Geochemical fingerprinting of Monegros cherts: re-defining the origin of a prehistoric tracer

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Abstract

The geographical name of Monegros has traditionally been used to describe a high-quality dark-coloured chert originating in a carbonate lacustrine environment, being one of the most important long-distance tracers in SW Europe during Prehistory. This chert type outcrops in Monegros region, situated in the Ebro Basin (NE Iberia), but not only there, as cherts with same descriptions are found in other regions, some far from the homonymous area. Nevertheless, prehistorians working on the characterisation of lithic sources have frequently used the term Monegros to define this chert basing their attributions solely on macroscopic descriptions. These are not sufficient as they do not allow cherts from Monegros region to be distinguished from cherts from other regions.

In this study, the area where Monegros cherts outcrop has been delimited and field work has been carried out to identify the origin of the geological formations and the preserved outcrops. The classic approach has proved to be insufficient for this purpose, so geochemical fingerprinting using energy-dispersive X-ray fluorescence (ED-XRF) has been performed. The results obtained after this first geochemical approach show that some differences can be found between the different formations in the Monegros region in terms of their major and minor components.

Keywords

Geochemistry, ED-XRF, Monegros cherts, lithic procurement

1. Introduction

The geographical name of Monegros has traditionally been used by prehistorians from Iberia and France to describe a high-quality, dark-coloured chert originating in a carbonate lacustrine environment that usually contains Liesegang rings as well as charophyte algae and some gastropods. Their supposed outcrops have frequently been located loosely in the middle Ebro Basin, without specifications of the geological formation of origin (Roy-Sunyer, 2016; Roy-Sunyer et al., 2013; Vaquer and Remicourt, 2006; Vaquer and Vergély, 2006).

This Monegros chert type has been seen by prehistorians as one of the most exploited chert sources in NE Iberia during Prehistory and it is supposed to have been a long distance tracer for some specific periods. Some authors have attributed this origin to cherts appearing as far away as the South of France for materials ascribed to the Neolithic and Chalcolithic periods (Gandelin et al., 2006; Perrin et al., 2006; Vaquer and Remicourt, 2006; Vaquer and Vergély, 2006) and for the Upper Palaeolithic (Foucher, 2015). Although a handy denomination, it has to be highlighted that most of the studies that have used the term Monegros to describe a specific chert type lack a detailed analytical basis. Thus, these associations are only based on visual characteristics, and no specific study has been done integrating microscopic or geochemical analyses to directly relate the supposed Monegros cherts appearing in the archaeological record with the geological formations of origin.

With the aim of shedding light on this issue, in recent years our team from the University of Zaragoza has been working to redefine the term “Monegros”. We have first tried to delimit the area where Monegros cherts outcrop and subsequently undertaken field work to identify the different geological formations containing chert and to determine the nature and characteristics of the preserved outcrops.

Our recent studies, some of which have been published, examine the different geological formations containing chert attributed to the Monegros type and describe the main macroscopic and petrographic features of each type (García-Simón, 2018; García-Simón and Domingo, 2016). Nevertheless, despite efforts to accurately determine the main characteristics of each chert type, macroscopic and petrographic studies are not sufficient as in most cases the main features appear to be regularly repeated in the different formations under study. Given the impossibility of distinguishing between formations by classic studies, geochemical fingerprinting by the use of energy-dispersive X-ray fluorescence (ED-XRF) has been applied to determine the major and minor components for each chert formation and outcrop.

2. Monegros cherts: geographic and geologic context

Monegros is a large natural region located in Aragón (Spain), in the heart of the middle Ebro Basin, occupying part of the provinces of Zaragoza and Huesca. In recent decades, massive irrigation works in the area have partially transformed its landscape. Nevertheless, despite these newly cultivated spots, the Monegros region is still one of the most arid areas in Western Europe.

The geological history of the Ebro Basin was systematized following the stratigraphy documented in the Alcubierre ranges, the highest elevation of the middle Depression (Pardo, 2004; Quirantes, 1978). These ranges organise the Monegros area, whose name (Monegros – Montes Negros - Black Mountains) derives from the traditional abundance of pines and kermes oaks. In geological terms, the Monegros boundaries lie in the north with the PrePyrenean Ranges, in the south with the Iberian Ranges, in the east with the Catalan Coastal Ranges and in the west with the Bureba Corridor which links the Ebro Basin to the Duero Basin and the Northern Plateau. Its geological features are simple: from the Late Eocene (Upper Priabonian) to the Upper or Middle Miocene (Upper Vallesian or Turolian), the centre of the basin was an endorreic system that received water from the rivers flowing from the surrounding ranges. In the margins of the basin, various systems of fluvial and alluvial fans deposited the current lithologies, while in the central area there were carbonated and evaporitic lakes. These lakes were gradually displaced southwards when the Pyrenean rising (Alpine Orogeny) took place (Arenas et al., 1999).

Between 12 and 8 million years ago, there was an exoreic episode that caused the erosive removal of the basin deposits to the Mediterranean and started the shaping of the present-day landscape, as a result of the activity of a growing fluvial network under semiarid climatic conditions. Nowadays, several hydrological systems structure the territory and delimit the Neogene geological formations. The Ebro River is the main watercourse, some of the most important tributaries being the Gállego and Alcanadre (a secondary tributary to the Ebro, because it belongs to the Cinca river sub-basin) in the north and the Jalón, Huerva, Martín, Guadalope and Matarraña in the south. These tributaries have modelled the Tertiary deposits and the Quaternary detrital formations that cover them.

The top part of the reliefs (Castellar, Alcubierre ranges, Monte Oscuro and Sigena to the north of the Ebro River and La Muela and La Plana to the south) are formed by structural calcareous platforms, where the siliceous nodules are nowadays embedded. The highest elevations barely surpass 800 m asl. But prior to the setting of the carbonate lacustrine systems that led to the formation of the massive limestones, the centre of the basin was occupied by several evaporitic lacustrine systems framed by others of carbonate character. Besides limestones and gypsums, marls, sandstones and conglomerate layers and clay formations make up the Monegros lithology (Quirantes, 1978).

3. Materials and Methods

3.1 Materials: surveys and previous analyses

In this study, four geological units containing lacustrine cherts and outcropping in the Monegros region were considered for analysis: 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. In order to collect chert samples and to characterise their outcrops, some field surveys were systematically done. Several outcrops were located and analysed, with more than 80 chert outcrops points identified in the Middle Ebro Basin. Eight of these were selected for specific study, as they possessed the suitable features for use by past societies – an abundance of good knapping chert nodules together with their easy extraction from the bearing rock – (**Figure 1**). Each of the eight selected outcrops was described in a specific database, outlining their main characteristics. Samples were then collected with the aim of obtaining a broad representation of the internal variability of the outcrop. After macroscopic observations and petrographic characterisations that are briefly presented here (**Figure 2**), a total of 139 samples from the 8 different outcrops were selected for geochemical analysis. In order to improve analysis time and avoid surface alterations, all the samples were prepared in squares of 5 x 5 mm without cortex surfaces.

Figure 1 – The Monegros region, in NE Iberia, with the location of the chert outcrops selected for this study (geographical map). Geological map: *Quaternary:* 174- Conglomerate, sandstone, gravel, sand, silt and clay; 173- Conglomerate, gravel, sand, lutite, marl, calcarenite, travertine limestone and toba. *Miocene*: 164- Conglomerate, sandstone, lutite, limestone, marl and gypsum; 163- Conglomerate, sandstone, lutite, limestone and gypsum. Source (for the geological map): (IGME, 1998).

The **Sierra de Pallaruelo – Monte de la Sora Unit** (Lower Aragonian, Miocene) takes its name from the Sierra de Pallaruelo for its terrigenous lithofacies, while the name Monte de la Sora is preferred for the carbonate lithofacies. This unit is constituted at its base by pelitic sediments of distant alluvial origin, between which are interspersed some sandstone and limestone levels. The top of the unit is almost completely constituted by margo-carbonate levels of lacustrine origin that usually contain lacustrine chert nodules (Quirantes, 1978).

Cherts from the Sierra de Pallaruelo – Monte de la Sora Unit have been identified in two areas of the studied region. First, we have detected some chert outcrops at the top part of the calcareous relief where nowadays stands the hermitage of Santa Quiteria, near the town of La Almolda (Zaragoza, Spain). Several siliceous outcrops were identified, one of which (*Santa Quiteria –SQ-)* was selected for the geochemical analysis. Moreover, cherts from this unit were also detected in outcrops near the towns of Muel and Mezalocha (Zaragoza, Spain), in the hills of San Borombón. Chert outcrops are found all over the hill, with more than 20 outcropping points being identified. Most of these show evidence of past knapping –mostly from the 18th and 19th centuries- directly related with the production of gunflints. One outcrop from this area was selected for geochemical analysis: *San Borombón –SB*–. Thirty-three samples from the two studied outcrops –SQ and SB– were selected for geochemistry. Macroscopically, cherts from this unit possess calcareous cortex and nodular morphologies, with fine grain and high knapping aptitude. They have a wackestone type texture, with inclusions of metal oxides, organic matter and some carbonate relicts. Charophyte algae are abundant, as well as some gastropod sections. In some samples Liesegang rings appear. Under the microscope, it can be seen that these cherts are mainly composed of a mosaic of cryptoquartz, which is the main texture. Some length-fast chalcedony is observed mostly filling porosities.

The **Sierra de Lanaja – Montes de Castejón Unit** (Upper Aragonian, Miocene) lies in the Eastern area of the Ebro Basin and is mainly composed of distant alluvial facies and fan border facies at the base, and margo-carbonate series originated in a lacustrine environment at the top. This is the carbonated unit par excellence, being responsible for shaping the reliefs that characterise the Tertiary of the Middle Ebro Basin. This unit inludes limestones from the Montes de Castejón, La Muela, La Plana de Zaragoza, Alcubierre and Sigena areas. Among their lithologies, chert is detected, being one of the best quality chert types from the Monegros region (Quirantes, 1978).

Cherts coming from the Sierra de Lanaja – Montes de Castejón Unit appear embedded in the silty limestones that form the San Caprasio hill, next to the village of Farlete (Zaragoza province) (*La Torraza –LT–* outcrop) as well as in the hills next to the villages of Muel (Zaragoza province) (*Campo de las Horgas –HOR–* outcrop) and La Muela (*La Muela 1 –LM1–* and *La Muela 2 –LM2–).* In the La Muela outcrops, evidence of past knapping was found during our surveys, these being mostly related to the production of gunflints during the 18th and 19th centuries (Barandiarán, 1974; Tarriño et al., 2016). Four outcrops located in different areas of the studied region were selected for geochemical analysis. A total of 63 samples from this unit were studied. Macroscopically, these cherts are almost identical in the four selected outcrops, possessing thin calcareous cortex and good knappability. Their texture can be defined as a wackestone type, with abundant carbonate relicts, some fragments of Charophyte algae and scarce lacustrine gastropods. In some samples Liesegang rings appear. Microscopically, cryptoquartz is the main texture, with other silica forms also present such as length-fast chalcedony filling ancient porosities. Carbonate relicts are still abundant, mostly in the form of micrite and in some cases calcite or dolomite rhombohedral crystals.

The **Bujaraloz – Sariñena Unit** (Agenian - Aragonian, Miocene) is fundamentally constituted by evaporitic facies present in a large extension of the central sector of the Ebro Basin. Towards the northern margin, this unit is constituted by an alternation of reddish-brownish clays, sandstone palaeochannels and, locally, layers of limestones, which together represent distal fluvial fan facies. In the southeastern area of the Ebro Basin, these materials are crowned by carbonate levels of lacustrine origin. Towards the northwest, they progressively pass to marls with gypsum nodules from salt-lake margins. Cherts are embedded within the carbonated sediments of lacustrine origin, appearing mostly at the southeast of the basin (Quirantes, 1978).

Cherts were recovered within the stratified limestones and marls intercalated with clay outcropping between the towns of Peralba and Candasnos (Zaragoza province). The outcrop selected for this study was named *Puente Candasnos –PC–*. Cherts from this outcrop possess irregular nodular morphologies with homogeneous textures with metal oxides, carbonate remains, probably organic matter and detrital quartz crystals. Charophyte algae and gastropod sections configure the micropalaeontological content. In some samples Liesegang rings appear. At microscopic scale, a cryptoquartz mosaic is the main texture, with some length-fast chalcedony and macroquartz cementations. 23 samples coming from the same outcrop were selected for geochemical analysis.

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 Ebro Basin, the unit represents an evolution of fluvial facies of alluvial fan at the bottom towards lacustrine conditions at the top. The unit is mainly carbonated, composed of limestones, marls, shales and, in a lesser quantity, sandstones and lignites. These carbonated sediments originated in shallow lacustrine conditions in the central area of the basin. Two cartographic parts have been differentiated as components of this unit: a lower one mostly composed of clays and sandstones, and an upper one dominated by limestones and marls that frequently contain chert. An important complex of several chert outcrops has been identified in Valcuerna (Peñalba, Huesca). This is a long ravine that links the eastern foothills of the Alcubierre ranges to the Ebro River watercourse, being a clear passageway providing access different territories. The calcareous steps that frame the ravine hold massive quantities of isolated chert nodules and siliceous layers embedded in the matrix where they originated. Erosive processes set them loose from the limestone levels, being the rocks easily accessible at the bottom of the flat-floor valleys (Luzón et al., 2002).

From the 15 outcrops identified in the Valcuerna ravine, one has been selected for analysis in this fingerprinting geochemical study. It has been named *Valcuerna* –VA–, and 20 samples representing the main diversity observed in the outcrop have been selected for geochemical analysis. From the macroscopic perspective, these cherts possess a thin calcareous cortex, they are fine-grain and have excellent knappability. The texture, which is a wackestone type, has inclusions of metallic oxides, possible organic matter and abundant carbonate relicts. Bioclasts in the form of Charophyte algae and lacustrine gastropods are abundant. In some samples Liesegang rings appear. Microscopically, a cryptoquartz mosaic is the main texture, with cementations of length-fast chalcedony filling ancient porosities. Micrite carbonates are still frequent, as well as some calcite rhombohedral crystals.

Figure 2 – Main views of the outcrops selected for geochemical analysis.

3.2 Methods: ED-XRF

ED-XRF (energy-dispersive X-ray fluorescece) was applied to analyse major and minor elements in the samples. The analyses were carried out at the Research Centre for Applied Physics in Archaeology, IRAMAT, Bordeaux, France. Nine elements were quantified (Na, Mg, Al, Si, P, K, Ca, Ti, Fe) using an X-ray fluorescence spectrometer SEIKO SEA 6000VX (Orange et al., 2017). Fundamental parameters corrected by the granodiorite GSP2 from the U.S Geological Survey (USGS) international standard (Wilson, 1998) were used. A 3 x 3 mm collimator was employed and the analysis time was set to 400 live seconds for each measurement condition (3 conditions with air or He environment and Cr or Pb filter were established). To check the machine calibration and accuracy, the JCh-1 chert standard from the Geological Survey of Japan (GSJ) international standard was used (Imai et al., 1996). To prove and validate the receipt and to check the machine accuracy, several measurements with the JCh-1 chert standard were established. The results showed that the standard deviation was always lower than 0.08 w%, validating the accuracy and precision of the receipt. More detailed information can be found in an already published paper (Sánchez de la Torre et al., 2017). Statistical works were carried out using XLSTAT software (Addinsoft, 2017).

4. Results and discussion

Despite having quantified nine elements using energy dispersive X-ray fluorescence (Na_2O, MgO, Al_2O_3, SiO_2, P_2O_5, K_2O, CaO, TiO_2 and Fe_2O_3), only five were used for interpretation (Al_2O_3, SiO_2, K_2O, CaO and Fe_2O_3) as the values obtained for Na_2O, MgO, P_2O_5 and TiO_2 were below the detection limits of the instrument used. The main raw data can be found in the Supplementary Material. With the five well-dosed elements, whose averages were almost always beyond the detection limits of the device, boxplots were elaborated with the aim of determining the existent variability for each outcrop and the relation with the other outcrops and formations (**Figure 3**) (**table 1**).

Table 1 – ED-XRF analytical data (in w%) with the mean, median and standard deviation (std. dev.) obtained for each outcrop.

In the Al_2O_3 boxplot (top left corner) it can be observed that outcrops from the Lanaja-Castejón formation have the greatest internal variability, the most evident case being the LM1 outcrop. In contrast, the outcrops from the Cinca formation (VA) and the Bujaraloz-Sariñena formation (PC) are the most homogeneous, with low internal variability. As regards the differences between the formations, it seems that cherts from the Cinca formation (VA) are the only ones that are clearly separate from the rest, having the lowest quantities of Al_2O_3 quantities. The other cherts had similar higher values.

In the SiO_2 boxplot (top middle), the LT outcrop from the Lanaja-Castejón formation can clearly be differentiated from the other groups as it has high internal variability and the lowest SiO_2 averages, below 97w%. The lowest internal variability is observed in the LM2 outcrop, being the most homogeneous. The outcrops from the Pallaruelo-Sora formation possess different SiO_2 values, being possible to distinguish between outcrops from the same formation.

In the K_2O boxplot (top right corner), despite some differences in the same formation –in the Lanaja-Castejón group, where LM1 and LT have higher averages and LM2 and HOR lower proportions, no clear differences can be established between the groups as they all possess quite homogeneous values.

The boxplot representing CaO values (bottom left corner) is interesting as it clearly separates the LT outcrop from the other outcrops in the Lanaja-Castejón group and also from the other formations. Moreover, this outcrop possesses high internal variability, while the other outcrops from this group are quite homogeneous. Differences can also be established between the SB and SQ outcrops in the Pallaruelo-Sora group, the former being more similar and the latter possessing higher internal variability.

Finally, the boxplot representing Fe_2O_3 values (bottom right corner) seems to be quite different for each outcrop, differences also being observed in the frame of a same group. Some variations could be established between formations, the Pallaruelo-Sora group having the highest values and the Cinca formation having the lowest.

Figure 3 – Boxplots with the different values (in wt%) obtained after ED-XRF analyses. From top left to bottom right: Al_2O_3, SiO_2, K_2O, CaO and Fe_2O_3 values.

K_2O, CaO and Fe_2O_3 provided some interesting data in terms of the differentiation between geological formations and, in some cases, within the frame of a specific formation. For this reason, a scatterplot of Log Fe_2O_3/K_2O vs Log CaO/K_2O was elaborated. In the left-hand side of **Figure 4**, the scatterplot is organised by the main groups (Bujaraloz-Sariñena, Cinca, Lanaja-Castejón and Pallaruelo-Sora) and some differences can be seen between them. A large number of samples from the Pallaruelo-Sora formation are well-distinguished from the other groups, as they possess higher amounts than the rest. Nevertheless, some samples are overlapped with other formations. Similarly, there is an area where most of the samples from the Lanaja-Castejón group are placed, despite some of the samples being placed in an overlapping zone. This overlapping area is composed of samples from the Cinca formation, some samples from Bujaraloz-Sariñena and some from the remaining groups.

The scatterplot on the right in figure 6 shows the samples separated by outcrops, the idea being to distinguish clearly between the outcrops. In this case it can be observed that samples from SQ are mostly separated from the other outcrops and clearly separated from the SB outcrop which comes from the same geological formation. Nevertheless, in the case of the Lanaja-Castejón group, the outcrops overlap and are not well-delimited. Finally, the samples from PC outcrop –from the Bujaraloz-Sariñena formation–are divided into two groups, one separated from the rest and the other located in the overlapping area.

Figure 4 – Scatterplots of Log Fe_2O_3/K_2O vs Log CaO/K_2O for all the analysed geological samples. Left image is organised by formations and right image by outcrops.

Given that it was not so easy to clearly distinguish between the four geological formations, principal component analysis (PCA) was applied. In this case, the five well-dosed elements were taken into account (Al_2O_3, SiO_2, K_2O, CaO and Fe_2O_3) and only the median values of each outcrop were considered. Applying these criteria, a good degree of separation between the outcrops and geological formations was obtained (**Figure 5**). The principal component analysis revealed that 86.3% of the cumulative variance in the samples could be explained. Component 1 covered 56.56% and component 2 29.74%. In the plot, the groups can be clearly distinguished from each other, especially those from the Bujaraloz-Sariñena formation (PC) and the Cinca formation (VA). Moreover, outcrops from the Pallaruelo-Sora formation (SQ and SB) are distinguished from each other and also from the other formations. Finally, the outcrops from the Lanaja-Castejón group can be clearly distinguished, the LT and LM1 outcrops being placed far away from the others in the same group and from the other formations.

5. Discussion

After having delimited the Monegros region and its siliceous sources, four geological formations containing chert nodules were identified. Macroscopically and petrographically they possessed similar features, making it difficult to distinguish between formations and impossible to directly connect archaeological cherts with similar features to any specific geological formation. For this reason, geochemical characterisations were undertaken and, after quantifying major and minor components by energy-dispersive X-ray fluorescence (ED-XRF) and applying statistical procedures, some differences were observed between the formations and outcrops.

Boxplots and scatterplots were useful mainly for observing the internal variability within each outcrop and also identifying which elements were more interesting for recognising differences between the groups. The scatterplots with the most suitable elements allowed us to determine some specificities for the Pallaruelo-Sora and Lanaja-Castejón groups, which were easily distinguished from the rest. Nevertheless, an overlapping area was identified, mostly with samples from the Cinca and Bujaraloz-Sariñena formations but also some specimens from Pallaruelo-Sora and Lanaja-Castejón. For this reason, statistical procedures were applied to distinguish the formations more easily. Principal component analysis (PCA) using the median values from each outcrop proved to be the most suitable manner to easily separate outcrops and formations. In this case, a good degree of separation was achieved and the overlapped areas disappeared.

Having observed that PCA was a useful statistical procedure to distinguish between the groups, we considered adding other geological formations with outcrops in NE Iberia containing cherts macroscopically and petrographically similar to the Monegros type. Samples from the Castelltallat, Tremp and Tartareu-Alberola groups were incorporated. These formations outcrop in the eastern margins of the Ebro Basin and the first Pre-Pyrenean borders in the provinces of Lleida and Huesca, at almost 150-200 km from the Monegros region, and have recently been geochemically characterised following the same procedures (Sánchez de la Torre et al., 2017) . With the data already published, a new PCA plot was elaborated, including the median values from each analysed outcrop (**Figure 5**).

The resulting plot allows the three new geological formations to be distinguished from the previous ones, only the Tremp formation being close to the Cinca group. The quantification of major and minor elements by ED-XRF and the application of statistical procedures have emerged as good techniques for easily distinguishing not only between the different geological formations outcropping in the Monegros region, but also with the main similar geological cherts outcropping in NE Iberia.

Figure 5 – Principal Component Analysis (PCA) plot with the median values from each outcrop including other lacustrine cherts outcropping in NE Iberia.

6. Conclusions

This study has achieved a first geochemical approach of Monegros cherts. The results confirm the value of geochemical methods and statistical procedures to establish differences between outcrops and formations. Energy-dispersive X-ray fluorescence (ED-XRF), despite being a good technique to determine major and minor components in chert samples, is limited as it does not possess a sufficiently high resolution to quantify trace elements, which are always below the device's detection limits. The next step will be the application of other analytical techniques, such as laser-ablation inductively coupled plasma mass spectrometry (LA-ICP-MS), that has already been proved as valuable when quantifying chert trace elements. In future studies we will analyse archaeological cherts that have been described macroscopically as probable Monegros cherts to verify if they are originate from this region, to identify the geological formation of origin and, if possible, the main outcrops.

Acknowledgements

The research leading to these results has received funding through a post-doctoral contract from the Spanish Government under the Juan de la Cierva program (FCJI-2016-27911) held by M. Sánchez de la Torre. L.M. García-Simón was a junior researcher at the University of Zaragoza hired thanks to the Additional Funding RyC 2013-12613. R. Domingo is a Ramón y Cajal senior researcher (RyC2013-12613). This research program has also been financially supported by the Spanish project HAR2017-85023-P and the ANR (French National Research Agency; nº ANR-10-LABX-52). We are grateful to J. Fanlo for his valuable help during the sampling of Muel outcrops.

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