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Multi-Element Analysis of Soils from Scottish Historical Sites. Interpreting Land-Use History Through the Physical and Geochemical Analysis of Soil

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Arable farming in the Highlands and Islands of Scotland by the 18th century was characterized by labour-intensive techniques of husbandry and the transfer of fertilizing resources from their extensive non-arable sector to their arable land. Such practices had a considerable impact on soil development and characteristics. The use of inductively coupled plasma-mass spectrometry (ICP-MS) in this study has allowed the geoarchaeological significance of a broad range of elements to be investigated on soils collected from a former settlement site on the Trotternish peninsula, Isle of Skye. The on-site geochemical data are expressed as ''enrichment factors'' relative to the mean concentration of the control samples. Of the elements investigated in this study Ba, La, Ce, Pr, K, Cs, Th, and Rb were found to be indicative of human settlement at Greaulin, whilst the spatial enrichment patterns for Ca and Sr suggest additions of shell sand to the arable land over the central region of the site. Vanadium, Co, Zn, Pb, Ni, Ga, Sc, Mg, Cu and U all showed minimal or no on-site enrichment. The generally lower on-site levels of percentage loss-on-ignition (relative to the control soils) are interpreted as reflecting the former use of the fields for cultivation. Interpretation of the P results, in the light of documentary evidence, suggest that some of the 19th century croft farmsteads may have reoccupied sites formerly inhabited during pre-clachan times. Hence, the clachan at Greaulin may not have been as long-standing a settlement feature as traditionally envisaged.

Keywords: GEOARCHAEOLOGY, ICP-MS, SCOTLAND, SOIL ANALYSIS.

Introduction

Human occupation and cultivation of an area may profoundly influence the local environment and the physical and chemical properties of soils may be significantly altered as a direct result of this human activity. In order for a chemical element to be of use for archaeological diagnostic purposes it must fulfil three criteria. Firstly, human activity must alter the natural concentration of this element across the site. Secondly, this influence must be readily apparent in comparison with natural background concentrations and variability of the element. Thirdly, any alteration needs to be fixed in the soil in a persistent form so as to ensure a lasting record of human occupation. Given favourable soil conditions, and provided recent land-use has not obscured prehistoric and historic patterns, variations in the concentration of certain chemical elements across a site have been shown to be of value to archaeologists in a number of different situations, for example, in the identification of settlement sites when surveying a large area (e.g. Sjoberg, 1976) and to locate and delimit site boundaries (e.g. Heidenreich & Navratil, 1973; Konrad, Bonnichsen & Clay, 1983; Cavanagh, Buck & Litton, 1988). Soil analysis has also been used to distinguish different functions or land-use activity over a site, and to aid identification and interpretation of settlement features such as hearths, mounds and livestock enclosures (e.g. Provan, 1971; Conway, 1983; Konrad, Bonnichsen & Clay, 1983; Craddock *et al.*, 1985; Davidson, Harkness & Simpson, 1986; Bethell & Carver, 1987). However, in spite of the wide diversity of problems to which soil geochemistry has been applied, only a limited number of chemical elements are routinely analysed in this context. Phosphorus (P) is generally considered to be the most important element in the identification and interpretation of settlement sites. However, other elements typically determined include calcium (Ca), magnesium (Mg), potassium (K) and the trace elements copper (Cu), zinc (Zn) and strontium (Sr).

Modern instrumentation with multi-element capabilities now allows a greater range of elements to be routinely considered in geoarchaeological work. An inductively coupled plasma-mass spectrometry (ICP-MS), housed at the Institute of Earth Studies at Aberystwyth, has allowed the geoarchaeological significance of a broad range of elements to be investigated.

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Rationale

Occupation and settlement of an area will tend to enrich the soil in certain elements, such as K, Mg and P, due to the accumulation of general site occupation debris and excreta, whilst other soil parameters, such as organic matter content, may be depleted through associated anthropogenic activity such as cropping. Investigation of the spatial variation of a range of elements and soil properties should therefore allow different types of land-use activity to be distinguished over an area. In this paper, we describe how the multi-element analysis of soils from a former settlement site on the Isle of Skye is indeed of use in interpreting the site's land-use history.

Traditional Highland and Hebridean farming

As previously mentioned, human settlement and associated agricultural activity can cause marked changes in the nature and properties of soils. Such changes are most distinct in marginal environments where humans have to cope with soils which pose severe limitations and restrictions on population growth and expansion (Davidson & Simpson, 1984). Extensive areas of high, rugged ground combined with high rainfall, low accumulated growing temperatures, short ripening periods, frequent storms and exposure, all work against farming in the Highlands and Islands of Scotland. Work by historical geographers and historians on the nature of farming practice in the Hebrides prior to crofting and the clearances has suggested that the density of settlement, coupled with the techniques and husbandries used, had a profound and widespread impact on the environment (Dodgshon, 1988, 1993*a*).

Arable farming in the Highlands and Islands by the 18th century was characterized by labour-intensive techniques of husbandry with vast inputs of labour needed to help transfer manurial supplements like seaweed, peat and turf (Dodgshon, 1988, 1992). The widespread use of hand-cultivation tools like the spade and caschrom* also required considerable investments of labour. Bound up with the use of the spade and caschrom was the construction of lazy-beds. Their construction involved the heaping of the soil into narrow ridges or rigs. The technique was ideally suited to difficult and marginal ground as it created a deeper, better drained soil and readily allowed the incorporation of manurial supplements (Glentworth, 1979). Indeed, a key objective in the construction of lazy-beds was to improve soil drainage. These practices employed by Highland townships had a considerable impact on the soil environment, most obviously on the physical characteristics of the soils (e.g. topsoil depth), although the chemical status of the soils is likely to have been affected as well.

Study Site

Greaulin (grid reference NG 390674–397684) is located about 3·5 km north of Uig on the Trotternish peninsula, Isle of Skye (Figure 1). Former settlement and farming activity at Greaulin has resulted in a complicated pattern of walls, trenches and lazy-beds derived from various phases of occupation. The detailed site plan of Greaulin (Figure 2) indicates the main field boundaries visible at the site today which include the head dyke (a large earth and stone bank which delimits the township's arable land and part of the better pasture from the rough grazing or common pasture), and the croft boundaries.

Site layout prior to 1811

A plan of the area by Mathew Stobie in 1764 (Figure 3) reveals that the settlement of Greaulin was organized in a manner typical of a nucleated Highland clachan, with houses and huts grouped closely together and surrounded by open arable fields that were probably organized on a runrig basis. The paucity of data documenting this type of settlement prior to the 18th century has led to assumptions concerning the history of such farming townships. In the context of the Western Highlands and Islands, the accepted interpretation has long been that the clachan is an archaic form of settlement, with some workers even suggesting a late prehistoric origin. However, recently Dodgshon (1993*b*, 1993*c*) has questioned this early origin, suggesting instead that the clachan, together with its runrig layout, may not have developed until the 13th century or later, replacing an earlier pattern of more loosely dispersed settlement.

Although one cannot assume that Stobie's map of Greaulin provides an exact depiction of the settlement layout as it stood in the mid-18th century, it can be assumed that the map conveys the general character and former location of the settlement. There is no visible evidence on the site today to indicate the location of the former clachan, although the head dyke, also marked on Stobie's map (Figure 3), is still a prominent feature of the site.

Site layout from 1811 to the present

Reorganization of the site into six crofts occurred in 1811. The boundaries defining five of the six crofts are still visible today as regular stone walls running roughly east to west across the site (Figure 2). During the 1820s, the initial six crofts were each subdivided into two, making a total of 12 tenancies on the site. Eleven of the 12 croft houses are clearly visible today, most with adjoining kailyards (small enclosures attached to the farmsteads or located nearby, where

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*Caschrom: a heavy-duty spade with carved handle and thick, narrow blade.

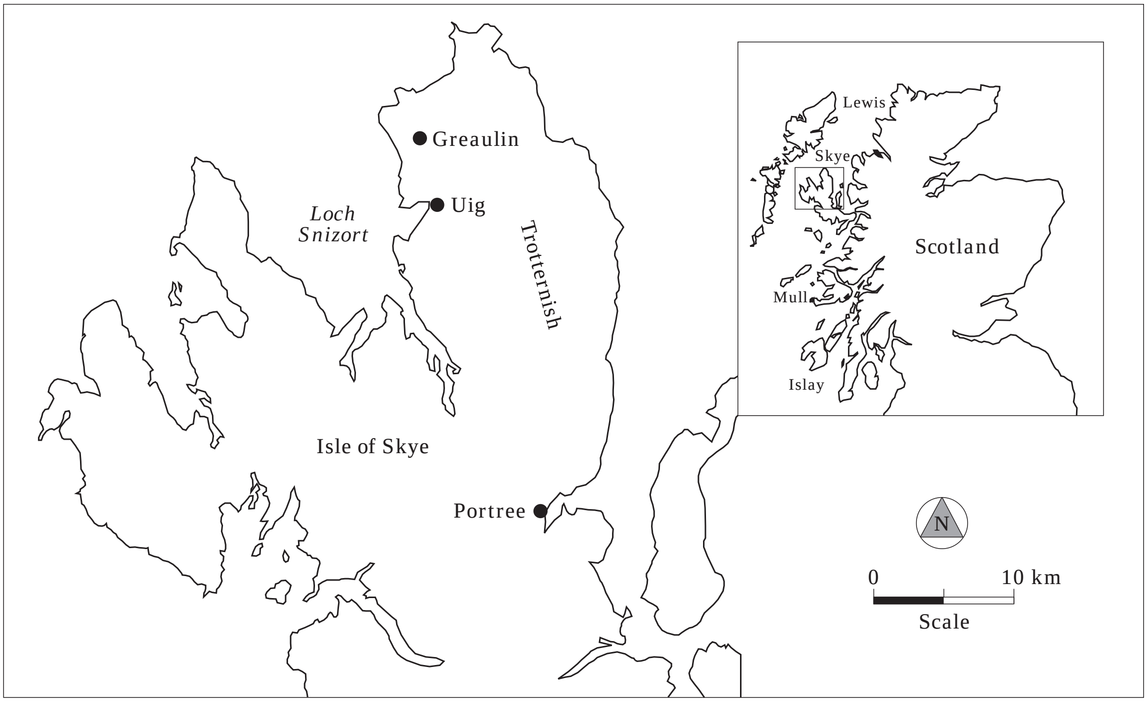

Figure 1. Location of Greaulin on the western side of the Trotternish peninsula, Isle of Skye.

vegetables were grown and compost was stored; Dodgshon, 1993*b*). The remaining croft farmstead and adjoining kailyard is thought to have been located further north, as indicated by the number 12 on Figure 2.

By the 1850s, the site had been cleared to become an extension of pastureland for the nearby farm of Monkstadt. During the 20th century the land has seen little modification and is currently used as rough grazing for sheep and cattle.

Methodology

Soil sampling

A random stratified sampling system (Page, Miller & Keeney, 1982) was undertaken at Greaulin in an attempt to maximize the study area covered by the investigation whilst collecting a minimum number of soil samples. The study site was divided into a number of subunits (Figure 4), in this case individual fields, and topsoil samples were collected from 60 fields using the W traverse method (Rowell, 1994). This is a widely used technique to obtain a representative composite sample from an area. Obviously, this technique of bulking samples does not provide information regarding variability over an area and we recognize that our bulked samples may not be totally representative of the individual fields considered in this study.

The top horizon of the soil profile was sampled (field observation and subsequent analysis indicated that mineralized (A horizon) and humose (Ah horizon) material was collected). All samples were from freely drained topsoils and were collected using a hand-operated screw-type auger. According to field size, between five and nine individual auger borings were made in each field, with the topsoil collected and bulked together. Topsoil depth was recorded at each sampling point. The soils were stored in kraft paper bags prior to sample pretreatment in the laboratory.

In addition, systematic sampling was undertaken using a regular grid layout superimposed over two adjacent croft farmsteads (see dashed square on Figure 4). The results of this systematic sampling are presented elsewhere (Entwistle, 1994), and only the mean concentration of each of the soil properties investigated in the two craft kailyards are presented here.

In order to assess the extent to which these on-site soils have been affected by human activity, it was necessary to estimate the ‘‘natural’’ background concentration and the ‘‘natural’’ variability of the soil properties under investigation. Thirty control topsoil samples (i.e. A and Ah horizon material) were collected at regular intervals, about 15 m east of the head dyke (Figure 4). The location may not provide a wholly acceptable control site, but bearing in mind the difficulty of finding such a site, i.e. one which reflects the settlement area studied without human impact, the location selected was considered the best available. Indeed, these off-site control soils are not considered to be totally unaffected by human activity (although there is no evidence, either physical or historical, to

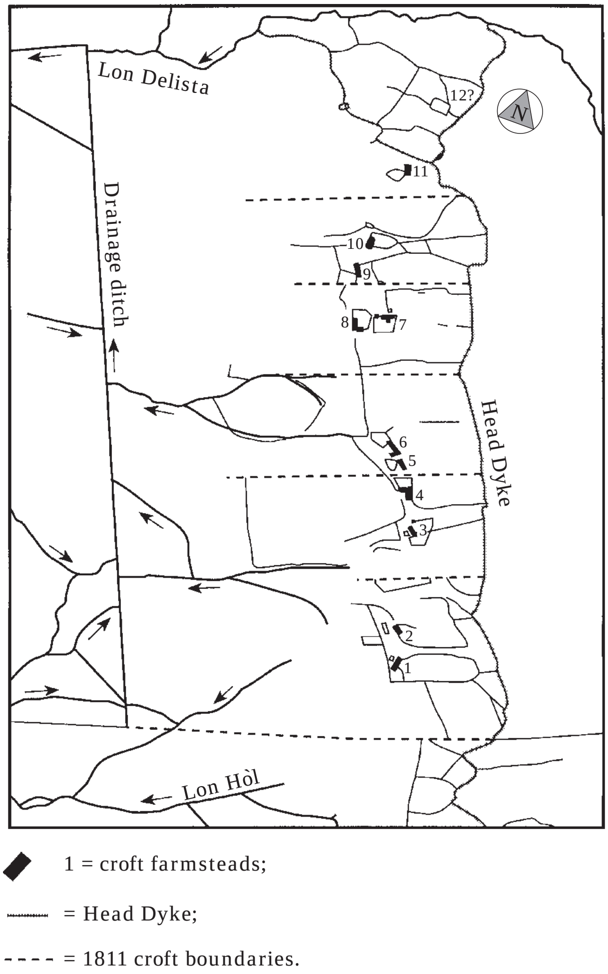

Figure 2. Field layout of the study site at Greaulin indicating the main lines of drainage and field boundaries which are visible today. The numbers refer to the 12 croft farmsteads at the site.

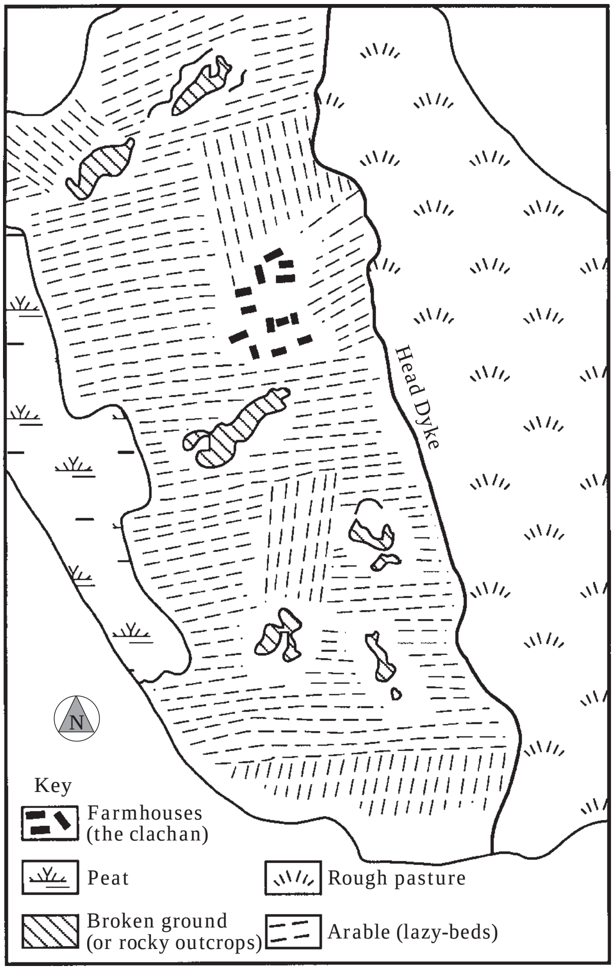

Figure 3. Plan of Greaulin taken from a map by Mathew Stobie (1764).

suggest that this area has ever been subject to stripping or cutting), but human impact is thought to have been minimal relative to the land west of the head dyke.

Soil preparation and chemical analysis

Determination of soil pH was undertaken immediately or shortly after sample collection, using a portable pH meter (Whatman International Ltd), with built-in pH probe and automatic temperature compensation. Measurements were made on a 1:2·5 (w/v) suspension of fresh soil and distilled water (Bascomb, 1987).

On return to the laboratory, the soil samples were air-dried, gently disaggregated with a pestle and mortar, and sieved through a 2-mm nylon mesh sieve. Subsequent analyses were undertaken on this <2 mm ‘‘fine-earth fraction’’ of the soil. Organic matter content was estimated gravimetrically (by loss-on-ignition at 375°C), following oven drying for 16 h at 105°C (after Ball, 1964). Subsamples for chemical analysis were powdered and homogenized in a tungsten-carbide rotating ball mill to facilitate acid digestion, and to ensure that a representative portion of the bulk sample was analysed.

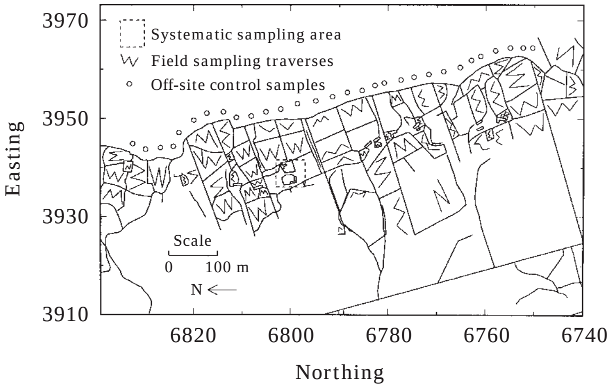

Figure 4. Field layout at Greaulin indicating location of the ‘‘W’’ soil-sampling traverses, off-site control samples and the systematic sampling grid.

The use of a nitric–perchloric (HNO_3–$HClO_4$, 4:1 v/v) digestion procedure was selected in our study for sample decomposition (after Thompson & Wood, 1982). Following this acid digestion procedure, 30 elements were determined semi-quantitatively, using the simultaneous multi-element capability of ICP-MS. Phosphorus was determined separately by ICP-MS in fully quantitative mode, and the elements K, Ca and Mg were determined by flame emission and atomic absorption spectrometry, respectively. These laboratory procedures (including quality control) are described in detail by Entwistle & Abrahams (1997).

Results

Off-site control soils and a comparison with on-site soils

As frequency distributions of geochemical data are often skewed (and estimations of the mean and the standard deviation of a data set are only valid when the data are normally distributed; Johnston, 1986), the Kolmogorov–Smirnov test was undertaken on the control data to check for normality of each of the 34 elements considered in our study, as well as the off-site pH, topsoil depth and percentage loss-on-ignition. Each of the soil properties investigated on the control samples had an observed distribution which was not significantly different from the normal distribution at the 0·05 confidence level (Table 1).

The Kolmogorov–Smirnov test was also undertaken on the on-site data to check for normality. Thirty of the elements from the on-site geochemical data, and the pH, topsoil depth and percentage loss-on-ignition data, had observed distributions which were not significantly different from the normal distribution at the 0·05 confidence level (Table 2). For K and Rb, a logarithmic transformation was required for normality, whilst the stronger reciprocal transformation was required for Cs and Th (Table 2).

The mean values (calculated using the transformed data sets where necessary) of the analyses undertaken on the control soils and the on-site soils are presented in Table 3. The mean values for the control soils indicate an average topsoil depth of 20 cm, being typically slightly acidic (mean = 5·7 pH units) and peaty (mean percentage loss-on-ignition = 25). In comparison, for the on-site soils, the mean topsoil depth is 30 cm, the mean level of percentage loss-on-ignition is 16 and the soils are slightly acidic (mean = 5·9 pH units). Of the elements determined in this study (on both the on-site and off-site soils) many fall within the typical range for soils (Kabata-Pendias & Pendias, 1984; Allen, 1989; Alloway, 1995), with the exception of Rb, Sr, U, Th and the rare earth elements (REEs), which are slightly lower than typical.

The Student's *t*-test was undertaken (using transformed data where necessary for K, Rb, Cs and Th), to establish whether there was a significant difference between the on-site and the control data (Table 3). With the exception of V, Cr, Ga, Pb and U, the calculated value of *t* was greater than the critical value, indicating that there is a significant difference (at the 0·05 significance level, using a two-tailed test) between the on-site and off-site soils for all but the aforementioned elements.

Table 1. The Kolmogorov–Smirnov Z value and associated probability (P) for the off-site (control) data. The Kolmogorov–Smirnov procedure was used to test for goodness of fit of the off-site data to a normal distribution (at a significance level of 0·05)

Element	Kolmogorov –Smirnov *Z* value	Two-tailed *P*
Off-site pH	0·8583	0·0500 ns
Off-site topsoil depth	1·3661	0·8573 ns
Off-site % loss-on-ignition	0·7305	0·6600 ns
Magnesium (Mg)	0·8853	0·4133 ns
Potassium (K)	0·5125	0·9554 ns
Calcium (Ca)	0·8297	0·4967 ns
Phosphorus (P)	0·9815	0·2903 ns
Scandium (Sc)	1·0362	0·2332 ns
Vanadium (V)	1·1555	0·1384 ns
Chromium (Cr)	1·1315	0·1545 ns
Cobalt (Co)	0·8860	0·4123 ns
Nickel (Ni)	0·8042	0·5374 ns
Copper (Cu)	0·5280	0·9432 ns
Zinc (Zn)	0·7137	0·6883 ns
Gallium (Ga)	1·0644	0·2073 ns
Rubidium (Rb)	0·9088	0·3808 ns
Strontium (Sr)	1·2660	0·0811 ns
Yttrium (Y)	0·5585	0·9141 ns
Cesium (Cs)	0·5095	0·9576 ns
Barium (Ba)	0·9896	0·2813 ns
Lanthanum (La)	0·4954	0·9668 ns
Cerium (Ce)	1·0274	0·2418 ns
Praseodymium (Pr)	0·7917	0·5576 ns
Neodymium (Nd)	0·6828	0·7397 ns
Samarium (Sm)	0·5965	0·8689 ns
Europium (Eu)	0·6827	0·7398 ns
Gadolinium (Gd)	0·7535	0·6212 ns
Terbium (Tb)	0·5880	0·8797 ns
Dysprosium (Dy)	0·8575	0·4540 ns
Holmium (Ho)	0·7471	0·6320 ns
Erbium (Er)	0·6582	0·7792 ns
Thulium (Tm)	0·8765	0·4260 ns
Ytterbium (Yb)	0·5222	0·9480 ns
Lutecium (Lu)	0·5086	0·9582 ns
Lead (Pb)	0·6658	0·7671 ns
Thorium (Th)	0·8005	0·5433 ns
Uranium (U)	0·6920	0·7245 ns

ns = not significant at the 0·05 level, therefore accept the H_0.
H_0 = the data are drawn from a normally distributed population.
H_1 = the population from which the data are drawn is not normally distributed.

Data analysis

The statistical difference observed between the on-site and off-site (control) samples for the majority of parameters studied was investigated further by the calculation of "enrichment factors". The on-site geochemical data are expressed as enrichment factors relative to the mean value of the control (off-site) samples. Thus, the enrichment factor of each element for each on-site sample was calculated by dividing the concentration of the on-site sample by the mean

Table 2. The Kolmogorov–Smirnov Z value and associated probability (P) of the on-site data. The Kolmogorov–Smirnov procedure was used to test for goodness of fit of the on-site data to a normal distribution (at a significance level of 0·05)

Element	Kolmogorov –Smirnov *Z* value	Two-tailed *P*
On-site pH	0·6052	0·4528 ns
On-site topsoil depth	0·8699	0·4356 ns
On-site % loss-on-ignition	0·5949	0·8709 ns
Magnesium (Mg)	0·9491	0·3286 ns
*Potassium (K)	1·2379	0·0933 ns
Calcium (Ca)	1·1983	0·1132 ns
Phosphorus (P)	1·3321	0·0575 ns
Scandium (Sc)	0·5472	0·9256 ns
Vanadium (V)	0·6148	0·8442 ns
Chromium (Cr)	0·3547	0·9996 ns
Cobalt (Co)	0·5638	0·9083 ns
Nickel (Ni)	0·4185	0·9948 ns
Copper (Cu)	0·6877	0·7316 ns
Zinc (Zn)	1·0473	0·2227 ns
Gallium (Ga)	0·9790	0·2932 ns
*Rubidium (Rb)	0·9760	0·2966 ns
Strontium (Sr)	1·0544	0·2162 ns
Yttrium (Y)	0·7709	0·5921 ns
†Cesium (Cs)	0·7923	0·5568 ns
Barium (Ba)	1·2969	0·0692 ns
Lanthanum (La)	1·1065	0·1727 ns
Cerium (Ce)	0·9945	0·2760 ns
Praseodymium (Pr)	0·5896	0·8778 ns
Neodymium (Nd)	0·7268	0·6663 ns
Samarium (Sm)	0·6072	0·8546 ns
Europium (Eu)	0·5014	0·9630 ns
Gadolinium (Gd)	0·6634	0·7710 ns
Terbium (Tb)	0·7785	0·5795 ns
Dysprosium (Dy)	0·7094	0·6955 ns
Holmium (Ho)	0·6458	0·7985 ns
Erbium (Er)	0·5346	0·9375 ns
Thulium (Tm)	0·7375	0·6482 ns
Ytterbium (Yb)	0·5252	0·9455 ns
Lutecium (Lu)	0·8014	0·5418 ns
Lead (Pb)	0·8527	0·4612 ns
†Thorium (Th)	0·8064	0·5338 ns
Uranium (U)	0·9740	0·2989 ns

ns = not significant at the 0·05 level, therefore accept the H_0.
H_0 = the data are drawn from a normally distributed population.
H_1 = the population from which the data are drawn is not normally distributed.
*Kolmogorov–Smirnov *Z* value and probability following logarithmic transformation of the data.
†Kolmogorov–Smirnov *Z* value and probability following reciprocal transformation of the data.

Table 3. Results of analyses undertaken on the 30 off-site control soils and the 60 on-site soils at Greaulin. Elemental concentrations are in µg/g air-dried soil

Soil property/ element	Off-site soils Mean	On-site soils Mean	Student's *t*-test*
Topsoil depth (cm)	20	30	7·25
pH	5·7	5·9	4·56
% loss-on-ignition	25	16	5·78
Ca	1,840	3,930	7·85
Mg	16,300	24,100	4·05
K	430	818	5·06†
P	1,010	1,440	5·09
Sc	7·56	12·0	6·95
V	140	115	ns
Cr	283	189	ns
Co	35·1	50·5	4·53
Ni	157	213	3·67
Cu	70·3	102	5·40
Zn	107	126	2·28
Ga	16·5	16·5	ns
Rb	3·13	6·69	5·77†
Sr	43·2	123	9·51
Y	4·30	8·71	14·63
Cs	0·65	1·12	4·56‡
Ba	50·2	115	10·30
La	4·61	10·3	11·46
Ce	9·80	21·7	12·28
Pr	1·08	2·43	13·56
Nd	4·85	9·94	13·39
Sm	1·10	2·15	12·64
Eu	0·36	0·62	11·45
Gd	1·29	2·32	11·58
Tb	0·19	0·33	11·60
Dy	0·96	1·77	12·09
Ho	0·16	0·30	11·92
Er	0·44	0·86	12·81
Tm	0·05	0·11	11·33
Yb	0·34	0·68	13·78
Lu	0·05	0·09	11·42
Pb	10·2	7·79	ns
Th	0·48	1·15	9·91‡
U	0·45	0·47	ns

*H_1 On-site ≠ off-site; all *t*-values quoted are significant at the 95% confidence interval.
ns = not significant at the 95% confidence interval.
† = calculated on logarithmically transformed data.
‡ = calculated on reciprocally transformed data.

off-site value. An estimation of what level of enrichment constitutes a minimum significant enrichment was arbitrarily set at two standard deviations (2σ) above the control mean. This value (off-site control mean + 2σ) was divided by the mean off-site value to give an estimation of the minimum significant enrichment factor for each element.

Any on-site enrichment factors which exceed the minimum significant enrichment factor (i.e. control mean + 2σ/control mean) are considered to be significantly enriched. The calculation of minimum significant enrichment factors enables the identification of areas which are enriched in one or more element, and allow comparisons to be made between the various elements, which typically occur over a wide range of concentrations.

Cluster analysis was undertaken on the on-site soil geochemical data in order to simplify the data set and group together those elements with similar spatial enrichment patterns. There is a great proliferation of clustering techniques and consideration must be given not only to the choice of method but also to the choice of which similarity or distance measure to use (Everrit, 1980). Each method has some distinctive features and one important consideration is often the nature of the resulting clusters (Krzanowski, 1988). In this study, the analysis was carried out using the multivariate statistical package MVSP (Kovach, 1990) and Ward's minimum variance method was undertaken on a squared Euclidean distance dissimilarity matrix, following conversion of the data to enrichment factors to standardize the data set. This agglomerative clustering

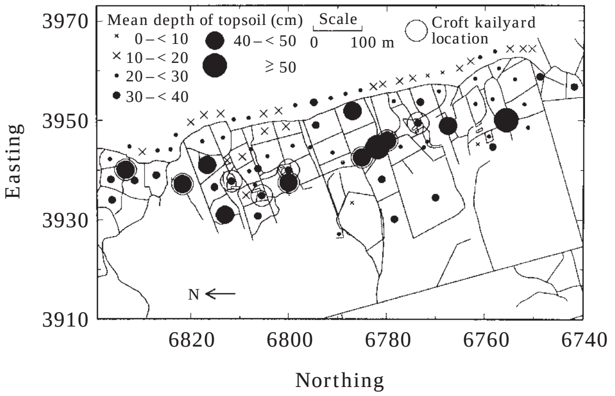

Figure 5. On-site and off-site topsoil depth (cm). The off-site control samples were collected along a transect positioned above the head dyke.

technique was selected as it exaggerates differences in the data and produces a dendrogram with large changes in level and clearly identifiable clusters.

Table 4 shows the Pearson product moment correlation coefficients between each of the elements investigated in this study. Owing to the nature of geochemical data, correlation between elements is often statistically significant. For example, as might be expected, there is significant correlation (at the 0·05 level) between each of the elements comprising the REE group (i.e. Lanathanum (La)–Lutecium (Lu)). As Pearson's product moment correlation is a parametric test of correlation between variables, for each of the elements which did not exhibit a normal distribution (see Table 2) correlation was undertaken using a non-parametric test, in this case Spearman's Rank (Table 5). However, with regards to the application of cluster analysis in this study, as the overall objective of clustering was simply to group together those elements with similar spatial enrichment patterns, the fact that certain elements are significantly correlated is not believed to be a problem.

On-site spatial variability of the parameters under investigation

Topsoil depth, pH and percentage loss-on-ignition The majority of on-site fields have a topsoil depth greater than the mean off-site value of 20 cm (Figure 5), whilst the on-site soils show slightly higher soil pH values, mostly associated with the kailyards, compared to the control sites (Figure 6). Topsoils sampled over the southern end of the site indicate levels of percentage loss-on-ignition similar to those found in the off-site control soils, although generally over the site there is a lower level of percentage loss-on-ignition relative to the control samples (Figure 7).

Geochemical data The calculated value of ''minimum significant enrichment'' is used to define the upper limit of the lowest category on the resulting geochemical data plots. Any enrichment between 1·0 and this value is considered to be of minor significance, whilst a value of less than 1·0 indicates a depletion. Four main groups of elements were identified by cluster analysis of the geochemical data and represent the four main patterns of spatial enrichment observed at Greaulin (groups I, II, III and IV on Figure 8). These four groups form the basis of the following discussion.

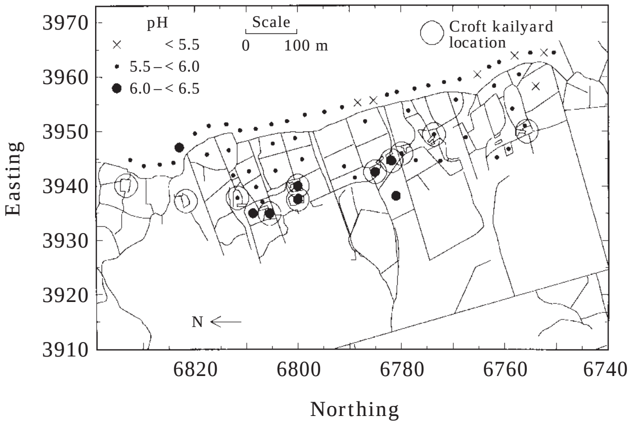

Figure 6. On-site and off-site pH values determined for the soils sampled at Greaulin. The off-site control samples were collected along a transect positioned above the head dyke.

Cluster group I contains the elements Ca, Ba, Y and the REEs (i.e. Lanathanum (La)–Lutecium (Lu)). These members of group I can be further divided into three subgroups on the basis of the results shown in the dendrogram (Figure 8) and their levels of enrichment.

(a) Represented by Ca (Figure 9). The spatial pattern of Ca enrichment is notably different from the other elements of cluster group I. The highest levels of Ca enrichment (>3·5) occur within the central region of the site, to the south of what is believed to be the site of the former clachan (as indicated on Stobie's map of 1764). Low levels of enrichment are observed in the northern and southern extremities of the site and in several of the croft kailyards.

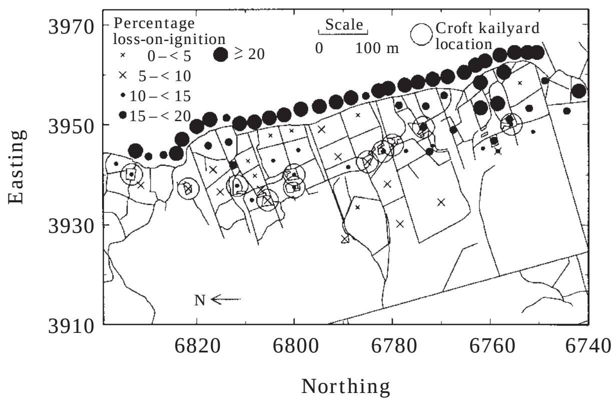

Figure 7. On-site and off-site percentage loss-on-ignition in the soil sampled at Greaulin. The off-site control samples were collected along a transect positioned above the head dyke.

Table 4. Matrix of Pearson's product moment correlation coefficients between all of the elements, determined on the on-site samples, which exhibit a normal distribution. Numbers in bold indicate significance at the 0·05 level

	Mg	Ca	P	Sc	V	Cr	Co	Ni	Cu	Zn	Ga	Sr	Y	Ba	La
Mg	*1·0000*														
Ca	−0·0233	*1·0000*													
P	**0·5021**	0·1552	*1·0000*												
Sc	0·0786	0·1818	**−0·2778**	*1·0000*											
V	0·0721	−0·1228	**0·2636**	−0·0471	*1·0000*										
Cr	0·0346	0·0346	0·2158	−0·1923	**0·7098**	*1·0000*									
Co	0·2500	**0·3486**	−0·1595	**0·5848**	0·0917	0·0168	*1·0000*								
Ni	**0·2921**	−0·0994	−0·1309	**0·2879**	0·0130	0·2409	**0·8344**	*1·0000*							
Cu	0·2476	0·0180	−0·0154	**0·5129**	−0·0008	0·0138	**0·7247**	0·7226	*1·0000*						
Zn	**0·3509**	0·0652	**0·4719**	0·1647	**0·3522**	0·3299	0·4568	0·3888	0·4565	*1·0000*					
Ga	**−0·3659**	0·1889	**−0·4355**	0·5655	0·0784	0·1387	**0·4772**	0·3015	0·3816	0·1845	*1·0000*				
Sr	**0·4003**	0·0636	−0·1223	**0·6396**	−0·0156	−0·1707	**0·5338**	0·3888	0·5152	0·1922	0·0675	*1·0000*			
Y	0·2010	0·1756	−0·0690	**0·8536**	−0·0297	**0·2669**	0·4789	0·1192	**0·3651**	0·2131	**0·3914**	0·6402	*1·0000*		
Ba	−0·0219	0·1730	0·1739	**0·6816**	−0·1228	**−0·3689**	0·3868	−0·0931	0·1181	0·1523	**0·4883**	0·3851	0·8016	*1·0000*	
La	0·0380	0·1823	−0·1501	**0·7103**	−0·0520	**−0·3565**	0·2872	−0·1287	0·1147	0·0548	**0·4109**	0·4427	0·8659	0·9380	*1·0000*
Ce	0·0438	0·1851	−0·1852	**0·7353**	−0·0914	**−0·3698**	0·3486	−0·0652	0·1468	0·0707	**0·4243**	0·4862	0·8618	0·9314	0·9861
Pr	0·0998	0·1772	−0·1444	**0·7781**	−0·0628	**−0·3224**	0·3699	−0·0008	0·1847	0·0871	**0·4037**	0·5442	0·9078	0·8733	0·9620
Nd	0·1559	0·1828	−0·0870	**0·7638**	0·0193	−0·2200	**0·4175**	0·0404	0·2353	0·2010	**0·3959**	0·6074	0·9313	0·8561	0·9276
Sm	0·0174	0·0974	−0·2153	**0·7823**	−0·0724	**−0·3032**	0·4670	0·1128	**0·3146**	0·1616	**0·4887**	0·5202	0·8986	0·8323	0·8820
Eu	0·1374	0·1136	−0·1069	**0·7087**	0·0411	−0·2073	**0·4127**	0·1115	**0·2779**	0·2396	**0·3833**	0·6032	0·8608	0·7258	0·7991
Gd	0·0935	0·2193	−0·1339	**0·7430**	−0·0331	0·2311	**0·4685**	0·1612	**0·3151**	0·2675	0·5087	0·5363	0·8799	0·7632	0·8136
Tb	−0·0173	**0·2827**	−0·0328	**0·6292**	0·0602	−0·1231	**0·3393**	0·0551	0·1796	**0·2951**	0·5241	0·3816	0·7716	0·7282	0·7946
Dy	0·0850	0·2348	−0·0604	**0·6376**	−0·0143	−0·1306	**0·4218**	0·1535	0·1868	**0·2850**	0·4794	0·4129	0·7691	0·6774	0·7150
Ho	0·1619	**0·3058**	−0·1106	**0·6849**	−0·0215	−0·1107	**0·4528**	0·2031	0·2266	0·2166	**0·4173**	0·5710	0·7725	0·6530	0·7271
Er	0·1739	0·2085	−0·0917	**0·7193**	0·0479	−0·0318	**0·4864**	0·2193	0·2412	**0·3232**	0·4883	0·5318	0·8076	0·7287	0·7579
Tm	0·0846	0·1713	−0·1086	**0·6352**	−0·0299	−0·2133	**0·4855**	0·1762	0·2068	**0·2915**	0·4760	0·4627	0·7757	0·7377	0·7631
Yb	**0·3083**	0·1277	0·0002	**0·6466**	0·0630	−0·0602	**0·4805**	0·2287	**0·2546**	0·3263	0·3245	0·5365	0·8161	0·6591	0·7381
Lu	0·1408	0·0978	−0·1049	**0·6223**	−0·1161	−0·2086	**0·5320**	0·2103	**0·2692**	0·2140	**0·2908**	0·5204	0·7607	0·6287	0·6567
Pb	**−0·2628**	0·2539	−0·0468	**0·3589**	0·1688	−0·0834	0·0086	**−0·3251**	−0·0253	0·0787	**0·3325**	0·0119	**0·3679**	0·5400	0·5181
U	−0·2451	0·1025	−0·0565	**0·4431**	0·0602	−0·1032	0·0252	**−0·3798**	−0·1931	−0·0130	**0·4495**	0·0245	**0·5553**	0·7558	0·7287

	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Pb	U
Ce	*1·0000*														
Pr	**0·9713**	*1·0000*													
Nd	**0·9282**	0·9607	*1·0000*												
Sm	**0·8912**	0·9195	0·9077	*1·0000*											
Eu	**0·8109**	0·8534	0·8636	0·8631	*1·0000*										
Gd	**0·8209**	0·8766	0·8999	0·8849	0·8933	*1·0000*									
Tb	**0·8020**	0·8386	0·8538	0·8133	0·8201	0·8882	*1·0000*								
Dy	**0·7358**	0·7851	0·7886	0·8075	0·8065	0·8448	0·8531	*1·0000*							
Ho	**0·7485**	0·8117	0·8377	0·7996	0·7830	0·8573	0·8455	0·8492	*1·0000*						
Er	**0·7722**	0·8206	0·8553	0·8117	0·7977	0·8671	0·8364	0·8552	0·8631	*1·0000*					
Tm	**0·7844**	0·8286	0·8336	0·8283	0·8108	0·8780	0·8022	0·7949	0·7949	0·8001	*1·0000*				
Yb	**0·7569**	0·8257	0·8593	0·8190	0·8366	0·8574	0·7791	0·7765	0·8215	0·8683	0·8269	*1·0000*			
Lu	**0·6791**	0·7383	0·7637	0·7638	0·7485	0·7563	0·6443	0·6814	0·7451	0·7162	0·7915	0·7847	*1·0000*		
Pb	**0·5240**	0·4632	0·4279	0·4121	0·3034	0·3327	0·4227	0·3555	0·2878	0·3397	0·2874	0·2265	0·2323	*1·0000*	
U	**0·6921**	0·6420	0·6318	0·6091	0·5152	0·5311	0·6155	0·5115	0·4492	0·5152	0·4970	0·4256	0·3656	0·5940	*1·0000*

Table 5. Matrix of Spearman's rank correlation coefficients between all of the elements, determined on the on-site samples, which do not exhibit a normal distribution. Numbers in bold indicate significance at the 0·05 level

	Mg	K	Ca	P	Sc	V	Cr	Co	Ni	Cu	Zn	Ga	Rb	Sr	Y	Cs	Ba
K	− 0·1594	*1·0000*	0·2473	− **0·2773**	0·6123	− 0·0528	− 0·2390	0·2047	− 0·1305	0·1179	0·2203	**0·4492**	0·7833	0·2912	0·6070	0·7662	0·6964
Rb	− 0·0121	**0·7833**	0·2520	− 0·1637	**0·7579**	0·0222	− 0·2488	**0·3613**	0·0081	**0·2655**	0·3777	0·3858	*1·0000*	**0·5377**	0·7745	0·8274	0·8121
Cs	− 0·1143	**0·7662**	0·2433	− 0·1083	**0·6095**	− 0·0-63	0·1554	0·1554	− 0·2430	− 0·0040	**0·2880**	0·4300	0·8274	0·2629	0·6935	*1·0000*	**0·7862**
Th	0·0405	**0·7490**	0·2250	− 0·1209	**0·7338**	− 0·0576	− 0·1695	**0·3422**	− 0·0324	0·1540	**0·3345**	0·3975	0·8223	0·3836	0·8081	0·7986	0·9050

	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Pb	U	Th
K	**0·7240**	0·7009	0·6845	0·6688	0·6694	0·5839	0·6191	0·6304	0·5619	0·5598	0·5434	0·5265	0·5603	0·4721	0·5328	0·5089	0·7490
Rb	**0·8740**	0·8773	0·8502	0·8270	0·7735	0·7644	0·7440	0·7518	0·6397	0·6499	0·6481	0·6721	0·6358	0·6032	0·5229	0·4631	0·8223
Cs	**0·8034**	0·7857	0·7841	0·7555	0·6957	0·6962	0·7296	0·7663	0·6375	0·6079	0·6133	0·6971	0·6489	0·5472	0·5057	0·6365	0·7986
Th	**0·9101**	0·8761	0·8662	0·8375	0·8456	0·6985	0·7154	0·6840	0·7180	0·6745	0·7412	0·6881	0·7107	0·6374	0·5196	0·6574	*1·0000*

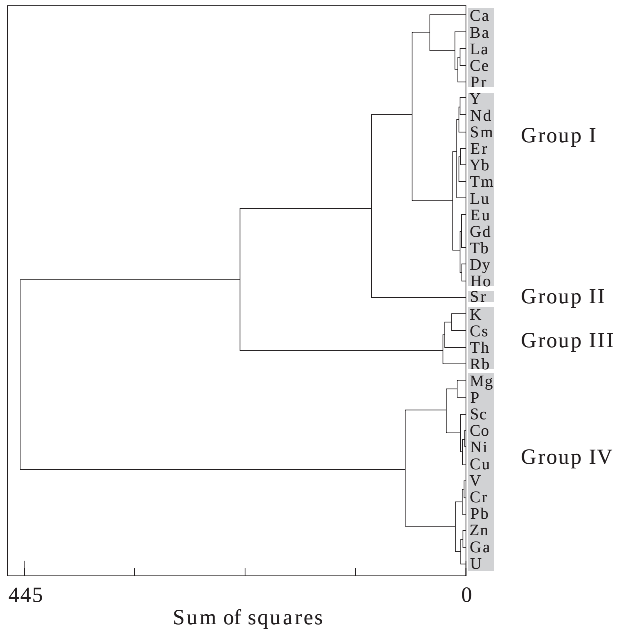

Figure 8. Dendrogram to show results of the cluster analysis of the geochemical data obtained from the analysis of the on-site soils at Greaulin. Four main groups were identified (groups I, II, III and IV).

(b) Represented by Ba (Figure 10), but also containing the elements La, Ce and Pr. All four elements indicate significant enrichment in what is believed to be the general area of the former clachan and on the lower slopes of the study site (NG 39306770–39356790), with enrichment values up to 4·5 times those of the control soils. The spatial patterns indicated by subgroup B are similar to those shown by subgroup C, though levels of enrichment are generally higher in the former.

(c) Represented by Nd (Figure 11), but also containing the elements Y, Sm, Er, Yb, Tm, Lu, Eu, Gd, Tb, Dy and Ho. The majority of fields and kailyards indicate significant on-site enrichment compared to the control soils, though levels of enrichment are less than 3·5. Most of the elements in this group show an indistinct pattern of enrichment associated with the presumed location of the former clachan and the fields on the lower slopes of this study site.

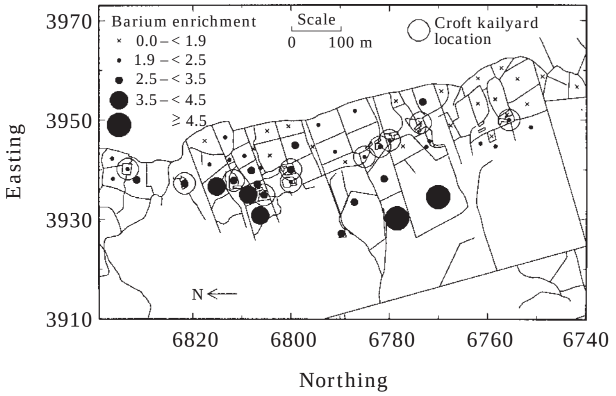

Figure 10. On-site enrichment of barium (Ba) at Greaulin. Ba belongs to cluster group I(B). The calculated minimum significant enrichment for Ba is 1·9.

Cluster group II is represented by Sr (Figure 12). The spatial enrichment pattern of Sr is similar to that of the cluster group I elements, as low enrichment values are grouped in the southern part of the site with higher concentrations evident within the central region. However, although Sr indicates enrichment around what is believed to be the general area of the former clachan, the pattern is not fully coincident with that shown by the group I elements. Only minor Sr enrichment is associated with the majority of croft kailyards sampled in this study.

Group III contains the elements K (Figure 13), Cs, Th and Rb. All four elements indicate similar spatial enrichment patterns, with high levels of enrichment (up to and greater than 3·5) associated with the presumed location of the former clachan. In addition, high levels of enrichment are also observed on the lower slopes of this study site, although this enrichment is less

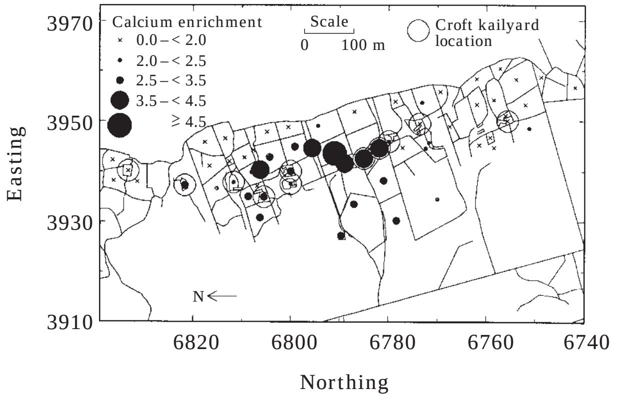

Figure 9. On-site enrichment of calcium (Ca) at Greaulin. Ca belongs to cluster group I(A). The calculated minimum significant enrichment for Ca is 2·0.

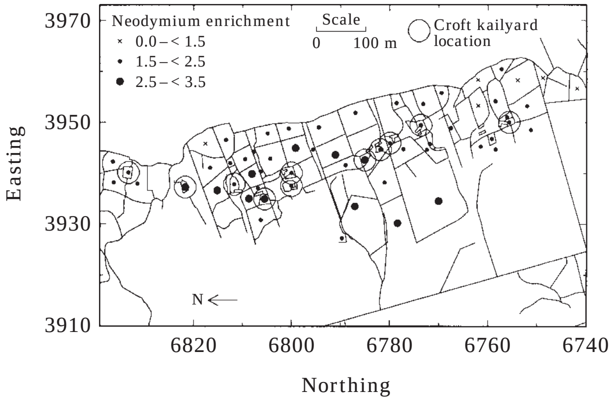

Figure 11. On-site enrichment of neodymium (Nd) at Greaulin. Nd belongs to cluster group I(C). The calculated minimum significant enrichment for Nd is 1·5.

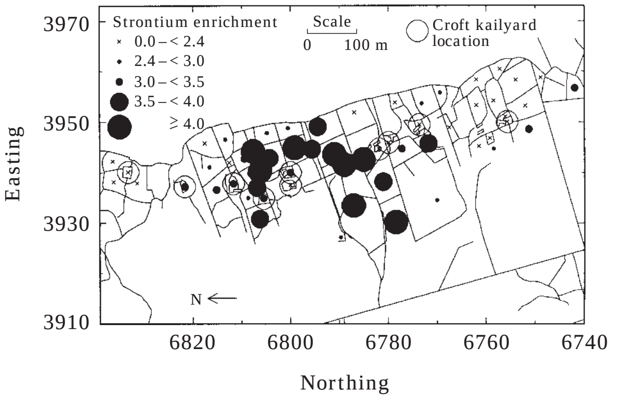

Figure 12. On-site enrichment of strontium (Sr) at Greaulin. Sr belongs to cluster group II. The calculated minimum significant enrichment for Sr is 2·4.

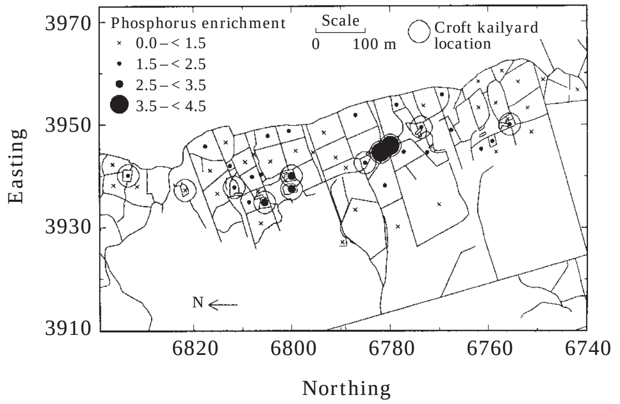

Figure 14. On-site enrichment of phosphorus (P) at Greaulin. P belongs to cluster group IV. The calculated minimum significant enrichment for P is 1·5.

consistent between the elements. The remaining parts of the site indicate little enrichment relative to the control soils.

Group IV contains 12 elements which have a wide range of geochemical affinities. These elements are Mg, P, Sc, Co, Ni, Cu, V, Cr, Pb, Zn, Ga and U. The patterns of enrichment shown by P and Zn are indicated in Figures 14 and 15, respectively, and are described below.

Phosphorus. Phosphorus indicates only minor on-site enrichment in the fields over the study site, although significant enrichment (i.e. greater than 1·5) is associated with 10 out of the 11 croft kailyards sampled. Only minor P enrichment is associated with the soils sampled over the presumed location of the former clachan.

Zinc. Zinc, along with other group IV elements (with the exception of P), indicates minor on-site enrichment, with no samples showing a concentration above the minimum significant value determined using the control soils.

Discussion

Soil is effectively a palimpsest, with each successive phase of human activity modifying the soil record. The spatial patterns observed for the soil properties investigated at Greaulin may therefore result from a whole series of anthropogenic influences which complement any natural variability brought about by, for example, variations in underlying parent material, or fluctuations in processes such as leaching and sorption.

Virtually all of the soils at Greaulin (both on-site and off-site) fall within a relatively narrow pH range (5·6–6·3 pH units; i.e. medium to slightly acidic; Woods, 1983). Soil pH affects many chemical processes, such as bioavailability and vulnerability of elements to leaching (Wedephol, 1969; Kabata-Pendias & Pendias, 1984), and the narrow range of values

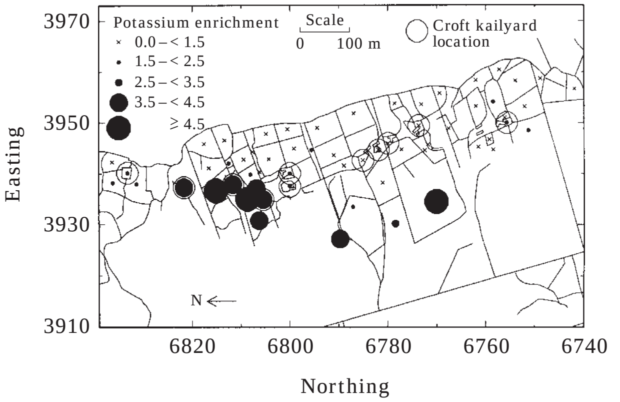

Figure 13. On-site enrichment of potassium (K) at Greaulin. K belongs to cluster group III. The calculated minimum significant enrichment for K is 1·5.

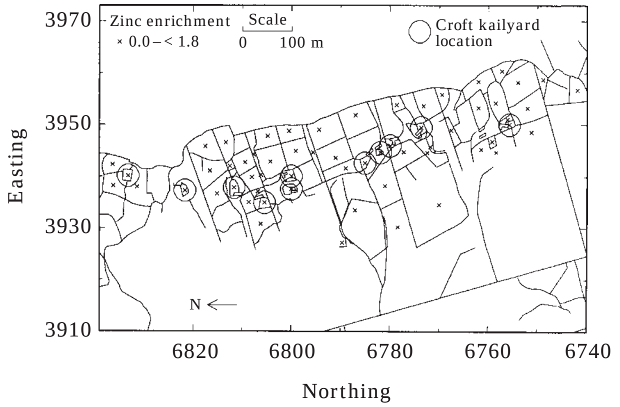

Figure 15. On-site enrichment of zinc (Zn) at Greaulin. Zn belongs to cluster group IV. The calculated minimum significant enrichment for Zn is 1·8.

observed at Greaulin indicates that the elemental variations observed within the soils of the study site may largely be due to other factors, such as human influences, rather than variations in pH.

Geochemistry of the soils of the presumed former clachan settlement

Anthropogenic modification of the soil by human habitation may differ from that caused by arable cultivation and livestock grazing. Modification of the soil record due to human habitation may result from the accumulation of organic and mineral-rich waste. Analysis of the soils at Greaulin revealed a significant enrichment (up to and greater than 3·5 times the mean concentration of the control soils) of K, Cs, Rb and Th (the group III elements) associated with the presumed location of the former clachan. Previous studies have found a similar link between elevated levels of soil K and settlement activity (e.g. Lutz, 1951; Jefferies & Woods, 1983). Although no work has been published regarding the behaviour of Cs, Rb and Th in relation to human settlement activity, a link is suggested by the results from Greaulin.

The geochemical characteristics of Cs and Rb are similar to those of K (Wedephol, 1969; Kabata-Pendias & Pendias, 1984). Therefore it is not too surprising to find similarities in the spatial patterns of these three elements. Specific anthropogenic sources of Rb and Cs are not known, but Wedephol (1969) suggests that plants may concentrate Cs in their tissues. Thus additions of food scraps, animal fodder and bedding may result in Cs enrichment. Once incorporated into the soil, Cs may be rapidly and strongly sorbed by solid soil material rendering it relatively immobile (Kabata-Pendias & Pendias, 1984). As such, Cs may be a reliable record of settlement activity, and its correspondence with areas of K enrichment and the presumed location of the former clachan seems to substantiate this.

The literature relating to Th geochemistry suggests that once present in the soil environment, the subsequent mobility and loss of Th through leaching may be limited by the formation of only slightly soluble precipitates and by adsorption onto clays and organic matter (Wedephol, 1969; Kabata-Pendias & Pendias, 1984). However, Th is not known to be an essential element in any organic process and is not known to accumulate to any significant degree in animal or vegetable matter. Hence, the reasons for the correspondence between elevated levels of Th and the presumed location of the former clachan site are unclear.

Elements of group I(B) (i.e. Ba, La, Ce and Pr) also indicate enrichment over the presumed former clachan site. Barium is usually associated with K in geochemical processes due to the similarity of their ionic radii (Kabata-Pendias & Pendias, 1984), whilst La, Ce and Pr all belong to the lanthanide series of elements, commonly referred to as the REEs. Bone and teeth contain REEs in trace amounts (Steinnes, Dahm & Furseth, 1984; Tuross, Behrensmeyer & Eanes, 1989) and the enrichment of these elements over the former clachan site may therefore be related to the accumulation of general occupation debris. Bone ash also contains appreciable amounts of Ba (Sowden & Stitch, 1957; Grupe & Hummel, 1991) as does the ash of marine plants (e.g. seaweed, algae), wood ash, mollusc shells and the ash of legumes (Wedephol, 1969).

Despite minor intra-element variation, an additional area of enrichment, common to all the elements discussed in this section, is evident on the lower slopes of the study site (i.e. NG 39306770–39356790). By implication this pattern of enrichment may be indicative of former settlement in this area. However, the paucity of sample points delimiting this region prevents further interpretation.

Soil modification associated with the post-1811 croft kailyards

It was hypothesized earlier that the kailyards, located adjacent to the croft farmhouses, post-dated the clachan settlement and were created following the 1811 reorganization of the area. Significant enrichment of P (i.e. greater than 1·5 times that found in the control soils) was found in 10 out of the 11 kailyards sampled, with minor levels of P enrichment associated with the former clachan site, and negligible enrichment over most of the remaining fields (Figure 14). Phosphorus enrichment is associated with additions such as bone, animal bodies, excreta and general occupation debris (McCawley & McKerrell, 1972; Proudfoot, 1976). For example, P enrichment in the kailyards is not too surprising given that these areas were used for the storage of manure, compost and general occupation debris, and the loss of nutrients from such heaps is inevitable during storage. Phosphorus losses due to inefficient storage and redistribution of manure are estimated to have been as high as 20% in farming townships such as that at Greaulin (Dodgshon & Olsson, 1988).

The occurrence of only minor P enrichment associated with the site of the former clachan is somewhat surprising given that such settlements are believed by some workers to be archaic settlement features and that cultural additions of P are relatively stable and long-lived in the soil environment. In contrast, it is thought that the croft farmsteads and kailyards were only used for a comparatively short period of time (about 40 years) and yet higher levels of P enrichment are observed in several of the croft kailyards than over the presumed clachan site. The chemical evidence appears to raise new questions about patterns of settlement on the site. Stobie's map of 1764 may provide some explanation. At first sight, the shaded areas marked on Stobie's map may be interpreted as rock outcrops or ''broken'' ground (Figure 3), but there is no evidence of such features on the site today.

Several of these shaded areas roughly correspond with the location of several of the 19th century croft farmsteads. Indeed, enrichment of those elements indicative of human settlement at Greaulin (i.e. K, Rb, Cs, Th) is observed on the lower slopes of the site which roughly corresponds with one of these shaded areas. There is a case for arguing that what Stobie was indicating by these shaded areas are the sites of abandoned houses, probably occupied before Greaulin became nucleated as a settlement (Dodgshon, 1993*c*). The reoccupation of some of these sites following the 1811 reorganization would account for the correspondence between some of the 19th century croft farmsteads, Stobie's shaded areas and the patterns of P enrichment. Such a theory would explain why some of the croft kailyards have higher P levels compared to those of the former clachan site, the higher P values being a relic of pre-clachan settlement activity in addition to the post-1811 activity. Interpreted in this way, the clachan at Greaulin would need to be seen as only one period or phase of occupation in a complex history of settlement.

On-site maxima in topsoil depth are largely associated with the croft kailyards, where depths are generally in excess of 40 cm (Figure 5). Typically, under natural soil conditions the A horizon (topsoil) rarely exceeds a thickness of 40 cm (Van de Westerningh, 1988). According to the classification system of the Soil Survey of England and Wales (Avery, 1973), soils with anthropogenically modified topsoils greater than 40 cm thick are referred to as ''man-made humus soils'', although such soils are often referred to as ''plaggen'' (e.g. Conry, 1971, 1974; Staines, 1975; Van de Westerningh, 1988). The genesis of such thick topsoils at Greaulin, largely associated with the kailyards, possibly reflects the inevitable build-up of material given the use of such areas for the storage of manure and general occupation debris, such as discarded and used building materials like turf and thatch. The use of additives such as turf and peat as fertilizer would also increase on-site topsoil depths relative to the off-site soils, and numerous studies have suggested a link between the anthropogenic additions of such materials and the occurrence of deep topsoils and farm mounds (Pape, 1970; Conry, 1971; de Bakker, 1980; Simpson, 1985).

Previous studies (Zabel, 1976; Weymouth & Woods, 1985) have highlighted the potential of pH determinations to locate and delimit sites of human habitation. However, at Greaulin the pH of the soil is similar in both the on-site and off-site samples, although areas of slightly decreased acidity are generally associated with the croft kailyards (Figure 6). Many workers have attributed on-site pH anomalies above the ''natural'' soil reaction to additions of refuse and ash, particularly wood and hearth ash (Weide, 1966; Hurley & Heidenreich, 1971; Woods, 1983). Such additions are likely to have been associated with human activity at Greaulin and may account for the slight decrease in acidity observed in the croft kailyards.

Soil modification associated with farming activity

The highest levels of P enrichment at Greaulin are associated with the croft kailyards, while the surrounding fields generally show low to negligible P enrichment. The low P enrichment observed in the majority of fields is at first sight surprising given that traditional Hebridean farming relied heavily upon additions of livestock manure, rich in P, to maintain soil fertility. However, a similar pattern was observed by Edwards (1983) in the Old Kinord area, Aberdeenshire. Edwards suggested that the lower concentrations of P in the fields, as compared with the settlement area, resulted from the use of the land for arable cultivation as such areas will experience P depletion due to the removal of the crop at harvesting (Provan, 1973; Limbrey, 1975; Weymouth & Woods, 1985). Traditional Highland farming was characterized by poor yields and nutrient deficient soils. A large proportion of any applied P (e.g. from manurial inputs) was probably utilized by the growing crop or else depleted by erosional loss of the P-rich topsoil, thereby reducing the amount of P to be converted into an insoluble, immobile form in the soil (Limbrey, 1975; Proudfoot, 1976). Work by Morel *et al.* (1989) has indicated that crop utilization of applied P is greater following application of P in spring compared to utilization following autumn or winter applications. As most P additions to the soil occurred in spring (Davidson & Simpson, 1984; Dodgshon, 1988) it is likely that a large proportion of this would be removed by the growing crop.

High levels of Sr enrichment are evident in the fields over the central area of the site, with lower levels associated with the croft kailyards (Figure 12). The pattern of Ca enrichment is less distinct, though again less enrichment occurs in the croft kailyards compared to the fields over the central area (Figure 8). The greater susceptibility of Ca to leaching, relative to Sr, may account for the observed differences between the elements at Greaulin (Mitchell, 1971; Ottaway & Mathews, 1988). As both Ca and Sr are constituent elements in shells, their enrichment at this site is thought to result from the application of shell sand to the cultivated fields. The use of shell sand as a fertilizer is well documented for many regions in the Hebrides (Darling, 1955; Geddes, 1955). The lower levels of Ca and Sr enrichment in the croft kailyards at Greaulin, compared to the fields, is interpreted as evidence that the shell sand was applied directly to the fields without prior storage in the kailyards.

Soil properties exhibiting depletion or minimal modification

Previous work has highlighted the potential use of Cu and Zn to indicate areas of human habitation (Sokoloff & Carter, 1952; Cook & Heizer, 1965; Bintliff *et al.*, 1990). However, neither of these elements indicates an enrichment above the minimal significant value. These elements occur as trace constituents in cultural additions such as manure, and as the off-site control soils contain on average 70·3 μg Cu/g and 107 μg Zn/g, deposition over long periods of time would be required for significant enrichment to occur. In addition, Cu and Zn are relatively mobile in acidic soils, such as those at Greaulin, and hence are susceptible to loss by leaching (Pickering, 1986). Both these factors may play a role in explaining the lack of anthropogenic enrichment of Cu and Zn over the site.

Limited or no on-site enrichment above the minimum significant value was observed for Sc, Ga, U, V, Co, Cr, Pb, Ni and Mg. The lack of on-site Mg enrichment above the minimum significant value is not too surprising given that it is one of the major elements in soil and typically has a high natural spatial variability. Hence a large change in concentration would need to occur before any anthropogenic impact is recognized. As in this study, Ottaway & Mathews (1988) found no enrichment of Cr, Pb and Ni in an area of former settlement (Gomolava, central former Yugoslavia), and concluded that little archaeological information was to be gained through their analysis.

The level of percentage loss-on-ignition in the on-site soils at Greaulin is generally lower than that of the off-site control sites (Figure 7). On the basis of the historical evidence and the chemical evidence discussed in the preceding sections, it is believed that this site was predominantly used for arable cultivation until the 19th century clearance. Sandor (1992) examined soil changes initiated by prehistoric cultivation in S.W. New Mexico and found lower levels of organic matter in the cultivated soils compared to the off-site control soils. Approximately 90% of the dry matter in livestock manure is organic, and additions of organic matter to the farmed fields at Greaulin would largely be associated with the application of manure. However, as well as being high in organic matter, manure also contains sufficient N to enable the rapid microbial breakdown of the readily decomposable organic matter. As more organic matter is added to the soil, more micro-organisms can be supported and this population increase results in faster decomposition of the organic matter (Limbrey, 1975; Stein, 1992). Consequently, the organic matter in manure is not persistent in the soil environment and any enrichment is likely to disappear over a relatively short space of time (Provan, 1973; Menzies & Chaney, 1974). Similarly, organic material with a high content of the alkaline earth elements, particularly Ca, is more readily decomposed than organic material low in these elements (Brady & Weil, 1996). Therefore, any coincident additions of Ca-rich material (e.g. shell sand at Greaulin) would help maintain high levels of organic matter decomposition. Any additions of seaweed at our study site may also have had a short residence time in the soil environment. Davidson & Simpson (1984) found no trace of seaweed in deep topsoils from around Quinni farm, Orkney, where historical sources document the application of such material.

Conclusions

Eight of the elements investigated in this study (Ba, La, Ce, Pr, K, Cs, Th and Rb) were found to be significantly enriched over the presumed site of the former clachan, and hence to be indicative of the former human settlement site at Greaulin. The patterns of enrichment for Ca and Sr are thought to result from additions of shell sand to cultivated fields over the central region of the study area. Similarly, depletion in the levels of percentage loss-on-ignition observed in the majority of fields is interpreted as reflecting their former use for arable cultivation, with consequent enhanced rates of organic matter decomposition.

On-site enrichment was observed for the group IA elements (Y, Nd, Sm, Yb, Er, Tm, Lu, Eu, Gd, Tb, Dy and Ho), particularly over the central area. However, the level of enrichment was relatively low and did not form any readily interpretable pattern in terms of anthropogenic activity. Of the remaining elements determined, V, Co, Zn, Pb, Ni, Ga, Sc, Mg, Cu, Cr and U indicated minimal or no on-site enrichment above the minimum significant value. Only minor variations of pH were observed over the on-site study area, with most samples falling within the pH range of the control soils.

The P results are difficult to interpret, although we suggest that some of the 19th century croft farmsteads may have reoccupied sites formerly inhabited during pre-clachan times, and that the clachan at Greaulin may not have been as long-standing or as persistent a settlement feature as traditionally envisaged.

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