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Gurova & Bonsall – Highlights:

- Prehistoric communities in northern Britain and Ireland used exotic pitchstone from sources in western Scotland
- Exchange networks persisted for millennia
- Non-destructive pXRF analysis shows that archaeological pitchstone derives from multiple geological sources
- Determining the function of pitchstone artefacts poses a challenge for conventional use-wear analysis

Author statement

Maria Gurova, Clive Bonsall: Conceptualization, Methodology, Data acquisition and curation, Writing-Original draft preparation, Reviewing and Editing.

Use-wear analyses and provenance determination of pitchstone artefacts: a pilot study from western Scotland

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ABSTRACT

Pitchstone (a volcanic glass similar to obsidian) from geological outcrops on the west coast of Scotland circulated widely among Neolithic communities in northern Britain and Ireland, representing an exchange network that in its areal extent rivalled those that developed around obsidian sources in continental Europe and the Mediterranean. While the archaeological distribution of pitchstone within the British Isles is now well documented, rather less is known about which sources were used or the functions of pitchstone tools found in archaeological contexts.

Here we report the results of a pilot study, the main objectives of which were to: (1) assess if we could discriminate between pitchstone sources and assign archaeological finds to sources using non-destructive chemical analysis, and (2) determine how use-wear manifests itself on pitchstone artefacts.

We undertook pXRF analyses of pitchstone samples from two geological outcrops on the Isle of Arran and five archaeological assemblages from Neolithic sites on Arran and mainland Scotland. We also conducted analyses of wear patterns on pitchstone artefacts from all five archaeological assemblages. To aid interpretation of the wear patterns observed on archaeological pieces, experiments were conducted in which natural pitchstone flakes and replicated artefacts were used in various tasks (processing of dry and fresh wood, pig hide, reed, and fresh meat and bones) followed by microscopic examination to assess the kinds of use-wear produced.

The results of the pXRF analyses suggest that the pitchstone found at each of the archaeological sites examined was obtained from more than one source. The results of the use-wear analyses were equivocal. Microfractures and striations were identified on both archaeological and experimental artefacts. But it proved difficult to distinguish taphonomic from use damage on the archaeological pieces.

Keywords: pitchstone; Scotland; Neolithic; experiments; use-wear analysis; portable X-ray fluorescence (pXRF)

1. Introduction

Pitchstone is a glassy acidic igneous rock formed by the rapid cooling of lava or magma. It resembles obsidian in its chemical composition but differs in having a higher water content (typically 5–10% by weight), a duller (waxy) lustre and, often, an irregular or hackly fracture.

Within the British Isles, pitchstone occurs in a zone encompassing the Inner Hebrides and adjacent Scottish and Irish mainland that witnessed intense igneous activity during the Palaeocene and early Eocene c. 64–52 Ma, known as British Tertiary Igneous Province (BTIP) (Emeleus and Gyopari 1992; **Fig. 1a**). Pitchstones are found in all the major igneous centres of the BTIP in the form of sills, dykes and lava flows. They vary in texture, from virtually aphyric to highly porphyritic, and in mineral chemistry.

Like obsidian, pitchstone was used in prehistory for artefact production. In a landmark study, Williams Thorpe and Thorpe (1984) recorded over a hundred archaeological sites with pitchstone artefacts in Scotland and northern England and attempted to identify the geological provenance of the pitchstone by elemental analysis of samples from 13 source outcrops within the BTIP and 22 archaeological sites, supported by petrographic studies. From this, they concluded that all archaeological pitchstone originated from sources on the Isle of Arran and had been ‘traded’ across northern Britain throughout the period from the Mesolithic to the Bronze Age.

Torben Ballin conducted an extensive review of the archaeological evidence for pitchstone use in northern Britain (Ballin 2009, 2013, 2015; see also Ballin and Faithfull, 2009). He increased the number of archaeological sites with pitchstone to c. 350. He concluded that: (i) Mesolithic use of pitchstone was probably confined to Arran¹; (ii) during the Neolithic pitchstone was distributed across northern Britain via a wide-ranging exchange network reaching as far north as Orkney and as far south as Cumbria; (iii) Early Neolithic pitchstone artefacts are predominantly aphyric, while both aphyric and porphyritic varieties were used in the Late Neolithic; and (iv) the distribution became more restricted in the Early Bronze Age, possibly reflecting significant social change.

Two questions left largely unresolved by these previous studies are:

1. Which source outcrops on the Isle of Arran supplied the pitchstones that circulated among Neolithic communities in northern Britain?
2. How were the pitchstone artefacts found in Neolithic sites used – did pitchstone fulfil a role that was primarily ‘functional’ or ‘symbolic’ (cf. Ballin 2013)?

To begin to address these questions, we devised a low-cost pilot project with three broad objectives. Could we: a) discriminate between pitchstone sources and assign archaeological finds to sources using non-destructive pXRF analysis; b) determine how use-wear manifests itself on pitchstone artefacts; and c) from the wear patterns observed, identify the function of individual artefacts.

2. Materials and methods

2.1. Sample selection

Nearly a hundred individual pitchstone outcrops have been recorded on Arran (Ballin and Faithfull 2009). During a two-day visit to Arran in April 2017, we collected pitchstone samples from three locations for pXRF analysis and use-wear experiments. These were Clauchland (Corrygills) shore (55°33′28.50″N, 5°05′30.62″W), Clauchland hills

¹ In a subsequent publication Ballin reported the first evidence of Mesolithic use of pitchstone from mainland Scotland (Ballin et al. 2018, 3–4).

(55°33'25.02" N, 5°07'47.38"W) and south of King's Cave (55° 31' 32.87" N, 5° 21' 20.09" W) (**Figs. 1b & 2**).

At Clauchland shore on the east coast of Arran, we collected angular fragments of pitchstone broken from blocks and boulders on the slope and foreshore below the "The Great Pitchstone" sill, which crops out in the raised beach cliff (**Figs. 2a-d**). Ballin and Faithfull (2009, 19) described this pitchstone as, "*generally aphyric but [with] a few well-shaped crystals of quartz ... dark bottle-green*".

The Clauchland Hills 'outcrop' consists of an area where small pitchstone pebbles have been exposed by footpath erosion (**Fig. 2e**). Ballin and Faithfull (2009, 19) describe this variety as "*... from a prehistoric knapper's point of view – beyond doubt the best-quality pitchstone to be found on [Arran], being absolutely homogeneous, and resembling obsidian greatly. Aphyric, almost black*".

To the south of King's Cave on the west coast of Arran "*five separate exposures of pitchstone occur on the shore and in the cliff bounding the raised beach*" (Ballin, Faithfull 2009, 27). Pitchstone blocks deriving from these exposures are abundant on the foreshore and provided samples for our pilot study. This pitchstone variety is dark green and highly porphyritic (**Fig. 2f**).

The archaeological samples included in our study came from two localities on Arran – Allt Lebnaskey and Whiting Bay, and two localities in mainland southwest Scotland – Auchategan (Argyll) and Luce Bay (Dumfries and Galloway) (**Fig. 1b, Table 1**). The finds from all four localities are thought to date to the Neolithic (c. 4000–2500 cal BC):

- Among the material examined from Luce Bay were artefact series labelled 'Glenluce' and 'Luce Sands', which were surface finds (possibly from different locations) within the dune systems around the bay (A. Sheridan, pers. comm.). They were part of a much larger collection in the National Museum of Scotland that consisted mainly of blades/bladelets of varying dimensions between 19 and 26 mm in length, 5 and 13 mm in width, and 2 and 7 mm in thickness.
- The lithic assemblage from the later of two Early Neolithic occupations at Auchategan (Marshall, 1978; Ballin, 2006) included over a hundred pitchstone artefacts. The 83 pieces examined in our pilot study were comprised mainly of debitage – blades, flakes and fragments, and 10 single platform bladelet cores.
- The sites at Whiting Bay and Allt Lebnaskey were discovered in surveys by Torben Ballin (Ballin and Faithfull, 2009). Two distinct concentrations of artefacts were identified at Whiting Bay, 75–100 metres apart. *Assemblage 1* included 46 pieces of pitchstone comprising both aphyric and porphyritic varieties. *Assemblage 2* (19 pieces) and the collection from Allt Lebnaskey are made from aphyric pitchstone. From the raw material and typological characteristics, it has been suggested that Whiting Bay 2 dates to the Early Neolithic, and Whiting Bay 1 to the Middle or Late Neolithic (Ballin Smith and Ballin, 2009, 126).

2.2. Experimentation

A limited series of replication experiments were undertaken to test the efficiency of pitchstone artefacts for various activities. The raw material for these experiments was obtained from the outcrop at Clauchlands shore on Arran and comprised six natural flakes with sharp edges plus three flakes and one blade knapped by Prof. Bruce Bradley (**Figs. 3 & 8**) from blocks of material supplied by the authors. Bradley's debitage (hereinafter "BB debitage") comprised mainly flakes and chips, several irregular bladelets and a fine pressure flaked bifacial point (**Fig. 3**). One pebble of porphyritic pitchstone from the foreshore near

King's Cave was also knapped by Bradley (**Fig. 3, no. 3**), but the material is of very poor quality and none of the debitage was used in our experiments. Similarly, the pitchstone pebbles collected from the Clauchland Hills source were not used to produce experimental artefacts.

The 10 experimental tools were used in various tasks that involved the processing of dry and fresh wood, reed, and pig hide, meat and bones, the duration of the experiments ranging from 10–50 minutes (**Table 4**). The purpose of these experiments was to: (1) gauge the efficiency of pitchstone in the performance of the different tasks, and (2) investigate the nature of wear formation on pitchstone.

2.3. *Use-wear analysis and recording*

In total 269 pieces of worked pitchstone from the archaeological sites included in our pilot study were subjected to techno-typological analysis, microscopic observation and detailed photographic documentation. The pieces analyzed included cores, flakes, blades/bladelets, and typological tools comprising burins, end-scrapers, perforators and notched pieces (**Tables 1 & 2**). Each artefact was drawn and photographed before analysis. Microscopic studies of wear traces were conducted under low-power magnification using a zoom stereomicroscope with an external light source. Two different microscope and camera combinations were used. The pitchstone artefacts from Auchategan and Luce Bay are kept at the *National Museum of Scotland's Collections Centre* (NMSCS) in Edinburgh and were studied there using a Leica S8 APO microscope connected to a digital camera. This particular microscope had a 10x eyepiece and low power objective, resulting in a maximum magnification of 25x, which is insufficient for lithic use-wear analysis. The assemblages from Allt Lebnaskey and Whiting Bay were obtained on loan from Dr Torben Ballin and were examined using an Olympus SZ40 microscope connected to a Canon EOS 70D camera – a combination that allowed microscopic observation and microphotography at magnifications up to 100x.

The use-wear replication experiments were carried out in Bulgaria and microscopic observation and photo documentation were done in the Conservation Laboratory of the National Institute of Archaeology with Museum in Sofia using a Keyence VHX-100k digital microscope at magnifications between 25x and 150x (a magnification range of 80x–150x is usually sufficient to detect the whole complex of wear traces including fine microchipping, polishes, striations, abrasion and attrition). A detailed microphotographic record of the surfaces of the experimental artefacts was made at various stages in the experiments – after the initial cleaning, after use but before removal of surface residues, and after removal of the residues. The photomicrographs (**Figs. 9–13**) were taken at magnifications between 60x and 100x.

2.4. *XRF analyses*

Non-destructive ED-XRF analyses of pitchstone artefacts and source samples were carried out using a Thermo Scientific *Niton XL3t Ultra* (portable) analyzer. This particular instrument is equipped with an Ag anode X-ray source (capable of a maximum voltage of 50 keV, current of 200 μ A and power of 4W) and a 45 mm² Silicon Drift Detector (SDD). Analyses are performed using beam filters to improve the detection of particular elements. The 'XL3t Ultra' has an analytical range of up to 38 elements from Mg to U, although this varies according to the 'mode' (calibration model) selected – the analyzer is supplied with several in-built factory 'calibrations' optimised for analysis of specific materials. The calibrations/modes provided with the 'XL3t Ultra' that are most suitable for the analysis of volcanic glasses (and other bulk samples) are the 'Mining' and 'Soil' calibration models.

We analyzed pitchstone samples from two source outcrops and all five archaeological assemblages (**Table 5**). Two sets of measurements were performed on the pitchstone samples, one set with the instrument operated in the *Fundamental Parameter* (FP) ‘Mining’ mode and the other set using the *Compton Normalization* ‘Soil’ mode. FP and Compton Normalization represent different mathematical approaches to the quantification of XRF spectra from samples. Experience shows the latter approach can reduce problems with the measurement of ‘thin’ samples and can provide data for heavy elements (e.g. Th, U) at low concentrations.

A third type of calibration, known as *Empirical Calibration*, can be achieved by obtaining readings on samples of known elemental composition (Certified Reference Materials), then using the correlation between the readings obtained on the CRMs and their known values (using linear regression analysis) to derive a calibration factor for each element. In this way, the values generated for archaeological samples by the analyzer can be ‘recalibrated’ using the empirically derived calibration factors. While recalibrating against reference standards (in theory) produces more accurate results, it is unlikely to have much impact on one’s interpretation of the data.

3. Results

3.1. Techno-typological and use-wear analyses

The techno-typological attributes of the pitchstone series from Auchategan, Allt Lebnaskey, Luce Bay and Whiting Bay 1 & 2 are summarized in **Tables 1 & 2**. The wear patterns observed on the same pieces through microscopic examination of edges and surfaces are presented in **Tables 3 & 4** and **Figs. 4, 6 & 7**, while the results of the replication experiments and wear patterns observed are summarized in **Table 4** and **Figs. 9–13**.

As noted above, the microscope equipment available in the National Museum of Scotland Collections Centre was unsuitable for detailed observation and use-wear analysis of the archaeological material from Auchatagen and Luce Bay (**Fig. 5**), although wear traces (striations, scarring, smoothing and rounding of edges and dorsal ridges) were visible on some pieces (**Fig. 4**). Nevertheless, over 300 low-magnification photomicrographs were taken and archived at the NMSCS for the benefit of future researchers.

Better results were obtained for the archaeological assemblages from Allt Lebnaskey and Whiting Bay:

- a) Five artefacts (all typological tools) from Allt Lebnaskey possess edge and surface modifications in the form of matt/dull polish, chaotic striations and fresh scarring, which indicate heavy alteration and attrition (**Figs. 6.3 & 6.4, right**). It would be speculative to pronounce on whether the modifications are due to post-depositional surface modification (PDSM) or to use followed by PDSM. Without doubt, some scarring (as illustrated in **Fig. 6.4, right**) has occurred *after* the smoothing of the surfaces and could indicate either re-use (cutting/sawing a medium-hard material) or deliberate resharpening for further use. It should be emphasized that, in general, the surfaces of artefacts from Allt Lebnaskey are less well preserved than in the Whiting Bay assemblages. The striking contrast between the abraded and matt surfaces of the blanks and the fresh retouch, suggests the use and/or intentional resharpening of the artefacts.
- b) Six artefacts among the Whiting Bay 1 assemblage (three blanks and three typological tools) exhibit two main complexes of wear: (i) fine irregular microchipping/scarring (e.g. **Fig. 7.5**) indicating in two cases cutting of soft to medium material and in one

case more likely scraping of a medium-hard material; and (ii) heavily abraded areas resembling the matt generic ‘polish’ and attrition observed on used obsidian artefacts by some researchers (e.g. Kononenko, 2011: 8) accompanied by pronounced striations (**Figs. 7.4 & 7.6**). This attrition could have resulted from sawing and scraping of siliceous hardwood or shell, for example (as illustrated in Kononenko, 2011, plates 151–2, 224).

- c) Four artefacts from the Whiting Bay 2 assemblage (two blanks and two typological tools) show features (retouch, microchipping/scarring, edge smoothing and striations) that could result from utilisation (**Figs. 7.1–7.3**). In two cases cutting and sawing a medium-hard material is presumed (**Fig. 7.1**), in the other two cases, a transverse chisel-like motion is possible, producing ‘stepped’ splintered retouch (**Fig. 7.3**) and large flake removals from the proximal edge (**Fig. 7.2**).

On the experimental artefacts only irregular scarring and residues were evident on the active edges after use, as follows:

- a) The first experiment involved cutting bamboo-like reeds using a bidirectional movement for 50 minutes. This caused no perceptible change to the cutting-edge either at the macro or micro level, apart from reed residue observable before cleaning. No attrition of the natural lustre and no striations were detected (**Fig. 9.1**).
- b) The second experiment was designed to test the resilience of a sharp natural edge when scraping hard, dry wood. After 15 minutes of continuous use, irregular dorsal (micro-)scarring was produced, but tool efficiency was unaffected (**Fig. 9.2**).
- c) Cutting a fresh twig of hazel for 30 minutes resulted in fine irregular bifacial scarring unassociated with any striations or attrition. The surface after use but before cleaning with acetone appeared greasier than after cleaning (**Fig. 10.1**).
- d) An extension of this experiment involved scraping the twig with the small retouched piece of pitchstone for 10 minutes. The short use duration did not result in any detectable microscopic edge modification apart from light smoothing of the edge, and the retouched part remained unchanged during the process. The efficiency of this operation was negatively affected by the high moisture content of the wood (**Fig. 10.2**).
- e) A series of experiments were performed on damp (soaked) pig hide – a material that traditionally in Bulgaria was used for making a particular type of shoe. The first experiment involved scraping for 20 minutes. Although scraper use was brief it generated weak smoothing of the microtopography on both faces along the edge, as well as residues that were easily removed by cleaning of the tool (**Fig. 11.1**).
- f) The same piece of pig hide was then cut into thin strips for 30 min using the distal part of a massive but sharp natural flake. The edge broke at the very beginning, but this did not seem to affect the efficiency of the operation. In total, six strips were cut; the edge was re-sharpened incidentally during this work but apart from the resulting scars no other traces were observed (**Fig. 11.2**).
- g) A blade selected from among the BB debitage was used for 20 minutes sawing a dry calf bone. Scarring quickly appeared on the working edge and pitchstone chips were detached progressively, while bone powder covered the microtopography and the negatives of the utilisation retouch. A 2-mm deep groove with a U-shaped profile was made in the bone, and a white ‘band’ of bone residue on the working edge of the blade was visible before cleaning. However, no use-striations were observed (**Fig. 12.1**).

- h) The experiment was repeated after soaking the bone in water and urine for 29 hours. For this experiment, a Janus (Kombewa) flake with a very sharp distal transverse edge (selected from the BB debitage) was used. Four parallel incisions (grooves) were made in 15 minutes and scarring and macro-damage of the edge developed very quickly. The bone remained more fibrillose than in its dry state with less bone powder produced. A further incision was made linking the parallel grooves but using the distal part of the left edge on the convex Janus flake surface. This operation lasted just 4 minutes resulting in breakage of the edge. In general, micro-scarring of the working edge was as pronounced as in the case of sawing dry bone, and there were no diagnostic micro features to distinguish between the two operations (**Fig. 12.2**).
- i) The same piece of bone was soaked for 24 hours then scraped using a flake with a transverse edge (BB debitage). The scraping was efficient for the first 5 minutes but became less efficient during the next 10 minutes as the bone became progressively drier. The working edge of the flake acquired regular scalar retouch with a small quantity of fibrous bone residue; the latter was easily removed by cleaning. The most evident result of the bone processing was the utilisation retouch, which on archaeological specimens could easily be mistaken for intentional marginal retouch (**Fig. 13.1**).
- j) The butchering experiment involved a flake with a very sharp (shallow-angled) edge selected from the BB debitage. A piece of pork meat with cartilage was cut into small pieces in 28 minutes. Under the microscope, the working edge showed delicate irregular micro scars. Greasy residues were removed on cleaning the tool (**Fig. 13.2**).

3.2. XRF analyses

Elemental concentration data were obtained for 38 elements, although the ‘light’ elements (Mg, Al, Si, P and S) were not measured in all cases. For all measurements, the measurement window of the analyzer was set to the 8-mm diameter spot size. Each sample was analyzed for a total of 180 seconds in ‘Mining’ mode – 60s using each of the ‘Main’, ‘High’ and ‘Low’ range filters that optimize the analyzer’s sensitivity for various elements, and 120 seconds in ‘Soil’ mode. To improve accuracy, the measurements obtained with the factory-set ‘Mining’ and ‘Soil’ calibrations were *recalibrated* against data for 23 CRMs (all pressed powder samples of various rock types) obtained using identical instrument settings.

The recalibrated data for 10 elements (Mg, K, Ti, Mn, Fe, Rb, Sr, Y, Zr, Nb) were analyzed statistically using *PAST Statistical Software v3.26* (Hammer et al., 2001) to investigate patterns of geochemical variation within archaeological and source pitchstone samples. The results of the pXRF analyses of archaeological and source samples are presented graphically in **Figs. 14 & 15**.

4. Discussion and Conclusions

The results of our pilot study lead us to the following preliminary observations:

4.1. Use-wear analyses

Our replication experiments did not produce identifiable use wear signatures – that is no regular or distinctive microwear patterns were observed that could serve as reliable indicators of function in comparative studies of archaeological collections. No match was found between the experimentally obtained wear traces and those observed on pitchstone artefacts from selected Neolithic sites. The former consisted principally of simple uni- or bifacial scarring on the edges, while some archaeological pieces exhibited very obvious

(micro) wear in the form of pronounced smoothing, abraded zones/attrition and ‘polish’ of the whole surface, chaotic striations, and areas of abrasion, as shown in **Figs. 4, 6 & 7**.

The question of ‘polish’ on pitchstone artefacts needs clarification. We are not aware of any previous descriptions of utilization polish on pitchstone or replication experiments. It is possible that utilization polish on pitchstone appears differently than on obsidian and devising experiments to address this problem is a challenge.

The use-wear analyses had some fundamental limitations that undoubtedly impacted the results:

- 1) The equipment used for the archaeological and experimental use-wear analyses was not standardized in terms of magnification range, lighting characteristics or photographic resolution. Three different microscopes and corresponding illumination and photographic systems were employed.
- 2) Our lack of experience in use-wear analysis of volcanic glasses, compounded by the lack of a comprehensive experimental reference collection, was an equally severe constraint. We attempted to compensate for this by reference to the numerous published experimental use-wear studies of obsidian artefacts (e.g. Astruc et al., 2012; Beyin, 2010; Hurcombe, 1992; Lewenstain, 1981; Kononenko, 2007, 2011; Kononenko et al., 2015, 2016; Nesbitt et al., 2019; Schousboe, 1977). However, pitchstone typically has a less regular fracture and duller lustre than most obsidians which, along with the common presence of phenocrysts and microcrystallites, may affect how use-wear develops and appears on pitchstone.
- 3) Although we conducted replication experiments as part of the pilot study, these were restricted to the processing of skin, bone and muscle tissue and did not attempt to simulate other activities that may have been practised regularly by Neolithic people in northern Britain, such as fish processing and pottery manufacturing. Moreover, the duration of the experiments was variable and, in some cases, perhaps too short.
- 4) As will be evident from **Figs. 4–8**, the archaeological artefacts included in the pilot study differed in their morphometric and typological attributes from artefacts available for our experiments.

Despite these limitations, one positive result of the use-wear analyses relates to the assemblage from Auchtagen. Based on his analysis of the way in which different lithic raw materials (chert, quartz and pitchstone) were treated, Ballin suggested that pitchstone held both utilitarian and symbolic value for the Neolithic inhabitants of the site (Ballin 2006, 27). Our use-wear analyses offer some support for this idea, in that a large proportion of the pitchstone artefacts are well preserved without edge modifications that could be interpreted as traces of utilization. However, in the absence of comparative use-wear data for the chert and quartz assemblages from the site, this remains speculative.

4.2. pXRF analyses

Previous chemical provenance studies of archaeological pitchstones in the British Isles concluded that the raw material originated from sources on the Isle of Arran, most likely from the Clachland area where pitchstone associated with igneous intrusions crops out along the modern shoreline as well as inland. Thorpe and Thorpe (1984) used ED-XRF to measure the concentrations of major elements, and INAA for trace elements. Preston et al. (1998, 2002) performed WD-XRF analyses on whole-rock samples for major and trace elements and

electron probe microanalysis (EPMA) for major elements in residual glass from the same samples.

As noted above, the Niton *XL3t ultra* pXRF analyzer used in our pilot study has an analytical range from Mg to U. **Table 6** compares our pXRF data (average values) with the corresponding results reported by Thorpe and Thorpe (1984) and Preston et al. (1998, 2002) for whole-rock pitchstone samples from the Clauchland area.

A number of individual pitchstone outcrops have been recorded in the Clauchland area (Ballin and Faithfull, 2009) and it is unlikely that our samples come from exactly the same locations as those analyzed previously. This conclusion is supported by the observed differences in the mean concentration values for Fe, K, Zr and Sr between the three studies.

To compare our pXRF results for archaeological and source samples we used two forms of statistical analysis – bivariate analysis and hierarchical cluster analysis. For the bivariate analyses, we applied a modification of the method proposed by Kobayashi and Mochizuki (2007) for discriminating Anatolian obsidians, using the following indices: $Rb \times 100 / (Rb + Sr + Y + Zr)$ and Ti / Mn . In the bivariate plots (**Fig. 14**) the ranges of variation of the samples from the two source outcrops are represented by their convex hulls or envelopes (the smallest convex polygon that contains all the points of each set). The compositional data for the archaeological pitchstones from each of the 4 sites included in our pilot study are plotted against these source envelopes.

- The overlap between the convex envelope (CE) for Clauchland Hills and that for Clauchland shore is minimal, suggesting the two source outcrops can be regarded as separate chemical groups.
- Few archaeological samples overlap the Clauchland shore CE. This is unsurprising since none of the archaeological samples analyzed exhibited the irregular fracture characteristic of the Clauchland shore pitchstone.
- Some samples from each of the archaeological sites overlap the Clauchland Hills CE. Based on their chemical fingerprint, therefore, those samples *could* derive from the Clauchland Hills source. But most archaeological samples fall outside the CE and so are less likely to derive from the Clauchland Hills source.

In the bivariate plots the ‘spread’ of the archaeological samples from Auchategan, Luce Bay (and, perhaps, Allt Lebnaskey) exceeds the Clauchland Hills CE, suggesting these sites contain pitchstone from more than one chemical group (source). This is confirmed by the results of the cluster analysis, which indicate the presence of two chemical groups at Allt Lebnaskey, three at Auchategan, four at Luce Bay (one of which includes all the samples analyzed from Luce Sands), and three at Whiting Bay where groups I and II occur in both artefact concentrations (**Fig. 15**).

From these limited results we draw the following tentative conclusions:

- None of the archaeological pitchstone analyzed appears to originate from the Clauchland shore ‘great sill’ source outcrop, despite the abundance and easy accessibility of large pitchstone blocks on the raised beach and the modern foreshore.
- The pitchstone used at Allt Lebnaskey, Whiting Bay, Luce Bay and Auchatagen was not obtained from a single source, although some of that analyzed from each site could have originated from Clauchland Hills.

4.3. Possibilities for future work

Any future work will need to address the limitations of the pilot study.

Determining the sources of the pitchstone found in archaeological contexts across the northern and western British Isles will require geochemical fingerprinting of a much larger range of source outcrops, especially given the presence of pitchstone from different chemical groups at the same sites or neighbouring sites. The low cost and flexibility of pXRF make this the method of choice, but it may need to be used in conjunction with petrographic thin sectioning and more powerful techniques of elemental analysis with lower limits of detection.

A more comprehensive series of replication experiments is essential to directly compare wear patterns that develop on flint vs obsidian vs pitchstone, and microscopic examination of archaeological and experimental specimens requires the use of standardised equipment.

For various reasons, none of these conditions could be satisfied within the context of the pilot study.

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Figures & Tables

- Fig. 1 **a.** Map of Tertiary (62–55 Ma) volcanic and igneous complexes in the northwestern British Isles, known as the British Tertiary Igneous Province (BTIP). Localities in *italics* indicate where pitchstone outcrops are relatively abundant (redrawn from Preston *et al.* 2002).
b. Locations of pitchstone outcrops and archaeological assemblages considered in this paper: AG – Auchatagen, AL – Allt Lebnaskey, CH – Clauchland Hills, CS – Clauchland shore, KC – King’s Cave, LB – Luce Bay, WB – Whiting Bay (CB).
- Fig. 2 Pitchstone sources on the Isle of Arran referred to in the text: **a** – the ‘Great Sill’ at Clauchlands shore; **b** – fallen pitchstone block on the slope below the ‘Great Sill’; **c** – pitchstone boulders on the foreshore below the ‘Great Sill’; **d** – fractured boulder derived from the ‘Great Sill’ showing the aphyric character and irregular fracture of the pitchstone from the Clauchlands shore source; **e** – pitchstone pebble exposed by footpath erosion at Clauchland Hills; **f** – boulder of porphyritic pitchstone on the beach south of King’s Cave, Arran. (MG & CB)
- Fig. 3 Pitchstone samples used for experimental knapping and provenance determination: 1 – pieces from primary sources at Clauchlands shore; 2 – secondarily deposited pebbles from Clauchlands shore; 3 – pebble from the shore near King’s Cave; 4 – pebble from Clauchlands shore; 3–5 – debitage by B. Bradley (MG)
- Fig. 4 Auchatagan and Luce Bay sites – macrophotos of selected artefacts (MG)
- Fig. 5. Auchatagan (1) and Luce Bay (2) sites – drawings of selected artefacts (MG)
- Fig. 6 Whiting Bay (1 – Assemblage 1, 2 – Assemblage 2) and Alt Lebnaskey (3, 4) assemblages by drawings and photographs (MG)
- Fig. 7 Photomicrographs of artefacts from Whiting Bay: 1–3 – Assemblage 2; 4–6 – Assemblage 1(MG)
- Fig. 8 Natural pieces (1) and artefacts (2 plus the piece enclosed by the dashed circle from 1) used in the experiments – working edges before use. Photomicrographs in the magnification range 80x-100x (MG)
- Fig. 9 Experiments: 1– cutting reed (photomicrographs 100x [before cleaning]/60x [after cleaning]) 1 – scraping wood (photomicrographs – 60x/100x) (MG)
- Fig. 10 Experiments on wood: 1 – cutting hazel branch (photomicrographs 100x/60x); 1 – scraping hazel branch (photomicrographs 80x/100x) (MG)
- Fig. 11 Experiments on pigskin: 1– scraping (photomicrographs 100x/100x); 1 – cutting (photomicrographs 80x/80x) (MG)
- Fig. 12 Experiments on bone: 1 – sawing dry bone (photomicrographs 60x/60x); 1 – sawing soaked bone (photomicrographs 100x/60x) (MG)

Fig. 13	Experiments: 1 – scraping soaked bone (photomicrographs 80x/80x); 1 – butchering (photomicrographs 100x/60x) (MG)
Fig. 14	Scatterplot of $Rb \times 100 / (Rb + Sr + Y + Zr)$ and Ti/Mn data from X-ray fluorescence of pitchstone samples from archaeological sites in southwest Scotland, compared with the corresponding data for two geological sources on the Isle of Arran represented by their convex hulls (solid line – Clauchland Hills, dashed line – Clauchland shore) (CB)
Fig. 15	Cluster analysis of compositional data for Mg, K, Ti, Mn, Fe, Rb, Sr, Y, Zr and Nb from X-ray fluorescence analysis of pitchstone artefacts from archaeological sites in southwest Scotland, using the Paired Group (UPGMA) algorithm and Euclidean Distance similarity measure (CB)
Table 1.	The general structure of the archaeological assemblages.
Table 2.	The typological repertoire of the archaeological assemblages.
Table 3.	Summary of use-wear observations on archaeological artefacts.
Table 4.	Summary experiments and results.
Table 5.	Pitchstone sources and archaeological assemblages – numbers of samples analyzed by pXRF.
Table 6.	Mean elemental concentration values for pitchstone samples from geological sources in the Clauchlands area of Arran obtained by <i>a</i> – Williams Thorpe and Thorpe (1984), <i>b</i> – Preston et al. (1998), <i>c</i> – Preston et al. (2002) and <i>d</i> – this study. Major elements are reported as oxide values; trace elements are reported in parts per million (ppm). The r^2 value is a measure of the accuracy of the pXRF measurements based on empirical calibration using CRMs (this study).

Table 1

Artefact category	Allt Lebnaskey	Whiting Bay Assemblage 1	Whiting Bay Assemblage 2	Luce Bay	Auchategan	Total
precores	2	1	2		1	6
cores			2	1	10	13
blades		1	1	37	26	65
flakes	8	18	7	2	25	60
fragments	21	10	3		15	49
chips	25				4	29
tools	8	4	6	3	2	23
varia	1		1			2
pseudo artefacts	9	12	1			22
total	74	46	23	43	83	269

Table 2

Type category	Allt Lebnaskey	Whiting Bay Assemblage 1	Whiting Bay Assemblage 2	Luce Bay	Auchategan	Total
retouched blades	1	1		1	1	4
retouched flakes	4	1	1			6
splintered pieces	1	1	4			6
truncation		1				1
burin			1	1		2
endscraper	1					1
perforator	1					1
notched pieces				1	1	2
total	8	4	6	3	2	23

Table 3

Type of wear	Allt Lebnaskey	Whiting Bay Assemblage 1	Whiting Bay Assemblage 2	Luce Bay	Auchategan
scarring	+	+	+	+	+
striations	+	+	+	+	+
attrition zone/ polish	+	+	+		
smoothing					
PDSM	+	+	+	+	+

Table 4

Experm. No.	Worked Material	Mode of use	Tool	Duration (min.)	Scarring	Striations, Orientation	Edge Rounding	Attrition/polish	Residue (before cleaning)
1 (fig. 9 - 1)	reed	cutting/sawing	natural flake	50	single/sporadic	none detected	not detected	not detected	tissue particles
2 (fig. 9 - 2)	dry wood	scrapping	natural flake	15	irregular/ unifacial	none detected	not detected	not detected	tissue particles
3 (fig. 10 - 1)	fresh wood	cutting/sawing	natural flake	30	irregular/ bifacial	none detected	not detected	not detected	weak film
4 (fig. 10 - 2)	fresh wood	scrapping	natural flake with retouch	10	no additional scarring	none detected	light	not detected	tissue particles
5 (fig. 11 - 1)	pig hide (soaked)	scrapping	natural flake	20	sporadic	none detected	light	not detected	intensive tissue
6 (fig. 11 - 2)	pig hide	cutting	natural flake	30	bifacial + re-sharpening	none detected	not detected	not detected	tissue particles
7 (fig. 12 - 1)	dry bone	sawing	blade	20	intensive bifacial	none detected	light/ irregular	not detected	film, starch
8 (fig. 12 - 2)	soaked bone	sawing/engraving	flake	15+4	intensive bifacial	none detected	light/ irregular	not detected	particles, starch
9 (fig. 13 - 1)	soaked bone	scrapping	flake	15	intensive/unifacial/retouch-like	none detected	not detected	not detected	tissue particles
10 (fig. 13 - 2)	pig cartilage	butchering	flake	28	irregular	none detected	light	not detected	film, tissue

Table 5

Source	No. pieces
Clauchland Shore	10
Clauchland Hills	15
King's Cave	–
Archaeological assemblage	
Allt Lebnaskey	6
Auchatagen	10
Luce Bay – ‘Glenluce’	21
Luce Bay – ‘Luce Sands’	4
Whiting Bay 1	7
Whiting Bay 2	5
Total	79

Table 6

Source	Σ	TiO ₂	Fe ₂ O ₃	MnO	MgO	K ₂ O	Nb	Zr	Y	Sr	Rb	Ba	Reference
		r ² =0.9962	r ² =0.9978	r ² =0.9284	r ² =0.9705	r ² =0.9897	r ² =0.9908	r ² =0.9978	r ² =0.9319	r ² =0.9986	r ² =0.9985	r ² =0.867	
Clauchland Hills – sill	<i>n.d.</i>	0.15	1.48	0.03	0.05	4.34	–	–	–	–	–	–	<i>a</i>
Clauchland Hills	<i>n=19</i>	0.20	1.64	0.05	–	4.03	23	208	65	36	142	875	<i>d</i>
Clauchland Shore	<i>n.d.</i>	0.09	1.51	0.03	0.34	4.56	21	247	65	48	135	944	<i>b</i>
Clauchland shore – great sill	<i>n.d.</i>	0.12	(0.31)	0.05	0.00	4.22	–	–	–	–	–	–	<i>c</i>
Clauchland shore – great sill	<i>n=10</i>	0.14	1.46	0.04	–	3.75	23	197	63	35	136	874	<i>d</i>

Fig. 2

Bruce Bradley's debitage

Auchategan site

Glenluce site

Fig. 5

Fig. 6

Edges and surface modifications

Fig. 7

Fig. 8

Fig. 9

1

Cutting reed (*Phragmites australis*) – 50 min

Before cleaning

After cleaning

Scraping dry wood – 15 min

2

Before cleaning

After cleaning

Fig. 10

1

Cutting *Corylus avellana* L.
(hazel branch) – 30 min

Before cleaning

After cleaning

2

Scraping fresh hazel branch – 10 min

Before cleaning

After cleaning

Fig. 11

1

Scraping pig hide – 20 min

Traditional Bulgarian pointed shoes called "tsarvuli"

Before cleaning

After cleaning

2

Cutting pig hide – 30 min

Before cleaning

After cleaning

Fig. 12

1

Sawing dry bone (calf) – 20 min

Before cleaning

After cleaning

2

Sawing soaked bone (calf) – 15 min

Before cleaning

After cleaning

Fig. 13

Scraping soaked bone (calf) – 15 min

Before cleaning

After cleaning

1

Butchering (pork cartilage) – 28 min

Before cleaning

After cleaning

2

Fig. 14

Fig. 15