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Smartphone photos as a non-destructive approach to characterise siliceous artefacts

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ABSTRACT

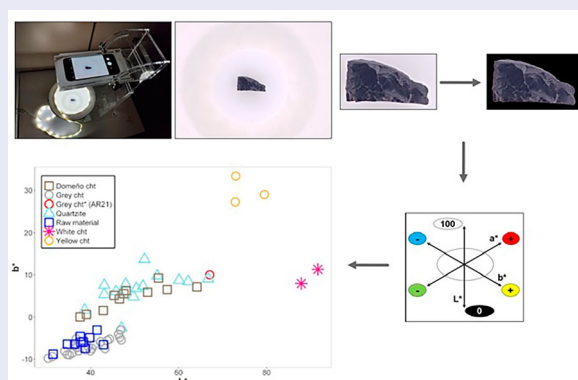
A methodological proposal was developed for the study of prehistoric siliceous artefacts which includes the use of smartphone photography as an analytical tool. Different kinds of materials (cherts, rock crystal and quartzite) were sampled from the lithic assemblage of La Calvera rock-shelter (Camaleño, Spain), chosen as a case study. Each sample was photographed with a smartphone and colour features were compared with portable and non-destructive standardised techniques including visible spectrophotometry, Raman, diffuse reflectance Fourier transform infrared spectroscopy and portable X-ray fluorescence. The obtained results suggest that smartphone image analysis is a reliable approach to characterise siliceous rocks and that it can be a valuable, cheap and fast method especially suitable for the first screening of large lithic assemblages.

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Smartphone; non-destructive analysis; imaging; siliceous artefacts



1. Introduction

Siliceous lithic artefacts are a unique source of information about ancient populations, especially for the Prehistoric period. The study and identification of the origins of the raw materials used for the manufacture of lithic tools permits the reconstruction of socio-ecological dynamics linked to the mobility of human groups (Hess and Riede 2021; Machado et al. 2017; Ramacciotti, García-Puchol, Cortell-Nicolau et al. 2022). Several analytical techniques have been used to characterise siliceous artefacts and raw materials including, for example, Fourier transform infrared spectroscopy (FT-IR), portable X-ray fluorescence

spectroscopy (pXRF), inductively coupled plasma optical emission spectroscopy (ICP-OES), inductively coupled plasma mass spectrometry (ICP-MS) and instrumental neutron activation analyses (INAA; Andersen et al. 2021; Chatzimpaloglou 2020; Delluniversità et al. 2019; Eixea et al. 2022; Newlander and Lin 2017; Ramacciotti et al. 2019; Ramacciotti, García-Puchol, Cortell-Nicolau et al. 2022). Nevertheless, many archaeological studies rely only on the analysis of macroscopic characteristics such as colour, translucency, or cortex presence to classify siliceous artefacts (Luedtke 1992). Although naked-eye examination permits a non-invasive and rapid examination of

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numerous samples, it is strongly conditioned by the experience and sensitivity of the researcher, and can lead to misleading or unreproducible results (Calogero 1992; Hess 1996; Milne, Hamilton, and Fayek 2009). However, there are several methods available for archaeologists to obtain data for artefacts colour properties, such as colour classification by Munsell Colour System charts (Chatzimpaloglou 2020; Hess and Riede 2021; Nazaroff, Baysal, and Çiftçi 2013; Newlander and Lin 2017). Munsell Colour charts allow a rapid and relatively straightforward characterisation of colour against standardised comparison charts, but this still risks influence from colour perception and environmental factors (Bloch et al. 2021). Furthermore, the attribution of a univocal colour code can be problematic in case of heterogeneous materials, being sometimes the primary chosen colour somewhat arbitrary. However, colour properties can be also measured by employing spectroscopic techniques, which measure the characteristics of the reflected light in the visible spectrum under controlled illumination conditions (Delluniversità et al. 2019; Zorzone-Indart, Lopez-Arce, and López-Polín 2021) providing reliable and robust data, but increasing costs with time-consuming operations and requiring training on the operators.

The present study shows a new methodological approach tested on La Calvera rock-shelter lithic assemblage (Diez-Castillo 1997), which includes the use of smartphone photography as an analytical tool. Smartphone-based methods have been developed in analytical chemistry for the colorimetric characterisation of different matrices such as human fluids or food (Rezazadeh et al. 2019; Sáez-Hernández, Antela, Mauri-Aucejo et al. 2022; Sáez-Hernández, Ruiz, Mauri-Aucejo et al. 2022). However, very few works deal with archaeological remains (Ramacciotti, Gallello, Lezzerini et al. 2022; Ramacciotti, Gallello, López-Montalvo et al. 2022; Sáez-Hernández, Antela, Gallello et al. 2022). Concerning siliceous artefacts, Marcos et al. (2022) characterised the colour properties of the artefacts from Los Canes cave (Sierra del Cuera, Spain) by image analysis of camera pictures but the artefacts were powdered prior to being photographed, which is a major flaw since in many cases siliceous artefacts cannot be destroyed due to their archaeological value. Furthermore, in the above-quoted study, the direct use of RGB or HSL parameters, which characterise primaries colours (red, green and blue) or points in a colour space (hue, saturation and lightness respectively), the lack of information related to the illumination conditions and the employed device, as well as the absence of any comparison and control with standard methods for colour analysis, compromise the experimental reproducibility and do not permit the evaluation of the quality of the data.

In the present study, each sample was photographed with a smartphone in a controlled environment and colour data were obtained by image analysis. This allowed the comparison of results obtained by imaging with those from more standard and widely used techniques in order to evaluate the reliability and potential of the smartphone method for characterisation and provenance studies of siliceous artefacts.

2. La Calvera rock-shelter and its geological and archaeological contexts

La Calvera rock-shelter (Camaleño, Spain) is a small cavity excavated in a bluff where alternating strata of quartzite and limestone outcrop. The site is located in the south-eastern end of Picos de Europa National Park at 1180 m above the sea level, within the Cantabrian Mountains. The current relief of the Cantabrian zone is the result of the superposition of the Variscan and Alpine orogeneses of the Carboniferous and Tertiary periods, respectively (Marquinez and Adrados 2000). The Carboniferous represents the key period in the formation of the Picos de Europa Unit (Bahamonde, Vera, and Colmenero 1997), in which carbonate sedimentation began on the marine platform, accumulating up to 1000 metres of carbonate sediments which today, after subsequent orogenic periods, form the calcareous rocks of the Picos de Europa, that are of greatest magnitude in the National Park and occupy the main area of park.

From a geographical point of view, it can be considered part of the area of Peña Oviedo megalithic complex, which is characterised by the presence of dolmens and megalithic structures dated up to the second half of the sixth millennium BP (Figure 1; Diez-Castillo 2010).

These monumental structures, which are among the archaeological evidence of the Vasco-Cantabrian region neolithisation (González Morales et al. 2004), were possibly aggregation places connected to the exploitation of the area for pastoral activities and were linked to open-air settlements such as those of the same Peña Oviedo and of La Calvera (La Campa), occupied from the second half of the sixth millennium BP to the Bronze Age (Diez-Castillo 2008, 2010). On the other hand, the survey excavations of La Calvera rock-shelter offered evidence of human activities prior to the introduction of the production economy and the appearance of the above-quoted megalithic complex, up to the ninth millennium BP (Diez-Castillo 2005). The stratigraphical sequence offered remains of hearts and charcoal, as well as of lithic industry, mainly composed by chips and flakes of dark grey chert and quartzite, similar to the rocks outcropping in the surroundings of the rock-shelter,



Figure 1. Localisation of La Calvera rock-shelter in the Iberian Peninsula.

although other chert types and fragments of rock crystal were found (Diez-Castillo 1997).

3. Materials and methods

3.1. Collected samples

Seventy-four samples were selected to carry out the archaeometric study of La Calvera rock-shelter lithic assemblage (the table is shown in the Supplementary Online Materials, SM1a).

Ten samples are chunks (RM01-05) of local grey chert raw material found in the rock-shelter and outcropping in its immediate surroundings (Figure 2(a)). Twenty-eight flakes of grey chert were analysed and, though almost all of them have macroscopic features similar to the above-quoted raw materials (Figure 2(b)), AR21 is different from the others, showing a lighter colour (Figure 2(c)). Other chert types were

collected from the lithic assemblage: two samples of whitish chert (AR09, AR23; Figure 2(d)) and three samples of yellow chert (AR37, AR38.1-2; Figure 2(e)). Fifteen fragments of quartzite (Figure 2(f)) and five of rock crystal (i.e. translucent and colourless quartz macrocrystal; Figure 2(g)) were collected from the lithic assemblage as well. Furthermore, in order to test the proposed method of colour analysis, eleven fragments of grey Domeño chert (Eixea, Villaverde, and Zilhão 2011) from a geological outcrop in Andilla (Valencia, Spain; Ramacciotti, García-Puchol, Cortell-Nicolau et al. 2022) were introduced in the sampling (Figure 2(h)).

3.2. Colour analysis and imaging

Each sample was brushed with ultrapure water to remove remains of earth and incrustations prior to the analyses. Images of the samples were captured



Figure 2. Picture of example sampled rocks: raw material chunk of dark grey chert (a: RM01), dark grey chert flakes (b: AR11; c: AR21), whitish chert (d: AR23), yellowish chert (e: AR38.2), quartzite (f: AR27), rock crystal (g: AR20) and Domeño chert fragment (h: AN01.2).

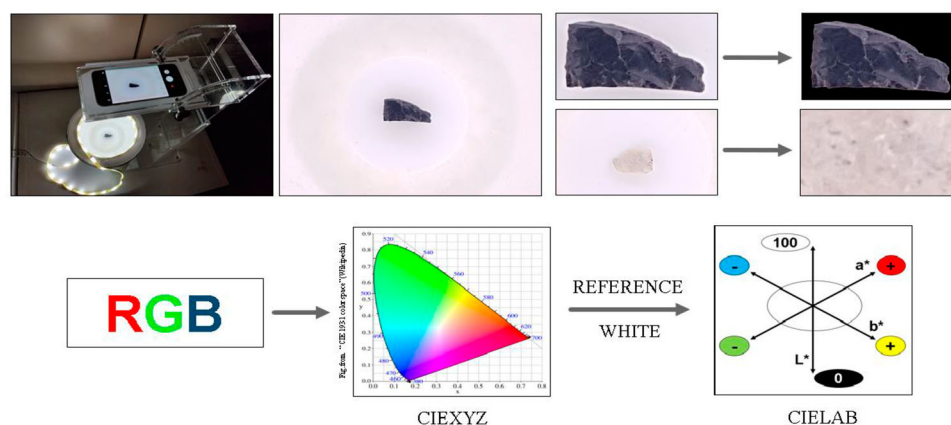


Figure 3. Flowchart for image analysis.

with a smartphone (Samsung Galaxy S7 Edge) to perform colour analysis. In order to validate the results, colour was also characterised by reflectance spectrophotometry in the visible region (Vis-SP) with a CM-26d portable spectrophotometer by Konica Minolta, calibrated with reference white and black to convert the signal to CIELAB colour space coordinates. Two up to five circular areas ($d = 5$ mm) were analysed and average values were taken into account. This last technique was considered as a reference method.

Flowchart for imaging process is shown in the Figure 3. In order to carry out image analysis, each sample was photographed with a smartphone in a controlled environment with constant illumination and equal camera setting (ISO: 100, f : 1/350 s, T : 5500 K, focus: manual, zoom: 3x). The camera settings were chosen to obtain the best quality pictures and avoid artefacts in the given photography environment. Imaging of smartphone pictures was performed with MATLAB (version: R2019b) by MathWorks and Colorlab toolbox (Malo and Luque 2002). In some cases, such as for most of the studied quartzites and dark chert samples, background pixels could be kept out on the basis of RGB parameters. For example, for most chert samples setting a mask to exclude pixels with R lower than ~ 200 was enough, while for quartzites B parameters was more effective (B lower than ~ 200). However, in other ones (e.g.: whitish samples) the sample surface could not be isolated in this way and a rectangular mask was applied. RGB data for each pixel were then converted to the CIEXYZ colour space coordinates and finally to CIELAB ones by a reference white photographed in the same condition of the samples. Average L^* , a^* and b^* were used which account for lightness, red–green and yellow–blue axes respectively (Fairchild 2004).

Both Vis-SP and imaging were performed on most of the collected samples (Supplementary Online Material, SM1b). However, rock crystals were not taken into account and some samples were excluded

from Vis-SP analysis due to their very small size and shape which did not permit to adequately cover the measurement spot of the device.

3.3. Atomic and molecular spectroscopy

Atomic and molecular spectroscopy analyses were carried out directly on the surface of intact samples with pXRF, Raman and diffuse reflectance infrared Fourier transform spectroscopy (DRIFT). Some of the smallest samples could not be analysed by pXRF and DRIFT due to their large measurement area requirements (Supplementary Online Material, SM1b).

Multielement analysis was carried out by a S1 Titan pXRF by Bruker equipped with a Rh X-ray tube (50 kV) and X-Flash® SDD (resolution: 147 eV; FWHM: 5.9 keV). Geochem-trace application was employed to obtain the concentrations of Al, K, Ca, Fe, Ti and Zr, since most of the other potential elemental markers could not be detected by the spectrometer. Average concentrations from two up to five measurements for each sample were taken into account.

Raman and DRIFT were used to obtain information on the main minerals and compounds. Raman spectroscopy analysis was performed in the region between 2000 and 100 cm^{-1} with an i-Raman® Plus spectrometer (model: BWS465-785S) by B&W Tek coupled with a microscope (20x magnification) and laser wavelength of 785 nm (max power at laser port: 495 mW). Each sample was analysed in two up to four points with an integration time of 1.5 s, 10 repetitions and laser set at 15% of power. Spectra were subsequently averaged. A portable spectrometer 4300 Handheld FT-IR by Agilent Technologies was used for DRIFT spectroscopy analyses. The analysis consisted in 50 scans in the mid-infrared region ($4000 - 650\text{ cm}^{-1}$). Each sample was analysed two up to four times in different spots and spectra were averaged. Background was measured between each analysis. Resolution of 4 cm^{-1} was employed.

Table 1. Average values and standard deviations of CIELAB colour space parameters obtained by imaging of smartphone photos.

Sample	Class	L*			a*			b*			Sample	Class	L*			a*			b*		
RM01	RM	34.7	±	8.5	2.3	±	1.0	−6.4	±	2.7	AR18.2	GC	44.3	±	7.4	3.2	±	1.2	−5.2	±	1.8
RM02	RM	36.4	±	7.7	2.1	±	0.9	−6.4	±	3.0	AR18.3	GC	46.8	±	7.7	4.1	±	1.6	−3.1	±	1.2
RM03.1	RM	31.5	±	5.6	2.2	±	0.7	−8.8	±	1.6	AR18.4	GC	40.8	±	11.6	4.1	±	1.9	−6.6	±	2.8
RM03.2	RM	42.9	±	8.2	2.9	±	1.3	−6.6	±	1.9	AR19	GC	43.3	±	9.3	2.7	±	1.2	−7.5	±	2.3
RM03.3	RM	38.7	±	6.9	2.8	±	0.9	−7.4	±	1.8	AR21	GC*	67.1	±	8.2	5.1	±	1.7	10.0	±	2.6
RM03.4	RM	38.3	±	6.4	2.2	±	0.9	−5.9	±	1.6	AR23	WC	91.7	±	2.7	0.6	±	0.5	11.3	±	1.2
RM03.5	RM	39.8	±	8.4	1.7	±	1.0	−4.9	±	1.7	AR24	QZ	66.7	±	16.2	3.2	±	1.5	9.1	±	3.8
RM03.6	RM	37.8	±	8.8	2.3	±	1.1	−6.0	±	1.7	AR26	QZ	52.2	±	11.0	7.1	±	2.3	13.7	±	3.9
RM04	RM	37.6	±	11.3	3.0	±	1.3	−4.6	±	3.1	AR27	QZ	43.1	±	11.1	4.5	±	1.5	5.4	±	2.3
RM05	RM	41.5	±	11.2	2.3	±	1.3	−3.1	±	3.1	AR28	QZ	38.6	±	13.6	4.3	±	1.4	1.7	±	2.9
AR01	GC	40.0	±	10.8	3.9	±	1.3	−8.0	±	2.2	AR29	QZ	51.9	±	14.6	6.2	±	2.0	7.3	±	3.0
AR02	GC	35.2	±	11.5	3.5	±	1.4	−9.0	±	2.7	AR30	QZ	49.8	±	9.7	4.7	±	1.2	4.8	±	1.5
AR03	GC	34.7	±	9.9	3.6	±	1.4	−8.9	±	2.7	AR31	QZ	50.4	±	13.2	5.4	±	2.4	6.8	±	4.2
AR04	GC	35.8	±	13.1	3.3	±	1.5	−8.6	±	3.7	AR32	QZ	62.2	±	9.7	6.5	±	2.7	8.4	±	2.7
AR06	GC	30.4	±	10.2	4.2	±	1.4	−9.8	±	2.7	AR33	QZ	43.1	±	11.5	4.6	±	2.0	7.5	±	5.7
AR07	GC	43.8	±	8.2	3.7	±	1.4	−5.6	±	1.9	AR34	QZ	46.8	±	13.4	5.7	±	1.6	5.3	±	3.9
AR08	QZ	47.0	±	12.3	1.1	±	1.5	−2.6	±	2.8	AR35	QZ	45.7	±	12.3	4.5	±	1.7	6.1	±	3.2
AR09	WC	87.9	±	3.1	−1.0	±	0.7	8.0	±	0.8	AR36.1	QZ	55.2	±	11.7	5.3	±	1.9	9.8	±	2.5
AR11	GC	36.3	±	8.8	3.6	±	1.4	−6.8	±	2.5	AR36.2	QZ	60.1	±	10.3	5.4	±	1.7	8.7	±	2.4
AR12.1	GC	39.5	±	8.8	3.2	±	1.3	−7.0	±	1.8	AR36.3	QZ	48.0	±	10.6	7.3	±	1.7	7.8	±	2.5
AR12.2	GC	38.7	±	9.8	3.3	±	1.3	−6.2	±	2.1	AR37	YC	73.0	±	9.1	5.5	±	3.9	33.4	±	9.5
AR12.3	GC	37.1	±	7.9	3.3	±	1.3	−7.2	±	1.7	AR38.1	YC	72.9	±	9.4	7.8	±	5.0	27.3	±	7.3
AR12.4	GC	31.3	±	11.6	3.9	±	1.4	−9.4	±	3.3	AR38.2	YC	79.5	±	6.1	3.7	±	2.4	29.0	±	11.9
AR12.5	GC	36.7	±	13.4	4.4	±	1.7	−5.1	±	4.7	AN01.1	DC	37.6	±	11.7	4.5	±	1.4	0.0	±	1.6
AR12.6	GC	40.3	±	11.5	3.5	±	1.4	−7.4	±	3.0	AN01.2	DC	42.9	±	9.5	4.0	±	1.1	1.6	±	1.4
AR12.7	GC	46.6	±	12.9	4.1	±	1.6	−4.8	±	4.2	AN01.3	DC	39.1	±	9.9	4.1	±	1.0	0.6	±	1.2
AR13.1	GC	38.9	±	10.7	3.6	±	1.4	−7.8	±	2.4	AN02.1	DC	57.4	±	7.6	3.9	±	0.9	6.5	±	1.2
AR13.2	GC	36.7	±	11.0	3.5	±	1.5	−7.1	±	2.9	AN02.2	DC	64.2	±	5.8	3.2	±	1.1	7.1	±	1.0
AR13.3	GC	42.2	±	9.3	3.6	±	1.4	−5.9	±	2.0	AN02.3	DC	55.4	±	4.2	5.2	±	0.9	9.3	±	0.7
AR14	GC	37.3	±	10.8	3.7	±	1.5	−8.0	±	2.3	AN04.1	DC	48.1	±	9.4	4.4	±	1.0	6.3	±	1.3
AR15	GC	47.0	±	7.2	2.7	±	1.1	−5.4	±	1.7	AN04.2	DC	47.6	±	11.5	3.9	±	1.1	5.5	±	1.8
AR16	GC	46.4	±	9.8	3.2	±	1.3	−4.1	±	2.5	AN04.3	DC	45.3	±	8.5	4.3	±	1.1	5.1	±	1.1
AR17.1	GC	36.4	±	9.8	2.8	±	1.2	−6.7	±	2.5	AN05.1	DC	46.6	±	9.9	4.5	±	1.0	4.3	±	1.5
AR17.2	GC	34.1	±	9.8	3.4	±	1.5	−8.1	±	2.7	AN05.2	DC	53.0	±	5.0	4.2	±	0.6	5.9	±	0.8

Note: RM = chunks of raw material; GC = grey chert; GC* = other grey chert; WC = white chert, DC = Domeño chert; YC = yellow chert; QZ = quartzite.

3.4. Data analysis

Statistical analysis was performed in R (version: 4.1.2; R Core Team 2021). Different principal component analysis (PCA) models were built to explore pXRF data, and both pXRF and imaging (L^* and b^*) ones. In each case, data were previously autoscaled (i.e., z-scores were used instead of raw data). Two cluster analysis (CA) models were built with the samples from La Calvera rock-shelter (except for rock crystal) using as variables previously autoscaled colour parameters (L^* and b^* from imaging) in the first, and concentrations from pXRF in the second (method: average linkage). The following R packages were employed for data analysis and visualisation: dendextend (version: 1.15.2; Galili 2015), factoextra (version: 1.0.7; Kassambara and Mundt 2020), ggplot2 (version: 3.3.5; Wickham 2009).

4. Results and discussion

4.1. Colour analysis

The results of colour analysis for imaging are reported in Table 1.

Raw materials chunks and local grey cherts show similar values for the three colour parameters with L^* ranging between 30.4 and 47, a^* between 1.7 and 4.4 and b^* between -9.8 and -3.1 . All the other cherts from La Calvera show higher L^* , being AR21 (light grey) the darkest one ($L^* = 67.1$) and the whitish the brightest ones (L^* is 87.9 and 91.7 for AR09 and AR23 respectively), while the yellowish cherts have intermediate values ($L^* = 72.9$ – 79.5). For all these, a^* values are from -1.0 (AR09) up to 7.8 (AR38.1). These cherts are also characterised by higher b^* ($b^* = 8.0$ – 33.4), indicating higher degree of yellowness. In particular, yellowish chert artefacts are the highest for this parameter. Concerning dark quartzites, they show intermediate values of L^* compared to the dark cherts and the others ($L^* = 38.6$ – 66.7), while a^* ranges between 1.1 and 7.3. As regards b^* , quartzites show in most of the cases positive values (from -2.6 to 13.7). Domeño chert fragments, introduced to test the method, have in most of the cases slightly higher lightness ($L^* = 37.6$ – 64.2) than grey local cherts and higher b^* as well ($b^* = 0.0$ – 9.3), while a^* ranges between 3.2 and 5.2. Comparing the results obtained by imaging and by Vis-SP (Table 2) shows that the variance of L^* , a^* and b^* in each sample is relevantly higher for imaging than for Vis-SP analysis (Tables 1 and 2). Furthermore, imaging gives slightly higher values for L^* and a^* , and slightly lower values for b^* than Vis-SP.

Figure 3(a) shows the heatmaps of the three colour parameters on the surface of sample AR03. The scale is based on mean and standard deviation, and each pixel is coloured according to its z-score. As can be observed, surface irregularities determine relevant

deviations from the average, caused by the different light reflection angle (Figure 4(a)). This issue can be partially avoided with Vis-SP by working with small measurement areas and choosing the most suitable spots. This fact can explain the different variance for the three CIELAB parameters obtained by the two methods.

Although imaging data from the three parameters deviate from those obtained by the reference technique (Figure 4(b)), L^* and b^* are highly correlated, being R^2 equal to 0.83 and 0.86, respectively. On the other hand, a^* data show a low correlation ($R^2 = 0.16$), possibly due to the presence of outliers (i.e., white and yellow cherts). The high correlations indicate that the differences among the classes of samples highlighted by imaging are actually linked to their own colour features, suggesting that smartphone photography could be a useful method to obtain objective colour data in order to carry out a first classification that can provide useful information for raw material provenance studies.

Based on the aforementioned observations, the samples were plotted in a L^* vs b^* scatterplot (Figure 4(c)). The grey chert artefacts from La Calvera fall together with the chunks of raw material, indicating that the two class of samples have very similar colour features, while the yellow and white cherts are characterised by the highest levels for b^* and L^* respectively and are plotted far from all the other samples. The other grey chert (AR21) has intermediate L^* and b^* . Finally, the grey Domeño chert from Andilla, introduced as control samples, and the quartzite flakes are plotted between AR21 and the other grey cherts from La Calvera.

4.2. DRIFT and Raman spectroscopy

Characteristic DRIFT (Figure 5(a)) and Raman (Figure 5(b,c)) spectra for each rock type are shown. The most relevant features for DRIFT are the Reststrahlen bands for quartz at ~ 1158 and ~ 788 cm^{-1} (Parish, Swihart, and Li 2013). The wide unresolved band between ~ 3700 and ~ 2700 cm^{-1} can be attributed to stretching vibrations of OH, possibly linked to the presence of clay phases (Yeasmin et al. 2017). Except for rock crystal, in which the OH band is barely visible, the different chert types from La Calvera cannot be clearly differentiated on the basis of mid-IR spectral characteristics. On the other hand, Domeño chert samples show bands close to 2880 and 2520 cm^{-1} , and an inverted band close to 1500 cm^{-1} , which are linked to the presence of carbonate minerals (Arrizabalaga et al. 2014; De Lorenzi Pezzolo, Colombi, and Mazzocchin 2018).

Raman spectra for samples of the different types were shown in Figure 5(b,c). Most of the bands shown by the samples correspond to quartz vibrational

Table 2. Average values and standard deviations of CIELAB colour space parameters obtained by Vis-SP.

Sample	Class	L*			a*			b*			Sample	Class	L*			a*			b*		
RM01	RM	29.7	±	3.8	0.2	±	0.3	1.4	±	2.1	AR17.2	GC	28.0	±	1.6	0.1	±	<0.1	0.5	±	0.4
RM02	RM	28.8	±	2.5	0.1	±	<0.1	1.1	±	0.8	AR18.2	GC	28.7	±	1.3	0.2	±	0.1	1.0	±	0.4
RM03.1	RM	28.5	±	3.6	0.2	±	0.1	0.8	±	1.2	AR19	GC	31.0	±	2.0	-0.2	±	<0.1	0.6	±	0.4
RM03.2	RM	27.3	±	0.5	0.1	±	<0.1	0.5	±	0.4	AR21	GC*	48.8	±	0.8	1.5	±	<0.1	7.4	±	0.1
RM03.3	RM	29.7	±	1.5	0.1	±	<0.1	0.4	±	0.2	AR23	WC	83.1	±	0.3	1.9	±	0.1	13.8	±	0.9
RM03.4	RM	28.6	±	0.8	0.3	±	0.2	1.1	±	0.3	AR24	QZ	41.3	±	8.6	0.5	±	0.6	4.0	±	2.2
RM03.5	RM	30.2	±	4.5	0.0	±	0.1	1.1	±	0.8	AR26	QZ	39.3	±	0.3	1.5	±	0.4	6.9	±	0.8
RM03.6	RM	29.8	±	1.2	0.2	±	0.1	1.0	±	0.5	AR27	QZ	36.7	±	2.4	0.7	±	0.2	5.5	±	0.9
RM04	RM	29.8	±	1.5	0.6	±	0.1	1.5	±	0.4	AR28	QZ	33.6	±	0.9	0.3	±	0.1	2.4	±	0.7
RM05	RM	28.9	±	2.7	0.4	±	0.2	0.9	±	1.1	AR29	QZ	40.2	±	3.5	1.8	±	0.8	5.6	±	1.3
AR01	GC	27.9	±	0.6	0.3	±	<0.1	1.0	±	0.4	AR30	QZ	43.0	±	1.3	0.8	±	0.3	5.7	±	1.2
AR02	GC	27.1	±	0.8	0.3	±	0.1	0.4	±	0.3	AR31	QZ	38.2	±	1.5	1.2	±	0.6	5.5	±	1.5
AR03	GC	28.4	±	1.4	0.1	±	<0.1	0.7	±	0.7	AR32	QZ	48.9	±	4.3	1.8	±	0.4	6.6	±	1.2
AR04	GC	27.1	±	2.1	0.3	±	0.3	1.2	±	1.3	AR33	QZ	41.0	±	5.7	2.7	±	3.6	8.3	±	6.6
AR06	GC	25.8	±	0.4	0.2	±	<0.1	0.5	±	0.1	AR34	QZ	39.1	±	1.9	1.3	±	0.1	5.3	±	1.0
AR07	GC	33.2	±	0.4	0.1	±	<0.1	1.1	±	0.1	AR35	QZ	34.4	±	2.2	0.5	±	0.4	4.4	±	1.2
AR08	QZ	33.9	±	2.7	-1.0	±	0.2	2.8	±	0.9	AR36.1	QZ	50.9	±	1.3	1.2	±	0.3	6.0	±	1.0
AR09	WC	75.0	±	1.0	-0.1	±	0.5	9.1	±	2.2	AR36.2	QZ	39.8	±	2.2	1.8	±	<0.1	7.7	±	0.4
AR11	GC	28.2	±	0.4	0.4	±	0.1	1.4	±	0.1	AR36.3	QZ	39.2	±	1.0	2.3	±	0.2	5.5	±	0.4
AR12.1	GC	30.4	±	4.2	0.3	±	0.1	1.5	±	0.5	AR37	YC	56.6	±	5.3	8.3	±	2.2	25.4	±	2.1
AR12.2	GC	29.0	±	1.5	0.1	±	0.1	1.9	±	0.4	AR38.2	YC	45.6	±	2.3	7.9	±	1.8	17.3	±	2.8
AR12.3	GC	30.6	±	2.6	0.5	±	0.2	1.6	±	0.4	AN01.1	DC	34.6	±	0.1	0.9	±	<0.1	3.7	±	0.1
AR12.4	GC	27.7	±	0.7	0.1	±	<0.1	0.3	±	0.2	AN01.2	DC	34.2	±	0.8	0.5	±	<0.1	2.3	±	0.3
AR12.5	GC	28.7	±	0.3	0.2	±	0.1	0.8	±	<0.1	AN01.3	DC	36.3	±	0.2	0.7	±	0.1	3.2	±	0.4
AR12.6	GC	30.3	±	0.8	0.2	±	0.5	1.1	±	1.6	AN02.1	DC	49.0	±	1.2	1.5	±	0.3	6.8	±	0.8
AR13.1	GC	27.3	±	0.3	0.2	±	0.1	0.7	±	0.1	AN02.2	DC	54.9	±	0.4	1.5	±	0.1	7.4	±	0.2
AR13.2	GC	27.9	±	0.2	0.2	±	<0.1	0.7	±	<0.1	AN02.3	DC	45.6	±	1.7	2.3	±	0.3	8.4	±	0.6
AR13.3	GC	35.8	±	1.4	0.8	±	0.1	6.0	±	0.9	AN04.1	DC	42.7	±	1.9	1.4	±	0.1	5.9	±	0.7
AR14	GC	31.6	±	0.4	0.3	±	0.1	-0.2	±	0.2	AN04.2	DC	39.4	±	1.4	1.5	±	0.2	5.6	±	0.3
AR15	GC	33.0	±	0.4	0.1	±	0.5	1.3	±	1.4	AN04.3	DC	40.1	±	4.7	1.5	±	0.1	6.4	±	0.7
AR16	GC	29.5	±	0.5	0.2	±	0.1	1.3	±	0.4	AN05.1	DC	44.0	±	2.9	1.5	±	<0.1	6.4	±	0.3
AR17.1	GC	29.7	±	2.2	0.0	±	0.1	2.2	±	1.6	AN05.2	DC	47.1	±	3.8	1.7	±	0.1	6.7	±	0.5

Note: RM = chunks of raw material; GC = grey chert; GC* = other grey chert; WC = white chert; DC = Domeño chert; YC = yellow chert; QZ = quartzite.

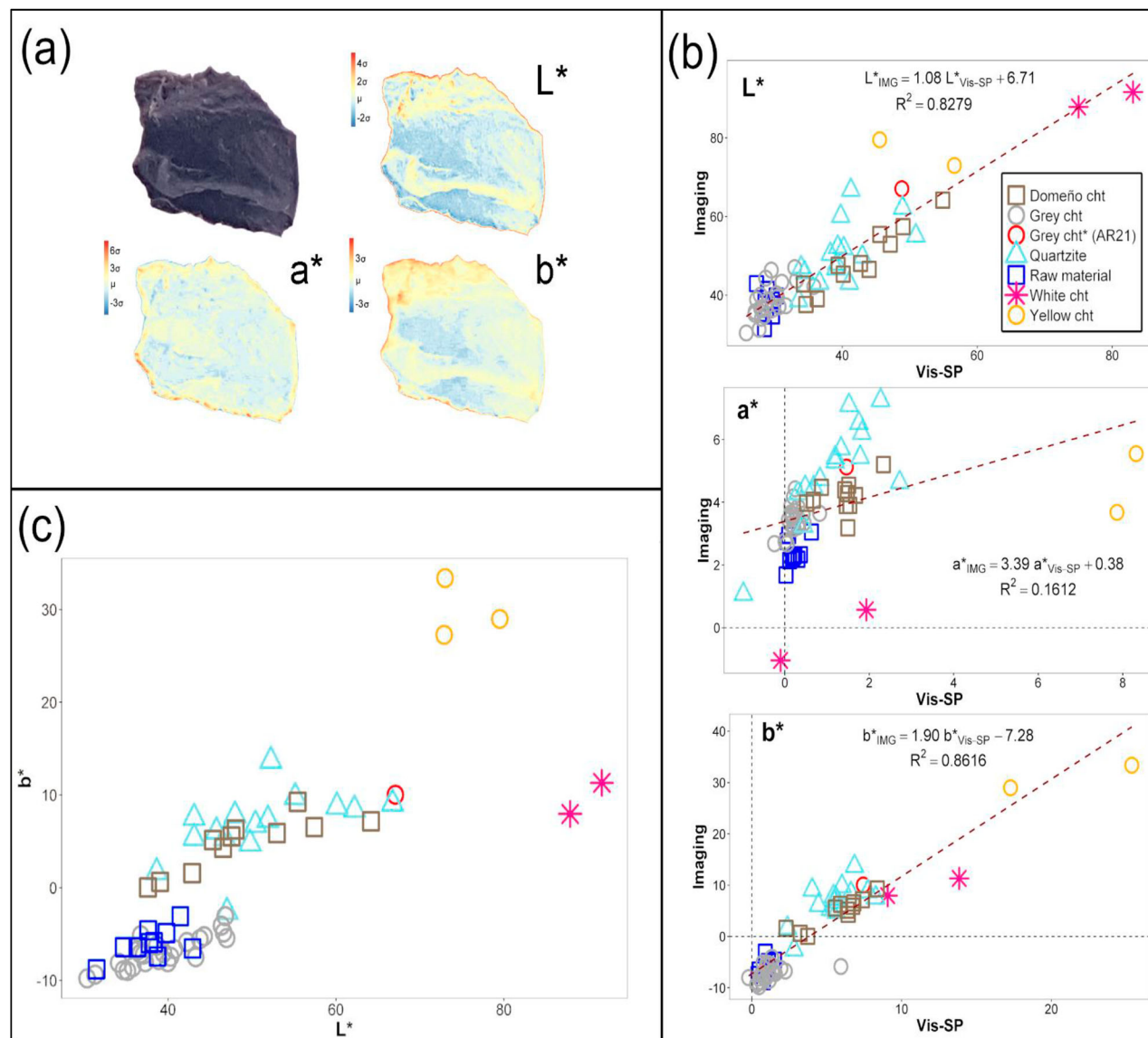


Figure 4. Results of colour analysis: (a) Heatmap of L^* , a^* and b^* for AR03 (see Table 1 for mean, μ , and standard deviation, σ); (b) comparison between colour analysis carried out by Vis-SP and imaging; (c) scatter plot for L^* vs b^* obtained by imaging of smartphone pictures.

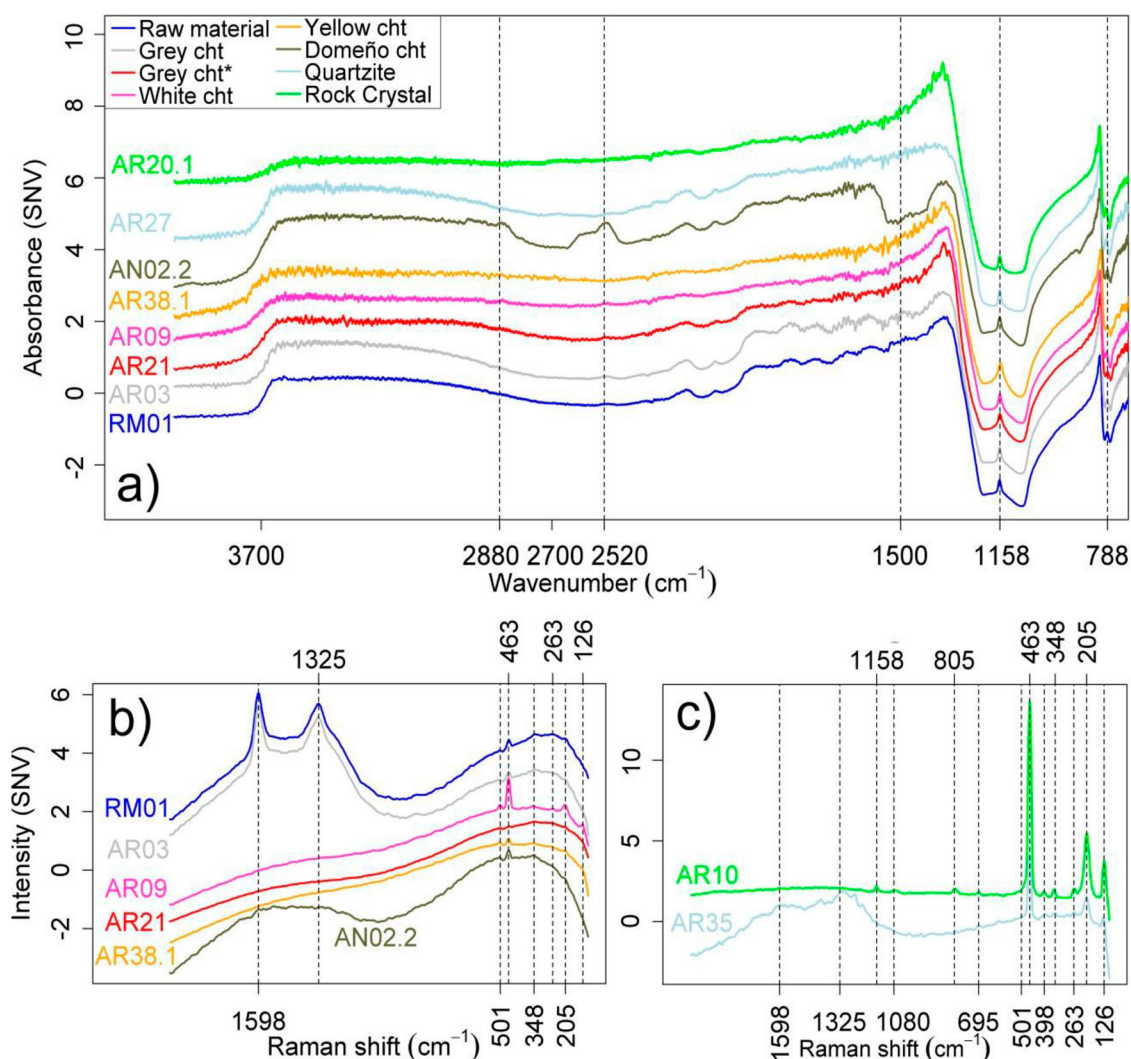


Figure 5. DRIFT (a) and Raman (b and c) spectra for the different types of samples. Spectra were scaled by standard normal variate (SNV) and they are offset to avoid overlapping.

modes (around 126, 205, 348, 398, 463, 695, 805, 1080 and 1158 cm^{-1} ; Chukanov and Vigasina 2020). However, it should be pointed out that quartz bands are more numerous and intense in rock crystals and in several quartzite samples than in chert, possibly due the higher crystallinity of these samples. The band at 501 cm^{-1} could mark the presence of moganite (Capel Ferrón et al. 2015) and is present with variable intensities in the different kinds of cherts. Except for AR21, all the grey chert samples show the typical D ($\sim 1325 \text{ cm}^{-1}$) and G ($\sim 1598 \text{ cm}^{-1}$) bands for carbonaceous materials (Sforza, van Zuilen, and Philippot 2014). Quartzite samples show also features in this region, though mostly unresolved, suggesting the possible presence of similar compounds, maybe characterised by more amorphous material (Sforza, van Zuilen, and Philippot 2014).

4.3. Multielement analysis

Elemental concentrations obtained by pXRF analyses are reported in the Supplementary Online Material

(SM3). In order to explore the analytical results and interpret them, PCA was carried out and the first two PC were taken into account, explaining 54.6% and 15.6% of the overall variance respectively (Figure 6).

Most local grey chert artefacts fall close to the chunks of raw materials and can be distinguished from the other cherts (Figure 7(a)), including the light grey sample (AR21), and to rock crystal due to the high PC1 scores of the latter. Indeed, the local grey cherts artefacts are characterised by higher levels of Al, K, Ti, Fe and Zr than the other cherts, as suggested by loadings plot (Figure 7(b)) and corroborated by the observation of the results (see the Supplementary Online Materials, SM4). Quartzite samples have scores similar to the local grey cherts, though they are scattered on both the axes. Finally, Domeño cherts from Andilla, introduced as test set for the method, can be distinguished from La Calvera materials due to higher PC1 and PC2 scores, especially linked to their high levels of calcium, which is positively correlated with both the PCs (Figure 7(c)), consistently with the presence of carbonates highlighted by DRIFT.

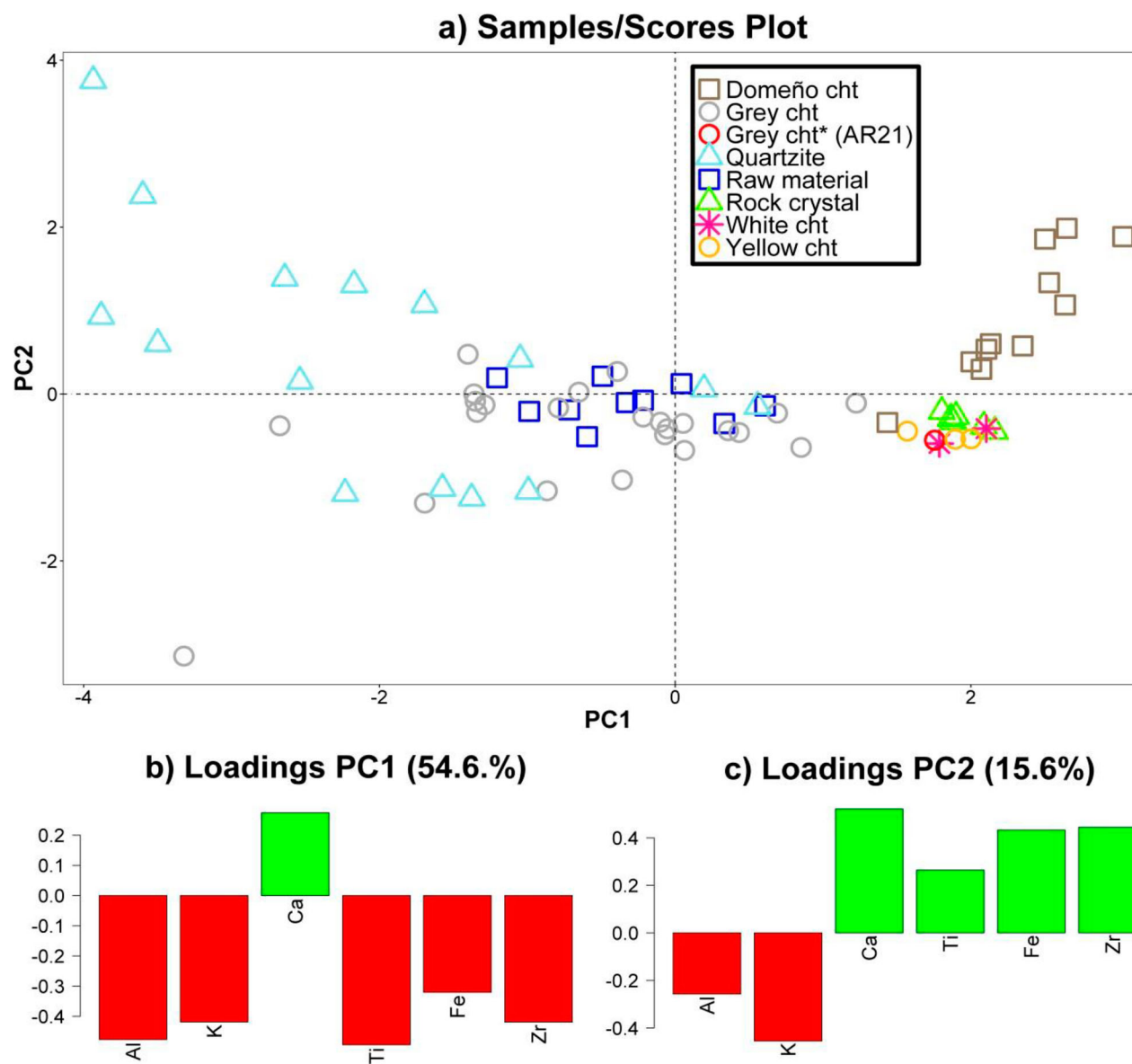


Figure 6. Samples/scores plot (a) and variables/loadings plots for PC1 (b) and PC2 (c).

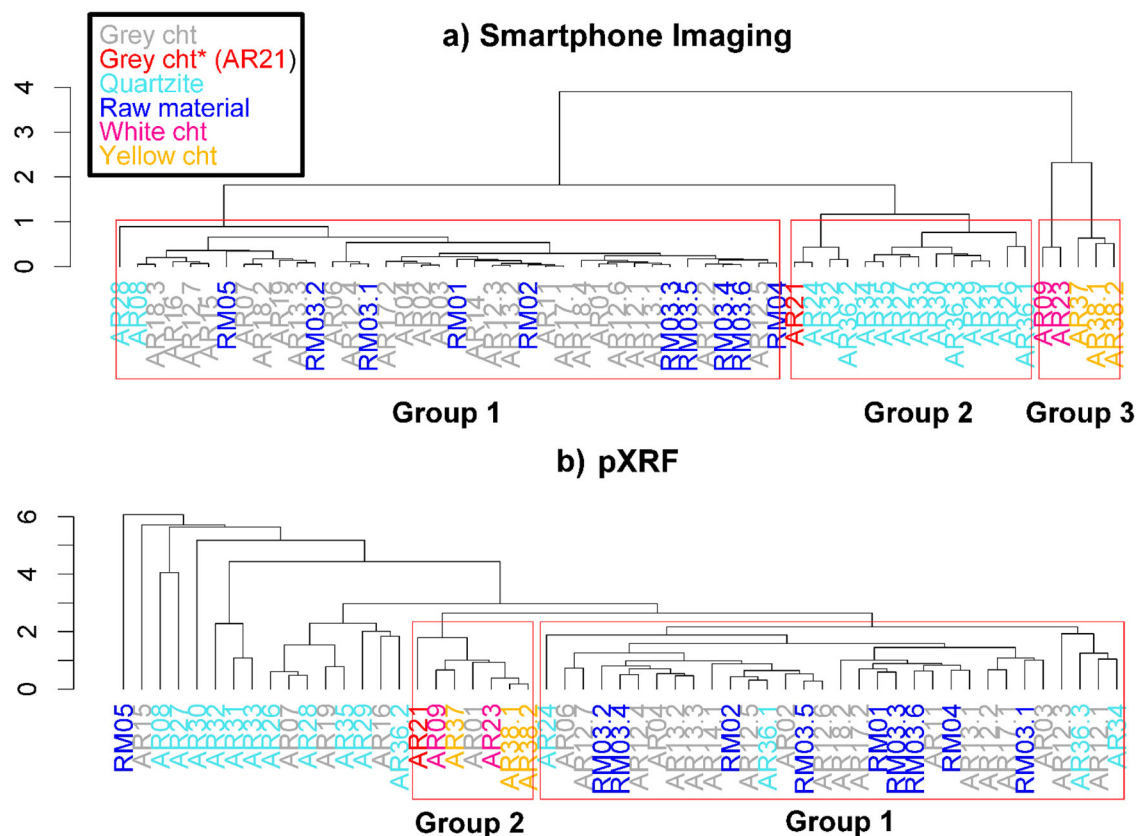


Figure 7. Cluster analysis of smartphone imaging (a) and pXRF (b) results.

4.4. Critical evaluation of the method

The tested smartphone photography approach permitted to obtain a wide range of useful information for the study of the different lithic materials employed at La Calvera rock-shelter.

Colour analysis of smartphone photos offered data which could have been used to discriminate among different rock types. The good quality of L^* and b^* parameters is suggested by the high correlations to those obtained with a Vis-SP, although it has been observed that imaging gave slightly higher values for L^* and lower one for b^* than the reference technique. Imaging data permitted to discriminate among the different cherts, in particular among the three grey chert types which were analysed: the local dark grey chert, the light grey chert (AR21) and the Domeño chert from Andilla, introduced to test the method (Figure 5). It should be pointed out that imaging can be applied on all the chert and quartzite samples, regardless to their size and shape. This is an important aspect since other techniques like pXRF, DRIFT and Vis-SP require a minimum surface area to carry out proper analyses, and X-ray fluorescence spectroscopy results can be also influenced by sample thickness (Williams, Taylor, and Orr 2020). Thus, imaging could be applied to challenging remains such as microdebitage. Furthermore, it is non-destructive, and also cheaper and more user friendly than laboratory devices.

Focussing on the materials from La Calvera rock-shelter, as can be observed in the CA (Figure 7), three groups are mainly present according to smartphone imaging (Figure 7(a)): local grey chert and chunks of raw materials fall in Group 1, most of the quartzite samples and grey chert AR21 cluster in a second group (Group 2) and, finally, the other allochthonous cherts fall in the third group (Group 3), in which they are divided in two sub-clusters. The clustering on the basis of pXRF data (Figure 7 (b)) is quite similar: Group 1 is mainly composed by local grey cherts and chunks of raw materials, and Group 2 by the non-local cherts, while most of the quartzite samples are in the remaining clusters together with few local grey cherts. Raman spectroscopy gave similar results, distinguishing the different lithologies, as well as between local and non-local cherts (Figure 5(b,c)).

These results suggest that smartphone pictures data could have been used as a previous step to identify the main groups in order to carry out a further in-depth characterisation on a representative subset. This would be particularly useful especially while working on large lithic assemblages or to select samples for destructive analyses, and could be easily implemented in the procedure of photographic documentation. Furthermore, colour data can be used as variables together with chemical ones in data analysis. Indeed, the comparison between pXRF PCAs with and without smartphone data suggests that colour properties

could increase the resolution of the model in order to carry out a provenance study (see the Supplementary Online Material, SM3).

4.5. Preliminary provenance hypothesis

The information obtained by smartphone imaging is in line with the study carried out by Columbu et al. (2023). In fact, the most common rock in the assemblage is a dark grey chert outcropping close to the site, found as chunks of raw materials and flakes in the rock-shelter. The possibility to distinguish this chert type from the other dark cherts from Cantabrian Range relies mainly on petrographic and mineralogical characterisation (Herrero-Alonso et al. 2021), the results of which suggest an attribution to Barcaliente Fm. chert (Columbu et al. 2023).

Similarly for quartzite, which is the second most common lithotype along the archaeological sequence. The area of La Calvera rock-shelter is characterised by the presence of different quartzite and quartzitic conglomerate outcrops (Rodríguez Fernández et al. 1994) and, as previously suggested (Prieto et al. 2021), a petrographic characterisation was carried out to determine the source but the precise type of rock could not be identified (Columbu et al. 2023).

The scarce occurrence of other chert types suggests a non-local supply. Concerning yellow chert (AR37, AR38.1-2), macroscopic features and elemental data are compatible with Piloña chert, outcropping in eastern Asturias about 50 km north-west to La Calvera rock-shelter (Tarriño-Vinagre et al. 2013; Tarriño-Vinagre, Elorrieta, and García-Rojas 2015). This chert was identified also in other Mesolithic sites of Picos de Europa area (León, Spain; Neira Campos, Fuertes Prieto, and Herrero Alonso 2016). As regards, the light grey (AR21) and white (AR09, AR23) cherts, any identification on the basis of the present data would be highly speculative.

Rock crystal is instead a possible local raw material. Indeed, macrocrystal quartz outcrops in the southern areas of Picos de Europa Fm. and in eastern Asturias, where it is commonly found in Mesolithic contexts (Arias et al. 2009; Neira Campos, Fuertes Prieto, and Herrero Alonso 2016).

Resuming, our approach employing smartphone imaging suggests a prevalence of local raw materials, especially dark grey chert and quartzite which outcrop in the surroundings of the site, as well as few cobbles of rock crystal, as confirmed by mineralogical and petrographic studies (Columbu et al. 2023). Allochthonous rocks are instead very scarce. This supply pattern is consistent with those observed in other sites of the area during the Mesolithic (Arias et al. 2009; Herrero-Alonso, Fuertes-Prieto, and Neira-Campos 2020).

5. Conclusions

The present study permitted to test for the first time the use of smartphone photography as an analytical tool for the characterisation of colour features of siliceous artefacts from a Prehistoric site. Smartphone imaging proved to be a reliable method to obtain colour data on this kind of material, as indicated by the comparison with the results from a reference technique (Vis-SP). It permitted to observe differences among rock classes, obtaining similar information to the standardised techniques such as Raman spectroscopy and pXRF, suggesting that this approach should be tested also on other types of rocks. These results indicate that colour analysis of smartphone photos could be a fast, reliable and cheap method to screen large lithic assemblages and to select representative samples for further investigation or destructive analyses.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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