Journal of Archaeological Science: Reports 30 (2020) 102237

Contents lists available at ScienceDirect

Journal of Archaeological Science: Reports

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

A new combined approach using confocal and scanning electron microscopy to image surface modifications on quartzite

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ARTICLE INFO

Keywords:
Laser scanning confocal microscopy
Scanning electron microscopy
Use-wear analysis
Polished surfaces
Quartzite

ABSTRACT

Confocal microscopy has been increasingly employed in the field of traceology to acquire metrological data of surface changes on a micro-scale. However, its advantages for a traditional visual inspection of use-wear are rarely highlighted.

As traditional optical microscopy (OM) has proven unable to entirely fulfil the prerequisites for an ideal observation of highly reflective and irregular materials, alternative ways for providing better observation conditions must be sought.

In this contribution, we explore the combination of laser scanning confocal (LSCM) and scanning electron microscopy (SEM) micro-graphs for the visual characterisation of wear on quartzite and evaluate the potential of both techniques.

1. Introduction

Lithic assemblages of the most varied chronologies are frequently composed of coarse-grained raw materials (e.g., quartz, quartzite, basalt). Hence, it is important to be able to retrieve functional information from such assemblages in a consistent way. However, analysts encountered technical problems when attempting to analyse reflective materials such as quartz and quartzite.

It has been shown that optical microscopy (OM) does not entirely fulfil the prerequisites for an ideal observation of use-wear on highly reflective and irregular lithic materials. The technical limitations of optical microscopy to image use-wear on coarse-grained and reflective materials (e.g., quartz) (Knutsson, 1988; Sussman, 1985, 1988), became clear with the first comprehensive experiments focused on the generation of use-wear (Grace, 1989, 1990; Hayden, 1979). While some researchers have documented a generally improved quality of observation of quartz surfaces when using microscopes equipped with differential interference contrast (DIC) (Igreja, 2009; Lemorini et al., 2014; Fernández-Marchena and Ollé, 2016; Márquez et al., 2016; Ollé et al. 2016), others did not see particular gain when analysing quartzite specimens (Pedergnana, 2017; Pedergnana et al., 2018).

Acquiring high quality optical microscopy images in a consistent way can be challenging for use-wear analysts, especially when dealing with highly reflective and irregular specimens. Parts of such samples can be out-of-focus, unevenly illuminated or poorly exposed. Despite the use of hardware autofocus systems, the resulting images can be noisy. While the capacity of optical microscopes to provide entirely in-focus images is inversely proportional to the depth of field of the sample; this issue can be partially overcome by obtaining extended depth of field (EDOF) images, even though the problem persists when the depth of field of samples exceeds certain limits. In fact, when large datasets are acquired on automated microscopes, images might still have out-of-focus areas. One of the consequences of employing OM alone to analyse use-wear on coarse-grained raw materials is that significant functional information can be missed (Fig. 1). If use-wear information is missed on a substantial number of samples, then the functional interpretation of the archaeological assemblages analysed, therefore of the sites' functions, will be incomplete or biased.

For example, polished areas on quartzite are not always visible with OM (Fig. 1: b&e). They are not highly reflective, as described elsewhere for flint (e.g., Keeley, 1980). On the contrary, they are generally of low reflectance (Fig. 1: b, c, e, f, m, n) and highly reflective areas observed under OM are actually the unmodified, original quartzite surface.

Also, edge rounding is not perceivable and 'polish' is not easily

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https://doi.org/10.1016/j.jasrep.2020.102237
Received 2 July 2019; Received in revised form 9 December 2019; Accepted 22 January 2020

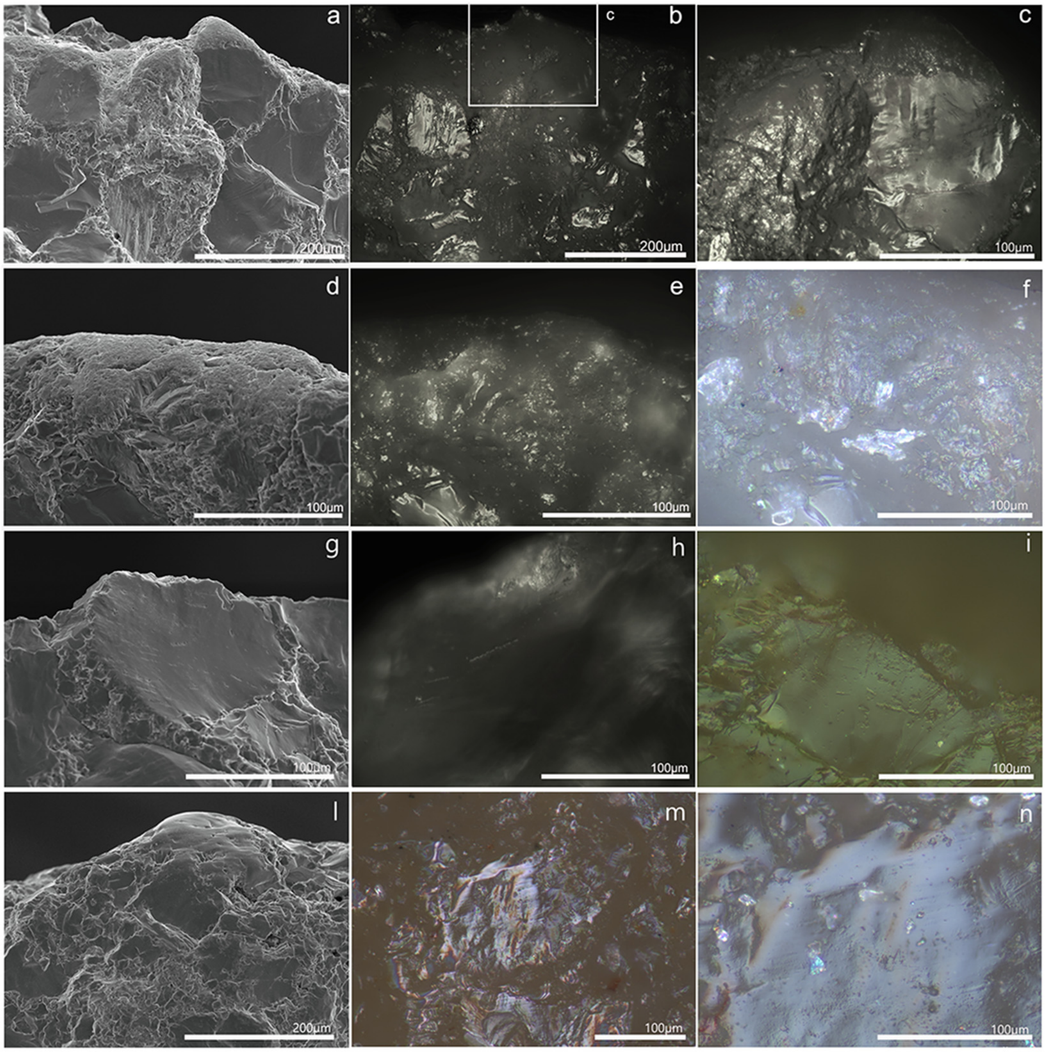

Fig. 1. Comparison of the same surface portions showing some use-wear of two experimental quartzite flakes imaged with traditional OM and SEM. a&b) The same surface portion showing polish originated from contact with wood. When imaged with SEM all attributes are visible, whereas under OM the polish is not visible; c) A close-up of the polished edge imaged in panels a&b seen under OM; d-f) Details of a polished area originated from contact with wood is better visible under the SEM, while the same spot does not seem to be polished under the OM; g) An entire crystal bearing multiple furrows imaged using SEM; h) Two furrows imaged under the OM. There are more furrows on the same crystal, but it was not possible to image them with OM; i) Entire crystals bearing furrows are imaged with OM only when observed at an adequate angle. Original magnifications (for OM, observation through oculars): a&b; l&m) 200 ×; c-i; n) 500 ×.

distinguishable (Fig. 1: c). The facets of quartz crystals reflect light differently, and some crystals appear particularly brilliant under the microscope (Fig. 1: e). Edge rounding is easier to appreciate with scanning electron microscopy (SEM), as this method provides higher depth of field. Striations on quartzite are also difficult to image with OM, due to the high irregularity of the surface (Fig. 1: h). Striations can be located on crystals' facets which cannot be imaged by OM because of technical constraints. Sometimes, it is not possible to overcome these constraints even with the aid of motorised stages and software for EDOF imaging (Fig. 1: h). This results in an inevitable loss of data, and therefore of functional information. In other instances, when entire quartz crystals are relatively flat and properly oriented to the light source, linear features can also be observed with OM (Fig. 1: i). These technical issues when observing quartzite have been recently overcome by employing scanning electron microscopy (SEM) on a large scale (Ollé, 2003; Vergès, 2003; Ollé and Vergès, 2008; Pedergnana, 2017; Pedergnana and Ollé, 2017; Pedergnana et al., 2018).

1.1. *Scanning electron microscopy (SEM)*

SEM has not been intensively used in the field of traceology, probably because of its high cost, time-consuming nature of observations, learning curve, and vacuum chamber limitations. It also requires a sample preparation procedure that adds to the overall sample preparation and analysis time. It has been used in some studies to image particular use-wear features or tiny details for which conventional microscopy was not powerful enough (Mansur-Franchomme, 1983; Yamada, 1993). When samples under study had irregular and reflective surfaces, the use of SEM has been more intensive and systematic due to its technical advantages over OM (Knutsson, 1988; Sussman, 1985, 1988; Borel et al., 2014). SEMs use a focused beam of electrons and detection of the reflection of this beam to magnify samples (Dunlap and Adaskaveg, 1997). They can be used both in high and low-vacuum conditions. Samples need to be coated with conductive materials (e.g., carbon, gold, platinum) in the high-vacuum mode only (although in any mode imaging is improved in this case). Electrons in the beam interact with samples and produce a range of signals that are collected by various detectors, yielding information about the surface topography

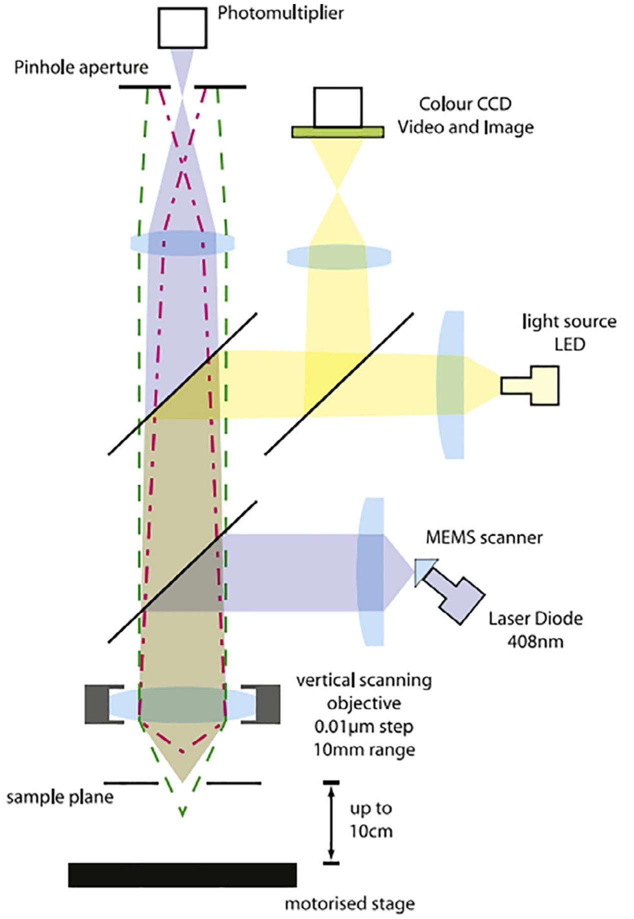

Fig. 2. Schematic view of a confocal microscope (After Evans and Donahue, 2008: 2225).

and composition of the scanned areas. For example, signals detected by SEM detectors are then converted into high-quality images which are shown on conventional computer screens. SEM produces greyscale images of surface relief; therefore no colour information is available. Secondary and backscattered electrons as well as x-rays are scattered by the sample allocated in a chamber that are detected by different detectors. Secondary electrons are generated when primary electrons displace specimen electrons from the specimen surface. A detector (called Everhart-Thornley detector in high vacuum conditions) reads the signals of the secondary electrons emitted by the surface. The contrast and soft shadows of micro-graphs obtained with this detector closely resemble those of specimens illuminated with light; therefore the images obtained are easily readable and interpretable. Backscattered electrons are incidental electrons reflected backwards from the scanned area. The resulting micro-graphs provide compositional data of the specimens observed. They provide both relative atomic density and topographical information. Elements with a high atomic number appear lighter in the resulting images, while elements having a low atomic number are darker. Moreover, X-rays generated from a sample bombarded with electrons provide detailed elemental information. The amount of energy released in the form of X-rays derives from the replacement of the secondary electrons emitted by the sample, with other electrons ‘jumping’ from more peripheral atomic orbitals (Dunlap and Adaskaveg, 1997).

Although it is usually acknowledged that SEM provides higher-quality images with larger depth of field than OM, it cannot offer methods for quantifying surface textural changes. Nevertheless, with last generation SEM software, it is possible to acquire multiple micro-graphs of a surface of interest scanned at different tilted angles, and then merge them in order to obtain 3D surface reconstructions (Tafti et al., 2015).

Generally speaking, quantification of wear is really important, as it provides numerical data of the worn surfaces which can be more easily ascribed to specific worked materials. Moreover, quantitative data collected in different studies could be directly comparable and analyses reproducible (as long as the equipment used is the same). These are the reasons why much effort has recently been put into developing techniques to quantify lithic use-wear (e.g., Evans and Donahue, 2005, 2008; Evans and Macdonald, 2011; Grace et al., 1985; Kimball et al., 1995; Macdonald, 2014; Stemp, 2014; Stemp and Stemp, 2001; Stemp et al., 2016). Particularly, laser scanning confocal microscopy (LSCM) has seen an incredible increase of popularity within the field of traceology due to the urgent need to provide quantitative data to better assess worn surfaces related to use (e.g., Evans and Donahue, 2008; Evans et al., 2014; Ibáñez et al., 2016; Stemp and Chung, 2011). However, the advantages of confocal microscopes in the traditional visual inspections of use-wear on knapped tools have been rarely highlighted (Evans and Donahue, 2008).

The main objective of this study was to compare micro-graphs of similar fields of view showing the same use-wear features (i.e., polish) on quartzite samples taken with SEM and LSCM. Both equipment types, which are not extensively used within the field of traceology, were evaluated and their potential to improve the method was assessed. Specifically, their potential to overcome the optical limitations of OM to image highly irregular and reflective lithic specimens is discussed.

1.2. Laser scanning confocal microscopy

Confocal microscopes are based on the earliest original idea advanced by Marvin Minsky in 1955 (and patented in 1957) (Minsky, 1988). Although there have been massive technological improvements on sophisticated digital acquisition and storage digital systems, the basic functioning principle has not changed at all. The development of confocal microscopes was based on the need to observe biological events *in vivo*, but they are now used in several other research fields, namely material sciences applications. Confocal microscopes are often built around a conventional light microscope and use either a laser (LSCM) or a light emitting diodes (LED) (imaging confocal microscope) to illuminate the sample (Leach, 2011; Paddok, 2000) (Fig. 2). The laser beam is scanned across the specimen point by point through the aid of the microscope’s objectives. The light reflected back by the specimen travels through a confocal aperture. Confocal images are built up during the beam scanning, when single pixels are illuminated. Large areas are scanned by rastering the beam by means of mirrors. The intensity of the signals corresponding to each pixel is then detected by a photo-multiplier which is found on the rear of the confocal aperture. The main function of the second pinhole is to impede light coming from above or below the focal plane of the microscope’s objective from entering the photo-multiplier. In this way, the out-of-focus light of images can be eliminated through spatial filtering. Z-series optical sections (confocal images taken from different z axis planes) are acquired using a motorised stepping motor and stored on computer media, being available for further processing into 3D representations of the sample.

Along with confocal images (i.e, maximum intensity maps), conventional optical images are also available. Due to the high quality of the objectives (high numerical aperture, NA), which is necessary to perform surface measurements, the resulting optical images have on average higher resolution than those obtained with conventional OM. In fact, all optical microscopes are limited by fundamental physical factors in the resolution that they can achieve (Calandra et al., 2019). Optical lateral resolution is defined as the minimum separation between two points that results in a certain contrast between them. It is influenced by the wavelength of the light used for illumination. The average wavelength of white light is 0.55 µm which results in a theoretical limit of resolution (not visibility) of OM in white light of about

0.2–0.25 µm (Tkaczyk, 2010:2). Regarding the objectives mounted on a microscope, resolution is also limited by the NA of the objectives.

On confocal microscopes, the highest NA available to be used in dry conditions (not immersed in either water or oil) in a LSCM is 0.95 (Leach, 2011:259). Based on this and on the violet to blue wavelength (between 0.4 µm and 0.29 µm) used by the LSCM here, the highest lateral resolution available for confocal microscopy is higher (between 0.25 and 0.29 µm) (Leach, 2011:277) than that obtained with conventional light-field microscopes. Vertical (axial) resolution is also higher in LSCM, as it filters out-of-focus points and thus achieves much higher vertical resolutions.

2. Methods

Experimentally generated worn areas on quartzite flakes knapped from a single fluvial cobble were scanned using SEM and LSCM. The micrographs obtained were compared and described. The experimental samples used in this study are part of a wider reference collection of use-wear on several varieties of quartzite which served for the analysis of two archaeological assemblages (TD10.1-Gran Dolina site, Northern Spain and the Payre site, Southern France) (Pedergnana, 2017, 2019; Pedergnana et al., 2017, 2018, 2019).

2.1. Experiments design

Experiments were performed employing 6 unretouched flakes obtained from the same cobble of metaquartzite (VHS4) in order to limit the intra-variability of the raw material (Fig. 3). The cobble was collected at Villasur de Herreros, a village near the city of Burgos (Northern Spain). The location was selected because of its proximity to the Sierra de Atapuerca archaeological complex. The six experimental replicas were used to work five different materials commonly associated with early prehistoric tasks: wood, bone, antler, fresh and dry skins, and cane (Table 1). All materials were worked for an hour by applying similar motions (i.e., transversal movement). The action was limited to whittling/scraping in order to control variables that may impact on polish development. The working angle was kept very low (40°), apart from when skin was scraped (60°), to maximize the surface of worn out areas. All the materials have been generally worked in a fresh state, except from the skin which was used in both fresh and dry states. The one red deer (*Cervus elaphus*) antler was soaked in water before the experiment for 48 h. The selected species of wood was a type of softwood, Aleppo pine (*Pinus halaepensis*). A long bovid bone (*Bos taurus*) and stems of giant cane (*Arundo donax*) were also used. All the experiments were performed by one author (A.P.), with the aim of maintaining all the variables involved in the experiment (such as the amount of exerted pressure, velocity, number of strokes per min) as constant as possible. The duration of experimental tool use was prolonged (60 min) to assure the formation of large, well-developed polished areas, knowing that polish takes longer to form on coarse-grained lithologies than on fine-grained ones (Clemente-Conte and Gibaja-Bao, 2009; Leipus and Mansur, 2007; Stemp et al., 2013).

Table 1
Experiment samples, action and duration of the experiments.

Reference	Worked material	Action	Duration min
VSH4-1	Soaked antler	Whittling	60
VSH4-2	Fresh cane	Whittling	60
VSH4-3	Dry skin	Scraping	60
VSH4-4	Fresh wood	Whittling	60
VSH4-5	Fresh bone	Whittling	60
VSH4-6	Fresh skin	Scraping	60

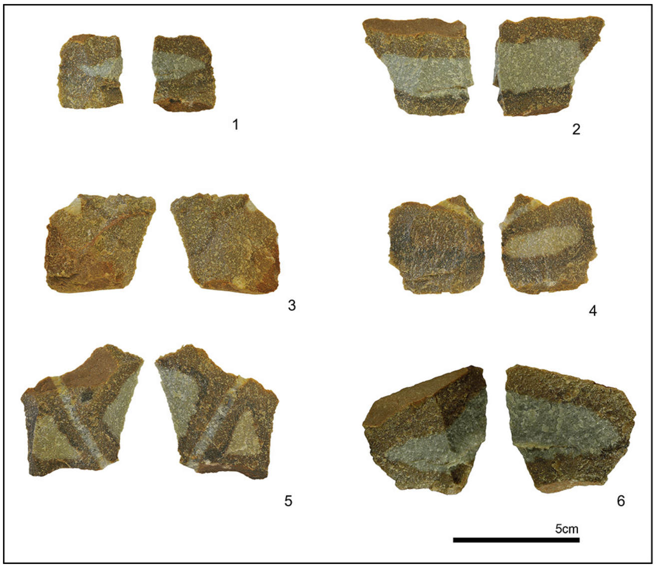

Fig. 3. The experimental quartzite flakes (VHS4) used in the whittling/scraping experiments. 1) VHS4-2, used on cane stems; 2) VHS4-5, used on bone; 3) VHS4-1, used on antler; 4) VHS4-4, used on softwood; 5) VHS4-6, used on fresh skin; 6) VHS4-3, used on dry skin.

2.2. Cleaning

Soon after the conclusion of the experiments, residues were removed by applying standard chemicals usually employed by analysists prior to microscopic scanning. The experimental replicas were firstly soaked in water and then subjected to ultrasonic baths in hydrogen peroxide (10%) for 15 min, in a neutral soap solution (Derquim-LM02) for 15 min and in acetone for 5 min. Once cleaned, the experimental tools were analysed with a SEM (Tarragona, Spain). In a second moment, LSCM scanning was done in order to measure tailored areas only (Bradfrod, UK). Prior to LSCM scanning and due to the very sensitive character of the analysis, the tools were additionally soaked in 10% NaOH for 10 min, and again in water for 10 min. Finally, they were rinsed with chromatography grade ethanol and dried immediately before analysis.

2.3. Microscopy

Each tool was first studied by means of two scanning electron microscopes used in both high and low-vacuum modes (JEOL JSM-6400; FEI quanta 600 SEM) to identify the areas which presented more widespread surface polished areas (Fig. 4: c). The same locations were then analysed with a confocal microscope (Olympus LEXT 4000) (Fig. 4: a). The samples were positioned so that the edge surface (i.e., the measured area) was as horizontal as possible (Fig. 4: b). The areas selected were imaged referring to similar horizontal fields of view. The relative differences in the magnifications of the different microscopes used were previously calculated (Table 2).

3. Results: polish on quartzite imaged through SEM-LSCM

Polish on quartzite is particularly visible when imaged with confocal microscopes, both in LEXT optical (Fig. 5: a) and laser (Fig. 5: b) images. When the analysis is coupled with the extraction of the topography layer of the analysed surface, it is possible to appreciate differences in depth regarding the position of polished and unpolished portions of the surfaces (Fig. 5: c). Some mineral inclusions other than quartz, which are typical of quartzite, are identifiable in both optical (Fig. 5: d) and confocal (Fig. 5: e) images. Changes in colour are not perceptible under SEM-secondary electron detectors (Fig. 5: f&g). Conversely, SEM-backscattered electron detectors can identify mineral inclusions in the rock, such as titanium (EDAX analysis provided the

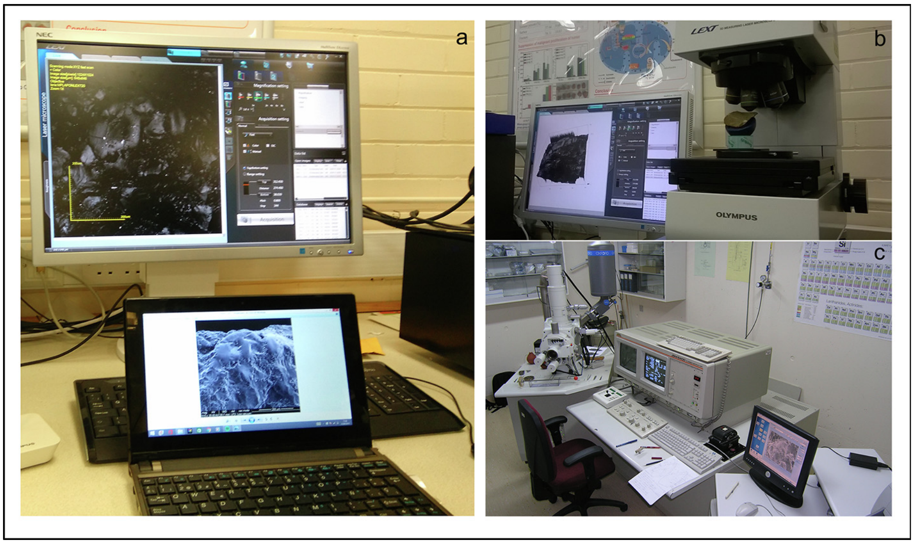

Fig. 4. a) LEXT software employed during LSCM analyses and in the front, a SEM picture used to localise each polished area during LSCM analysis; b) a quartzite experimental sample being analysed with LSCM; c) SEM (JEOL JSM-6400). The LSCM used is located at the University of Bradford (UK), while the SEMs used are located in the University Rovira i Virgili – URV (Tarragona, Spain), in the Servei de Recursos Científics i Tècnics.

elemental composition of the mineral), which are imaged in different shades on grey-scale images (Fig. 5: h). The optical microscope mounted on the LEXT was able to detect changes in colour of mineral inclusions (Fig. 5: a & d).

The highly reflective character of polish that is generally described for fine-grained lithologies observed under OM is not found on quartzite. However, on confocal images a sense of sheen can be perceived, especially contrasting with the original rock topography (Fig. 5: b; Fig. 6: b, d, f). Polished surfaces appear smoother than when imaged with SEM (Fig. 6: a, c, e). SEM has a tendency to reduce the real depth of field of the sample topography, resulting in a distorted perception of the differential heights of the analysed surfaces (Fig. 6: d, e; Fig. 7: a). Moreover, probably due to the high resolution of confocal images, tiny furrows and pits are clearly visible on the polished surfaces (Fig. 6: b, d, f), while they are less visible on the SEM micro-graphs acquired in this study (Fig. 6: a, c, e). Topographic layers of the analysed surfaces are extremely explicative, as they show the highest parts of the relief, where generally the polished surfaces are located (as in Fig. 8: c).

3.1. Polish originating from contact with antler

Working antler with quartzite, even for a prolonged time, does not cause polish on large surfaces (Pedergnana, 2017, 2019). The polishes we observed were all confined on single quartz crystals (Fig. 6). A more realistic tri-dimensional view of single crystals is obtained with LSCM (Fig. 6: b, d, f). As antler polish is generally not well-developed, it can be challenging to image it with both OM and SEM. Few polished locations were found on the analysed sample and were all located in the proximity of the used edge. Few striations in the interior of some quartz grains were observed and were more visible on confocal images (Fig. 6: f) than on SEM graphs (Fig. 6: e). The texture of the polished areas was quite smooth and was very similar to those found on the experimental flake used to work bone (see 3.1.2).

3.2. Polish originating from contact with bone

Polished areas on the sample used to whittle fresh bone are relatively larger than those observed on flakes used to work antler (Fig. 7: a, d, g). Also, the texture of this kind of polish is visually indistinguishable from antler polish. However, a few characters allow its discrimination on an experimental level. There are generally a higher number of linear features, mainly furrows, which are located on the flat surfaces of crystals (Fig. 7: b, c, l). Due to the high degree of smoothness, bone polish appears bright on confocal images (Fig. 7: e) and, although not shiny, smooth and directional patterns are clearly visible on optical images of the LEXT (Fig. 7: f). Small exogenous particles, which deposited on a sample after the acquisition of SEM micro-graphs, are detectable on confocal images (Fig. 7: h&i). This is extremely useful when attempting to measure the observed surfaces. In fact, by previously checking the surfaces to be measured through LSCM, one can verify whether the surfaces are visually clean and in this way, thereby guaranteeing that the acquired surface measurements (i.e., 3D surface data) are reliable.

3.3. Polish originating from contact with cane

Polish present on the tool used to work cane stems is easily recognisable due to its particular smoothness (Fig. 8). This kind of polish is extremely smooth, when visually compared with polishes related to other worked materials, and usually covers large areas located on the highest spots of the topography (Fig. 8: c). This polish type can appear highly reflective on confocal images (Fig. 8: b). Polish covers entire

Table 2
Comparison of the magnifications and settings necessary to image similar fields of view in the microscopes used in this study (two SEMs and LSCM). The pixel size for the images acquired with different microscopes were: JEOL and LSCM images = 1024 × 1024; FEI Quanta images = 1024 × 921 pixels.

Magn. SEM (JEOL)	Frame size (JEOL) – µm	Magn. SEM (FEI)	Frame size (FEI) – µm	Magn. LSCM (LEXT)	NA (LSCM)	Frame size (LSCM) – µm
50×	2312 × 1872	135×	2210× 1900	MPLFLN 5×-1×	0.15	2571 × 2579
100×	1156 × 936	260×	1150 × 989	MPLFLN 10×-1×	0.3	1281 × 1280
200×	578 × 468	510×	585 × 503	MPLAPONLEXT20×-1×	0.6	646 × 646
500×	231 × 187	1250×	239 × 205	MPLAPONLEXT20×-2×	0.6	323 × 323
500×	231 × 187	1250×	239× 205	MPLAPONLEXT50-1×	0.95	259 × 259
900×	128 × 103	2300×	130 × 111	MPLAPONLEXT50-2×	0.95	129 × 129

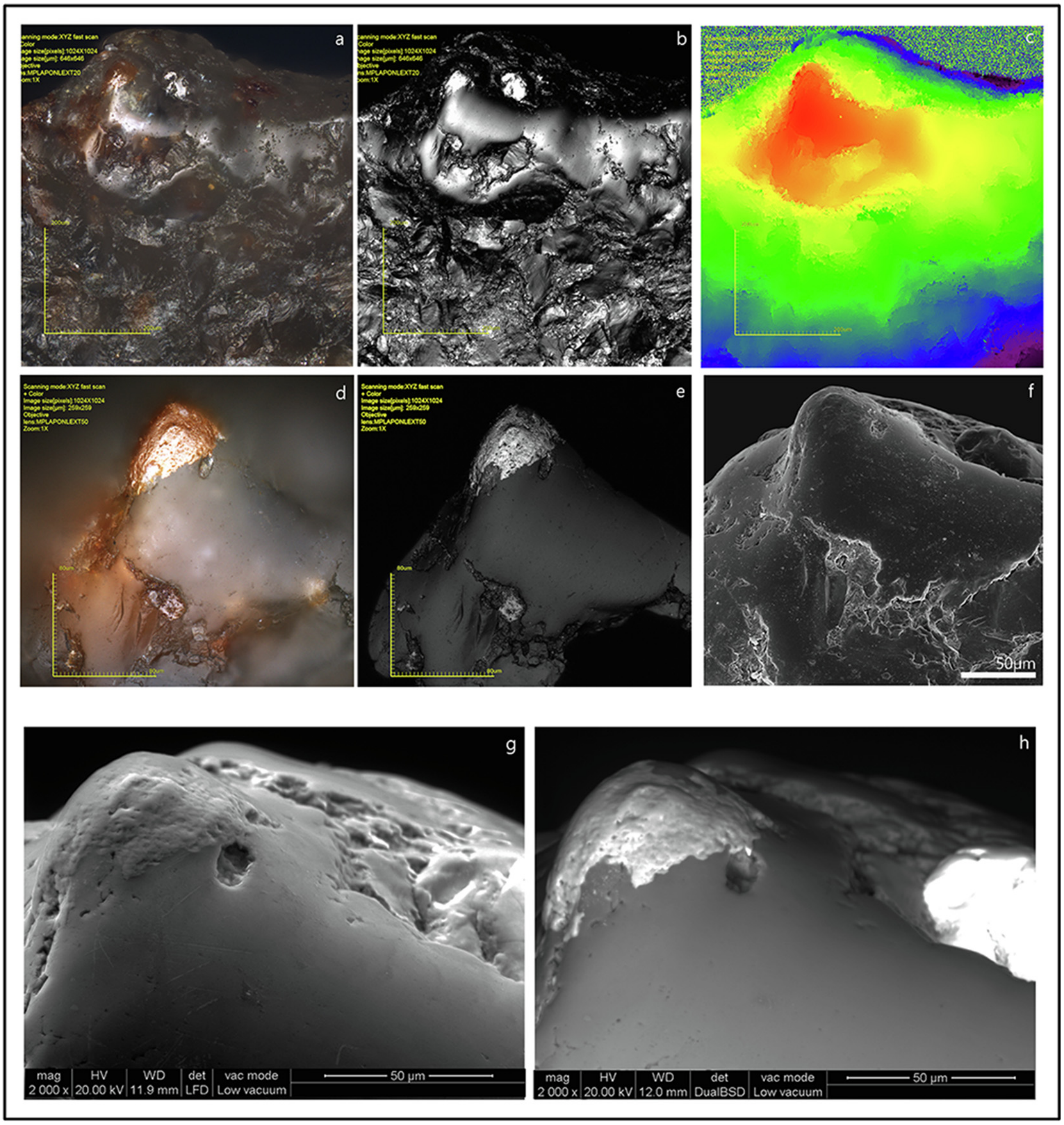

Fig. 5. Comparison of the same polished area generated after whittling a cane stem and observed under a LSCM (a-e: a&d = white field images, b&e = maximum intensity maps, c = height map), a high-vacuum SEM secondary electron detector (f), a low-vacuum SEM secondary electron (g) and back-scattered electron detector (h) images. An accessory mineral mainly composed of Ti is visible under the LSCM (d&e) and under the SEM-back-scattered elector detector, where it appears lighter than the rest of the surface (h). Original magnifications and lens: a-c) 20 ×, at 1 zoom; d&e) 50 ×, at 1 zoom; f) 450 ×; g&h) 2000 ×.

quartz crystals' surfaces, which are generally free from linear marks (Fig. 8: g-n). Nevertheless, marks resembling pits can sometimes be observed on confocal images (Fig. 8: m&n).

3.4. Polish originating from contact with dry and fresh animal skins

Tools used to scrape deer skins present diagnostic polishes, which are easily distinguishable from polishes formed after contact with other materials (Fig. 9). This kind of polish is found on the opposite extreme of cane's polish, as it is easily appreciable from its rough texture. Polish areas are in this case always very rough and they cover large surfaces (Fig. 9: a&b). The boundaries between crystals are erased as the surface is being worn down (Fig. 9: c-d). The contrast between the polished surfaces and the original, flat surfaces of quartz crystal is easier to see on confocal images (Fig. 9: b&d) than on SEM ones (Fig. 9: c). However, when the rough character of the polish is not well-developed, as in the case when fresh skins were worked, it can be difficult to determine the limits of the polished areas on confocal images (Fig. 9: e–f).

3.5. Polish originating from contact with wood

Extensive polished areas were not found on the sample used to whittle a wood branch. When present, polish was delimited on the highest surface portions of single crystals (Fig. 10: c-d). The flattest areas on the polish slightly resemble the polish obtained after whittling cane (Fig. 10: c). Pits and striating surfaces, as well as neoblasts (Pedergnana et al., 2017), are more readily visible on confocal images (Fig. 10: d & f) than on SEM micro-graphs (Fig. 10: e).

3.6. 3D surface modelling and polish formation

For each observed location, we obtained 3D models displayed as optical images, maximum intensity and height (i.e., topographic layer) maps. Topographic layers are themselves a valuable source of information, as they display different surface heights in different colours. Red is used to indicate the highest parts, that generally coincide with the areas where polish is present (Fig. 5: c; Fig. 8: c; Fig. 11: c, h; Fig. 12: c, l). Even if it is known that polish develops first on the highest parts of the topography, it might be useful to back up the analyst's

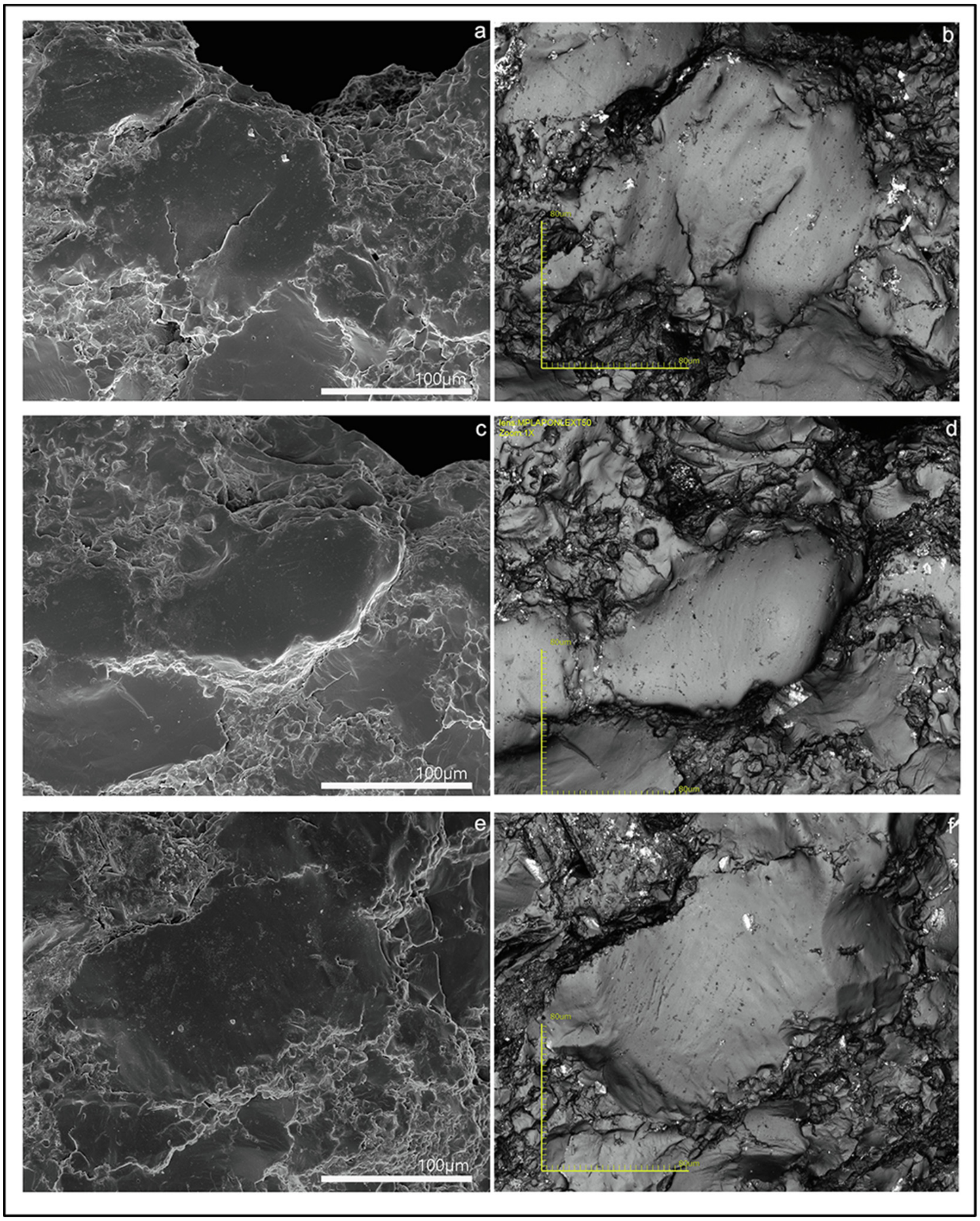

Fig. 6. Polish originating from contact with antler. Original magnifications: High-vacuum SEM micro-graphs, a, c, e) 450 ×; Max. intensity maps (confocal images), b, d, f) 50 ×, at one zoom.

interpretations by providing clear documentation of the topographic heights. Consequently, data from the topographic layer of confocal images can be very useful for understanding whether a surface is polished or not, when identifying polish on quartzite with both OM and SEM proves particularly difficult. Comparing optical and confocal images and the topographic layers obtained with LSCM of the same surface areas can be a valuable tool for overcoming the technical constraints of OM when searching for polish on quartzite (Figs. 11 and 12: a-c; f-h).

We saw that even after one hour of intensive use, the polished locations observed were still found on the highest parts of the surfaces, including for the hardest worked materials. This means that polish on quartzite is characterised by low rates of attrition. When 3D models of the worn surfaces are created, the location of polish is better perceived (Figs. 11 and 12: d-e; i-l). 3D models of topographic layers are indeed the most informative ones (Figs. 11: l; Fig. 12: e, l). They allow visually locating the polished areas within a topographic context, which is extremely useful when approaching the issue of use-wear formation on stone tools. They give some clues on where analysts should look when observing stone tools under a microscope. 3D models on both optical and confocal images are also useful, as they add information regarding colour and brightness of the polished areas to the models themselves (Fig. 11: d; I; Fig. 12: a; i).

4. Discussion

In the field of traceology, confocal microscopy has mainly been applied in attempts at quantifying use-wear on stone tools (e.g., Evans and Donahue, 2008; Evans et al., 2014; Evans and MacDonald, 2011; Stemp and Chung, 2011; Stemp et al., 2013; Ibáñez et al., 2016, 2018).

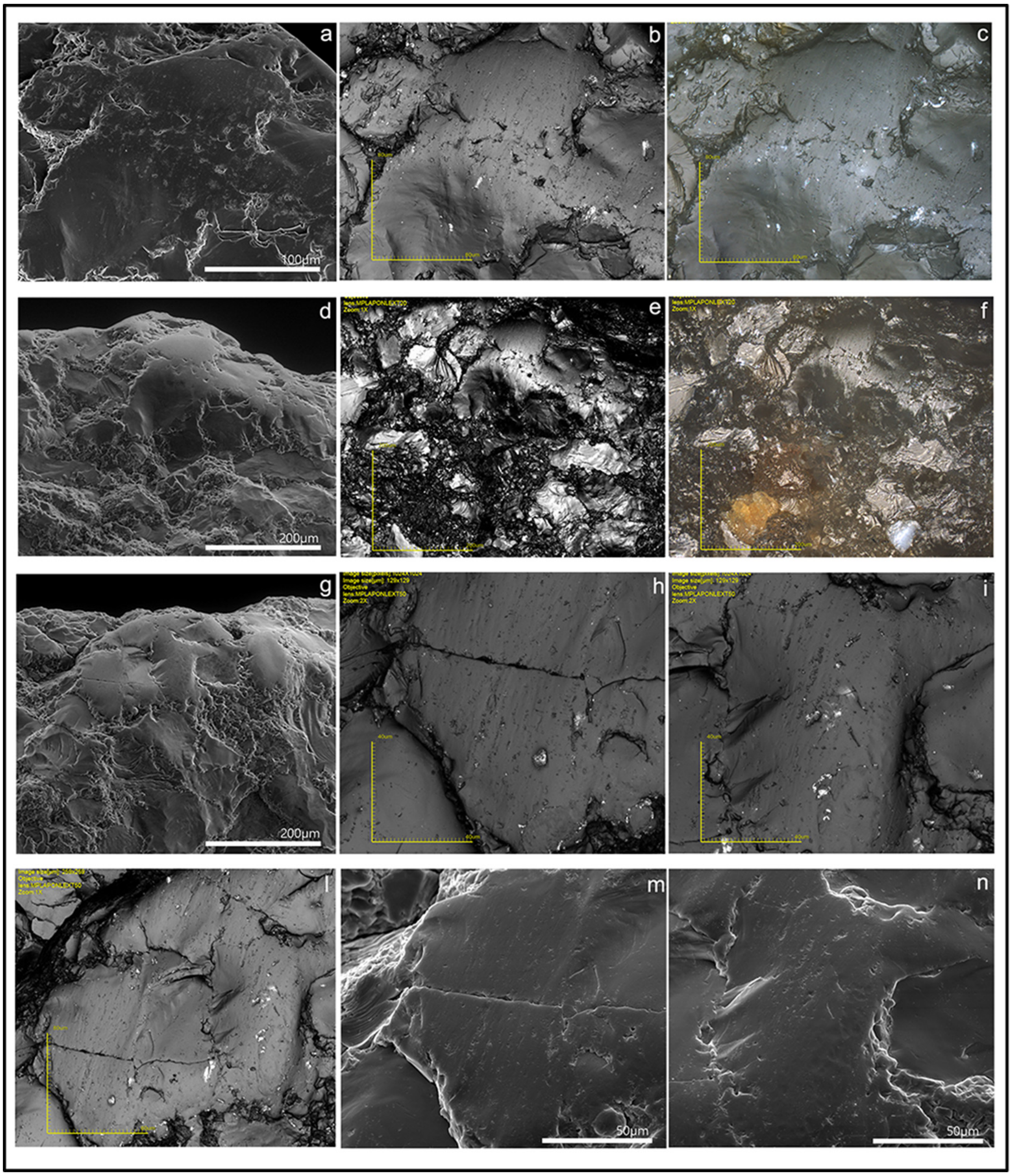

Fig. 7. Polish originating from contact with bone. a, d, g, m, n) SEM micro-graphs; b, e, h, i, l) maximum intensity maps (confocal images); c&f) optical images. Original magnifications: a) 450 ×; b, c, l) 50 ×, at 1 zoom; d&g) LFD detector, low-vacuum SEM, 510 ×; e&f) 20 ×, at 1 zoom; m&n) ETD detector, high-vacuum mode, 2000 ×.

A secondary, but not less important, feature of LSCM is the possibility of obtaining both optical and laser images. These allow better appreciation of the visual characteristics of the surfaces which are attempted to be measured. However, use-wear analysts have rarely taken advantage of this facet of LSCM and have only used it to obtain 3D-surface measurements of worn surfaces (Evans and Donahue, 2008; Xie et al., 2019). The relative ease with which confocal microscopes are used and the extremely high-quality images they provide make them really suitable to image particular features on reflective samples.

As polish on quartzite develops much more slowly than on flint and has clearly different visual characteristics (e.g., Clemente-Conte and Gibaja-Bao, 2009; Pedergnana, 2017), detecting polished areas on this and similar rock types (such as quartz-arenite) might be extremely challenging. In contrast to cherts, polish on quartzite is not bright; therefore its surface does not have enough contrast to be quickly detected when samples are observed with OM. On a general basis, polished areas on quartzite are matte and the brightest areas are random spots around the surface (Fig. 1: b, c, e). They correspond to the surfaces of single quartz crystals which are oriented in a way so as they reflect the white light used by the microscope to amplify the sample. This is one of the reasons why use-wear analysts need to be trained for a long time before being able to visually distinguish polished surfaces on quartzite from the original, unused surfaces. This is valid also when the microscope employed is a SEM. Under the SEM, smooth polishes (e.g. antler or bone polishes) on quartzite are visually analogous to the unused, flat surfaces of the largest quartz grains composing quartzites (for instance, Fig. 6: a, c, e). Therefore, the potential of other equipment to image polish on quartzite needed to be explored. The main objective of this study was to compare images taken with SEM and LSCM of the same polished areas on quartzite. Well-developed polish was created intentionally by performing prolonged actions on several worked materials (n = 5).

The observations of different polishes on quartzite by means of confocal imagining gave results consistent with the observations

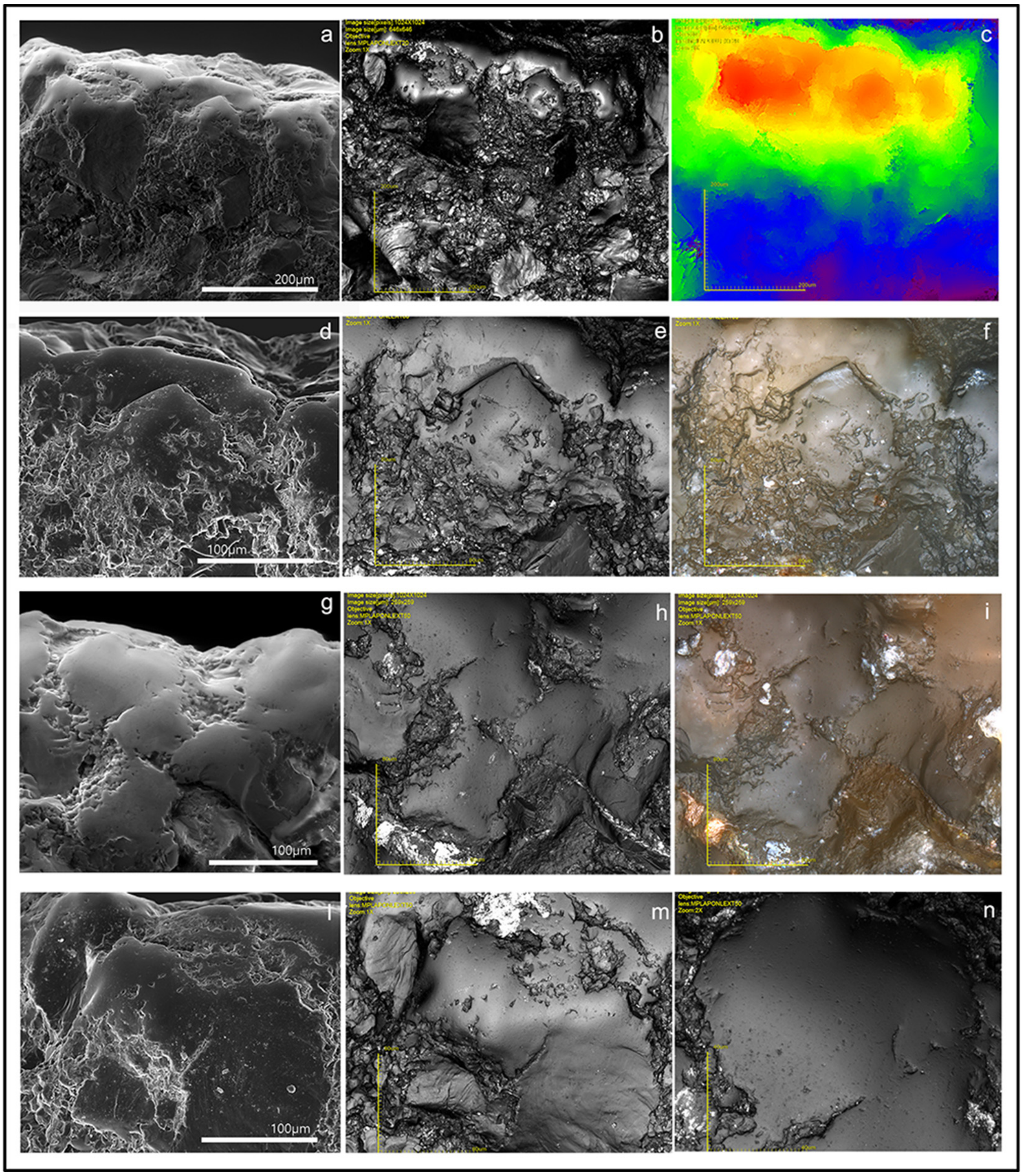

Fig. 8. Polish originating from contact with cane. a, d, g, l) SEM micro-graphs; b, e, h, m, n) maximum intensity maps (confocal images); c) height map (topographic layer); f&i) optical images. Original magnifications: a) LFD detector, low-vacuum SEM, 510 ×; b&c) 20 ×, at 1 zoom; d, g, l) high-vacuum SEM, 450 ×; e, f, h, i, m) 50 ×, at 1 zoom; n) 50 ×, at 2 zooms.

previously made using SEM. However, as was the case with SEM, confocal microscopy offers several advantages over conventional OM in the identification of polished areas on reflective materials such as quartzite. Experimental polishes originating from contact with different materials can be discriminated through visual analyses of optical and confocal images obtained with LSCM. Furthermore, 3D-models of the observed surfaces can be created and these contribute to a better understanding of how use-wear forms.

Hence, besides the common application in surface metrology, LSCM can be used to provide high-quality images (optical and confocal) of the samples analysed. Observing the surfaces to be measured is extremely important, as the surfaces should potentially be free from any extraneous particles before measuring. This allows for a rapid comparison with confocal images (i.e., maximum intensity maps) and therefore, documenting where the measurements are taken with exact precision. This can be very useful for showing the analysis workflow during the dissemination of results.

However, it has to be said that the true advantage of confocal microscopy over both OM and SEM is the possibility of getting quantitative data of the micro-surface of the sample. Features like smoothness, roughness, and waviness (and many others) can be measured and ordered in numerical categories. In this way, the correlation of the same numerical values of the parameters analysed is objective and comparable between different studies (if the same equipment is used). It has to be said that the available studies in the literature are limited to flint and little has been done to quantify use-wear on other raw materials. Larger datasets are needed, even for flint; comparison of data acquired with different pieces of equipment has to be done systematically and many tests are needed to define the most adequate parameters for assessing surface changes on different lithic raw materials. The surfaces of the samples presented in this study were all measured using confocal microscopy. Although not further discussed in this contribution, the results of this analysis are very promising as they allowed distinguishing different contact materials. This will have important implications as it demonstrates the value of confocal microscopy in the quantification of wear on raw materials other than flint.

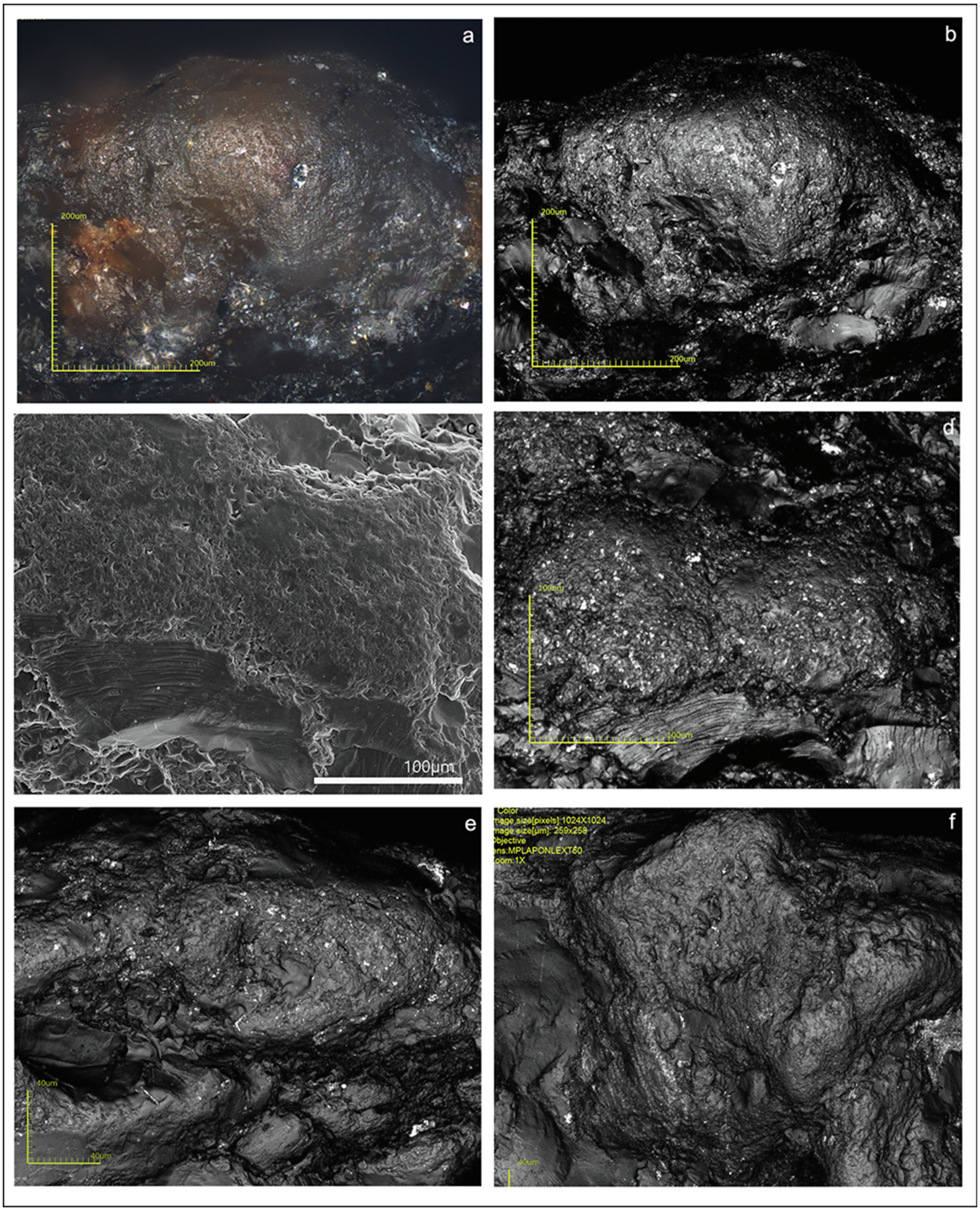

Fig. 9. Polish originating from contact with dry and fresh animal skins. a) Optical image; b, d, e, f) Maximum intensity maps (confocal images); c) high vacuum SEM micro-graph. Original magnifications: a&b) 20×, at 1 zoom dry hide; c) 450×; d) 20×, at 2 zooms; dry hide; e&f) 50×, at 1 zoom, fresh hide.

SEM is another piece of equipment which has not been used systematically in the field of traceology. The high-resolution and the three-dimensional effect of micro-graphs obtained through SEMs, which provide topographical, morphological and compositional information, make them invaluable for better assessing the nature of surface modifications of stone tools. One of the main advantages of SEM over OM is the detailed three-dimensional and topographical imaging and the versatile information gathered from different detectors. Although all samples must be prepared before being placed in the vacuum chamber (high-vacuum only), most SEM samples require minimal preparation actions.

The obvious disadvantages of both SEM and LSCM are size and cost. These microscopes are expensive compared to optical ones; they are large and must be housed in appropriate areas free of any possible electric, magnetic or vibration interference. Moreover, maintenance of both microscopes is necessary. Last, but not least, special training is required to operate both LSMC and SEM as well as to prepare samples for SEM observations. In addition, SEMs are limited to samples small enough to fit inside the vacuum chamber, and there are similar limitations of sample-height and weight of samples to be analysed with LSCM.

Conversely, OM is less expensive and time consuming, more user-friendly and more easily available at laboratories. It is a valuable tool for preliminary checking the surfaces to be analysed with other techniques, and could be used for selecting areas of interest. It also provides a real time representation of samples. Hence, a multi-scalar, complementary approach, if envisaged, can be widely flexible and be specifically tailored depending on one's own research questions.

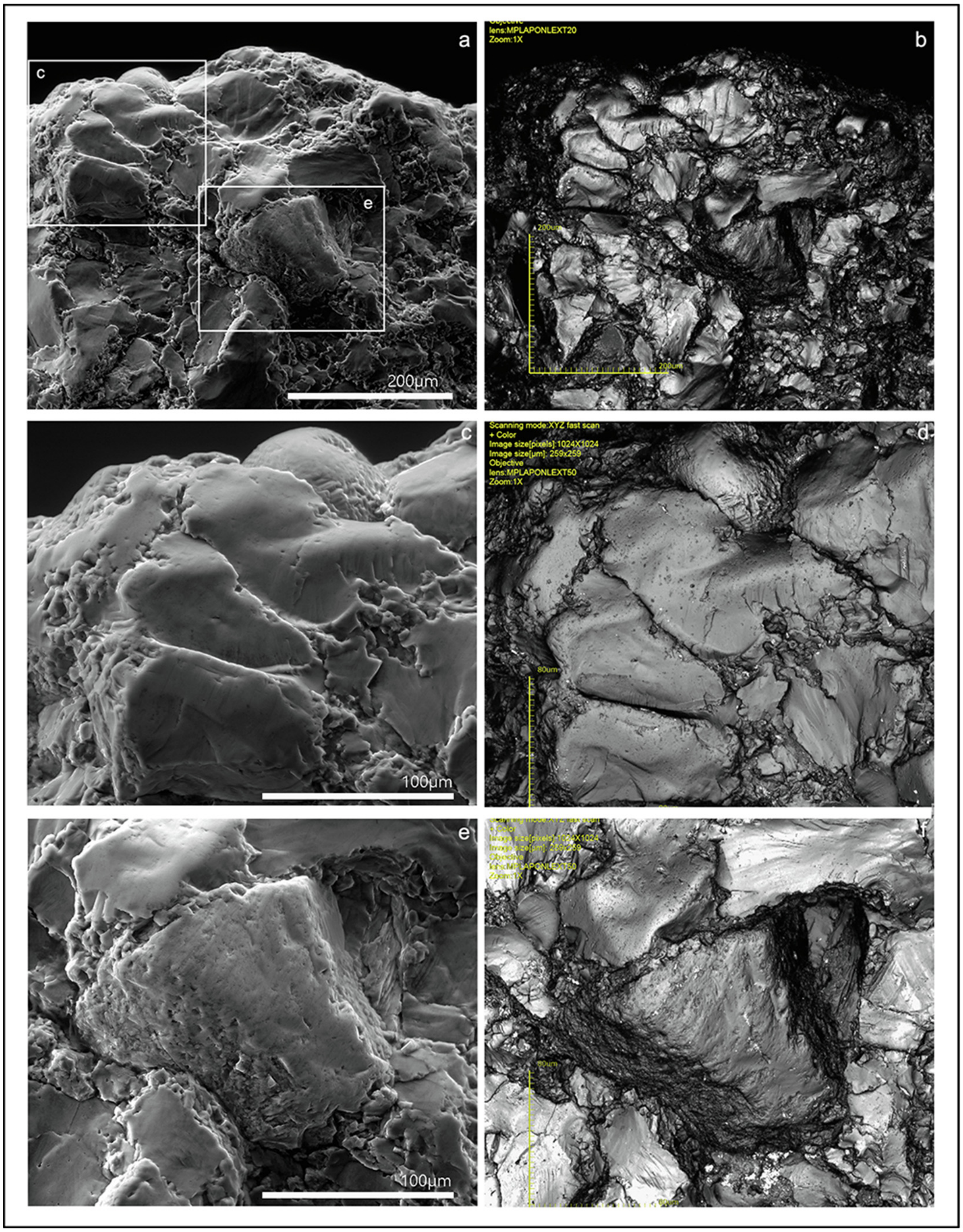

Fig. 10. Polish originating from contact with wood. Original magnifications: a) LFD detector, low-vacuum SEM, 510 ×; c&e) LFD detector, low-vacuum SEM, 1250 ×; b) Max. intensity map (confocal image), 20 × at 1 zoom; d&f) Max. intensity maps (confocal images), 50 × at 1 zoom.

5. Conclusion

The use of LSCM in use-wear analysis is relatively new and, therefore, still unexplored. The first preliminary studies investigating roughness changes of worn and unworn lithic surfaces have converted LSCM into an attractive analytical tool for use-wear analysists. The full potential of this equipment in traceology has not, however, been entirely assessed. In fact, besides providing quantitative topographical data, it can be also a powerful imaging tool. Both optical scans and confocal micro-graphs can be used to assess the visual appearance of worn surfaces on lithics.

This contribution aimed to show the largely unexplored possibilities of combining LSCM and SEM imaging modes to better assess the visual appearance of use-wear on quartzite. Although, this study focused on the analysis of well-developed polished areas originating from contact with different worked materials on quartzite experimental replicas, LSCM can also image other use-wear features, such as striations. Our results reflect two distinct aspects involved in the description of polished areas. First, LSCM grey-scale images add topographic information and, when used in conjunction with SEM images, contribute to a more thorough description of the visual aspect of the worn areas. Second, confocal microscopes can serve as high-resolution optical microscopes characterised by a large depth of field.

To conclude, both LSCM and SEM are very versatile and can offer multiple ways of analysing worn surfaces on stone tools. Both microscopes proved to be adequate for scanning irregular and reflective rocks, such as quartzite. They might be employed to analyse very reflective surfaces on materials other than quartzite (e.g., quartz),

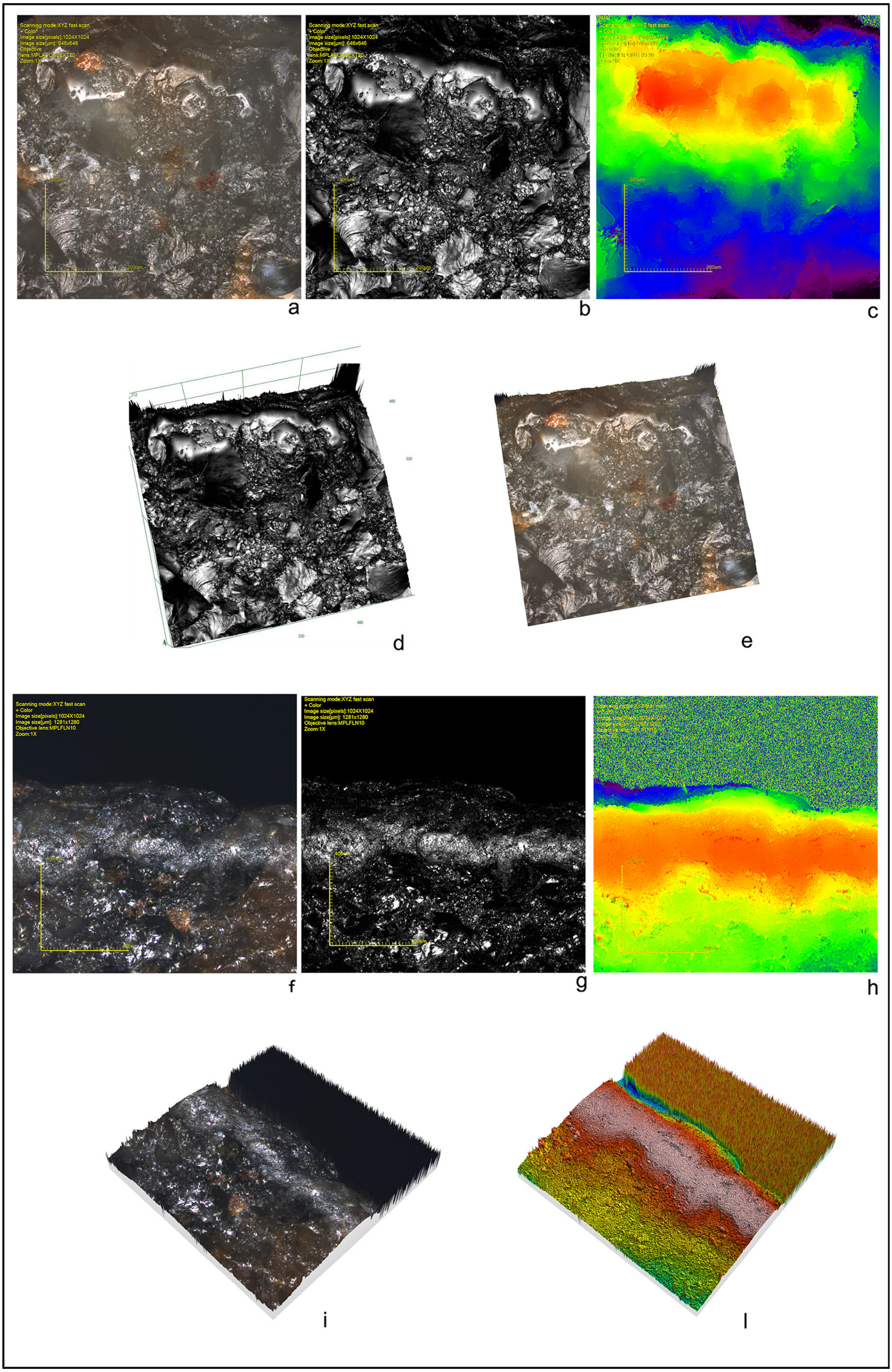

Fig. 11. a-e) Extensive, smooth polish on a tool used to whittle a cane stem. Comparison of the optical image (a), max. intensity map (b), topographic layer (c), 3D models on the max. intensity map (d) and the optical image (e); f-l) Extensive, rough polish on a tool used to scrape a dry skin of a deer. Comparison of the optical image (f), max. intensity map (g), topographic layer (h), 3D models on the optical image (i) and the topographic layer (l). a-e) Original magn: 20× at 1 zoom; f-l) 10× at 1 zoom.

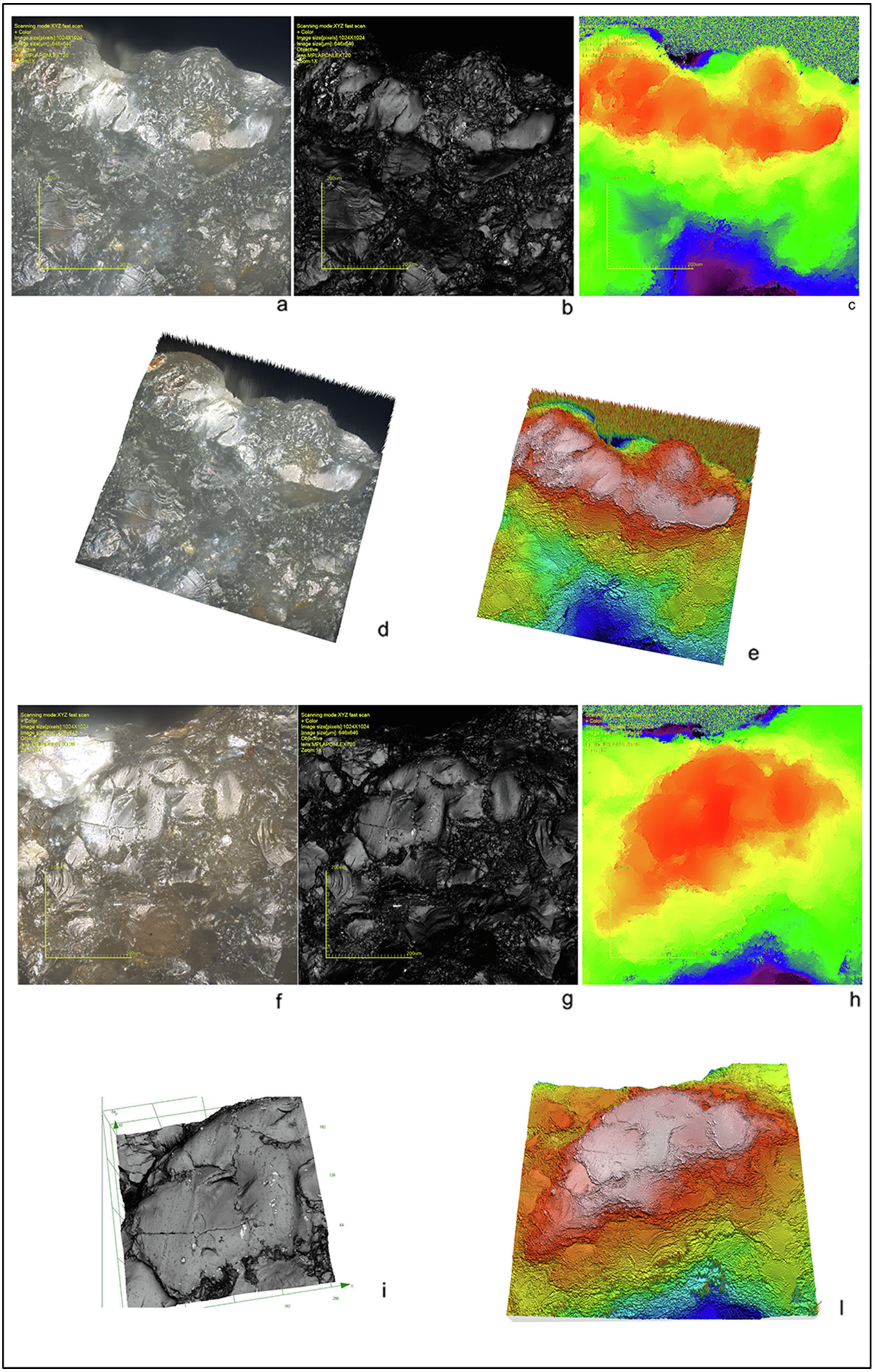

Fig. 12. a-e) Smooth polish on a tool used to whittle a deer antler. Comparison of the optical image (a), max. intensity map (b), topographic layer (c), 3D models on the optical (d) and the topographic layer (e); f-l) Smooth polish on a tool used to whittle a long bone. Comparison of the optical image (f), max. intensity map (g), topographic layer (h), 3D models on the confocal image (i) and the topographic layer (l). a-l) Original magn: 20× at 1 zoom.

especially when technical constraints of OM impede to adequately observe use-wear on them.

More efforts for developing this branch of study are needed, as the possibility of quantifying wear features (polish, striations, micro-scars) that are generally described subjectively, would allow taking a significant step forward in the development of the discipline.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This research was developed within the frame of the AHRC Fragmented Heritage project (AH/L00688X/1) at the University of Bradford, and of the MICINN-FEDER (PGC2018-093925-B-C32), the AGAUR (SGR 2017-1040) and the URV (2018PFR-URV-B2-91) projects at IPHES-URV. One of the authors (A.P.) was beneficiary of a Catalan pre-doctoral grant (2014FI B 00539), at the Rovira i Virgili University (URV), the IPHES and the Muséum national d'Histoire naturelle of Paris. The analyses were performed at the Servei de Recursos Científics i Tècnics of URV and at the University of Bradford School of Archaeological and Forensic Sciences. The authors want to thank the Editor-in-Chief and the anonymous reviewers for their very helpful comments. Thanks go also to Ivan Calandra and João Marreiros for their valuable comments to previous versions of this manuscript.

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