westward to a cliff near the head of the Lis drainage, we hypothesize that some nodules may also be present at the western face of the mountain, in areas not yet surveyed and sampled.

In the case of the cluster with the sample from the gravels of Arneiro da Milharia and Alcanena, it is also possible that its original outcrops may be at a higher altitude of the Serra d'Aire and Candeeiros, and even that other nodules coming from these outcrops could be available further downstream in the Tejo basin. The same logic applies to the sample collected at Casal da Brava, at more than 90 km towards the north, in this case linked to the Condeixa-Sicó-Alvaiázere mountain.

In other regions where raw material sourcing has been systematically investigated over the years, the chert outcrops are mapped in detail, and their petrography and geochemical characteristics are well established (e.g. Eixea et al. 2014, 2016; Rios-Garaizar and García Moreno 2015; Prieto et al. 2016; Vallejo Rodríguez et al. 2017; Abrunhosa et al. 2020; Gómez De Soler et al. 2020, 2023; Mayor et al. 2022; Sánchez De La Torre et al. 2023; Ramírez-Amador et al. 2025; Herrero-Alonso et al. 2025). Therefore, it is possible to link the Neanderthal sites to the sources with great confidence and distinguish when the acquisition was

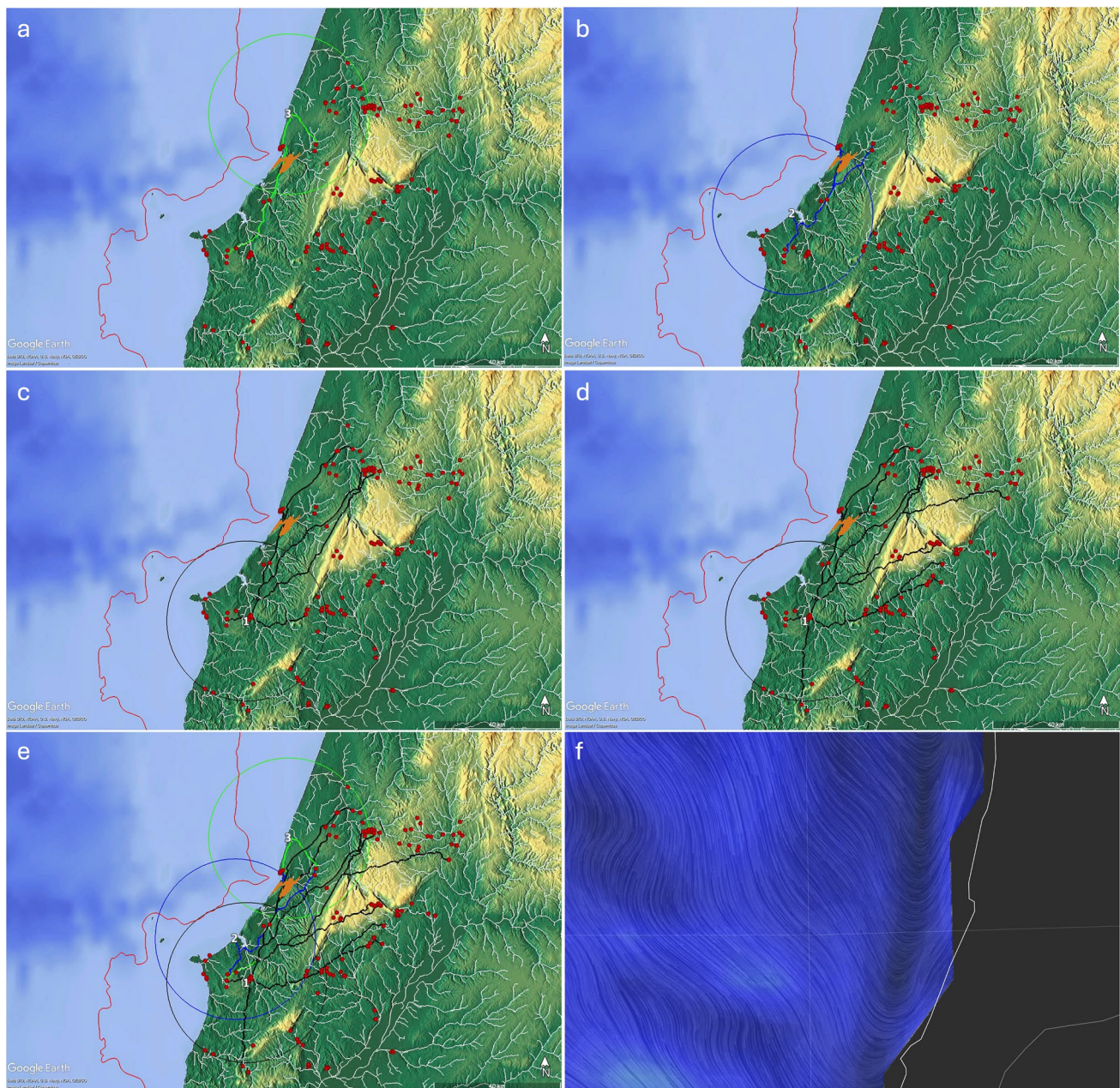


Fig. 6 Tracks linking the sites to the sources. Numbers: 1- Gruta Nova da Columbeira; 2- Praia Rei Cortiço; 3- Mira Nascente. Red dots: chert sources. Black lines and circle: Gruta Nova da Columbeira; Blue lines and circle: Praia Rei Cortiço; Green lines and circle: Mira Nascente. Orange polygon: Lagoa da Pederneira. **a)** Mira Nascente—PIXE; **b)** Praia Rei Cortiço—PIXE; **c)** Gruta Nova da Columbeira—PIXE; **d)** Gruta Nova da Columbeira—pXRF; **e)** All PIXE and pXRF results together; **f)** Details of the North-South direction ocean currents at 5 m below the surface (purple) along the coast of Portugal (Retrieved from: <https://earth.nullschool.net/#current/ocean/surface/currents/>)

made from primary or secondary sources. In the Portuguese Estremadura, it is still a bit difficult because this research is only at its beginning, with very few people working on it (Matias 2012; Aubry et al. 2012; Pereira et al. 2016; Jordão 2023). In fact, this was the first large batch of geochemical analysis ever performed in the region, using different methods at different times and a reference collection that was under construction and that grew afterwards.

For these reasons, but mostly because of the characteristics of the region (slopes and cliffs with outcrops exposed to the elements and streams at the bottom, along with a dense hydraulic network), we are cautious about assuming that such distances above 50 km correspond de facto to human transportation. Our hesitation is grounded in the geomorphological features of the region. Most of the geological nodules were collected from gravels located downstream

Table 4 Distance between the sites (GNC=Gruta Nova da Columbeira; MN=Mira Nascente; PRC=Praia Rei Cortiço) and the chert sources from where a geological sample clustered with an archaeological one, considering walking distance, time and orientation

Site	Method	Source	Km	Time	Orientation
GNC	pXRF	Runa	31,4	7h12	SSE
GNC	pXRF	Roliça	2,4	0h31	NE
GNC	pXRF	Nazaré—Sítio/Gruta	41,1	9h28	NNW
GNC	pXRF	Vale Comprido	27,2	6h26	E
GNC	pXRF	Arroteia	60,9	13h58	NNE
GNC	pXRF	Bajouco	61,1	14h30	NE
GNC	pXRF	Cabeça Vead	44,2	10h33	NE
GNC	pXRF	Moinho da Freira	60,1	14h14	NE
GNC	pXRF	Telhados Grandes	59,1	14h00	NE
GNC	pXRF	Areeiro Aeródromo Este	72,6	16h38	NNE
GNC	pXRF	Curvachia	73,9	17h21	NE
GNC	pXRF	Gato Preto	30,9	7h15	E
GNC	pXRF	Arneiro das Milhariças	56	13h02	ENE
GNC	pXRF	Alcanena	59,1	13h42	NE
GNC	pXRF	Martinela	74,4	17h24	NE
GNC	pXRF	Casal da Brava	93,2	21h46	NE
GNC	PIXE	Vidigal	70,3	16h30	NNE
GNC	PIXE	Areeiro Aeródromo Este	72,6	16h38	NNE
GNC	PIXE	Rio Maior	24,6	5h51	E
GNC	PIXE	Cezaredas	9,4	2h16	W
GNC	PIXE	Martinela	74,4	17h24	NE
GNC	PIXE	Nazaré—Sítio/Gruta	41,1	9h28	NNW
PRC	PIXE	Montes	48,3	11h23	NE
PRC	PIXE	Cezaredas	17,4	4h07	S
PRC	PIXE	Nazaré—Sítio/Gruta	41,3	9h33	NE
MN	PIXE	Montes	14,4	3h20	SE
MN	PIXE	Cezaredas	57,3	13h11	SW
MN	PIXE	Nazaré—Sítio/Gruta	12,7	2h57	N

from the sources and towards the sites. Of course, this means the original outcrop is even further away, but also that other nodules may have been closer. The same logic applies to the samples collected from primary outcrops, but where surface processes bring nodules closer to the sites.

The Serra d'Aire and Candeeiros mountains are cut by streams that run across chert outcrops, displacing, mixing, and transporting nodules across plains and terraces, towards the ocean. This includes Lagoa da Pederneira (literally “gun-flint lagoon”) south of Nazaré, the basin of the river Lis and the coast, making it one of the most prominent chert sources in the region. However, the thick dune field that covers this area, which reaches dozens of meters (Gonçalves 2007), the rise of the ocean during the Holocene, the steep landscape, and the thorny vegetation that covers it made it difficult to survey for chert nodules here.

The fact that the river Lis basin drains towards the north gives a misleading impression that it removes chert away from the archaeological sites in the study area. However, after the nodules are flushed into the Atlantic Ocean, the longshore ocean currents (that run in an NNW-SSE

direction) scatter them southward across the coast of the region (Fig. 6g). The dynamic graphic can be seen at (online source: <https://portal.emodnet-bathymetry.eu/>). These nodules would then be interspersed with those from fluvial gravels and Cretaceous conglomerates in the vicinity of the sites.

Therefore, although the geochemical analysis clusters the site with geological samples collected from the Lis, over 70 km to the north, nodules from the same original outcrop may have been brought and accumulated by ocean currents to places much closer to Praia Rei Cortiço or Gruta Nova da Columbeira. Therefore, such samples must be seen as a maximum distance from the site and not necessarily the place of acquisition.

This logic does not apply to the cluster between Gruta Nova da Columbeira and Runa or between the samples from this same cave and Praia Rei Cortiço with Rio Maior, however. This is because the natural movement of chert by the streams and rivers moves the nodules farther away to the south of the study area. Therefore, their presence at these sites may be better explained by human action. It is worth noting that the best passage between the western and eastern façades of Serra de Montejunto and Serra d'Aire e Candeeiros, that separate the coastal area from the Tejo basin, is exactly at Rio Maior.

We note that some archaeological and geological cherts do not cluster with any of the others. This could be a result of limited sample sizes, suggesting that more survey and sampling effort is needed to obtain a comprehensive comparison data set. Another explanation could be related to the sourcing from gravels presently located underwater on the continental platform, and/or covered by dunes. Pointing in this direction is the combination of small amounts of cortex (that is almost always waterworn, indicating transportation by water), high curation rates (that independently indicate distance decay and, in association, reinforce that possibility), and the fact that many of the archaeological samples not clustering are from Praia Rei Cortiço and Mira Nascente, both presently at the coast. Of course, the same process would impact the assemblage at Gruta Nova da Columbeira. Despite the coastline being only ca. 15 km away from the cave at present, during MIS5 and the MIS5/4 transition, sea level fall (Paine et al. 2024) may have increased that distance up to some 30 km or more (Alveirinho Dias et al. 1997; Benedetti et al. 2009). This could expose coastal gravels and bluffs as potential rich chert sources that could not be sampled in the present day.

The difficulties in disentangling some sources may be partially related to the surface-sensitive nature of the methods used. PIXE and pXRF have several advantages for chert provenance studies because they provide high-resolution, non-destructive and rapid data acquisition. However, the

shallow pXRF analysis, although previously used for raw material sourcing by Neanderthals can be influenced by stronger matrix effects, is more susceptible to surface alterations, weathering, and post-depositional contamination, and is limited to major and some minor elements. This makes many trace elements and rare earth elements (often critical for high-resolution provenance) remain undetected (Shackley 2011). Nevertheless, it allows to analysing large sample sets directly in the field or museum collections, making it particularly suitable for exploratory and comparative studies of lithic raw materials with rapid and relatively affordable costs.

On the other hand, PIXE allows for refining in greater detail each sample, particularly concerning their light to medium elements, but it is not portable, and the costs are very high. Despite its usefulness for raw material sourcing by Neanderthals (Mathis et al. 2014; Pitarch Martí and d'Errico 2018), these features limit its use. Therefore, the combination of these two methods seems to be a feasible approach, particularly if pXRF is used at a larger scale (not only, but also) for screening the samples. Still, these should also be combined with petrographic through thin section observation, and LA-ICP-MS to better identify trace elements, rare earth elements, matrix, post-depositional alterations, and fossils.

Nevertheless, there is a consistent geochemical overlap between artefacts from the three sites with the sampled geological sources, along with the absence of artefacts clustering outside the range of variability of the sampled geological sources. Together, this seems to support an interpretation of continuous procurement of chert in sources within the geochemical variability in the region. In other words, these three sites seem to have broadly shared the same raw material landscape over the entire time span of Neanderthals' occupation of this territory, with limited transport of raw materials from very long distances. This is particularly interesting because this territory is adjacent to that exploited by the Neanderthals who occupied Gruta da Oliveira (Matias 2012, 2016; Linscott et al. 2023). At this point, the exploitation of the chert-rich region of Rio Maior by both of those occupying Gruta Nova da Columbeira and Gruta da Oliveira should not be under- or overvalued. It may relate to transiting between these regions by the same community, contact between the communities living in the two territories, or the exploitation of the same chert-rich area by communities from each side of the mountains, without any significant relationship. This last hypothesis seems to align better with the isotopic analysis made on human remains from Gruta da Oliveira (Linscott et al. 2023).

From a paleoanthropological perspective, the pattern seen in this exploratory project is consistent with flexible and opportunistic procurement strategies, where selection

criteria may have been likely driven by availability and accessibility rather than strict preferences for narrowly defined raw material qualities and sources.

Overall, this exploratory study seems to confirm the validity of geochemical analysis for assessing raw material sourcing when combined with multivariate statistics, at least for chert artifacts in the study area. The geochemical analysis also shows intra-source heterogeneity and geological continuity across the region, that is, some sources have some internal variability, probably related to geological and sedimentary inputs, mineral inclusions, or diagenetic processes. This is not a problem, however, because it gives the compositional differences that characterise each source and, therefore, help in their identification. Nevertheless, the reasons for these internal variabilities should be explored further in the future.

Conclusion and future remarks

This multi-proxy study, although exploratory, gave some consistent results and opened future avenues of research.

First, it confirmed that raw material sourcing is a viable investigation in the region, including through geochemical methods, as it was already confirmed following previous works on assemblages from open-air, rock shelters and cave sites by previous works in contexts of more recent chronologies.

Second, it suggests that sourcing at Gruta Nova da Columbeira, Mira Nascente and Praia Rei Cortiço occurred predominantly between the north limit of Torres Vedras source and the NE façade of Serra Aire e Candeeiros (at most), and the western façades of Montejunto, and Serra Aire e Candeeiros mountains and the coast. This territory at the western façade of the Montejunto – Serra Aire e Candeeiros mountains is adjacent to that exploited by the Neanderthals occupying Gruta da Oliveira.

This exploratory investigation also did not address some aspects and opened new questions.

First, Neanderthals occupying Gruta Nova da Columbeira and Gruta da Oliveira exploited the area of Rio Maior, which is the richest chert area of Portugal. Rio Maior is situated precisely within the corridor between the two mountains that connect the western coastal region and the southeastern Tejo basin. However, the significance of such exploitation in these two sites warrants further investigation due to its potential economic and paleoanthropological implications.

Second, it is still unknown which were the driving forces of the economy and mobility of the western-most Neanderthals. This could be accessed by delving in detail into the raw material sourcing and economy of each occupational level, namely by focusing on volume, weight, production, curation

of each group and subgroups of raw materials, and their relationship with the time and energy costs of acquisition.

In summary, the exploratory chert sourcing of Gruta Nova da Columbeira, Mira Nascente and Praia Rei Cortiço suggests complex sourcing patterns within relatively delimited territories following other Iberian cases. This suggests a deep knowledge of the landscape and an elaborate management of its resources that may be behind the long Neanderthal survival.

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Data availability All data available in the article or in the supplementary Information.

Code availability Not applicable.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

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