Read at the Annual Conference of the Ussher Society, January 1998

GEOCHEMICAL FINGERPRINTING OF WEST CORNISH GREENSTONES AS AN AID TO PROVENANCING NEOLITHIC AXES

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M. Markham and P.A. Floyd. 1998. Geochemical fingerprinting of west Cornish greenstones as an aid to provenancing Neolithic axes. *Geoscience in south-west England,* **9,** 218-223.

Of the large number of Neolithic stone axes made of greenstone, some 392 (referred to as Group 1 axes) are believed to have been manufactured in west Cornwall around the Mount's Bay area. To aid the location of the greenstone that provided the materials for the axes, geochemical fingerprinting of the axes and greenstone outcrops was undertaken in the Mount's Bay area to both discriminate the greenstone localities and provide a basis for matching Group 1 axes. Non-destructive analysis of the axes was determined by a portable XRF Spectrometer unit that gave comparable results for selected greenstone samples to standard laboratory-based XRF techniques. Geochemical fingerprinting of the greenstone localities by portable XRF spectrometer provided a degree of discrimination between them, although preliminary data on the axes suggests that there is not a very strong correlation between axe composition and possible greenstone sites. Further work is required on other greenstone localities and axes as current data does not conclusively point to an origin in this area of west Cornwall.

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INTRODUCTION

The Devonian and Lower Carboniferous sequences of southwest England are characterised by episodes of volcanic activity with the production of largely submarine pillow lavas and high-level intrusive sills and sheets (Floyd, 1995). During the Variscan orogeny the rocks were subjected to low-grade alteration and the volcanics developed secondary assemblages indicative of the pumpellyite and greenschist facies of regional metamorphism (Floyd, 1995). Subsequently, on the intrusion of the granite plutons, contact metamorphic effects were locally superimposed with the production of new assemblages and textures. Overall, the effects of alteration on the predominantly basic volcanic rocks was two fold: (a) variably developed secondary assemblages superimposed on a still recognisable primary basic assemblage (pyroxene-plagioclase-ilmenite±olivine), and (b) the textural effects of deformation with the progressive development of a foliation and eventually a marked schistosity.

From the industrial viewpoint the mineralogical alteration effects on the basic volcanics has strengthened them and many of the massive (non-foliated) intrusive basic rocks are used for roadstone (Edmonds *et al*., 1969). Stone Age man also appreciated the durable qualities of the altered basic rocks and fashioned many hand axes from what is generally referred to as "greenstone", that is, low-grade metamorphosed dolerites and basalts.

STONE AGE AXES

Stone axes are grouped according to petrological, mineralogical and textural criteria. Over 40 different axe groups are recognised from a total of 7625 axes found in Britain (of which only 3546 have been grouped) (Clough and Cummings, 1988). Just over 1000 axes are made of greenstone, of which 392 members (referred to a "Group 1") have been identified as being manufactured in west Cornwall and possibly originating from the Mount's Bay area (Figure 1), largely on the basis of the site of origin and the local presence of greenstone (Stone, 1951).

Group 1 axes, originally catalogued in the 1940s (Keiller, 1941), are broadly described as uralitized gabbro with original pyroxene and feldspar, with the characteristic development of a uralitic fringe of blue-green amphibole around the primary pyroxene; epidote, sphene and chlorite are common accessory minerals. Because this assemblage and texture is a very common feature of many Cornish greenstones, the actual outcrops, representing the stone age factories used by Neolithic man, have never been positively identified. However, the assumed manufacture from this area is still largely based on the apparent similarity between a petrographic examination of thin sections of greenstone axes and the outcrops in the Mount's Bay area. To date no definite match has been achieved, such that archaeologists have suggested that the outcrops are actually underwater and have yet to be sampled! (Evans, 1962)

PROJECT OBJECTIVES AND APPROACH

The prime object of this work is to determine from where the Group 1 greenstone axes originated, using a combination of petrographic and textural data coupled with new geochemical discrimination techniques. Petrographic comparisons between axe and potential outcrop are a standard method to identify sources (Markham, 1997) although in this case the results have proved inconclusive. Geochemical fingerprinting of the axes, especially trace element distributions, was considered a more sensitive and selective approach. Also, there already exists a broad-based set of geochemical data on Cornish greenstones (both intrusives and lavas) that shows some significant chemical distinctions between different localities composed of broadly similar mineralogical assemblages (Floyd, 1995). If both axes and greenstone outcrops can be sufficiently discriminated geochemically this provides a sound basis for comparison and the strong possibility of linking axe to source.

However, in order to provenance axes by geochemical means, a non-destructive analytical method is needed to preserve the axe, although ideally should be a similar technique to that used to analyse field samples (to minimise inter-technique errors). In this connection we have used X-ray Fluorescence Spectrometry, both a portable instrument (PXRF) and a laboratory-based instrument (LXRF)(see below).

ANALYTICAL METHODS AND COMPARISONS

XRF Spectrometry determines the concentration of elements in a sample by measuring the variable intensity of fluorescent energies given off by a sample when illuminated by x-rays.

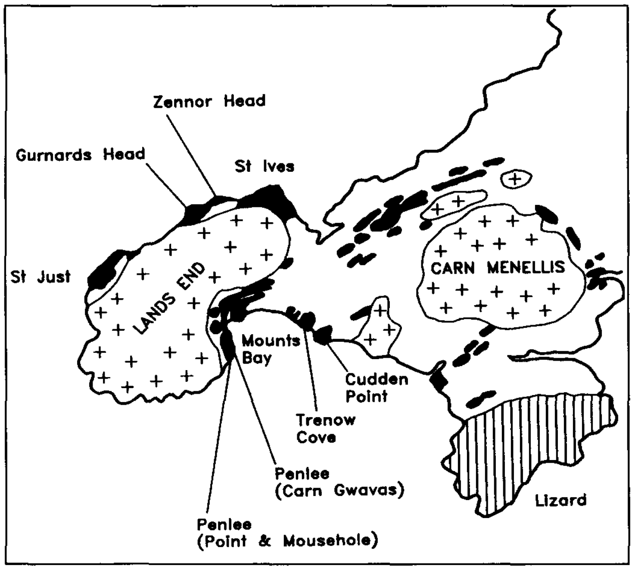

Figure 1
Map of west Cornwall showing the occurrence of greenstone outcrops sampled for this study. (Greenstones -solid ornament, Granite '+'ornament)

Considerable preparation is required prior to analysis, involving crushing to a fine powder and subsequent pelletizing, and thus destroying the original sample. This is standard procedure for laboratory-based instruments, but is not an option for valuable and irreplaceable axes. In this context the PXRF has a number of advantages - it is portable (carried by one person), is nondestructive, the samples need no preparation, and can work off mains or battery. Provided the sample has a reasonably planer surface greater than 25 mm in diameter it can be geochemically analysed. For PXRF the radiation energies needed to fluoresce the sample are obtained through the radioactive decay of isotopes of Fe, Cd and Am. The reflected energies are detected by a high purity HgI_2 detector which requires no external cooling. Table 1 summarises the main features of both PXRF and laboratory XRF (LXRF) equipment

Before meaningful geochemical comparisons can be made between the axes and potential sample localities, it is necessary to check that the PXRF instrument is capable of producing similar results to laboratory based XRF instruments (at Open University) and also existing data from the literature derived by similar means (at Keele University). A selection of greenstone samples collected from around Penlee Point (Penlee Lifeboat station and Mousehole foreshore) and from within and shore side of the Penlee Quarry at Cam Gwavas were analysed by LXRF and PXRF at the Open University and compared with existing data by Floyd (1976, 1983, 1984) and Floyd and Al-Samman (1980). The results for the three sets of data (PXRF, LXRF, literature) are shown in Figure 2 and Table 3, and clearly demonstrate that each data source can separately define and discriminate the two sampled sites (foreshore versus quarry). We consider that the overlap of geochemical data derived from the PXRF is sufficiently good to allow discrimination of the axes and sources, and also compares well with existing LXRF-derived data.

FEATURES OF GREENSTONE SITES

Figure 1 shows the sites subjected to preliminary evaluation as potential axe sources. Initially the choice of locations was determined by the proximity to Mount's Bay and environs (the suggested origin of Group 1 axes), and the amount of existing data available. Table 2 summaries the location and nature of emplacement for the greenstone bodies; detailed descriptions of some of the following sites may be found in Floyd *et al.* (1993).

Cudden Point [SW 548 275] - This is a relatively coarse, massive, sheet-like intrusive body of greenstone composed of meta-gabbro and

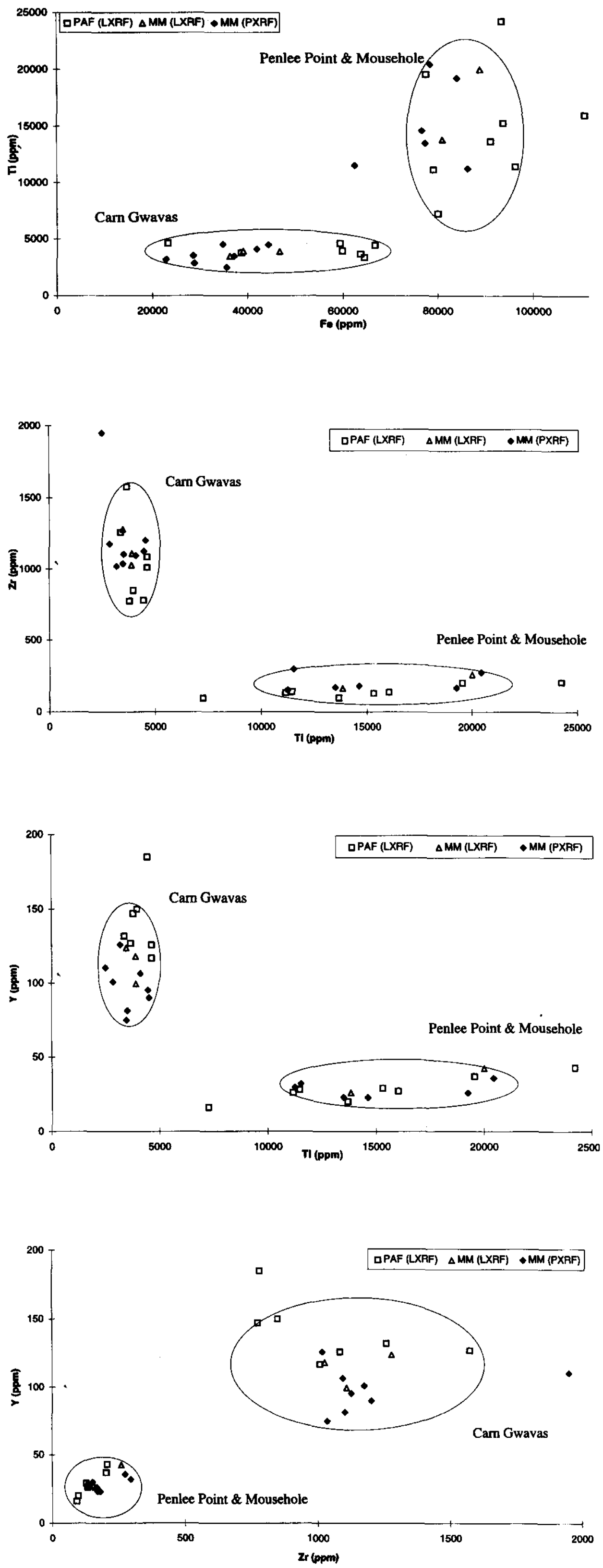

Figure 2
Envelopes enclose geochemical data from two major greenstone outcrops around Penlee Point and adjacent foreshore, and the Carn Gwavas quarry, derived by PXRF (MM(PXRF)), laboratory-based XRF by Markham (MM(LXRF)) and by Floyd (PAF(LXRF)). Note the general overlap of PXRF and LXRF data from the two localities.

meta-dolerite, with strongly foliated margins (Floyd and Lees, 1972). It shows the typical development of pale green uralitic actinolite fringing large clinopyroxene prisms, some of which may exhibit pigeonite lamellae. Petrographically it is characterised by rare crystals of brown primary amphibole, two generations of secondary amphibole (pale uralite and blue-green), and replaced ovoids of olivines surrounded by pyroxene.

Trenow [SW 529 303] - This site covers a series of scattered outcrops around Trenow Cove including a small quarry at Perranuthnoe. Periglacial "head" covers much of the area behind the wave-cut platforms and it is not clear if more than one massive intrusive body exists here. The greenstones here are similar to Cudden Point, but generally finer grained and composed of metadolerite and meta-basalt. The metamorphic mineralogical growth has developed further with a higher proportion of secondary minerals, especially amphibole, albite, epidote, chlorite and sphene. In order to decrease the potential variability of results all data presented below comes from the west end of Trenow Cove.

Penlee Point [SW 474 269] - This includes much of the Mousehole foreshore around the Penlee Life Boat station to Carn Gwavas to the north. It includes the two sills found in the inland quarries at Penlee Point proper. Although within the Land's End granite aureole, the greenstones here show similar mineralogical and textural features to low-grade regional meta-basites elsewhere in west Cornwall. They are mainly meta-dolerites with uralitic actinolite, grading to actinolite-plagioclase hornfelses with a weak foliation. Evidence of contact metasomatism is shown by the development of rare blue zoned tourmaline, and the patchy hydrothermal "bleaching" of some outcrops.

Carn Gwavas [SW 470 280]- This is restricted to the huge (now disused) quarry between the shore and the granite inland and the immediate foreshore east of the quarry. Although often considered and quarried as "greenstone", this rock type is not basic, but more intermediate in composition and thus quite distinct mineralogically and chemically from typical greenstones. It has an interlocking granular texture composed almost entirely of albite and ragged amphibole. It displays variable degrees of hydrothermal alteration with the development of kaolinite, biotite, chlorite, as well as a complex of sulphide mineralization veinlets (Floyd, 1965). Axes derived from this source would be very distinctive mineralogically and chemically.

Zennor [SW 450 394] - This forms a small resistant headland of greenstone west of St. Ives within the granite contact aureole. It is mainly a fine-grained amphibole-rich hornfels of massive aspect.

Gurnard's Head [SW 432 387] - This headland is largely composed of a massive sheet-like body that grades upwards into a pillow lava sequence and represents a high-level intrusion near the sediment-water interface. Again within the granite aureole, the original mineralogy and texture have been replaced by a hornfelsic matte of actinolite with subsidiary plagioclase and rare biotite replacing amphibole. Chlorite-filled amygdales in the pillowed section are still recognisable.

Table 2 shows the mineralogical features of the greenstones found at these sites, as well as summarising some of their geochemical characteristics.

GEOCHEMICAL DISCRIMINATION OF GREENSTONE SITES

In this section we attempt to discriminate the different selected sites on the basis of PXRF data alone prior to any comparison with axe data derived by the same technique. The PXRF has been set up to measure 13 elements such that a large number of binary plots can be generated. In this preliminary exercise, however, a number of diagrams plotting absolute abundances measured in ppm have been selected to illustrate the chemical diversity of the greenstone sites and provide a measure of discrimination (Figure 3 and Table 4). We have chosen to present the data as averages ± 1sd since this presents a clearer indication of the outcrops and reduces clutter on the plots. 2SD 'space' will be used in the final assessment although it is recognised that a multivariate statistical approach will be needed in the future.

As seen from the plots in Figure 3, a number of chemical features characterise the sites which can be summarised below:

(a) In all plots, the Cam Gwavas body is geochemically distinctive relative to the rest, being typified by high Zr and Y, coupled with low Ti, Fe, and Sr. Also, as mentioned above, it is also mineralogically and texturally distinctive. These features provide a good discrimination for this type of "greenstone", whereas some of the other greenstone locations have more overlapping characteristics.

(b) Cudden Point also has a distinctive chemistry relative to the other greenstones, with very low incompatible element contents, especially Ti, Zr, Y and Nb; the Zr/Y ratio is also low at between 2-6.

(c) The Trenow data shows the largest standard deviation of data often overlapping other greenstone sites, with the exception of Cudden Point and Cam Gwavas. There is a degree of chemical overlap between Penlee Point, although this locality is within the granite aureole and would be expected to show some mineralogical and textural differences.

(d) There are small chemical differences between the actinolite-bearing greenstones (hornfelses) within the granite aureole at Gurnards Head and Zennor. In general, Gurnards Head has systematically higher Fe, Y and Ti than Zennor.

As seen (Figure 3) discrimination is by no means clear cut for all greenstone compositions. The log plot of Ti versus Zr provides one of the better discriminations with different locations showing partial separation on a curved trend, starting with (i) Cudden Point, (ii) Zennor, (iii) Gurnards Head + Penlee, (iv) Trenow and finally (v) Carn Gwavas. The log plot of Y versus Zr, on the other hand, has an almost linear trend with the same progression.

	Portable XRF	**λd XRF**
Precision (relative error)	usually 10% or better	Major elements to 0.5% Minor elements to 1 - 5 %
		(of value measured)
Operation	One person portable, hand held or stand mounted sensor, battery or mains powered	Laboratory based, high power x-ray tube and cooling required
Detection limit	Major elements to 100ppm Trace elements to 3 ppm	Trace (rarely <1 -2 ppm)
Measurement time	Typically 3- 5 minutes depending on age of sources	5 -40 minutes depending on precision required
Sample Preparation	None, other than presenting a clean, dust free surface	Pressed powder discs Glass pellets
Cost	Machine £50,000 Loan £1,000 per week	£120,000
Range of Elements	Heavier than potassium	Heavier than boron

Table 1. *Comparison of portable and laboratory based XRF equipment (after Markham, 1997: Potts, 1995)*

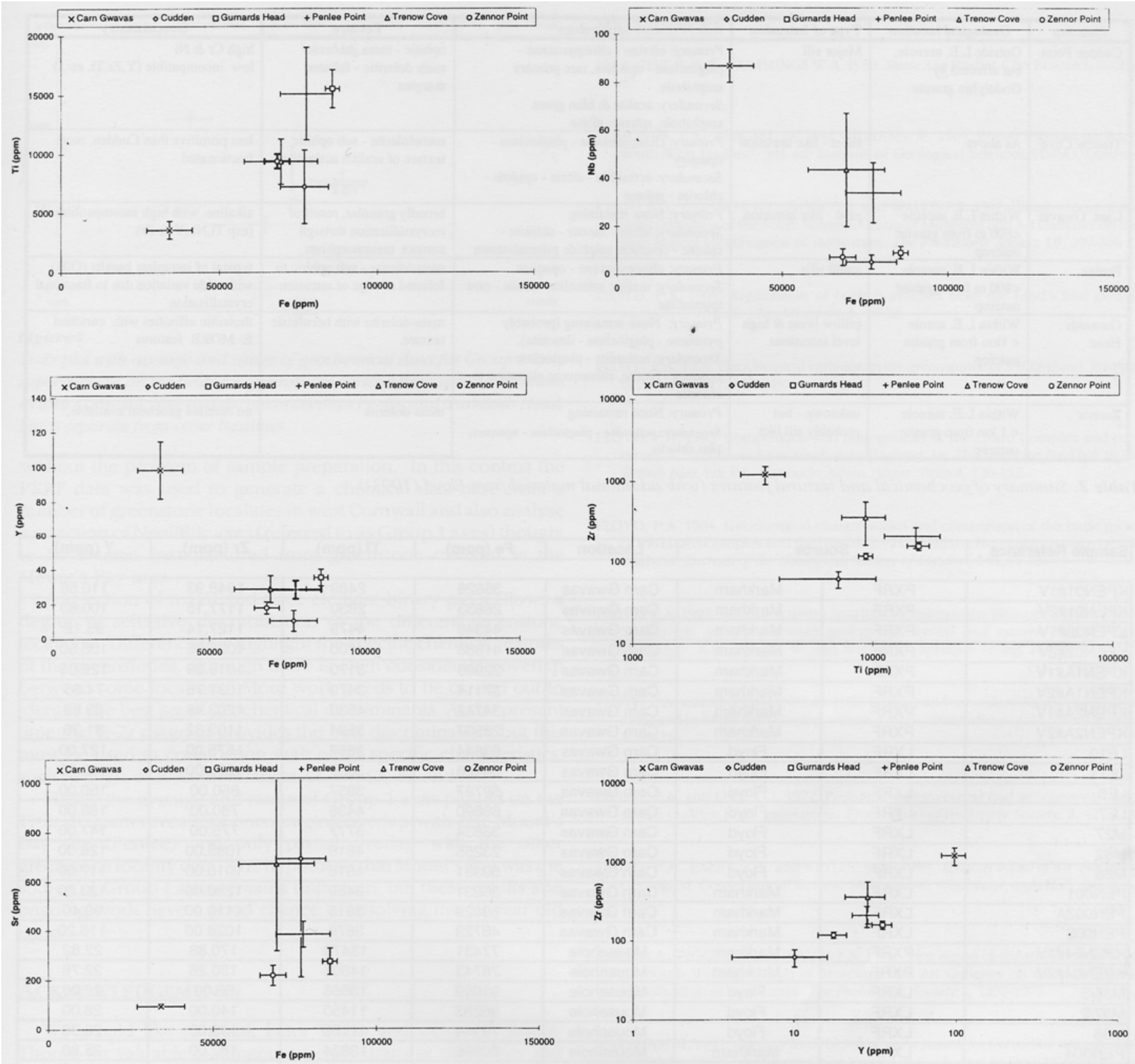

Figure 3
Geochemical data derived by PXRF data (averages ± 1 sd) from selected greenstone localities in west Cornwall. Note that some diagrams provide a degree of discrimination between selected greenstone localities.

PRELIMINARY AXE DATA

At the moment only a preliminary assessment and comparison between the greenstone sites and the axes can be made. This is a consequence of discovering that the geochemistry of the Group 1 axes was more heterogeneous than expected with some axes having compositions that lay well outside the normal range. Histogram plots of axe elements showed a non-standard distribution and upon investigation some of the previously classified axes may have to be regrouped. Further work on existing petrographic thin sections and accession records is being carried out and this will need to be concluded before axes are eliminated from Group I. Hence this feature suggests that all the so-called Group 1 axes may not belong to this group or indeed be derived from Cornish greenstones at all. Thus, some 50 Group 1 axes have been measured by PXRF and an average together with a characteristic range calculated. The range identified in Table 4 has been taken from the aforementioned frequency histograms of the axe data and does not represent a standard deviation for the full data set, that is, it excludes those axes with apparently non-Group 1 chemistry.

The axe average and range is plotted in Figure 4 (with data in Table 4) and compared with the geochemical fingerprints for the greenstone sites. The Ti versus Zr plot chosen to illustrate the comparison shows the axe range is comparable with both Penlee and Gurnards Head, with Carn Gwavas, Cudden and Zennor outside the axe range. Whilst encouraging, these are early results and more work will be required to build up a conclusive picture.

CONCLUSIONS

One of the most important conclusions to draw from this type of geochemical fingerprinting of archaeological artefacts is that a PXRF spectrometer can provide sensitive and accurate results without the destruction of the specimen. We have demonstrated that comparable data can be obtained from a PXRF instrument relative to a standard laboratory-based XRF spectrometer and without the problem of sample

Outcrop	Geological location	Type of Intrusion	Mineralogy	Texture	Geochemistry
Cudden Point	Outside L.E. aureole, but affected by Godolphin granite	Major sill	*Primary*: olivine - clinopyroxene - plagioclase - opaques, rare primary amphibole *Secondary*: uralite & blue green amphibole, sphene, albite	ophitic - meta gabbroic/ meta doleritic - foliated margins	high Cr & Ni low incompatible (Y,Zr,Ti, etc,.)
Trenow Cove	As above	sheet - like intrusion	*Primary*: clinopyroxene - plagioclase - opaques *Secondary*: actinolite - albite - epidote - chlorite - sphene	metadolerite - sub ophitic texture of uralitic actinolite	less primitive than Cudden, more fractionated
Carn Gwavas	Within L.E. aureole <600 m from granite outcrop	pipe - like intrusion	*Primary*: None remaining *Secondary*: albite - biotite - chlorite - sphene - common sulphide mineralisation	broadly granular, result of recrystallisation through contact metamorphism	alkaline, with high incompatibles (esp Ti,Nb,Y & Zr)
Penlee	Within L.E. aureole <800 m from granite outcrop	small sills	*Primary*: clinopyroxene - opaques *Secondary*: uralitic actinolite - albite - rare tourmaline	metadolerite - sub ophitic to foliated at edge of intrusion	typical of intraplate basalts (OIB), with wide variation due to fractional crystallisation
Gurnards Head	Within L.E. aurole < 1km from granite outcrop	pillow lavas & high level intrusions	*Primary*: None remaining (probably pyroxene - plagioclase - ilmenite). *Secondary*: actinolite - plagioclase - opaques - biotite, subsequent alteration to chlorite	meta-dolerite with hornfelsic texture.	tholeiitic affinities with enriched E- MORE features
Zennor	Within L.E. aureole < 1 km from granite outcrop	unknown - but probably sill like	*Primary*: None remaining *Secondary*: actinolite - plagioclase - opaques, plus chlorite	meta-dolerite	no detailed geochem available,

Table 2. *Summary of geochemical and textural features (with additional material from Floyd (1993))*

Sample Reference		Source	Location	Fe (ppm)	Ti (PPm)	Zr (ppm)	Y (ppm)
KPEN01#1V	PXRF	Markham	Carn Gwavas	35626	2489	1948.93	110.56
KPEN01#2V	PXRF	Markham	Carn Gwavas	28833	2830	1177.15	100.83
KPEN02#1V	PXRF	Markham	Carn Gwavas	44346	4479	1127.14	95.12
KPEN02#2V	PXRF	Markham	Carn Gwavas	41888	4100	1095.35	106.50
KPEN1A#1V	PXRF	Markham	Carn Gwavas	22900	3170	1019.09	126.05
KPEN1A#2V	PXRF	Markham	Carn Gwavas	37114	3479	1037.76	74.85
KPEN2A#1V	PXRF	Markham	Carn Gwavas	34742	4530	1203.46	89.89
KPEN2A#2V	PXRF	Markham	Carn Gwavas	28507	3524	1103.32	81.36
LE10	LXRF	Floyd	Carn Gwavas	63644	3657	1575.00	127.00
LE11	LXRF	Floyd	Carn Gwavas	64468	3357	1260.00	132.00
LE6	LXRF	Floyd	Carn Gwavas	59797	3957	850.00	150.00
LE7	LXRF	Floyd	Carn Gwavas	66690	4436	780.00	185.00
M27	LXRF	Floyd	Carn Gwavas	38634	3777	775.00	147.00
M62	LXRF	Floyd	Carn Gwavas	23255	4616	1085.00	126.00
M63	LXRF	Floyd	Carn Gwavas	59331	4616	1010.00	117.00
PEN001	LXRF	Markham	Carn Gwavas	36231	3459	1280.00	123.80
PEN002A	LXRF	Markham	Carn Gwavas	39029	3915	1110.00	99.40
PEN002	LXRF	Markham	Carn Gwavas	46723	3879	1028.00	118.20
KPEN04#1V	PXRF	Markham	Mousehole	77431	13478	170.88	22.82
KPEN04#2V	PXRF	Markham	Mousehole	76743	14605	180.26	22.76
M35/3	LXRF	Floyd	Mousehole	91099	13668	98.00	20.00
M37/2	LXRF	Floyd	Mousehole	96283	11450	140.00	28.00
M38	LXRF	Floyd	Mousehole	79144	11150	133.00	26.00
PEN004	LXRF	Markham	Mousehole	80996	13824	162.00	25.80
KPEN03#1V	PXRF	Markham	Penlee Point	62547	11518	296.58	31.95
KPEN03#2V	PXRF	Markham	Penlee Point	86293	11244	153.10	29.64
KPEN03#3V	PXRF	Markham	Penlee Point	78401	20440	274.85	35.66
KPEN03#4V	PXRF	Markham	Penlee Point	83990	19247	168.88	25.79
LE12	LXRF	Floyd	Penlee Point	77542	19543	203.00	37.00
M39	LXRF	Floyd	Penlee Point	93439	24219	208.00	43.00
M45/2	LXRF	Floyd	Penlee Point	110903	16006	137.00	27.00
M55	LXRF	Floyd	Penlee Point	79991	7254	92.00	16.00
M71	LXRF	Floyd	Penlee Point	93789	15287	128.00	29.00
PEN003	LXRF	Markham	Penlee Point	88830	20005	260.00	42.30

Notes: One reading per sample, except where annotated '#nV', etc, for PXRF data, where n represents the n th reading for that sample.

Table 3. Samples analysed by portable XRF (PXRF) and laboratory XRF (LXRF) in order to assess similarity of process.

Location	Number of Samples	Number of Readings	Fe		TI		Sr		Y		Zr		Nb	
			ppm	SD	ppm	SD	ppm	SD	ppm	SD	ppm	SD	ppm	SD
Carn Gwavas	4	8	34244.49	7189.14	3575.07	747.76	91.47	9.13	98.15	16.52	1214.03	303.44	86.88	6.82
Cudden Point	4	9	76919.88	7622.18	7209.62	3113.32	691.08	482.36	10.09	5.95	61.26	14.00	4.99	2.88
Gumards Head	4	8	85791.26	2239.18	15523.64	1619.35	272.28	53.85	35.07	4.89	157.17	19.17	8.85	2.56
Penlee Point	2	6	77567.47	8299.36	15088.50	3906.26	382.76	52.22	28.10	5.22	207.42	61.65	33.85	12.48
Trenow Cove	5	14	69485.64	11552.08	9345.47	1918.02	671.02	354.91	28.49	7.82	357.46	197.31	43.42	23.73
Zennor Point	3	7	68293.94	3987.44	9365.85	624.37	216.02	40.90	17.57	3.52	116.93	10.27	6.99	3.47
Group I Axes	50	118			14026.00	4421.00					184.60	37.60		

Table 4. Summarised data used in Figures 3 and 4.

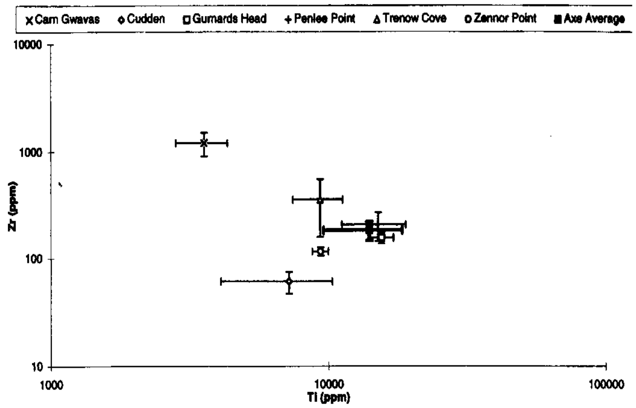

Figure4

Ti-Zr plot with average and range of geochemical data for Group 1 axes superimposed on the fields for greenstones derived from various localities in west Cornwall. Note that the mean overlaps Penlee and Gurnards Head, but is separate from other localities.

preparation. In this context the PXRF data was used to generate a chemical data-base from a number of greenstone localities in west Cornwall and also analyse a selection of Neolithic axes (referred to as Group 1 axes) thought to have been manufactured from greenstone material in the Mount's Bay area.

A selection of major and trace element binary plots allows a degree of selective discrimination of the different greenstone localities. Different plots highlight the specific chemical features of the greenstones, although there is often considerable overlap between some localities. More work needs to be carried out to identify the best set of geochemical discriminants. At the present time the Ti-Zr diagram provides the best discrimination, but this must be used in conjunction with other specific characteristics, such as high or low abundances of Ni, Cr, Fe and Y.

Finally the average and range of Group 1 axes plotted on the Ti-Zr diagram reveals an encouraging overlap with one Mount's Bay site (Penlee) and only partial overlap with the other greenstone locality data. The hypothesis that Mount's Bay was the source of Group I axes remains unproven, but these results and ongoing work have a good chance of resolving the 50 year old issue.

ACKNOWLEDGEMENTS

We would like to thank Peter Webb and Olwen Williams-Thorpe for valuable comments on early drafts of this paper and to The Ian Gass Fund of the Open University for assistance with the provision of the PXRF.

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