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To cite this version:

Caroline Hamon, Solenn Réguer, Ludovic Bellot-Gurlet, Yvan Pailler, Vérane Brisotto. Tracking the first bronze metallurgists of Western Europe: combined use-wear analysis and X-ray fluorescence synchrotron spectroscopy of a stone toolkit from Ploneour-Lanvern (Brittany). Archaeological and Anthropological Sciences, 2020, 12 (1), pp.14. 10.1007/s12520-019-00999-x . hal-02444675

HAL Id: hal-02444675

https://hal.science/hal-02444675v1

Submitted on 21 Feb 2021

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Hamon C., Réguer S., Bellot-Gurlet L., Pailler Y., Brisotto V. (2020). Tracking the first bronze metallurgists of Western Europe: combined use-wear analysis and X-Ray Fluorescence synchrotron spectroscopy of a stone toolkit from Ploneour-Lanvern (Brittany), *Archaeological and Anthropological Sciences*, **12**, 14. https://doi.org/10.1007/s12520-019-00999-x

Tracking the first bronze metallurgists of Western Europe: combined use-wear analysis and X-ray fluorescence synchrotron spectroscopy of a stone toolkit from Ploneour-Lanvern (Brittany)

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Abstract

At the turn of the second millennium BC in Europe, the development of copper and bronze metallurgies stimulated major economic shifts, with the emergence of elites and far-reaching diffusion networks. However, the exact organization, techniques, processes, and equipment employed in this early copper and bronze metallurgy still remain poorly understood. In this context, our aim has been to identify the first stone toolkits used by Early Bronze Age metallurgists on a site in western Brittany (France), a region connected to the so-called Atlantic complex. The combination of use-wear analysis and Synchrotron X-ray fluorescence spectroscopy (XRF) imaging highlights a correlation between the distribution of diagnostic use-wear traces (smoothing and micropolishes) related to metal ore processing or metal objet shaping and elemental residues of copper and zinc. These results demonstrate the existence of copper ore processing in western Brittany during the Early Bronze Age (2200-1900 BC). They also highlight the diversity of the stone tools involved in metal production. Finally, this comprehensive methodology underlines the potential of functional analysis of macrolithic tools to reveal the existence of metal production. This is particularly relevant for archeological sites where the structures of metallurgical production are barely visible and often unrecognizable and where metallic objects are rare due to their intensive recycling and diffusion.

Keywords: Early Bronze Age, stone tools, use-wear analysis, X-ray fluorescence imaging, synchrotron source.

Introduction

The emergence and development of copper and bronze metallurgy is a key step in the evolution of protohistoric societies. At the turn of the second millennium BC in Europe, copper and Early Bronze Age societies were characterized by major economic shifts. Emerging powerful elites enriched themselves by controlling the exploitation of ore sources (copper, tin, silver, and gold), mastering metal production, and developing extensive diffusion networks. The intensification of long distance exchanges of valuable objects and know-how stimulated the emergence of new cultural complexes.

Along the Atlantic coast, the rise of copper alloy metallurgy has been dated to the middle of the third millennium BC, within Late Neolithic to Early Bronze Age contexts, two millennia after its emergence in south-eastern Europe (Radivojevi et al. 2010).

Bell-Beaker populations appear to have been instrumental in stimulating the spread of these new technologies which accompanied their phenomenal cultural expansion throughout continental and littoral Europe between 2800 and 1900 BC. In these contexts, the deposition of stone implements within so-called metallurgists' graves throughout Europe highlights the intrinsic mobility, craft skill, increasing specialization, and particular social status of these craftsmen (Bátora 2002; Butler and van der Waals 1967; Drenth et al. 2014; Fitzpatrick 2002). Between 2100 and 1800 BC, in Western Europe, the Armorican Tumulus culture in Brittany and the Wessex culture in southwest England developed an intense and novel interregional exchange network across the Channel based on metallurgical innovations (Marcigny et al. 2017), often termed a system of "maritories" (Needham 2009).

However, the exact organization, techniques, processes, and equipment employed in bronze metallurgy still have to be identified within these cultures (Pernot 1998). In fact, these workshops generally left only ephemeral archeological traces, such as hearths, rare crucible fragments, or various kinds of stone tools (Armbruster 2010; Boutoille 2015; Freudenberg 2010; Webb 2015). Consequently, the reliable identification of metallurgists' stone tools on different types of archeological sites (open-air settlements, specialized workshops, enclosures) appears to be a promising and challenging new approach to identifying the practice of bronze metallurgy.

To date, most of the archeometric analyses of copper and bronze metallurgy have focused on the characterization of the composition and structure of metal objects (axes, daggers, swords, etc.), in order to answer questions regarding ore origin, places of production, and circulation of metal. However, archeometric studies have rarely been applied in order to identify the possible role of stone implements within the metallurgy of copper, bronze, silver, or gold on the European continent. But as a matter of fact, stone tools undoubtedly played a major role at several stages of these metallurgical processes, from the extraction of ores to their processing by crushing and grinding, the casting/forging of metal objects, and finally their sharpening/finishing. In southern France, a combination of microscopic use-wear analysis and X-ray fluorescence spectroscopy (XRF) of stone tools has been used to identify metallurgists' stone toolkits which were involved in processing copper ores at La Capitelle du Broum in Cabrières (Hérault, France), one of the oldest metal processing sites in the western Mediterranean Basin dating to 2900 cal BC (Hamon et al. 2009). In the Iberian Peninsula, combined use-wear analysis and synchrotron-based XRF have recently identified stone tools involved in copper and silver processing in the Middle Bronze Age culture of El Argar (Delgado-Raack and Risch 2008; Delgado-Raack et al. 2014). On the Atlantic coast, microscopic use-wear analysis conducted on two Middle- and Late Bronze Age series from Brittany has led to the identification of a complete percussion and abrasion toolkit which was involved in different steps of the production of metal objects (Hamon and Blanchet 2015).

This paper presents a methodology which combines microscopic use-wear analysis at low and high magnification and Synchrotron XRF analysis using fast imaging, in order to identify a metallurgist's stone toolkit on a recently excavated Early Bronze Age site at Kersulec in Ploneour-Lanvern (Brittany, France, 1950-1690 BC). The goal of this combined approach was to obtain detailed information regarding the functioning of the macrolithic tools (kinetics, hafting, use-gesture) and the material being transformed (nature, state, texture, composition). Ultimately, this would allow us to (1) identify the tools involved in metallurgical processes, (2) identify the material that they had been in contact with, and finally (3) determine in which stage of the *chaîne operatoire* of metallic production they were used. This case study aims to present the most suitable combination of archeometric methods in order to identify the metallurgical stone tools within larger macrolithic assemblages found on Bronze Age sites. The generalization of such functional studies of stone tools offers an opportunity to identify the equipment used in the different stages of metal production (mining, processing, finishing) and to highlight the existence of copper and bronze production workshops within the various kinds of Early Bronze Age archeological sites (open-air settlements, specialized workshops, enclosures) known throughout Europe.

Materials and method

The site of Kersulec in Ploneour-Lanvern (Finistère)

The Early Bronze Age occupation site of Kersulec in Ploneour-Lanvern (Finistère; Fig. 1) is located in a flat area on the northwestern slope dominating the valley which ends in the Trunvel Lake. The substratum of the northern part of the site consists of Trunvel micaschists, while to the south, it consists of Ploneour orthogneiss. These formations are overlain by clayish silts in which the archeological remains were discovered in the course of a rescue excavation carried out in 2015 by the *Institut National de Recherches Archéologiques Préventives* (INRAP) (Pailler 2016).

Despite the fact that dug features tended to be scarce and poorly preserved, it was possible to identify several structures. A cist burial and a pit burial, which had been cut by a large waste pit, were revealed. These burial types are well-known in the Early Bronze Age of lower Brittany. The relative chronology indicates that these burials predated the domestic occupation. The occupation itself is composed of a small building with a surface area of approximately 10 m^2, two storage pits and two light structures each consisting of four posts; the latter were interpreted as possible workshops or granaries.

Fig. 1: Localization of the Early Bronze Age site of Ploneour-Lanvern (Brittany), among the Atlantic coast.

Several isolated structures, 2 small wells and 13 hearths complete the settlement. All of these structures suggest a rather small-scale domestic occupation of short duration. This is confirmed by the homogeneity of the ceramic assemblage and by two C14 dates which date the occupation to the first third of the second millennium cal. BC (1955-1775 cal BC and 1885-1690 cal. BC). In this context, the discovery of a crucible used for smelting copper or bronze, together with a series of stone tools, under one of the possible post-built structures in the northern part of the site raised the possibility of the existence of a metallurgical workshop. Such structures remain difficult to identify on Early Bronze Age sites as they generally leave little or no traces, except for some preserved stone and clay tools. Thus, the site of Kersulec in Ploneour-Lanvern offers a rare opportunity to explore evidence for metallurgical production and related craft activities dating back to the Early Bronze Age in western France.

Sampling and use-wear analysis of macrolithic tools

Following their discovery, the macrolithic tools from Kersulec were analyzed according to the principles of technological analysis, i.e., in terms of their raw materials (types, cohesion, granulometry, and mechanical properties), typology (morphometry and morphology of the tools), and technology (shaping processes and schemes, possible hafting, localization of the active surfaces).

Macro- and microwear analysis was conducted at the UMR 8215 Trajectoires laboratory using a stereomicroscope SMZ 800 (up to 80 x) and an Olympus BX51M microscope (up to 200 x).

Fourteen stone tools were analyzed in order to better understand their function and how they were actually used within a wide range of possible food preparation and craft activities. The toolkit consists of 14 tools including hammerstones made from quartz fragments and cobbles, rare grinding tools used in food production, and burnishers and polishers involved in ceramic production. Only five of these tools displayed possible use-wear related to metal production. Among these, three were selected for residue analysis because of the good state of preservation of their surfaces.

Since its development in the 1990s, use-wear analysis on macrolithic tools has focused on the development of a suitable terminology adapted to the diversity of rocks employed, particularly for coarse and heterogeneous rocks, and on the need for highly integrated low and high power observation of the traces. The terminology employed in this paper follows the generally accepted framework for use-wear analysis of macrolithic tools. At a macroscopic level (Hamon 2008; Adams et al. 2009), the topography and microtopography of the surfaces are described in terms of their morphology (flat, sinuous, rough) and regularity. The leveling of the surfaces is characterized by its distribution (*loose*, *covering*, or *concentrated*) and density (pattern of leveling, i.e., *separated, close, or connected*), incidence, morphology (*flat, sinuous or rounded*), and texture (*rough or smooth*).

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Linear traces follow more or less the same guidelines for distribution and density, but orientation and dimensions are also considered. Grain edge rounding, grain extraction, and microfractures are also described at this scale of observation. At a microscopic level, descriptions of micropolish follow the terminology that is generally used, with some adaptations to take account of the heterogeneous character of the raw material and its granular structure (Clemente Conte 1997; Hayes et al. 2018).

In parallel, experimental reference data has been compiled focusing on a variety of technical fields, from food processing (cereals, nuts, plants, etc.) to craft production including the shaping of pottery, the manufacture of ornaments, the production of stone or bone tools, the treatment of skins and vegetable fibers, the grinding of pigments, etc. (for a general overview, see Dubreuil and Savage 2013). In this field, very few studies describe specific use-wear traces left on experimental stone tools involved in metal processing as most of the experiments that focus on ore processing or the production of metallic tools tend to be aimed at describing the products obtained (Timberlake et al. 2007) and the traces left on metallic objects (Gutiérrez Sáez and Martín Lerma 2015). Our own reference data includes more than 130 experiments encompassing all fields of food processing and craft production including food grinding (cereals, plants, nuts), pottery production, the manufacture of ornaments, the manufacture of stone and bone tools, the treatment of skins and vegetable fibers treatment, the shaping of metal objects, and the crushing of pigments (Hamon 2008, Hamon and Plisson 2008). Recently, experimental tests were conducted on the crushing of copper ore and on the shaping of copper and bronze objects by percussion and abrasion using 18 stone tools (Fig. 2). A summary of the related use-wear traces at macro- and microscopic scales is presented in Fig. 2. Contact with metal is mainly characterized by rough leveling of the surfaces, microbreakage of the individual grains, and shiny and compact to micropitted micropolish. These observations corroborate the description of "a shiny surface" on hammers and anvils surfaces employed for the manufacture of metallic objects (e.g., Delgado-Raack and Risch 2008, p. 239).

X-ray fluorescence imaging using the DiffAbs beamline at Soleil Synchrotron

Use-wear analysis led us isolate three tools (n°13, 15, 18) from among the stone tool assemblage; these displayed specific use-wear suggesting possible processing of metallic ores and metal objects. These three tools were mapped through X-ray fluorescence using the DiffAbs beamline at the Soleil Synchrotron facility (France). The equipment used allows rapid XRF imaging and yields elemental mapping over large surfaces of the tools in quite a short time span (approx. 1 h) and allows us to identify specific areas of interest, e.g., metallic residues. It seeks to identify and estimate the concentrations and distributions of copper, arsenic, iron, zinc, and lead residues which are used in different bronze alloys.

The DiffAbs beamline of the Soleil Synchrotron facility (France) is a versatile setup providing multi-technique and multi-scale characterization tools capable of analyzing a large variety of materials. X-ray diffraction, absorption, and fluorescence spectroscopies can be applied in local probe or imaging (raster maps) modes to reveal chemical and structural information.

The so-called macrobeam setup at DiffAbs consists of two cylindrical, vertically-focusing mirrors, and a Si (111) double crystal monochromator, which allows monochromatization and focusing of the beam to about 285 μm × 216 μm (horizontal × vertical, full width at half maximum FWHM).

The acquisition of pluricentrimetric elemental maps with a resolution of several hundreds of microns is possible, thanks to the combination of the focused X-ray beam, the efficiency of the detectors used, and the new continuous *Flyscan* acquisition mode (Leclercq et al. 2015). This fast, efficient scanning of the samples leads to a reduction in the acquisition time per map (with frequencies of up to several hundreds of Hz) allowing much more data to be collected from bigger samples and from a larger corpus (Esteve et al. 2017). In this work, a spatial resolution of 300 μm and a dwell time of 30 ms were chosen. This spatial resolution was adopted to allow the analysis of large surfaces within a reasonable time compatible with the available beam time and the number of samples to be analyzed.

A major advantage of this setup is that no sampling or sample preparation is required. The samples are directly positioned on the beam trajectory with an orientation of about 25°. All of the measurements were performed using an incident beam energy of 18 keV. XRF spectra were recorded with a four-element silicon drift detector (SDD, Vortex-ME4®, Hitachi, with a total active area of 120 mm^2-200 mm^2), placed in the same horizontal plane at 90° from the beam to minimize the X-ray scattering contribution.

All XRF experiments were performed at room temperature and atmospheric pressure (Ar K lines from ambient air are visible in the XRF spectra) blocking. Due to air absorption, the sensitivity to the detection of fluorescence from light chemical elements such as Si, P, S, etc. is limited.

Analysis of each recorded XRF spectrum was performed using the PyMCA software suite (Solé et al. 2007). Energy calibration and a fitting procedure of the fluorescence lines for each spectrum allowed us to identify and evaluate semiquantitatively the contribution of each element. The images of elemental distribution correspond to the plotting of the calculated area of the main fluorescence lines fitted by Gaussian shapes.

Fig. 2: Experimental tests for the use of stone tools in metallurgical processes with summary of the macro and micro use-wear traces observed: (a) copper ore crushing with stone anvil and crusher on the quartzite crushing tool; (b) bronze axe hammering on the dolerite hammer; (c) bronze axe abrading on the sandstone abrader (experiments and photo: C. Hamon, H. Gandois, A. Burlot; microphotos: C. Hamon).

The detection of trace elements must take into account the inherent limitations of the method: the possible overlapping of the Sn L peak (3.44 keV) and the major Ca Kα peak (3.6 keV) and the possible overlapping of the Ag peak and the Ar Kα peak (2.96 keV). For this reason, Sn and Ag peaks are difficult, if not impossible, to detect when quantities of residues and preserved signals are very low, and at the quite high energy required for the detection of the Cu and Zn peaks. Metallic trace elements of Pb residues were also sought. The mapping of the complete surface of the stone tools allows us to isolate elements involved in rock composition on unused areas and elements composing residues related to the processed matter. Black and white images clearly highlight the elemental distribution, especially that of copper residues.

For each area imaged, the same data interpretation process was followed. Whole maps and distributions of main elements are observed and described in light of the areas pinpointed by use-wear analysis. Then, a merging of the XRF spectra is carried out on selected zones, i.e., on areas representative of the original raw material on the one hand and on zones showing clear use-wear traces according to the microscopic analysis on the other. This is done so as to highlight the chemical contrasts between the rock itself and possible residues arising from the use of the artifact. When evidence for copper working activities is being sought, then a particular focus is placed on the intensity of Cu Kα peak. Comparison of spectra from the rock and working zones allows us to confirm the intensity difference of the Cu Kα peaks and thus to confirm the presence of Cu linked specifically to the use-wear traces.

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Results

The analyzed tools correspond to a restricted spectrum, in terms of action modes as well as activities represented. Among the stone tool assemblage, the combination of particular raw materials and specific macroscopic use-wear traces on three tools led us to interpret them as metallurgical tools which were used with different kinematics and actions on the material.

The first (tool n°13), a microgranite cobble, broken lengthwise, presents numerous zones of percussion on several of its faces (Fig. 3). Both ends of the artifact display fine percussion zones on beveled flattened surfaces. Two percussion cupules occupy the center of face 1, while the opposite face, face 2, features a single percussion cupule. They are surrounded by highly shiny areas. Use-wear analysis has shown that the cupules correspond to a series of coarse and transversal and deep linear percussion impacts. The peripheries of the cupules present concentrated, close, smooth leveling with shiny aspect, together with covering, oblique, parallel striations that attest to the grinding of hard mineral material in fine fractions (fragments, powder) (Fig. 3a). An open, smoothed, and rough micropolish, with strong metalized shine, is visible on these surfaces. The main cupule (Fig. 3b) associates fine percussion impacts and covering and smooth leveling on its low topography. Besides the three shiny zones localized between the percussion zones and the sides of the tool, covering, surface, smooth leveling at the bottom of the cupules along with fine percussion impacts, further demonstrate the simultaneity of crushing and grinding actions. The sides of the tool exhibit covering, close, smooth leveling associated with short individual striations on the faces, all orientated following a transverse

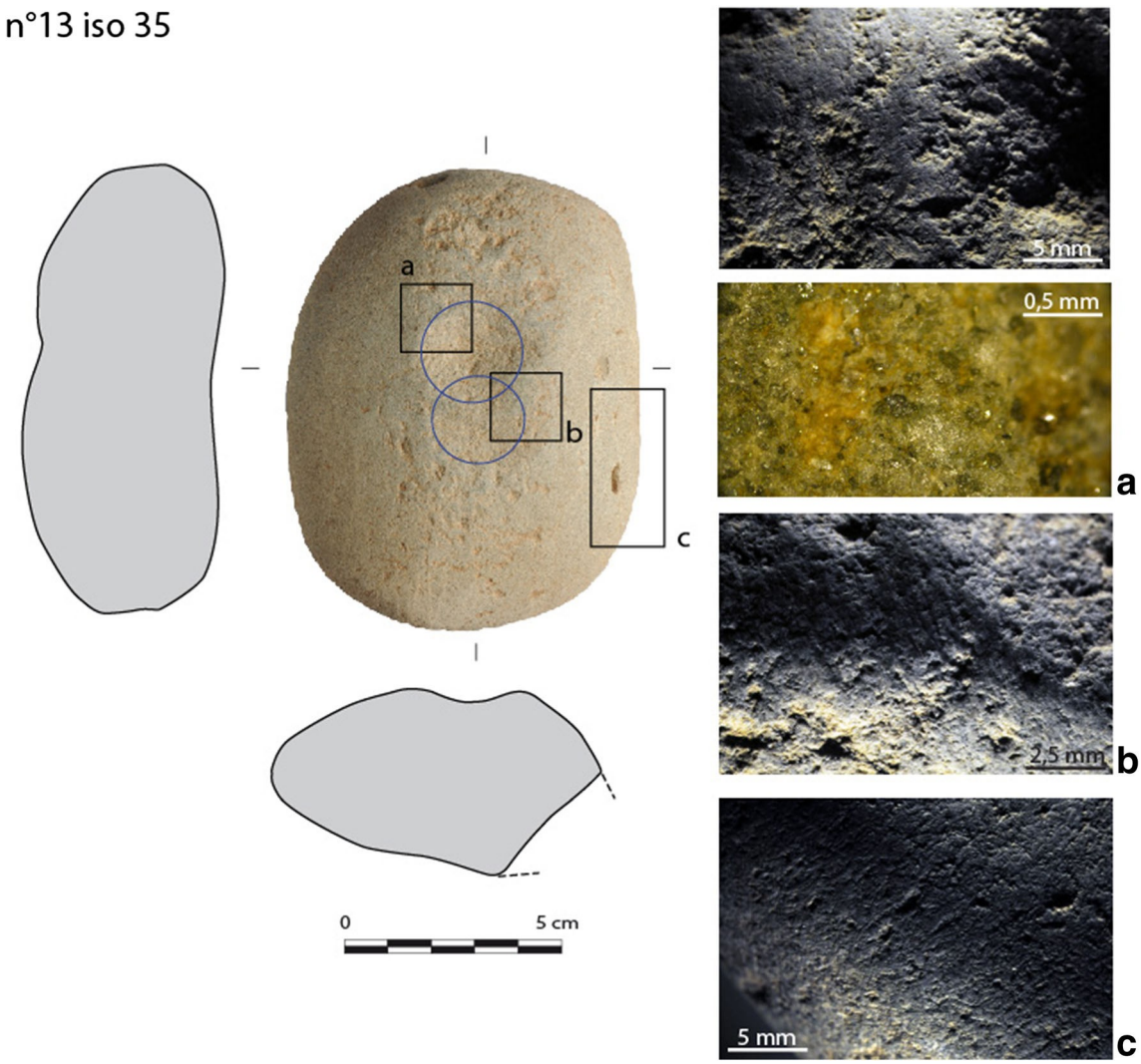

Fig. 3: Ploneour-Lanvern «Kersulec», crushing tool on microgranite cobble, n°13; in blue, localization of two cupules on face 1: (a) interface between cupule 1 (with percussion impacts) on the right and reflexive smooth leveled zone on its periphery (with detailed of microtraces); (b) interface between cupule 2 (with percussion tracesl on the left) and microstriated and leveled zone on its periphery (in the center); (c) macrotraces of tool sides (photo: E. Collado, drawings V. Brisotto, microphotos: C. Hamon).

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gesture (Fig. 3c). Based on this association of traces, we can say that this tool would have been used alternately for reducing hard mineral matter - using its ends and the center of its face - and then for finer grinding of the mineral matter in smaller fraction. For this tool, XRF maps were acquired in order to analyze the two faces bearing visible cupules. The distribution of the main chemical elements is displayed in Figs. 4 and 5. These results clearly show that Fe, Sr, Ca, Rb, and Ti are constitutive elements of the rock itself. Edges of higher intensity are visible on the Ca and Sr maps, and these two elements are collocated. Two other elements (Ti, Rb) seem related to the distribution of some specific minerals composing the rock: Ti appears as small spots or stars and Rb as diffuse stains. The distribution of these five different elements does not show any correspondence with the distribution of use-wear traces and seems more related to the structure of the rock itself. Nevertheless, the XRF map draws attention to two other specific elements distributed on a specific zone (Fig. 4): Cu and Zn are collocated on the periphery of the cupule where smooth leveling has been detected. Furthermore, the XRF spectra show a difference between the measurements for these specific zones and the rock itself, where Cu is clearly less present (Fig. 6). Cu is distributed mostly as very small dots at the periphery of the cupules and not over their entire periphery.

The second tool (tool 15; Fig. 7) was made from a mesial fragment of a polished eclogite ax. Trapezoidal in shape, the object displays use-wear on its ends and sides. The microscopic observations indicate that the faces show neither distortion of their morphology nor use-wear traces: the uniform polishing, as well as the shiny coating, is solely the result of the final shaping and polishing of the initial ax. Both ends were used in the same manner: their plano-convex surfaces show a covered close smoothing associated at its periphery with a group of short oblique to transverse striations (Fig. 7a right). The peripheral edges of these surfaces display overlapping close, smooth leveling, with shiny aspect, as well as covered, short, transversal, and parallel microstriations, generally associated with longitudinal facets (Fig. 7a left). One of these facets, on one side, bears clear longitudinal striations. Although end 1 does not display visible micropolish, end 2 bears considerably bright, semi-closed, rough, and micropitted micropolishes in places (Fig. 7b). Edges and facets on the ends and sides of the tool constitute the most solicited zones of use, consistent with longitudinal abrasive actions on flat metallic surfaces (Fig. 7c). No traces of percussion have been observed. A similar combination of traces has already been reported by Delgado-Raack and Risch (2008:239), which confirms that the tool probably functioned as a hammer. XRF maps were acquired for both ends of the artifact (Figs. 8 and 9). They present a very particular elemental distribution: Fe, Mn, Ti, and Ca are most likely related to the rock composition of the tool. Ca and Ti are quite homogeneously distributed within the volcanic paste, while Fe and Mn show a more "granular" appearance in relation to specific minerals. On the two ends of this tool, the XRF Kα lines of Fe, Mn, and Ti are less intense, and their distribution seems to be the exact inverse of that observed on the polished zone, suggesting that these elements relate instead to the rock itself (Fig. 10). In contrast, this polished zone seems to be enriched by Cu and Zn. Despite the low quantity measured, this particularity is noteworthy as the distribution of Cu and Zn is clearly correlated with the polished facets visible on end (2).

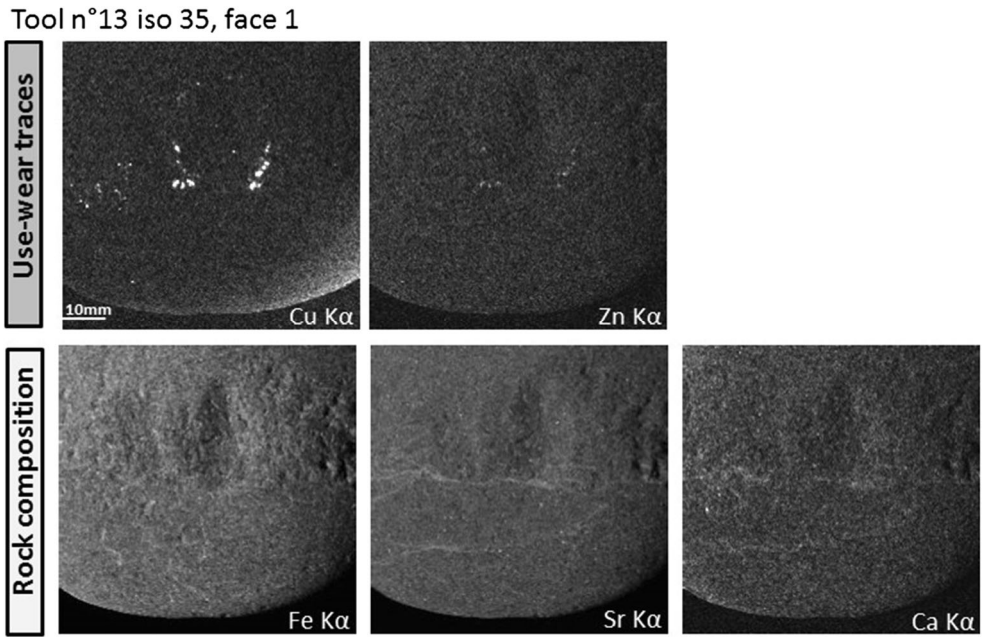

Fig. 4: Tool n°13, face 1. XRF elemental maps at 18 KeV with acquisition step of 300 µm, showing distribution of use-wear and major rock elements.

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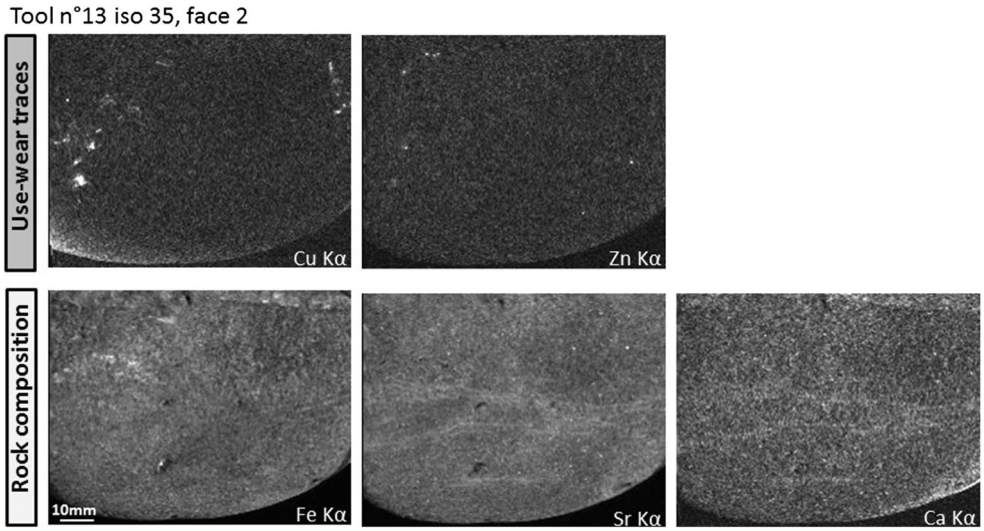

Fig. 5: Tool n°13, face 2. XRF elemental maps at 18 KeV with acquisition step of 300 µm, showing distribution of use-wear and major rock elements.

The third implement is made out of a small ovoid cobble of diorite whose shape has been completely modified by use (tool 18; Fig. 11). Intensively used, its ends and sides show facets of percussion, and there are two percussion cupules on each of its faces. The use-wear analysis highlighted four groups of working zones on the tool. The two pointed ends show covering smooth leveling with a transversal orientation, a high degree of leveling of the surface, important

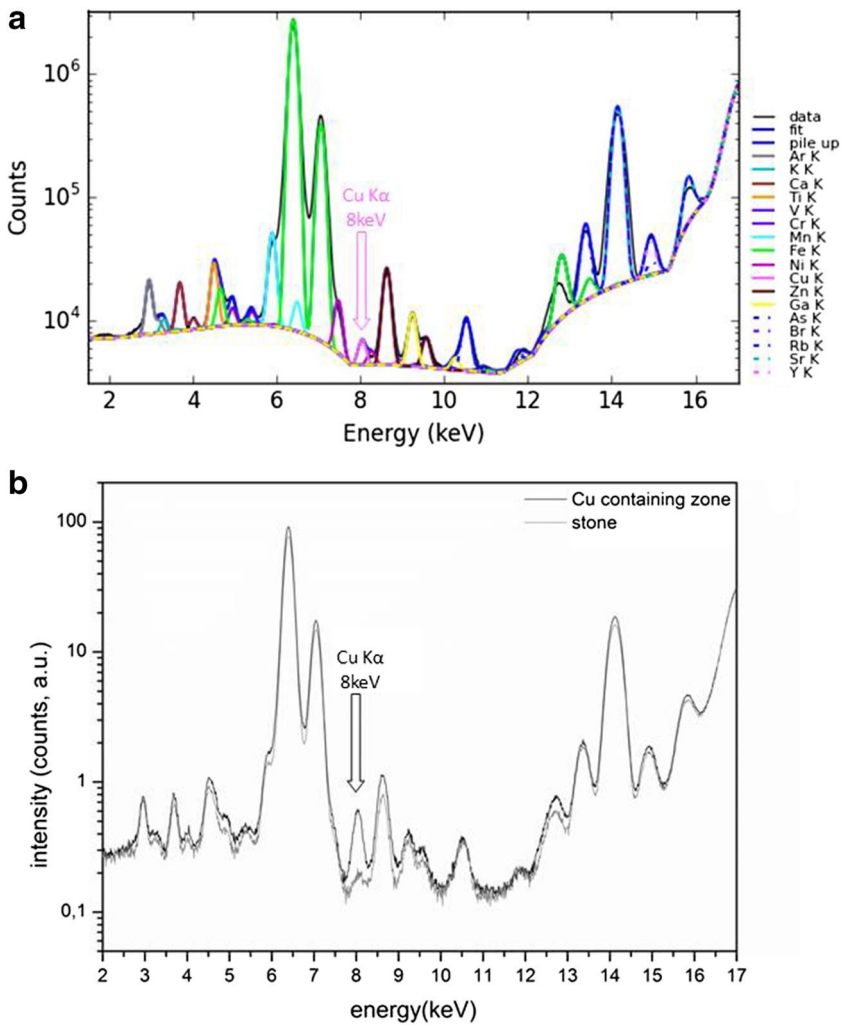

Fig. 6: Tool n°13, XRF spectra - 18 KeV; (a) fit from PyMCA to deconvolute the contribution of the different elements; (b) comparison of spectra from used and non-used (natural) surfaces of the tool.

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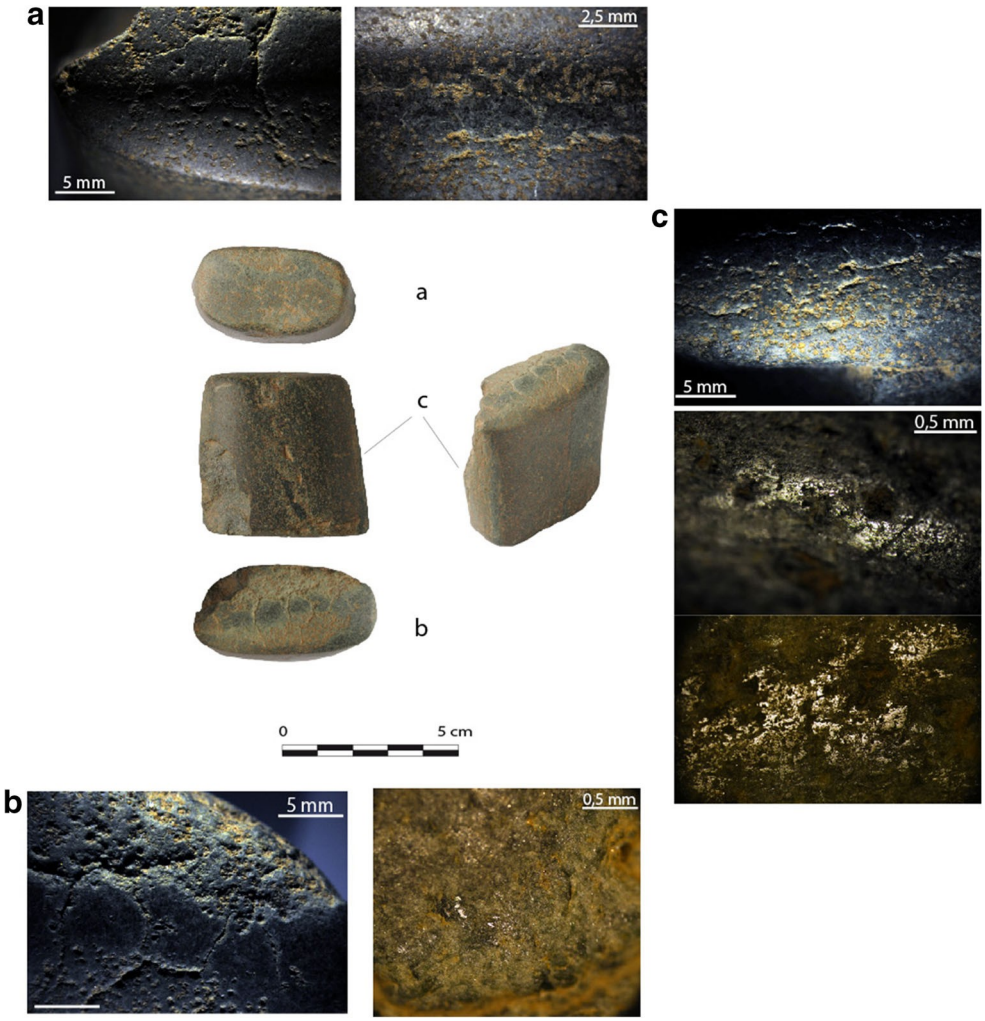

Fig. 7: Ploneour-Lanvern «Kersulec», ax fragment in eclogite reused as hammering and polishing tool, n°15 (a) macrotraces on end 1 (left, micropolish visible on the edges; right, transversal microstriations); (b) macro- and microtraces on end 2 (with characteristic micropolish); (c) macro- and microtraces on side 1 (with characteristic micropolish) (photo: E. Collado, drawing Y. Pailler, microphotos: C. Hamon).

microbreakage of the grains, and a bright, semi-closed, rough, and micropitted micropolish (Fig. 11d). Three facets, directly related to the smoothed ends, show evidence of fine percussion: the impacts display very regular distribution and diameters together with considerable rounding of the grains (Fig. 11a-c and e). Four narrower, circular zones occupy one side of the tool in particular: they bear coarser, lozenge-shaped impacts. The two opposite faces exhibit impacts or cupules created by strong percussion (Fig. 11f).

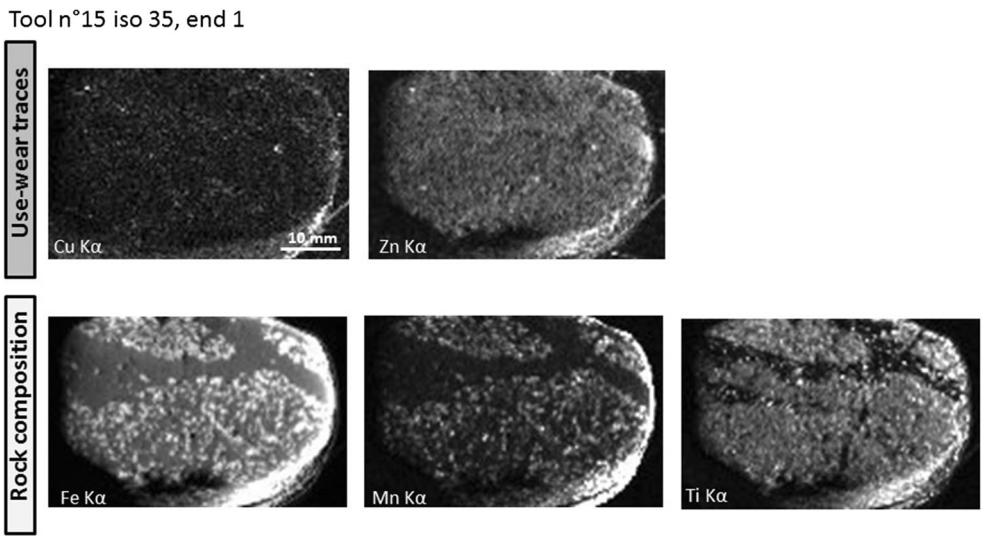

Fig. 8: Tool n°15, end 1. XRF elemental maps at 18 KeV with acquisition step of 300 µm, showing distribution of use-wear and major rock elements.

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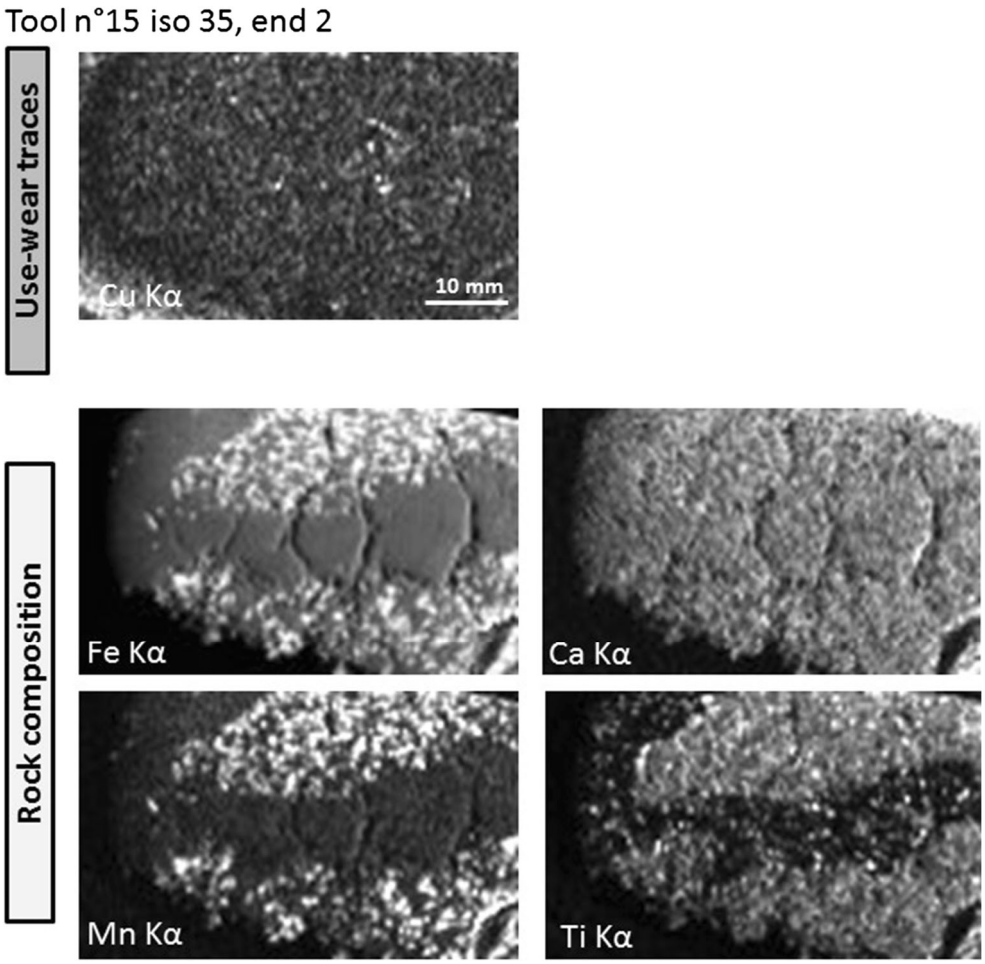

Fig. 9: Tool n°15, end 2. XRF elemental maps at 18 KeV with acquisition step of 300 µm, showing distribution of use-wear and major rock elements.

In between these zones, natural areas present covering, smooth leveling, and random percussion impacts, which attest to the simultaneous, rather than successive, use of these zones. Each of these four zones is related to a specific action mode on the material being transformed: percussion of the material by strong impacts on the faces and sides, regularization by fine percussion on oblique areas, and finishing by abrasion with the ends. All of these use-wear traces are compatible with the shaping of metallic objects, whereby percussion and abrasion were carried out alternately. An XRF map was acquired in order to reveal the distribution of elements on face (1) of the tool, where a cupule is visible. The distributions of the main elements are presented in Fig. 12. The XRF maps clearly show that Fe, Ti, Ca, and Sr are constitutive elements of the rock itself. In general, these elements are homogenously distributed; Ti has a more granular aspect. Two Cu spots correlated with Zn that are visible on each side of the cupule are of particular interest. Their localizations correspond to that of a well-developed, shiny, smooth leveling. This presence of Cu is underlined by the XRF spectra in Fig. 13 which highlight the difference in the intensity of the Cu Kα peak between the spots containing Cu and the rock itself.

These three tools share several notable features. First, they are all made of hard, cohesive, heat-resistant raw materials that are particularly suitable for the treatment of ores or metallic items. Secondly, they have all been intensively used as indicated by the significant deformation of the morphology of several of their faces. The macroscopic traces clearly suggest diversity in the handling of these tools, with evidence for crushing, shaping, and abrading actions. The microscopic use-wear analysis points to the processing of a semi-hard mineral material, and comparisons with the experimental data suggest that this material was metallic. The elemental XRF imaging underlines a correlation between the localization of these use-wear traces and the distinctive XRF lines of copper and zinc, two elements known to be involved in copper metallurgy and in bronze alloys. However, it remains difficult to distinguish whether these elements are the residues of copper production (with Zn possibly present naturally) or whether they are residues of bronze alloy production. Despite the spectral interferences that prevent the detection of tin traces, the processing of copper ore and copper-based alloys (i.e., bronze) is demonstrated. The combination of these four levels of analysis allows us to propose the following interpretation of the functions of these stone tools: one crushing-grinding tool

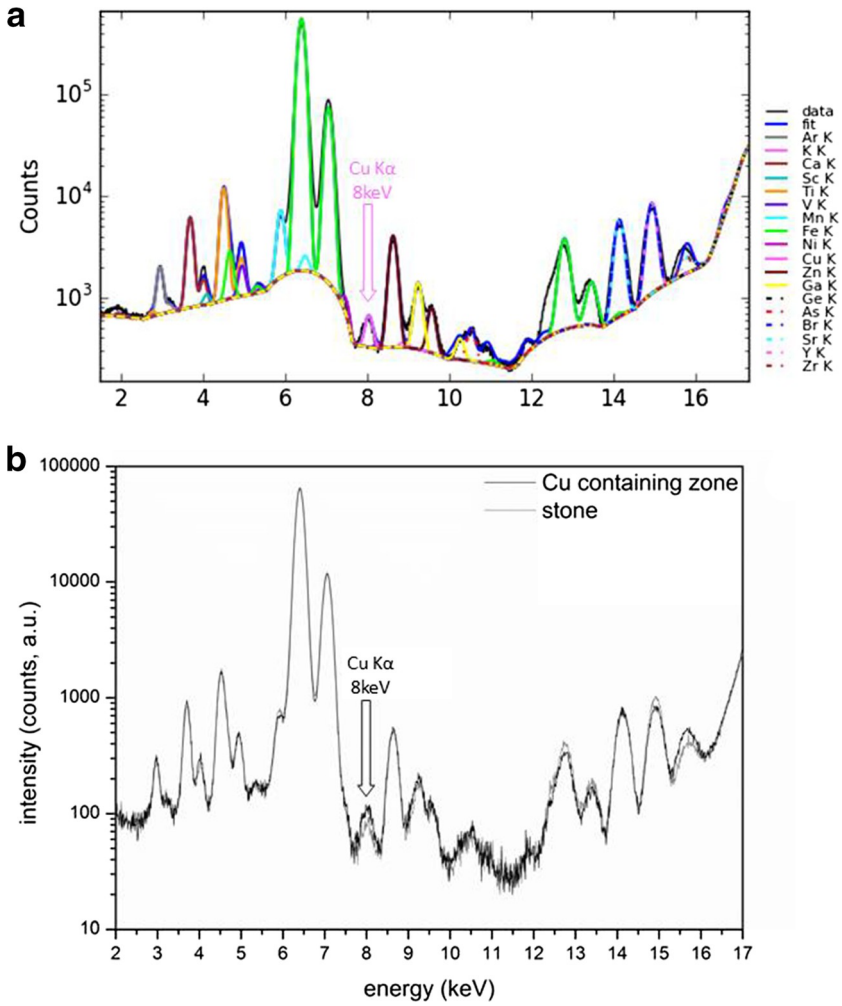

Fig. 10: Tool n°15, XRF spectra - 18 KeV (a) fit from PyMCA to deconvolute the contribution of the different elements; (b) comparison of spectra from used and non-used (natural) surfaces of the tool.

(n°13) used for the processing of copper ore, a small hammering and polishing tool (n°15) used for the shaping of metallic objects, and one tool (n°18) used for fine percussion and abrasion in the manufacture of metallic objects.

Discussion

A comprehensive methodology based on microscopic use-wear analysis and XRF imaging for the detection of stone tools used for metallurgical activities

The results clearly demonstrate the very close association between copper residues and metallic micropolishes on the working faces of the three stone tools selected. Consequently, a number of points can be made regarding the methodology.

In view of previous published tests (Hamon et al. 2009; Delgado-Raack et al. 2014), the amount of metallic residues detectable using XRF measurements was expected to be particularly low. Several combined taphonomic and alteration processes can explain the generally poor preservation of copper and bronze residues on the surfaces of stone tools. In particular, this can be due to their high tendency to migrate into the soil or surroundings objects. For this reason, copper and bronze residues are generally difficult to identify on archeological artifacts using standard technologies, such as portable XRF techniques, even XRF measurements based on the previous optical detection of localized macroscopic residues. The use of the XRF raster map technology on the DiffAbs beamline of Soleil Synchrotron offered several advantages for optimizing the detection of copper residues. First, the ability to choose the most suitable X-ray microbeam energy allows the measurement settings to be adjusted to achieve the best possible level of detection. Secondly, the use of the continuous *Flyscan* mode makes it possible to analyze and map the complete working surface of the tools and to get comparable elemental spectra on a large grid including natural surfaces and use-wear surfaces. The common distribution of micropolishes (characterized by microscopic analysis) and copper residues (highlighted by the XRF Flyscan imaging) appears to exclude the possibility that these phenomena are the result of external contamination or microvariations in the mineral composition of the rock itself.

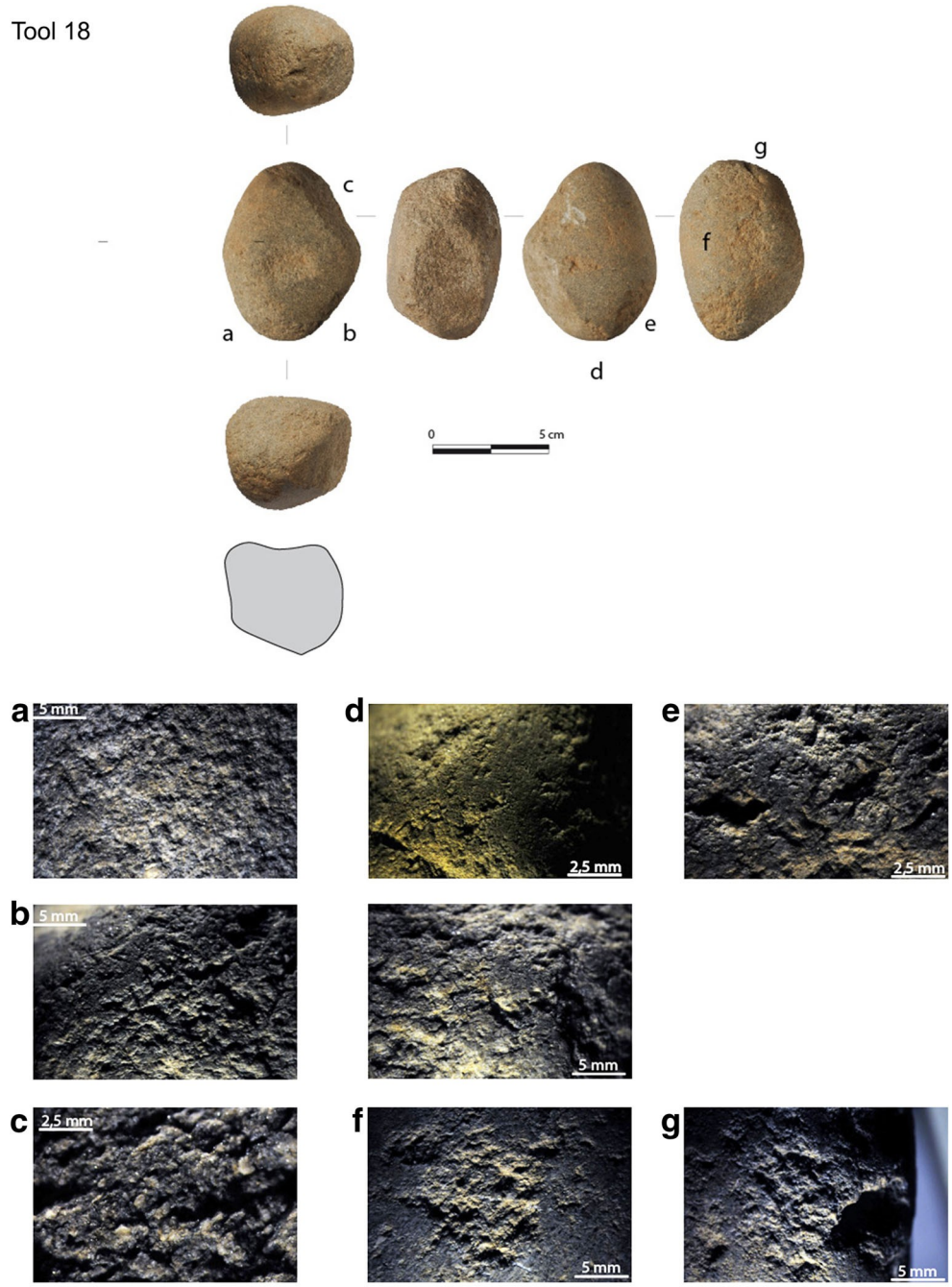

Fig. 11: Plonéour-Lanvern «Kersulec», small multifaceted hammering and crushing tool on a diorite cobble, n°18: (a-c and e) small percussion impacts on some facets in the ends; (d and g) combined smooth leveling and fine percussion on the ends; (f) percussion impacts in main cupule on face 1 (photo: E. Collado, drawing V. Brisotto, microphotos: C. Hamon).

Despite the tiny amounts of copper detected, their co-distribution with micropolishes definitely links their presence to the use of the tools for metallurgical operations. This common distribution suggests that the copper residues were trapped in the interstices of the rock and then covered by the micropolish or that the copper micro-residues were included within the micropolish itself during its formation. Our integrated analysis has clearly shown that only some of the areas bearing use-wear traces also display copper residues. In particular, the cupules did not contain any residues of copper although the location of these percussion impacts at the center of the main area of micropolish and copper micro-residues indicates that they were almost certainly used for the crushing of ore. For this reason, our observations would suggest that good preservation of the micropolishes has favored the good preservation of the copper residues and that micropolish formation integrates - at least partly - micro-residues of the transformed material. This assertion is entirely in line with previous observations that highlighted the presence of cereal phytoliths within the micropolishes of flint sickles (Anderson-Gerfaud 1992) or of apatite residues incorporated within micropolishes observed on bone sharpeners (Christensen and Valla 1999). Our results support the idea of a common origin for the two kinds of traces, micropolishes, and residues, linked to the purpose for which the tool was used.

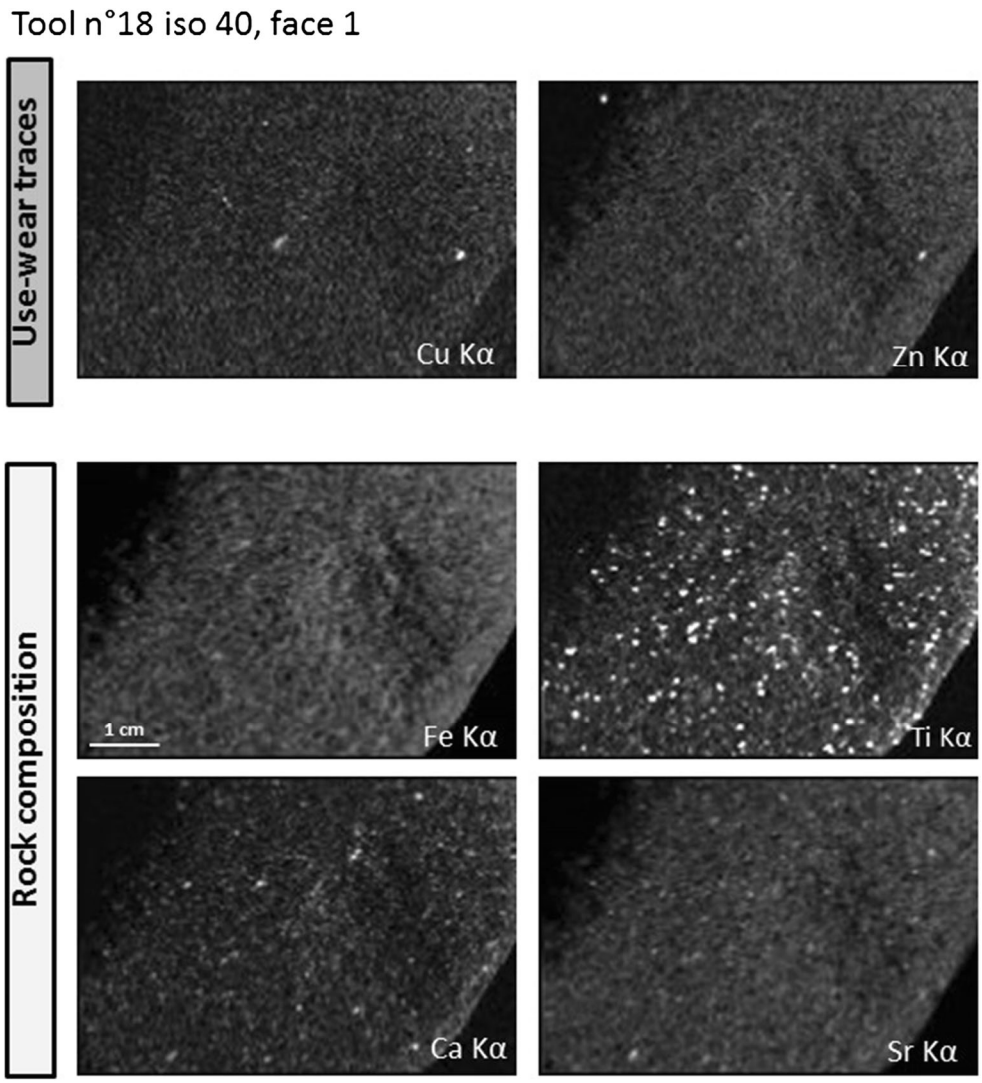

Fig. 12: Tool n°18, face 1. XRF elemental maps at 18 KeV with acquisition step of 300 µm, showing distribution of use-wear and major rock elements.

In this sense, our observations provide new evidence for discussing the nature and mechanisms at the origin of the formation of micropolishes, which was the subject of robust exchanges between use-wear analysis specialists in the 1980s and 1990s (Anderson-Gerfaud 1986; Christensen 1998; Unger-Hamilton 1988; Yamada 1993).

A bronze metallurgist's toolkit in Kersulec

The comprehensive techno-functional approach developed for the macrolithic tool assemblage has allowed us to identify new evidence for the existence of a metallurgical workshop, which up to now was only suspected on the basis of the presence of a single crucible fragment. It also demonstrates the diversity of the types of tools and actions involved in the treatment of metallic objects. Crushing, hammering, abrading, and sharpening of tools all occurred on the Kersulec settlement. The large spectrum of actions represented suggests the use of stone tools at different stages of the *chaine operatoire* of metal processing (Fig. 2). These observations fit with previous observations and use-wear analyses conducted on different types of metal objects (axes, daggers, points) produced during the Copper and Bronze Ages in western and southern Europe (Dolfini 2011; Gutiérrez Sáez et al. 2010; Gutiérrez Sáez and Martín Lerma 2015; Quilliec 2007). These studies reveal that hammering, abrasion, and polishing actions were an integral part of the shaping and sharpening of metal objects prior to their use and deposition.

In Kersulec, crushing tools were used in ore processing and in the reduction of ore into powder before casting. Hammers were used for the shaping of metallic objects by plastic deformation, especially for the thinning and elongation of metal sheets. Finally, abraders and polishers were used to regularize the surface of metal objects up to their finishing. Hence, despite the relatively low number of tools potentially associated with the metallurgical process, all stages of the production of metal objects are represented on the Early Bronze Age settlement of Kersulec.

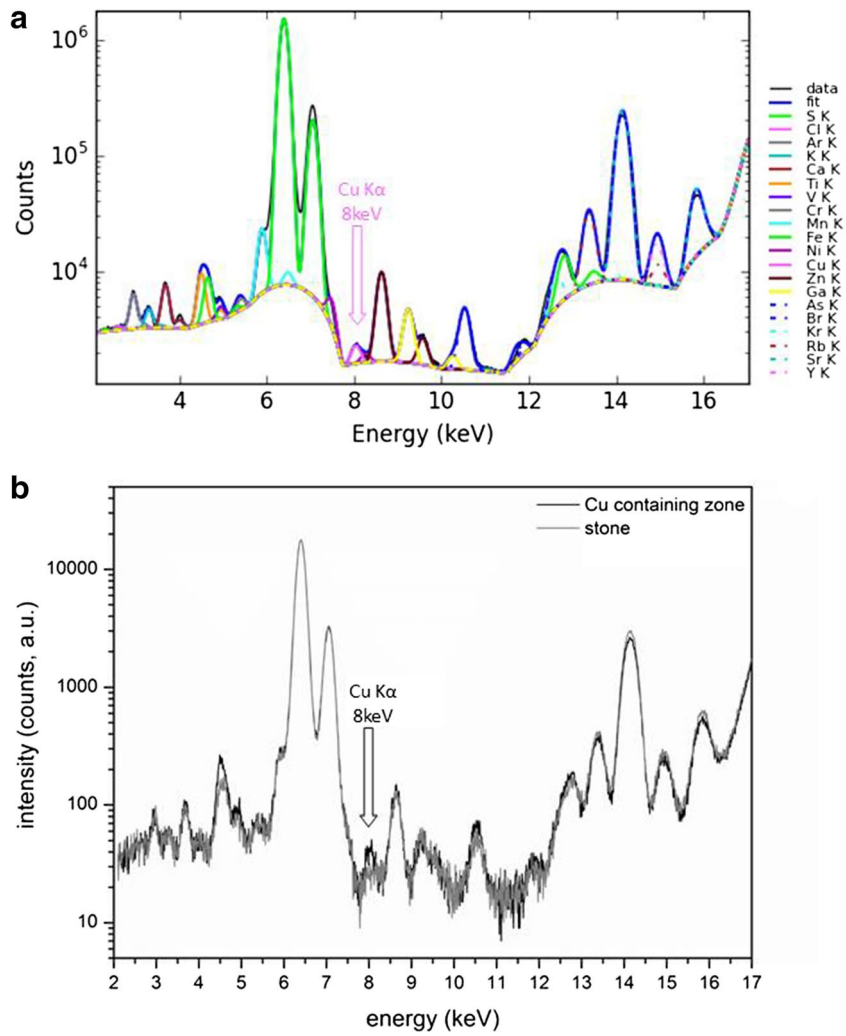

Fig. 13: Tool n°18 face 1, XRF spectra - 18 KeV (a) fit from PyMCA to deconvolute the contribution of the different elements; (b) comparison of spectra from used and non-used (natural) surfaces of the tool.

These tools must have been used alongside metal and wooden tools, as has been suggested in various other archeological contexts (Thevenot 1998).

Furthermore, these tools were all manufactured from dense, fine-grained rocks, specifically chosen for their hardness and resistance to impacts and to the effects of heat. The blanks selected were generally cobbles, offering naturally homogeneous surfaces, a low internal fissuring rate and smooth edges. The crushing tools are also of quite small dimensions, which would suggest precision crushing activities on small quantities of precious metallic compounds. It is also remarkable that the recycled Neolithic ax is probably related to the metallurgical activity. These choices indicate that raw materials with very particular mechanical properties were being deliberately selected for finishing operations requiring high precision. The Kersulec tools attest to the intensive use of all faces of the tools and an optimization of particular blanks. This is evident in the combination of several action modes (up to three) on one single tool and by the intensive use of almost all of the available faces of the tools.

Implications for the emergence of metallurgy in Northwestern Europe

The identification of crushing tools used for copper ore processing in Kersulec attests to metal production in Brittany, a question that has long been the subject of intense debate between specialists in the Bronze Age of Western Europe.

Based on the evidence for copper-based metallurgy in Kersulec, which is one of very few sites to produce such evidence, it would seem that, in Brittany, this early metallurgy was not organized around structured workshops or specialized sites. Apart from mining contexts, it is generally difficult to identify these metallurgical sites as they do not involve complex organization, complicated logistics, or structured production areas: a hearth, some charcoal, a handful of stone tools, and some ore, which generally disappears after it has been smelted, are the only remains that can be expected from this activity.

Kersulec perfectly illustrates this situation, and the fact that that the metalurgists' stone tools were not found in a dedicated area, or within a well-organized workshop, is not exceptional for the period. Indeed, the low visibility of this activity on the Early Bronze Age sites of Brittany and its association with domestic settlements suggest that such workshops existed in different kinds of contexts: open-air settlements, domestic contexts, enclosures, etc. Such an organization of the production clearly contradicts the notion of specialized sites dedicated to metal production.

Although the very discrete copper outcrops in Brittany have not yet revealed any clear evidence for extraction, this discovery of copper ore processing tools in Kersulec fuels the hypothesis of widespread small-scale copper or bronze production workshops in the region. This small-scale production at local level would fit with the exploitation of the small copper outcrops known in Brittany, especially in Finistère. The possible exploitation of these copper deposits has long been a subject of debate because of the relatively poor quality and low volumes of the ores present and their limited accessibility (Briard 1984, p. 93). This production could have been controlled by the elites buried in the Armorican Tumuli, elites who would have contributed to the development of an economy of prestige goods based on privileged exchanges with the British Isles, the Channel, and the North Sea coasts between 2150 and 1950 BC. As in many other contemporaneous funerary contexts, the deposition of stone tools related to metallurgy highlights the particular status of metallurgical crafts and of the people who mastered these technologies (Nicolas et al. 2015; Turek 2004).

Conclusion

Our identification of a metallurgical stone tool kit through use-wear analysis conducted on objects from the Early Bronze age site of Kersulec appears to represent a promising and challenging new approach to understanding the earliest copper and bronze working in the area. Our comprehensive analysis is based on the best combination of techniques so as to limit the biases intrinsic to each individual methodology. It allows us to propose highly reliable functional interpretations that could not have been reached by applying each method - use-wear and synchrotron micro-XRF analysis - separately. It underlines the potential of macrolithic tool use-wear analysis to reveal the existence of metal production on different types of archeological sites where structures associated with metallurgical production are barely discernible or difficult to recognize and where metallic objects are rare due to intensive recycling and diffusion. Although copper and bronze residues remain difficult to detect due to taphonomic factors, the multiplication of use-wear studies on macrolithic tools in Copper and Bronze Age contexts completes our knowledge of the *chaine operatoire* for the production of metal objects.

By allowing us to identify the metallurgist's stone tools and the different stages in the production of metal objects, this approach offers us the possibility to study in detail the techniques and processes respectively involved in the various regional productions of metal objects. It also offers us an opportunity to identify the contexts in which copper and bronze were produced, those who mastered these crafts and those who controlled the production, and thus it can help us to understand the major changes that occurred in the Early Bronze Age societies of Europe at the turn of the second millennium BC.

Acknowledgments - We acknowledge Soleil for providing access to their synchrotron radiation facilities, and we would like to thank the whole team for their assistance in using the "DiffAbs" beamline. The experimental tests have been undertaken in collaboration with Henri Gandois (UMR 8215, Trajectoires) with the participation Aurélien Burlot; a great thanks to them for accepting us to use photos and results.

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