Journal of Archaeological Science: Reports 37 (2021) 103041

Contents lists available at ScienceDirect

Journal of Archaeological Science: Reports

journal homepage: www.elsevier.com/locate/jasrep

Reflectance spectroscopy in combination with cluster analysis as tools for identifying the provenance of Neolithic flint artefacts

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ARTICLE INFO

Keywords:
Flint
Quarries
Provenance
Reflectance spectroscopy
Cluster analysis

ABSTRACT

The provenance of flint artefacts found in archaeological excavations is very important for archaeologists to better understand artefact displacement, exchange networks, mobility patterns, prehistoric migrations and various cultural processes such as commerce, procurement strategies, territorial boundaries, etc. Reflectance Spectroscopy is an analytical technique that measures the radiation reflected by any type of surface, as it is sensitive to electronic and vibrational processes, both at the atomic and molecular level, which results in spectral signatures and absorption bands of the material. In this study the flint material of 16 quarries with evident signs of having been used to manufacture artefacts during the Neolithic period have been characterized by means of their spectral signatures and used to identify the origin of 5 flint blades found in a dolmen close to one of the quarries. The application of cluster analysis to compare the spectral signatures of both the quarries and the archaeological blades has ruled out one of them as not being sourced from the studied quarries, as well as assigning a very probable origin to the other four. Interestingly, none of the blades studied have been sourced to the nearest quarry, as one might presume. The results of this study confirm that reflectance spectroscopy combined with the multivariate statistical analysis could provide a powerful tool for determining the origin of flint artefacts which have been deposited in museums and other institutions.

1. Introduction

Inheritance, imitation, transmission, dissemination and assimilation of cultural technologies constitute some of the important factors for understanding the process of neolithization in the Iberian Peninsula. Specifically, lithic technology and its diffusion allow us to reconstruct a substantial part of the technical system and therefore allow us to draw conclusions about patterns of cultural transmission. Specifically, evidence for circulation of goods as seen through the study of objects, both as raw material or as manufactured goods, constitutes the main field for the analysis of the exchange of diverse materials during the Neolithic. The dissolution of homogeneity in the technological tradition of elaborate large flint blades, the renewing of the circulation networks of raw materials, the emergence of new ways to knap the flint, the apparition of new procedures (such as heat treatment) and new utensils, constitute relevant clues to address the problems of the continuity or rupture between the Mesolithic and Neolithic worlds. Thus, knowledge about the origin and circulation of lithic artefacts in the Iberian Peninsula and by extension in Europe is of extraordinary importance for understanding social functions in the Neolithic.

Therefore, the identification of provenance (or source) of a geological material utilized by prehistoric people is of primary importance within the field of archaeology and has led to the development of many qualitative and analytical techniques within interdisciplinary approaches. Specifically, in flint sourcing studies, both theory and techniques from the fields of geology, chemistry, nuclear physics, remote sensing, biology, physics, and mathematics have been utilized and, therefore, may be found in current archaeological literature (Ciberto and Spoto, 2000; Rodriguez-Tovar et al., 2010; Parish, 2011; 2018; ten Bruggencate et al. 2017; Newlander and Lin, 2017).

Flint (or chert) is defined as a sedimentary rock composed of 70% to 99.9% micro to crystalline quartz (SiO_2), occurring in bedded or nodular deposits within limestone or dolomite with oxides of calcium, carbon, iron, potassium, aluminum and magnesium as its major impurities (Luedtke, 1992). The colour of the flint is mainly due to the presence of organic matter with elements H, C, N and O, which induce the formation of nodules of flint in the early diagenesis processes. Precisely, it is the variation in these other minerals and textures present in minor

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https://doi.org/10.1016/j.jasrep.2021.103041
Received 24 May 2020; Received in revised form 10 April 2021; Accepted 22 April 2021

concentrations that provides an opportunity for distinguishing flint samples from different source localities. Prehistoric people would have had an understanding of these material types and knowledge of where to locate them.

All the flint resources utilized by prehistoric people have undergone some degree of chemical weathering or human alteration. Chemical weathering is produced by the precipitation of soluble materials from the surface regions of the flint, and a patina can be formed across the surface of some flint types due to this weathering process (Luedtke, 1992). The wear on flint under natural conditions is mainly due to the bleaching by ultraviolet radiation and the chemical precipitation of soluble materials such as iron outside the crystalline matrix. As a result of this bleaching or patina, the spectrum of the material could be affected, especially in the visible region of the spectrum (400–750 nm). Some types of flint are more resistant to these natural weathering processes and appear to be only slightly modified, whilst other types exhibit a more whitish appearance. The higher degree of wear appears on or near the surface of the material and can extend into the material at varying depths. Leaching of soluble materials can also mask potentially diagnostic absorption features of the flint type. Additionally, thermal alterations by humans are a constant in the manufacture of flint and can alter the spectrum. The prehistoric thermal alteration of flint was a common practice and was often a necessary process to produce stone instruments, especially in low quality flints (Schmidt et al., 2012). This procedure modifies the material at the molecular level, since water-free molecules enclosed within the crystalline matrix are boiled, producing hematite due to the oxidation of ferrous iron (Hubbard et al., 2005; Schmidt et al., 2012). This change can sometimes be seen as pinkish nuances in the material.

For all these reasons, flint can have a great deal of visual variability. Even pieces of flint from the same source can present a high degree of diversity in terms of their visual appearance, for instance varying from a dark or almost black chocolate colour to pale bluish grey in less than a few centimetres above. This explains that visual identification alone is inadequate for any serious study of flint material types (Luedtke, 1979; Parish and Werra, 2018).

Until recently, analytical techniques for the characterization of prehistoric flint objects and for the identification of the quarries where the raw material used in their manufacture came from have been based mainly on petrological and geochemical methods (Church, 1994). Nevertheless, these techniques present the drawbacks of time, expense, destructiveness or some combination of these three. However since 1999, reflectance spectroscopy for the identification and characterization of minerals and rocks has begun to be used more and more frequently (Hubbard et al., 2005; Parish, 2016; Parish and Werra, 2018). Depending on the chemical composition and optical properties of the samples, after illuminating with an appropriate light source, each material returns a characteristic reflectance curve called a spectrum or *spectral footprint*. VIS-NIR reflectance spectra contains extensive information about the physical and molecular composition of a material like flint, because the technique detects molecules that include phosphates, nitrates, carbonates, sulphates, metal oxides and hydroxides. In fact, infrared spectroscopy has been proposed (Schmidt et al., 2013) as a new technique for detecting the heat-induced effects within lithic artefacts and for quantifying –OH bonds and water in archaeological flints.

Cluster analysis is a statistical method which aims to classify the multivariate data of objects measured by a set of variables to obtain groups maximizing both the intra group homogeneity and the inter group heterogeneity. One of the most common methods of analysis is based on the similarity measure called Euclidean distance from quantitative measures and a clustering agglomeration algorithm between groups as the average linkage, the nearest neighbour or the Ward method. Therefore, the main benefit of cluster analysis is that it groups similar data together and helps to identify patterns between data which might not have been apparent previously. Results can be presented by means of a graphics tree or dendrogram to plot the linkage between groups, providing a classification in groups and subgroups based on the relationships obtained from the analysis. This method is used in taxonomic research to find meaningful conglomerates of regions, communities or neighbourhoods with similar profiles, to obtain significant results when natural grouping does not exist, helping to identify patterns between data which might not have been apparent previously or accept or refute a previous classification. It is the main tool for exploratory data mining and big data, and a common technique for statistical data analysis used in many fields, including machine learning, pattern recognition, image analysis, information retrieval, bioinformatics, data compression, and computer graphics (Hennig et al., 2015).

The main objective of this study is to test the applicability of reflectance spectroscopy in combination with cluster analysis to identify the provenance of flints artefacts found in museums or other institutions to the quarry source from where they were originally extracted and therefore contribute to a better knowledge of material exchange networks during the Neolithic period in the Iberian Peninsula. Since this method is fast, portable, reliable and non-destructive, it has advantages over other study methods such as those based upon macroscopic analysis, which is often of questionable utility with regard to identifying very similar types of flint (Ray, 2007). In addition, in this study the flint from 16 quarries located in Andalusia has been characterized and a high variability of spectral signatures related to the geological structure of each region has been found. Although reflectance spectroscopy has been extensively applied to the study of artistic heritage (Bacci, 1995; Piccolo et al., 2002), its application to the study of flint artefacts in combination with mathematical techniques of multivariate analysis is very recent and still remains in the early stages (Anuradha et al., 2013; Parish, 2016; 2018).

2. Material and methods

2.1. Quarries and flint samples

Material used for this study consists of 207 flint samples obtained from 16 representative quarries in the Andalusia region with clear evidence that they were used by Neolithic and Chalcolithic people to make specialized flint tools. Table 1 shows the geological period, province, name and acronyms used of the quarries sampled and Fig. 1 shows the geographical location of quarries in Andalusia.

Puerto del Zegrí (PZE) and Loma de los Pedernales (LPE) are quarries whose origin can be ascribed to the end of the Secondary Jurassic period, with a main geological domain of calcareous turbidites. The Malaver (MAL), Guaro (GUA) and Los Gallumbares (LGA) quarries belong to the lower Tertiary Cretaceous period with a predominant geological domain of flysh facies. The Ardite (ARD), La Galeota (LGA), Castillo del Turón (CTR), Puerto de los Martínez (PLM), Espíldora (ESP), Canchal de las Herrizas (CHC) and Los Reconcos (REC) quarries belong to the Tertiary-Eocene-Oligocene period with a principal geological domain of flysh facies. The Alcolea (ALC) quarry belongs to the Tertiary –Oligocene-Myocene period also with a principal geological domain of flysh facies. The Calañas (CAL) and Andévalo (AND) quarries belong to the Paleozoic period dominated by rhyolites. Finally, the Aznalcollar (AZC) quarry can be designated to the Upper Devonian and also has a main geological domain of rhyolites. The use of these quarries for the manufacture of specialized tools is dated between 3,400 to 2,400 years BC.

2.2. Sampling methodology

Prior to the collection of flint samples, an exhaustive examination of each quarry was carried out to assess the amount of variation present. Depending on the size and degree of visual heterogeneity due to weathering, a representative number of flint pieces per quarry site were taken for spectroscopic analysis. These samples ranged from 3 pieces for the CAL quarry (the shortest in extension and the most homogeneous)

Table 1
Geological period, denomination, acronyms, letters in Fig. 1 and number of samples analyzed of the studied quarries.

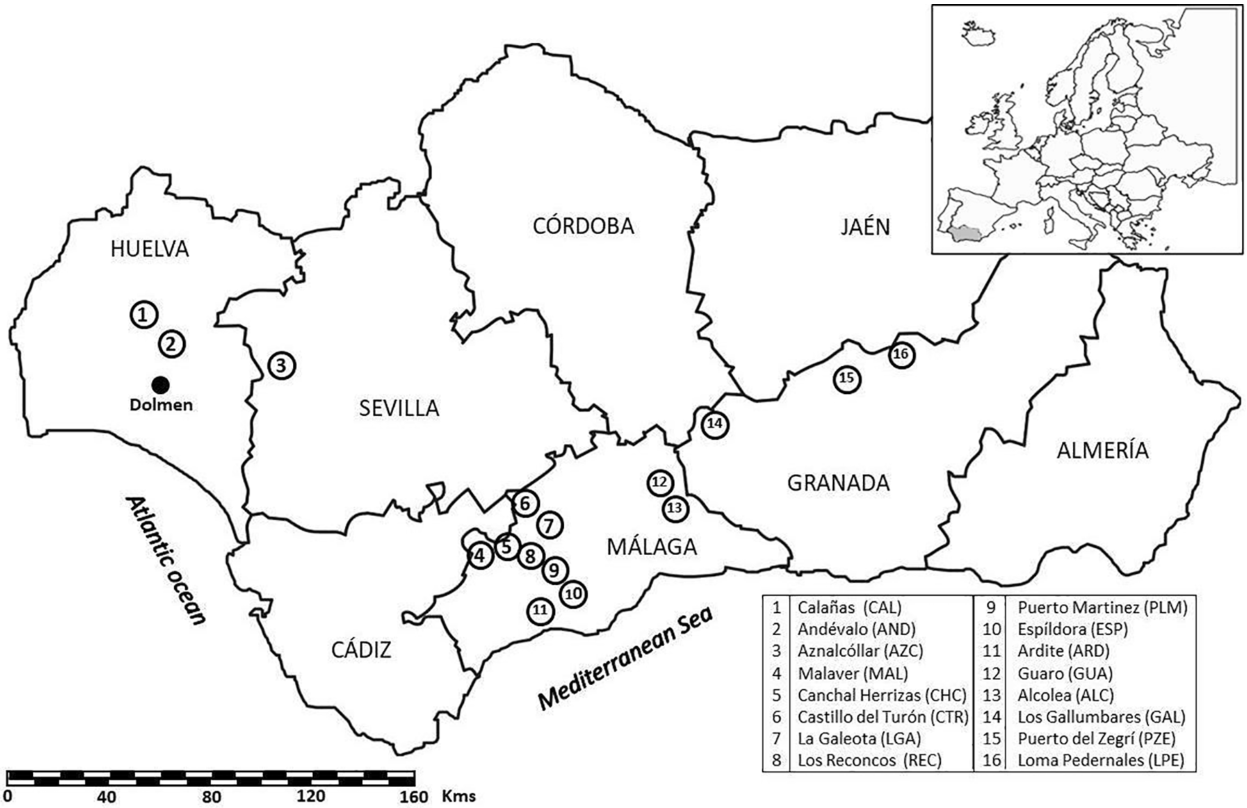
Geological period	Province	Denomination	Acronyme	Letter	Samples
Paleozoic	Huelva	Calañas	CAL	E	3
	Huelva	Andévalo	AND	B	7
Upper Devonian	Sevilla	Aznalcollar	AZC	D	7
Secondary Jurassic	Granada	Los Gallumbares	GAL	I	29
	Granada	Puerto del Zegrí	PZE	O	5
	Granada	Loma de los pedernales	LPE	L	7
Tertiary Oligocene-Myocene	Málaga	Alcolea	ALC	A	30
Tertiary Eocene-Oligocene	Málaga	Ardite	ARD	C	9
	Málaga	La Galeota	LGA	K	13
	Málaga	Puerto de los Martínez	PLM	N	4
	Málaga	Guaro	GUA	J	14
	Málaga	Espíldora	ESP	H	8
	Málaga	Castillo del Turón	CTR	G	15
	Málaga	Canchal de las Herrizas	CHC	F	8
	Málaga	Los Reconcos	REC	P	8
Lower Tertiary Cretaceous	Málaga	Malaver	MAL	M	40
				Total samples	207

and up to 40 for the MAL quarry (Table 1), for a total number of 207 samples. Flint material was collected by one of the archaeologist authors of this paper, who is an expert in Andalusian flint, through a series of systematic geo-archaeological surveys throughout the surface of each quarry

2.3. UV–VIS-NIR spectra collection.

The flint reflectance spectra were collected by using the portable Spectroradiometer FieldSpec4STD (Analytical Spectrometer Device), with spectral ranges 350–2500 nm. The spectral resolutions were 3 nm at 700 nm and 10 nm at 1400/ 2100 nm, with a total number of 2,151 spectral bands. The system use a high intensity contact probe A122307 (Analytical Spectral Devices Inc.) with a halogenous light source and a measurement surface area equivalent to a circle 2 cm in diameter and a maximum specular reflectance of 5%. The device was turned on and allowed to optimize for 30 min prior to use.

After optimizing the instrument, a white reference was taken as a baseline measurement using a Spectralon of 3.62″ diameter (Analytical Spectral Devices Inc.), material that provide a nearly 100% reflective Lambertian surface across the entire spectrum. A dark current correction (DCC) was applied to remove electrical current generated by thermal electrons (called a dark current), and is added to that generated by the incoming photons. The raw data returned are 16-bit numbers corresponding to the output of each wavelength in the VIS/NIRS detector array and each 2 nm sample of the spectrum to generate a relative reflectance (the reflectance factor), which is the quantity that the system measures. The system reduces noise in the desired spectral signal by a spectrum averaging technique (average of 20 spectrums per quantification). Spectra were acquired and processed by using the Indico Pro v6.5.6.1 software developed by ASD Inc. In each flint sample spectra were measured by directly applying the contact probe on three representative zones of its surface. For later calculations the average of all the spectra obtained from each quarry was used. According to the ASD



1	Calañas (CAL)	9	Puerto Martinez (PLM)
2	Andévalo (AND)	10	Espíldora (ESP)
3	Aznalcóllar (AZC)	11	Ardite (ARD)
4	Malaver (MAL)	12	Guaro (GUA)
5	Canchal Herrizas (CHC)	13	Alcolea (ALC)
6	Castillo del Turón (CTR)	14	Los Gallumbares (GAL)
7	La Galeota (LGA)	15	Puerto del Zegrí (PZE)
8	Los Reconcos (REC)	16	Loma Pedernales (LPE)

Fig. 1. Map of the region of Andalusia with the location of the flint quarries and the San Bartolomé de la Torre dolmen.

manual, the white level was calibrated after taking 25 spectra.

2.4. Statistical analysis

The statistical analysis of spectra was primarily based on the multivariate statistical methods by means of cluster analysis (Mukhopadhyay, 2009; Esbensen and Swarbrick, 2018). Visualization of complete spectra was made by the ViewSpecPro version 6.0 software (Analytical Spectral Devices Inc,) and all statistical applications described for spectral analysis were carried out by using the multivariate analysis software Unscrambler version 10.5, CAMO, 2018. Additional calculations were made by using the statistics package Statgraphics18 (Statgraphics, 2018). The data were not pre-processed before the statistical analysis. Cluster analysis was performed by using hierarchical average-linkage method and squared Euclidean distances.

3. Results and discussion

3.1. Mean spectral signatures of the quarries

The flint samples from the studied quarries can be described as light to dark brown or even black, medium to fine grained with varying dark to light lenses. The colour variation present in the material can be directly attributed to varying degrees of silicate replacement processes and meteorization to which the individual nodule or piece has been subjected.

In order to study the variability of spectral reflectance in the quarries studied, Fig. 2 presents the spectral signature of each quarry averaged across all the samples measured in each quarry and Table 2 shows those wavelengths at which the maximum (peak) reflectance were obtained in each quarry. From both the table and the figure, it can be seen that there is a great deal of spectral variability amongst the flint collected fromthe studied quarries. Thus, spectral reflectance varies from very low in PLM, ESP and LGA to high reflectance in ARD, GUA and MAL (Fig. 2 and Table 2). Similarly, the spectral signatures allow differentiating several quarries with a very low reflectance when averaged across all sampled in each quarry. Moreover, some quarries show the existence of wavelengths of strong absorption and reflectance in the studied range (e.g. LPE, GAL, MAL or AND) whereas in others no significant variations in absorption/reflectance were found (e.g. LGA, PLM and REC) (Fig. 2 and Table2). A file containing the raw reflectance data for each sample of the quarries and archaeological artefacts can be found in the online edition of the article.

3.2. Spectral absorption by water and OHs

The H_2O content and the H_2O/OH relationships present in silica-rich materials have significant effects on their VIS–NIR spectra. Typically, flint contains 0.5–0.8 wt% of SiOH and 0.3–0.6 wt% of H_2O, strongly retained in liquid inclusions, crystal defects and grain boundaries (Schmidt et al., 2013). For mineral diagnosis, water (H_2O) and –OH (hydroxyl ions) have a particularly important interest since water and bound OH overtones can be detected in their reflectance spectra (Schmidt et al., 2013), thus informing about the diagenetic processes under which the different types of flint were formed.

The first overtone due to the –OH of a mineral appears at approximately 1400 nm and indicates the –OH bonds with most frequently Si-OH, Al-OH, Fe-OH or Mg-OH or a mixture of these (Mg_3 at 1392 nm, Mg_2Fe at 1398 nm, $MgFe_2$ at about 1404 nm and Fe_3 at about 1412 nm) (Clark et al., 1990). One second overtone, due to the combination of (H_2O) and hydroxyls (OH), appears at around 1900 nm and characterizes the presence of water. For flint, this water is always in an inclusion or trapped in the crystalline mesh of the host crystal. The presence of this feature indicates that the crystallization took place in the presence of an

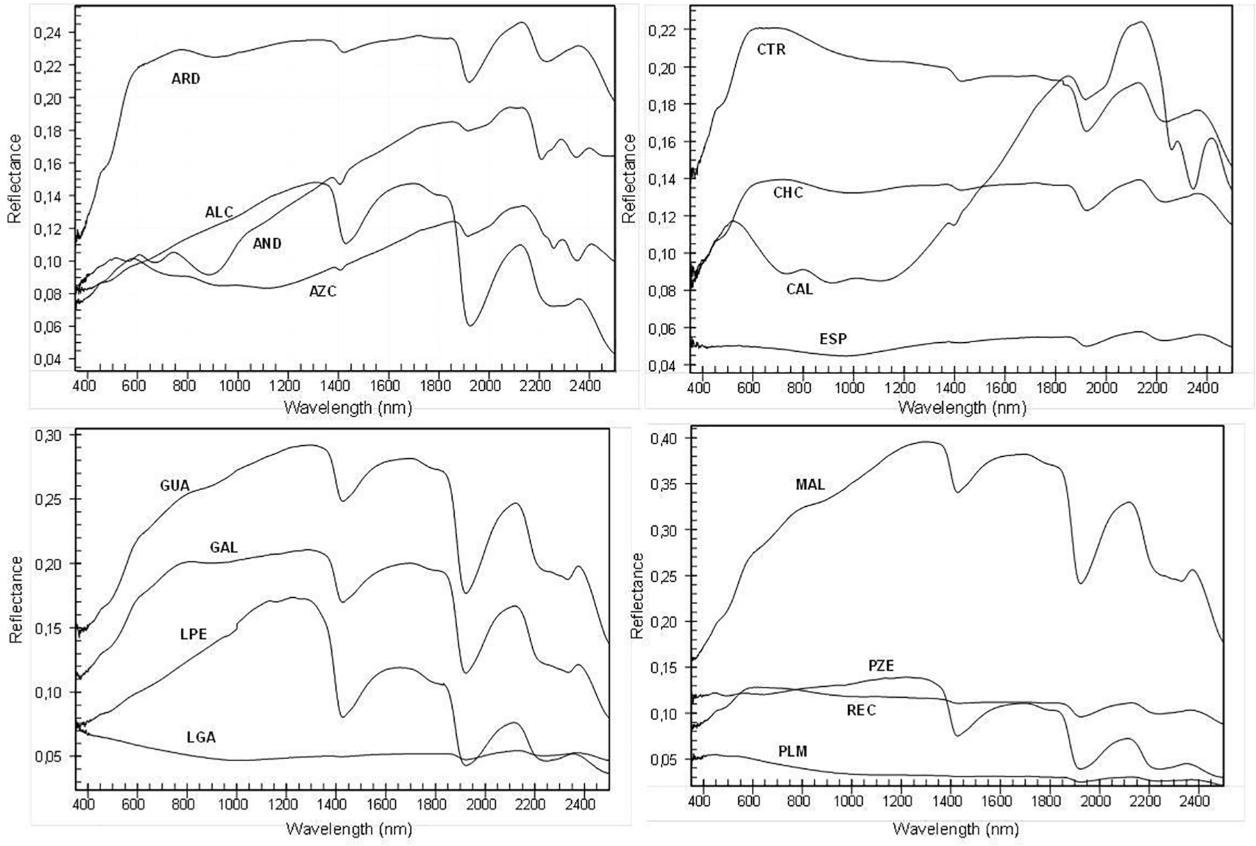


Fig. 2. Spectral signatures of each quarry averaged across all the samples measured in each quarry. The acronyms are described in Table 1 and Fig. 1.

Table 2
Wavelengths for maximum reflectance values (peaks) in each quarry. Values within brackets correspond to the value of reflectance. Acronyms as described in Table 1.

Quarry	Wavelength in nm and corresponding relative reflectance (between bracquets)								
ALC	452 (0.0872)	–	–	1291 (0.1478)	1698 (0.1474)	1804 (0.1402)	2123 (0.1087)	–	2356 (0.0768)
AND	515 (0.1064)	612 (0.1073)	743 (0.1080)	1376 (0.1513)	1855 (0.1833)	2085 (0.1940)	2131 (0.1919)	2284 (0.1725)	2397 (0.1670)
ARD	778 (0.2385)	–	–	1302 (0.2351)	1718 (0.2378)	1808 (0.2362)	2130 (0.2429)	–	2355 (0.2286)
AZC	581 (0.1016)	978 (0.0852)	–	1383 (0.0960)	–	1856 (0.1237)	2136 (0.1333)	2291 (0.1125)	2409 (0.1098)
CAL	522 (0.1171)	797 (0.0910)	1005 (0.0866)	1377 (0.1161)	–	1830 (0.1940)	2137 (0.2223)	2284 (0.1588)	2418 (0.1604)
CHC	715 (0.1399)	–	–	1368 (0.1367)	1717 (0.1375)	–	2130 (0.1380)	–	2363 (0.1308)
CTR	689 (0.2208)	626 (0.2208)	–	–	1711 (0.1952)	–	2127 (0.1914)	–	2362 (0.1769)
ESP	546 (0.0531)	612 (0.0523)	–	1376 (0.0525)	1722 (0.0549)	1831 (0.0560)	2130 (0.0588)	–	2365 (0.0573)
GAL	816 (0.2039)	–	–	1285 (0.2105)	1700 (0.2000)	–	2120 (0.1664)	–	2376 (0.1210)
GUA	–	–	–	1292 (0.2807)	1700 (0.2695)	–	2122 (0.2336)	–	2375 (0.1869)
LGA	–	–	1006 (0.0472)	1374 (0.0503)	1722 (0.0521)	1834 (0.0526)	2131 (0.0549)	–	2371 (0.0531)
LPE	–	–	1122 (0.1624)	1211 (0.1593)	1670 (0.0479)	–	2115 (0.0539)	–	2364 (0.0370)
MAL	–	–	–	1286 (0.3797)	1678 (0.3620)	–	2118 (0.2978)	–	2374 (0.2272)
PLM	455 (0.0721)	–	1170 (0.0494)	–	1711 (0.0479)	–	2122 (0.0463)	–	2375 (0.0434)
PZE	456 (0.1303)	821 (0.1361)	1071 (0.1408)	–	1677 (0.1167)	–	2114 (0.0817)	2356 (0.0573)	2363 (0.0569)
REC	606 (0.1317)	–	1092 (0.1180)	–	1590 (0.1120)	1822 (0.1108)	2127 (0.1147)	–	2367 (0.1067)

significant aqueous phase. The rocks that do not contain such inclusions are mostly metamorphic rocks formed under high temperature and high pressure or they are magmatic ones. A third overtone due to water is at around 2400 nm and in the case of flint also at approximately 2200 nm. Thus, a mineral whose spectral signature has an absorption band at 1900 nm contains water, but if only one band appears at 1400 nm, but no band at 1900 or 2200 nm, it indicates that it only contains hydroxyl ions. The hydroxyl ion has only one vibration frequency, and the position of its wavelength depends on the ion to which it is attached, so it is very interesting to diagnose the existence of certain ions in the studied mineral. In addition, the OH ion appears quite frequently at multiple crystallographic sites of a specific mineral, and typically binds to metal ions. Therefore, there may be more than one OH-linked overtone in the same mineral. Particularly interesting for diagnostic purposes is the vibration of metal-bound OHs in the 2200 to 2300 nm band (Clark, 1999).

In the material collected from the Andalusian quarries, the first overtone for OHs in all the samples analyzed appears between 1396 nm in CAL and 1430 nm in PLM and CHC, with reflectance values varying between 0.0476 in PLM and 0.3232 in MAL (Fig. 2). The second overtone in the material studied due to a combination of water and OHs appears between 1915 nm in AZC and 1926 nm in ALC and LPE, with reflectance values varying between 0.0327 in LPE and 0.2149 in MAL (Fig. 2). With respect to the third overtone, it appears between 2210 nm in AND and 2261 nm in CAL, with reflectance values between 0.0343 in LPE and 0.2306 in MAL (Fig. 2). Therefore, the characterization of the quarries using these bands informs us about the differences between the diagenetic processes in the formation of the different flint types in Andalusia.

3.3. Application to the identification of the probable origin of five archaeological blades

In order to test the reliability of the method for identifying the probable origin of archaeological artefacts, five flint blades found in the excavation of the Dolmen of San Bartolomé de la Torre (Andévalo, Huelva), deposited in the archaeological museum of Huelva under the accession numbers CE1356, CE1357, CE1358, CE1359 and CE1364, have been spectroscopically analyzed. This dolmen is geographically very close to the Andévalo quarry (see Fig. 1). Table 3 shows the relevant wavelengths at which the maximum reflectance (peaks) are obtained for each blade.

The results of the cluster analysis between the signatures of the five blades and those for the quarries (Fig. 3) allowed us to identify six sub-clusters in which the signatures of the blades under study were grouped. Blade CE1356 appears very near to the MAL quarry, blade CE1359 to ALC, blades CE1364 and CE1358 to the PZE quarry and blade CE1357 to the PLM, LGA and ESP quarries (Fig. 3). To statistically verify the similarity of two spectral signatures, one of the most used methods (Varmuza et al., 2003; Willemse-Erix et al., 2009; Parish, 2011; Craig et al., 2013) is to calculate the coefficient of Pearson's correlation between all of the values in each spectral signature. Thus, in our study, and since the spectral signatures provide polynomial curves, the correlation coefficients for a polynomial curve of degree 5 have been calculated for the quarry signatures in relation to the flint blades within each sub-cluster and the origin of which is to be established. The R-Squared coefficient indicates in percentage the degree of relationship between each pair of signatures (Steel and Torrie, 1980). The results indicate that CE1356 correlates to the MAL quarry in 94.8% of scores, CE1359 with the ALC quarry in 94.2% and CE1358 and CE1364 with the PZE quarry in 94.2% and 91.2%, respectively. In turn, CE1358 and CE1364 appears correlated in 99.2% of the scores, indicating that both artifacts were most likely to have been carved from the same flint core. Conversely, CE1357 is not closely correlated to any of the quarries in its subgroup. Fig. 4 shows the spectral signatures of each of the blades plotted together with the signatures of the quarries corresponding to the subgroup obtained in the cluster analysis of Fig. 3. The great advantage of multivariate statistical methods is their capability to extract required spectral information and to explore this spectral information for qualitative or quantitative applications (Ritz et al., 2011), including comparison and/or identification of materials of similar composition or provenance.

Since, as previously mentioned, the absorption due to the content of water and OHs is a diagnostic criterion for flint, Table 4 presents the wavelengths and their corresponding reflectance values for the 5 archaeological blades and the quarries most related to them according to the results obtained in the cluster analysis shown in Fig. 3. Therefore, we carried out a cluster analysis to check the relationships between them

Table 3
Wavelengths for maximum reflectance values (peaks) for each of the flint archaeological blades. Values within brackets correspond to the value of reflectance.

Quarry	Wavelength in nm and corresponding relative reflectance (between bracquets)							
CE1356	–	–	1136 (0.4501)	1228 (0.4546)	1678 (0.3635)	2110 (0.2487)	–	2357 (0.15774)
CE1357	591 (0.0615)	774 (0.0603)	1354 (0.0566)	1734 (0.0561)	–	2134 (0.0571)	2274 (0.0533)	2426 (0.0536)
CE1358	472 (0.0907)	1213 (0.1248)	–	1655 (0.1029)	–	2118 (0.0759)	–	2353 (0.0540)
CE1359	475 (0.0833)	–	1274 (0.1560)	1660 (0.1334)	–	2117 (0.0997)	–	2355 (0.0702)
CE1364	466 (0.0814)	950 (0.1466)	1125 (0.1542)	1642 (0.1105)	–	2115 (0.0713)	–	2357 (0.0468)

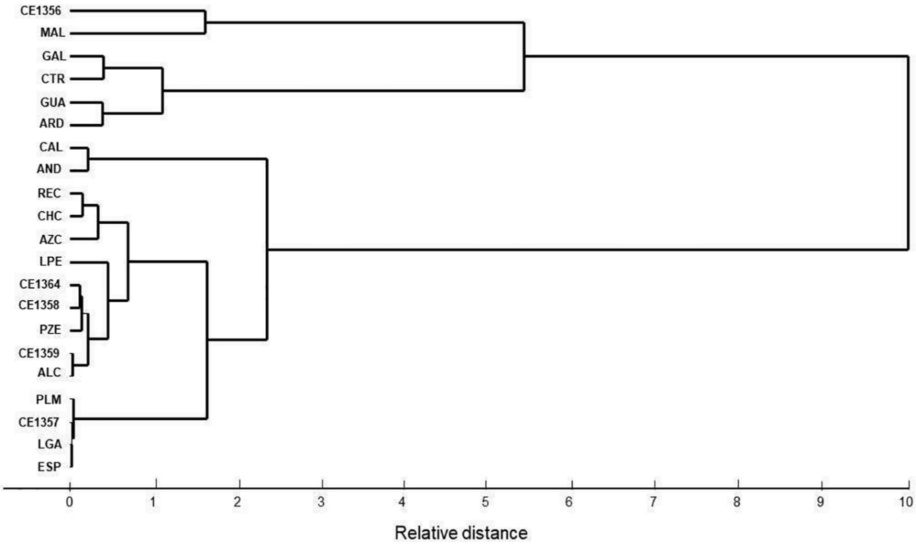


Fig. 3. Dendrogram obtained after performing a cluster analysis by using the hierarchical average-linkage method and squared Euclidean distances for the quarries along the spectral signatures of 5 archeological flint blades (see text for details). The acronyms are described in Table 1.

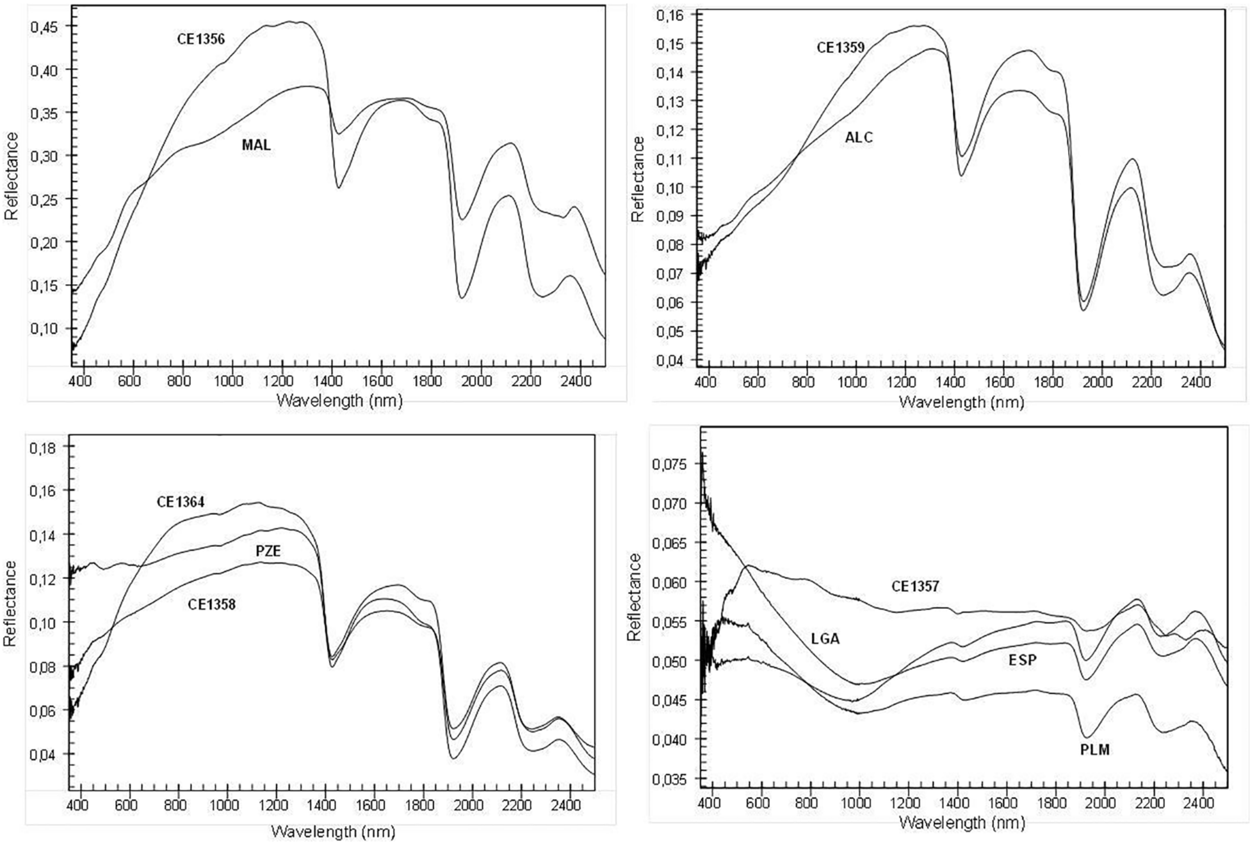


Fig. 4. Spectral signatures of 5 archaeological blades in relation with their relevant quarries as revealed by a cluster analysis (see text for details).

taking into account both the reflectance values and the wavelengths for the absorption of water and OHs of Table 4. The results are presented in the dendrogram of Fig. 5. Again, the results confirm the close relationship between CE1356 with the MAL quarry, CE1359 with ALC and CE1354 and CE1358 with the PZE quarry, whilst now CE1357 appears to be clearly separated from the rest of quarries described in the dendrogram of Fig. 3.

Therefore, it can be concluded that the blade CE1356 was probably manufactured from flint of the Malaver quarry (MAL) located approximately 180 km in a straight line from the dolmen where it was found, CE1358 and CE1364 from pieces of the Puerto del Zegrí quarry (PZE) located at 319 km, and CE1359 from flint of the Alcolea quarry (ALC) located at 256 km from the study site. The CE1357 blade does not seem to come from any of the Andalusian quarries characterized in this study and, quite possibly, comes from some quarry in southern Portugal. Interestingly, none of the 5 studied blades seem to be spectroradiometrically related to the Andévalo quarry (AND), despite being geographically the closest quarry (30 km) to the dolmen where the five

Table 4
Relevant wavelengths for water absorption (valleys) in each quarry and 5 archaeological blades. Values within brackets correspond to the value of reflectance. Acronyms as described in Table 1.

	Wavelength (nm) and reflectance values		
Quarry or blade	~1400 nm	~1900 nm	~2200 nm
ALC	1429 (0.1106)	1926 (0,0596)	2254 (0,0715)
ESP	1421 (0.0517)	1922 (0.0509)	2239 (0.0540)
LGA	1421 (0.0498)	1924 (0.0479)	2236 (0.0508)
MAL	1426 (0.3232)	1924 (0.2249)	2252 (0.2306)
PLM	1427 (0.0449)	1924 (0.0401)	2250 (0.0408)
PZE	1427 (0.0908)	1924 (0.0523)	2245 (0.0518)
CE11356	1426 (0.2623)	1924 (0.1350)	2252 (0.1375)
CE11357	1401 (0.0559)	1926 (0.0537)	2251 (0.0531)
CE11358	1427 (0.0808)	1923 (0.0444)	2247 (0.0481)
CE11359	1429 (0.1039)	1926 (0.0573)	2253 (0.0325)
CE11364	1427 (0.0795)	1922 (0.0380)	2250 (0.0415)

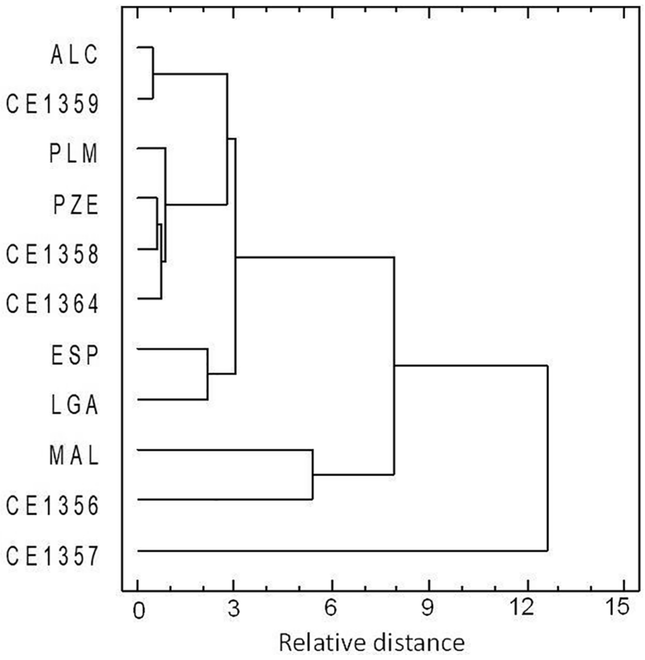


Fig. 5. Dendrogram obtained after performing a cluster analysis by using the hierarchical average-linkage method and squared Euclidean distances for both wavelengths and reflectance values for water absorption in the relevant quarries along with 5 archaeological blades (see text for details). The acronyms are described in Table 1.

blades were found.

The study and characterization of flint pieces has traditionally been accomplished involving the fields of ethnology, ethno-archaeology, typology, geology, mineralogy, experimentation and social archaeology. The petrological analysis of these rare flint artefacts is an especially important and necessary methodology for demonstrating their extraordinary value in terms of understanding how Neolithic societies functioned. In fact, the purpose of any archaeological lithic sourcing analysis is to allocate artefacts to their sources in order to better understanding artefact displacement, exchange networks, mobility patterns and various cultural processes (e.g. trade, commerce, procurement strategies, territorial boundaries, etc.) (Hubbard et al., 2005; Berrocal, 2012; Parish, 2018; Ten Bruggencate et al., 2017). Understandably, the results of these analyses are of great interest to archaeologists.

Reflectance spectroscopy is a non-destructive analytical technique, fast, portable, and low cost, once the instrument has been amortized. As in thin-bed petrographic analysis or X-ray diffraction, for example, reflectance spectroscopy is based on the comparison with a known base repository. The importance of having such a reference system or spectrotheque cannot be overstated, since given an artefact of unknown origin it can be identified regarding its particular spectrofacies by comparing it with reference samples. Since 1999, this technique has successfully been used on several thousand Neolithic objects (small and large hand axes, sheets, knifes, beads, bracelets, other tools, and objects of ornament), and it is often possible, provided with an adequate spectrotheque or spectral reference library, to accurately source the raw material. The advantages of the method are evident since the portability of the instrument allows *in situ* analysis without the need to transport the artefacts to be studied and without the need for insurance that to cover any possible damage of the material during transport.

Another advantage of reflectance spectroscopy is that whole-rock specimens and artefacts can be measured with minimum sample preparation. In addition, the artefacts are not altered or contaminated by the reflectance spectroscopy, so that they can still be subjected to further analysis by other techniques. Moreover, when the spectral reflectance is acquired by means of a contact probe with high light intensity as in this study, the penetration of the photons can reach up to almost one centimetre deep in the flint, returning a deeper spectrum in the thickness of the flint than those due to patina.

These points are particularly important for very valuable pieces in museums or other institutions. Obviously, for this purpose a fundamental requisite is to have a catalogue (a spectral library) as complete as possible of spectral signatures or spectrofacies of the quarries from where such lithic artefacts are suspected to have been procured. At present, some large open source spectral libraries are available for minerals such as the USGS Spectral Library version 7 (Kokaly et al., 2017) and the ASTER Spectral Laboratory. Commercial libraries are also available, notably the Specminone (Hauff, 1993).

In this study, 207 flint samples obtained from16 of the most important quarries in southern Spain have been spectro-radiometrically analyzed, and the peaks and minimum valleys of reflectance for each one of them have also been identified, as well as the presence or absence of water and OH ions. Multivariate analysis through main components and cluster analysis has allowed us to group the quarries into separate clusters according to their spectral reflectance, thus offering a first approximation to their general classification. Subsequently, the application of these results to the source of 5 flint blades found in the excavation of a dolmen near one of the quarries has allowed us to exclude one of them as not belonging to the quarries of Andalusia, as well as assigning a very probable origin to 4 others. Interesting, none of the blades studied have shown a strong spectroscopic relationship with the nearest quarry, as one might have presumed *a priori*.

4. Conclusions

The results of this study showed that reflectance spectroscopy combined with the Multivariate Analysis of VIS-NIR spectra could provide a powerful tool for classification and source identification of archaeological flints found in settlements and burials of prehistoric societies curated in museums and other institutions. Specifically, the application of this method to the sourcing of five archaeological artefacts of flint has proven to be an accurate and non-destructive technique to provide important information about the provenance of lithic artefacts found in tombs and other archaeological excavations. Moreover, another conclusion that we can propose in this study focuses on the main types of flint defined for the Cordillera Bética (flint-type flint and flint of the Milanos Formation). These Neolithic quarry sites have a distribution towards the west of the Iberian Peninsula. Thus, they appear in the Alentejo and Algarve necropolises (such as Aljezur, Alcalar or Marcella) and the regions of Évora in Portugal and Extremadura in Spain, with the analysis of the centre Peninsula still pending. At the opposite extreme is the Levantine region, since although geographically close to the main quarries of the mid Sub-Baetic System, the artefacts analyzed in funerary contexts differ from those we have studied. Therefore, we hypothesize a social separation in the relationships established between the Neolithic communities of the south and those in the Iberian Peninsula.

Acknowledgements

The authors thank the staff of the Huelva Museum for the facilities given to access the archaeological flint blades studied in this article. Likewise, the authors appreciate the suggestions and constructive criticisms provided by the two reviewers and in particular wish to thank Dr. Ryan M. Parish for his excellent review and recommendations that have notably improved the original manuscript. Angela Tate is thanked for reviewing the English version of the manuscript.

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