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Geochemistry and mineralogy of weathered basalt from Morvern, Scotland

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SUMMARY. The chemical, mineralogical, and textural changes involved in the weathering of basalt have been traced through various stages from fresh rock (which has a cation exchange capacity of 10 meq/100 g due to the presence of a swelling chlorite mineral) to reddened basaltic rubble consisting of interstratified montmorillonite-illite, hematite, and anatase. The cation exchange capacities of the rocks increase progressively with the formation of secondary clay from labradorite as Al, Fe, and Ti accumulate and Si, Mg, Ca, and Na are depleted—much of the K is retained in the secondary clay mineral. The weathering is considered to be contemporaneous with the formation of the Antrim bauxites but not so intense.

TERTIARY basalts outcrop over an area of 72 square km on the peninsula of Morvern, forming part of the much more extensive Thulean volcanic province which includes Northern Ireland, the Faeroes, Iceland, and the coasts of Greenland as well as the west coast of Scotland and the Hebridean Islands. On Morvern the lavas consist of some 450 m of basalt, the petrography and field relationships of which have been described in detail in the Geological Survey Memoir of the area (Bailey *et al.*, 1924). Most of the basalt is little altered by weathering and occurs as a hard, coherent rock but the upper parts of the flows are often stained red by weathering and sometimes are overlain by a red earth or bole (Richey *et al.*, 1961). Lightly weathered olivine-basalt in Northern Ireland contains large quantities of smectite with a high cation exchange capacity (CEC) (Alexander and Heal, 1974) and these materials contribute significantly to the high CECs of the soils derived from the Irish basalts.

This paper describes the mineralogy and chemistry of incoherent, weathered, basaltic material, less weathered basalt, and fresh basalt from Morvern, paying particular attention to the clay-size particles in these materials.

Samples

The samples were collected from a road cutting some 10 m high at Killundine, Morvern (National Grid Reference NM 588497), approximately 140 m above sea level in an area with an annual precipitation of about 1500 mm.

The most highly weathered sample consists of very friable brick-red coloured fragments up to 3 cm across. This material, which is overlain by younger basalt, occurs in an uneven, discontinuous layer of maximum thickness about 40 cm at the top of a flow and incorporates mottled material containing white patches and spots.

Samples were also taken of slightly weathered basalt, one (A) from near the more highly weathered material and another (B), for comparison, from some 20 m distant. To complete the sequence for tracing chemical and mineralogical changes, the fresh rock, which is a hard, coherent basalt showing no signs of weathering, was sampled from as near the weathered specimens as possible.

Methods

The weathered samples were disaggregated by gentle grinding under water, dispersed by ultrasonic treatment, and the fraction $< 2\ \mu m$ separated by standard sedimentation procedures. X-ray diffraction traces were obtained using a Philips 2 kW diffractometer with iron-filtered Co-$K\alpha$ radiation, divergent, receiving, and scatter slits of $1°$, 0.2 mm, and $1°$ respectively, and a scanning rate of $2°\ 2\theta$ per minute. Samples were presented in two forms—as oriented aggregates (for the clay fractions), to enhance the 00l reflections, and as random powders, to obtain the hkl reflections.

Prior to chemical analysis, all rocks were ground for 2 minutes in a Tema mill—in the case of the fresh rock, this ground material was also used for the separation of the clay fraction. The chemical composition of the total materials and their respective clay fractions was obtained by X-ray fluorescence spectrometry using the methods of Norrish and Hutton (1969) for sample fusion and for corrections for background and inter-element effects. The $K\alpha$ lines of all the elements were measured on a Philips PW 1540 spectrometer equipped with a 2 kW Cr tube (and a Ti filter for the determination of Mn) and a flow-proportional counter fitted with a 1 μm polypropylene window, using pulse-height discriminator settings appropriate to each element. Sodium was determined on pressed-powder pellets, international rock standards being used for calibration.

Differential thermal analysis (DTA) curves were obtained on an apparatus based on that designed by Mitchell and Mackenzie (1959), using a 150 mg sample equilibrated at 56% relative humidity and heated at 10 °C per minute in a nitrogen atmosphere.

CEC values were determined by the neutral ammonium acetate method as modified by Mackenzie (1951).

Results and interpretation

Rocks

Chemical composition. The chemical composition of the total materials is given in Table I along with their CEC and Parker's Index. This index (Parker, 1970), which is calculated on a hydrous basis and depends on the proportions of alkali and alkaline earths present in the sample, allowance being made for the bond strengths of these elements with oxygen, is a measure of the degree of chemical weathering that has taken place. Thus the decreasing value of the index from 66 in the fresh rock to 21 in the red material is due to the overall depletion of the sum of Na, Mg, K, and Ca, although the amount of K is in fact at a minimum in the basalt. Silica and MnO also are depleted on weathering, whereas Al_2O_3, TiO_2, and Fe_2O_3 apparently accumulate in the more weathered samples.

Depletion of the various elements from the weathering regime is related to the nature of the host primary minerals, the mobility of these elements once they are released by the weathering of these minerals, and the susceptibility of the elements to retention in new phases in their immediate environment. To calculate the true as opposed to the apparent chemical changes during weathering it is necessary to assume that one chemical component is stable and not removed during the weathering process. For this purpose TiO_2 was chosen since, although it has been shown that Ti is slightly soluble and is mobile over limited distances (Hutton, 1977), there is considerable evidence that, after solubilization, it is quickly reprecipitated *in situ* (Bain, 1976; Berrow *et al.*, 1978), the net overall effect being no change in the total TiO_2 content. The procedure used in calculation of chemical changes was to multiply the various values obtained in the analysis of the samples by a factor that equated the quantities of TiO_2 in the fresh rock and in the samples. These calculated values were then compared with the analysis of the weathered material and the percentage depletion calculated for each component. The results are summarized in fig. 1 from which an indication of depletion rates can be obtained.

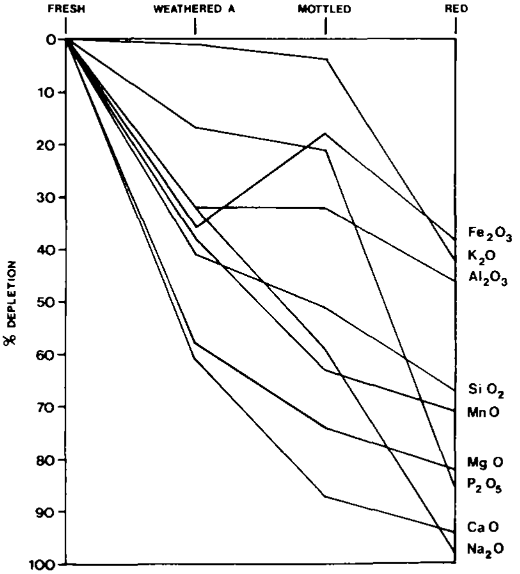

FIG. 1. Diagram illustrating the depletion of the oxides in the rocks assuming TiO_2 is immobile.

Between each successive weathering stage in the sequence fresh basalt, weathered basalt A, mottled sample, and red sample, every oxide except Fe_2O_3 is progressively depleted; Fe_2O_3 initially decreases but increases during the formation of the mottled sample (Al_2O_3 remains constant during this change) and becomes depleted again at the later stage. Weathered basalt B is not considered in this sequence as it is not in close proximity to the other samples but it is at a similar stage to weathered basalt A. No data are available to distinguish between ferrous and ferric iron, but presumably iron released in ferrous form from the primary minerals is oxidized to ferric which has a much lower mobility. The extent of depletion of the elements on weathering varies, nearly all Ca and Na but less than half the Fe, K, and Al having

TABLE I. *Chemical composition (in weight %) and cation-exchange capacity (in meq/100 g) of rocks (R) and clay fractions (C)*

	Fresh		Weathered A		Weathered B		Mottled		Red	
	R	C	R	C	R	C	R	C	R	C
SiO_2	44.3	37.9	42.2	37.6	39.3	43.1	38.3	41.7	34.0	39.5
TiO_2	1.33	0.13	2.13	0.51	1.95	0.59	2.34	0.63	3.06	1.69
Al_2O_3	14.3	10.7	15.6	16.3	15.5	20.3	17.3	18.9	17.9	18.8
Fe_2O_3*	11.8	15.4	12.3	11.4	13.4	6.17	17.1	6.41	17.0	11.0
MnO	0.13	0.18	0.13	0.56	0.14	0.22	0.09	0.28	0.09	0.49
MgO	10.3	15.8	6.92	9.71	7.54	4.96	4.76	9.11	4.20	4.84
CaO	9.1	2.51	5.75	1.53	4.49	1.37	2.08	1.69	1.14	1.11
Na_2O	1.4	1.5	1.5	1.6	1.1	1.2	1.0	1.0	0.1	0.1
K_2O	0.26	0.15	0.42	0.14	0.33	0.52	0.44	0.38	0.35	0.44
P_2O_5	0.15	0.11	0.20	0.22	0.18	0.13	0.21	0.23	0.06	0.08
SO_3	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.05
LOI†	6.70	16.04	13.45	20.41	16.11	22.00	16.84	20.35	22.04	21.31
Total	99.77	100.42	100.60	99.98	100.04	100.56	100.46	100.68	99.94	99.41
CEC	10.1	41.9	35.8	71.6	46.7	83.2	61.2	85.8	69.4	83.1
Parker's index	66.4		51.2		45.4		31.3		21.2	

* Total iron as Fe_2O_3.

† Loss on ignition at 1000 °C obtained from undried samples.

Clay fractions equilibrated at approximately 56% relative humidity.

been lost. In the initial stages of weathering Ca and Mg are removed most rapidly, the weathered rock containing less than half the quantities of CaO and MgO in the fresh rock. This reflects not only the dissolution of the small amount of calcite in the fresh rock but also the breakdown of labradorite and olivine. Potassium, like Ca and Na, is present in the plagioclase, but unlike the other elements in the feldspar, is removed only slightly during the first two stages of weathering as it is retained in the main clay mineral being formed.

There is a clear inverse relationship between CEC and Parker's Index (Table I), the CEC (and loss on ignition, LOI) increasing due to the formation of secondary clay minerals as the Index decreases with loss of alkalis and alkaline earths.

Mineralogical composition. The bulk mineralogy of the samples, as revealed by the X-ray diffraction traces in fig. 2, can be related to the changes in chemical composition of the samples (fig. 1). The diffraction trace of the fresh rock (fig. 2*d*) contains many peaks due to feldspar (F) and a few due to pyroxenes (P) with moderate peaks due to clay minerals (C). The weathered rock A (fig. 2*c*) also contains much feldspar but has a reduced pyroxene content and increased amounts of clay. Weathered rock B (not shown on fig. 2) is similar to A but has a slightly higher clay content.

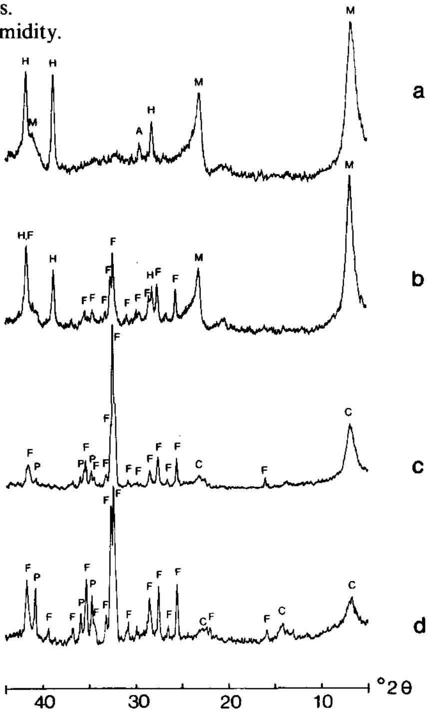

FIG. 2. X-ray diffraction traces of random powder mounts of the ground rocks, using Co-$K\alpha$ radiation. A—anatase; C—clay; F—feldspar; H—hematite; M—montmorillonite; P—pyroxene. *a*—red weathered sample; *b*—mottled weathered sample; *c*—weathered rock (A); *d*—fresh rock.

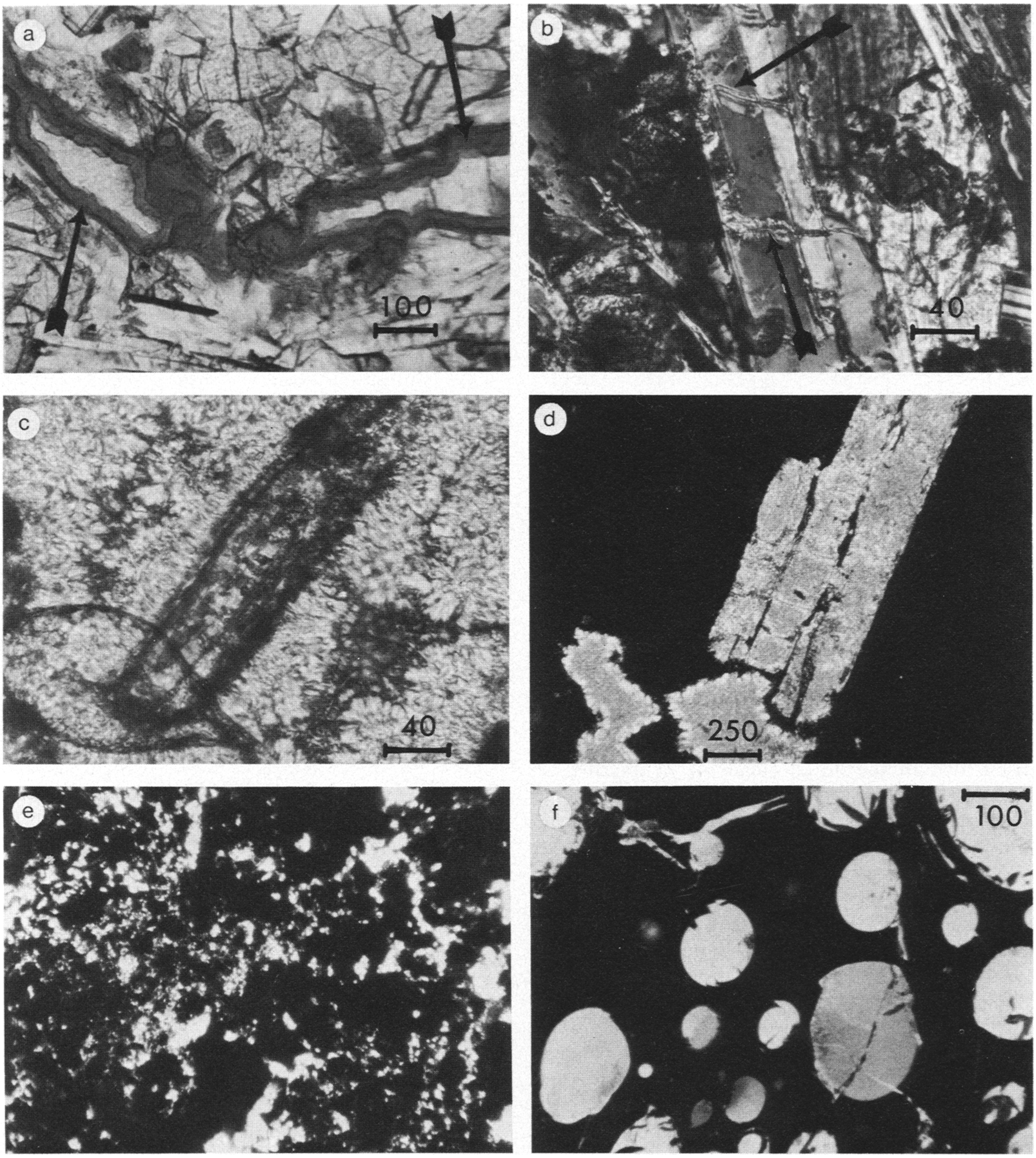

FIG. 3. Photomicrographs of thin sections. *a*—fresh rock showing chloritic material (arrowed) enclosing carbonate; *b*—weathered rock with feldspar showing incipient alteration to clay along cleavage traces (arrowed); *c*—extensive area of clay enclosing remnant of feldspar lath in weathered rock; *d*—feldspar phenocryst converted to clay, enclosed by hematite (black) in mottled material; *e*—intimate mixture of clay (white) and hematite (black) in red weathered material; *f*—spherules of clay in hematite matrix in red weathered material. Lines indicate scale in microns.

The major change in the mineralogy occurs at the mottled stage: in this (fig. 2*b*) all the pyroxene has disappeared, hematite (H) is present in significant proportions (reflecting the accumulation of Fe_2O_3) and the content of clay minerals increases markedly (reflecting the retention of Al_2O_3). Feldspar, which is still present at this mottled stage, is absent in the red material (fig. 2*a*) which is composed of 14 Å clay and hematite with a small amount of anatase.

Textural changes. Accompanying the chemical and mineralogical changes are major changes in the texture of the rocks. The fresh rock consists of a fine-grained, ophitic intergrowth of purplish-coloured augite and plagioclase feldspar: the latter is occasionally zoned and, from the extinction angles of the albite twins, is a labradorite of composition An_{60}. Irregular greenish patches of chloritic material are common, often enclosing carbonate (fig. 3*a*), and there is also a small amount of an iron-ore mineral. There is no indication of any glassy material. In the weathered rock some of the feldspar crystals show incipient alteration to clay along cleavage traces (fig. 3*b*) and some parts of the rock show more extensive areas of clay enclosing remnants of feldspar laths (fig. 3*c*). The mottled material contains areas of hematite with pseudomorphs of clay after feldspar (fig. 3*d*). In the most highly weathered red material the texture of the original basalt has been completely destroyed and the bulk of the rock is an intimate mixture of clay and hematite (fig. 3*e*), although some parts contain spherules of clay in a hematite matrix (fig. 3*f*).

Clay fractions

Chemical composition. There are significant variations in the chemical composition of the clay fractions (Table I). Since the clay from the fresh rock was separated from material ground in a Tema mill, it contains an appreciable proportion of non-clay minerals, such as labradorite, accounting for the high Ca content and lower LOI and CEC. Despite this, however, the contents of Al, Si, Ti, and K are higher in the clays from the highly weathered samples, whereas those of Mg, Ca, and Na are considerably lower. Iron is variable, decreasing in the initial stages of weathering and increasing in the final stage.

Mineralogical composition. The X-ray diffraction traces of all the clays display a strong 15.0 Å peak (fig. 4) which is displaced to 17–18 Å on addition of glycerol. In the 060 region there is a single peak at 1.537 Å for the clay from the fresh rock (fig. 4*e*), two peaks (at 1.53 and 1.50 Å) in the weathered rock clays (fig. 4*c* and *d*), and a single peak at 1.499 Å due to a clay mineral in the fraction from the mottled and red materials (fig. 4*a* and *b*)—the peak at 1.486 Å being due to the hematite. Thus the fresh rock contains a trioctahedral swelling mineral, the weathered rocks a mixture of trioctahedral and dioctahedral swelling minerals (sample B having a larger amount of dioctahedral type, reflecting its higher Al and lower Mg contents) and the mottled and red samples a dioctahedral swelling phase. The clays from all the weathered samples collapse to 9.8 Å at 550 °C and are therefore smectitic. The clay in the red sample is an interstratified montmorillonite-illite (thus explaining the K_2O content of 0.44 %) which has an infrared spectrum more akin to that of illite than to montmorillonite. The trioctahedral mineral in the fresh rock, which produces a broad reflection at 13.9 Å and a weaker, broad reflection at about 10 Å on heating at 550 °C, is a swelling-chlorite mineral. Both these minerals will be characterized in greater detail in a subsequent publication. The rock sample also contains a small amount of normal, non-swelling chlorite as revealed by the peak at 7.2 Å after glycerol treatment. The peak at 3.2 Å on the fresh and weathered rock traces indicates a small amount of feldspar.

The DTA curves of the clays from the weathered samples all display a low-temperature endothermic system with a large peak at 120–130 °C and a smaller one at 190–210 °C (fig. 5) characteristic of loss of adsorbed water from smectites saturated with divalent cations. A broad dehydroxylation endotherm at 500–550 °C is followed by a higher temperature endotherm (760–790 °C) and an exotherm (850–925 °C) due to structural change. These curves are consistent with smectite being the principal clay mineral. The curve for the fresh rock, on the other hand, is quite different, the adsorbed water being lost at 110 °C and well-defined endotherms at 525 and 730 °C being followed by a sharp

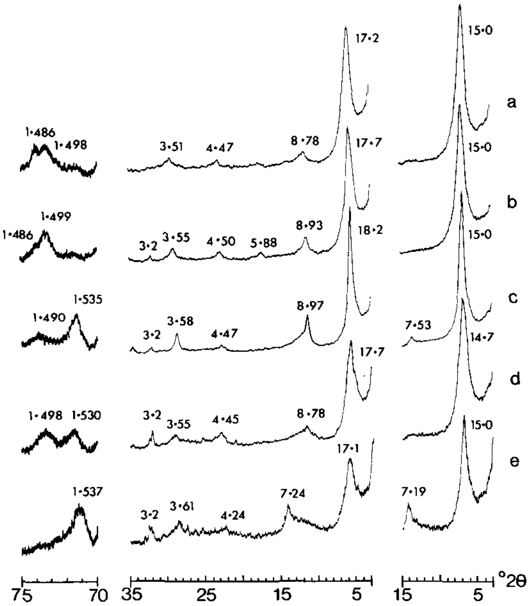

FIG. 4. X-ray diffraction traces of clay fractions from: *a*—red weathered material; *b*—mottled weathered sample; *c*—weathered rock (A); *d*—weathered rock (B); *e*—fresh rock. Spacings indicated are in Angstrom units. 3–15° segment—oriented aggregate; 3–35° segment—oriented aggregate after glycerol treatment; 70–75° segment—random powder mount.

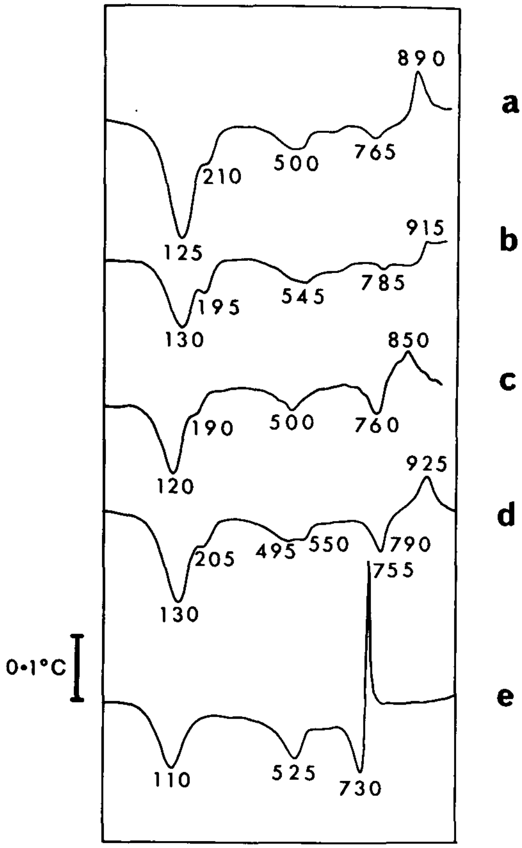

FIG. 5. DTA curves of clay fractions from: *a*—red material; *b*—mottled weathered sample; *c*—weathered rock (A); *d*—weathered rock (B); *e*—fresh rock. Temperatures indicated are in °C.

strong exothermic peak at 755 °C. This curve is reminiscent of that of a chlorite (Caillère and Hénin, 1957) or swelling chlorite (Lippman, 1954).

Discussion

The fresh rock is fairly typical of the basalts on Mull and Morvern; the contents of all the oxides fall within the limits of variation in chemical analyses quoted by Fawcett (1961) for Mull basalts, except that Na_2O is slightly below the minimum found on Mull. The tendency to weather with rusty red surfaces is common in the lavas outside the zone of pneumatolysis, which occurs around the central intrusion on Mull (Bailey *et al.*, 1924), and the predominance of montmorillonite in the fine fraction of red interbasaltic beds in the north-west of Scotland has been reported previously by Godard *et al.* (1961). Interbasaltic layers of red earth or bole up to a metre in thickness have also been recorded (Richey *et al.*, 1961), but there are no bauxitic clays like those formed from similar Tertiary basalts found in Antrim, Northern Ireland, where major interbasaltic residual deposits (up to 13 m thick) formed on the upper surface of flows during periods of volcanic quiescence. These deposits were formed by the leaching out of the alkalis and alkaline earths to produce an assemblage of kaolin minerals, principally halloysite, from feldspar followed by a later stage of desilication leading to a residue rich in gibbsite (Eyles, 1952). Smectite is less common and tends to occur either in specific areas where desilication was less intense (Patterson, 1955) or in the early stages of weathering (Alexander and Heal, 1974). These smectitic weathered materials confer high cation-exchange properties on the soils in this part of Ireland (Brown, 1954; Smith, 1957; McConaghy and McAleese, 1957; McAleese and Mitchell, 1958).

Another lateritic palaeosol found in Tertiary basalts during the drilling on the Iceland-Faeroe Ridge (Nilsen and Kerr, 1978) is considered to have been formed by subaerial weathering of basalt in a warm, humid climate during the early Tertiary. Except for potassium, the geochemical trends in this palaeosol are similar to those at Killundine; moreover, the mineralogy of the basaltic rubble in the middle of the profile is also similar to that of the red material at Killundine, consisting mainly of montmorillonite with some hematite and a small amount of anatase. In the upper part of this sequence, however, kaolinite progressively replaces montmorillonite.

Lateritic weathering was considered to have been responsible for the formation of a red bole from Tertiary basalt on Skye (Anderson and Dunham, 1966) and the chemical composition of this bole shows some similarities to the red material at Killundine. However, in the Skye material, the quantities of CaO and Na_2O are much higher (6.48 and 1.70% respectively), TiO_2 has not been concentrated to the same extent, and the basaltic texture is still visible in thin section, indicating that the basalt on Skye has not been so highly weathered as that at Killundine.

The conversion of the fresh basalt to the incoherent, friable red rubble at Killundine involves major changes in chemical composition, mineralogy, and texture. A comparison of chemical and X-ray diffraction information indicates a progression from a trioctahedral clay in the fresh rock through a mixture of di- and trioctahedral phases in the weathered rock to a dioctahedral phase in the most highly weathered samples. The trends in the chemical composition of the clays are generally similar to those for the bulk materials—namely, the loss of divalent cations and the relative accumulation of trivalent cations. The apparently obvious inference that the dioctahedral phase has developed from the trioctahedral mineral is, however, erroneous. Thin section observations indicate clearly that feldspar is being converted to clay and indeed is totally replaced by clay in the mottled material (fig. 3*d*) in which the clay is dioctahedral only. Thus the dioctahedral montmorillonite-illite, which is the dominant phase in the most highly weathered samples, forms from the labradorite, while the trioctahedral swelling-chlorite type mineral is being destroyed. When the small amount of K present in labradorite is released during the breakdown of the feldspar it is not removed by leaching but is retained in the illitic component of the dioctahedral phase.

The irregular nature of the occurrence of the swelling-chlorite in the basalt suggests that it is not an alteration product of any of the other ferromagnesian minerals but has formed from a late-stage fluid during the cooling of the lava. Fawcett (1965) found material with similar optical properties in analogous textural occurrences in the Mull lavas.

The possibility of the chemical, mineralogical, and textural changes being due to hydrothermal activity rather than to weathering must be considered as it could be argued that the reddening and development of montmorillonite is due to hydrothermal alteration along the joints of the lavas and between flows. Certainly a well-established hydrothermal aureole, in which a range of minerals has formed in amygdales, occurs in the basalts around the central intrusive complex forming the core of the Mull volcano (Walker, 1971), but the Killundine locality is several km from the outer limit of this zone and, although proof is impossible, the weight of evidence seems to indicate that weathering is responsible for the formation of these clays. The characteristics of the material are indeed very similar to those of ferruginous tropical soils developed on basalts under poor drainage conditions (Godard *et al.*, 1961). Furthermore the geochemical variations involved (enrichment of Fe, Ti, and Al and, to a lesser extent, Si relative to alkalis and alkaline earths) are strongly reminiscent of a weathering regime. In view of the characteristic lateritic weathering of similar rocks in Antrim and on the Iceland-Faeroe Ridge, it would seem that the Killundine samples have also been formed by weathering but in a less intense environment.

Montmorillonite has been recorded as the initial weathering product of plagioclase in basalt in various environments (Craig and Loughnan, 1964; Loughnan, 1969; Singer, 1970, 1973; Lunkad and Raymahashay, 1978) and even as pseudomorphs after plagioclase in saprolitic weathering (Singer, 1973). As weathering proceeds and leaching becomes more effective, the montmorillonite is replaced by kaolinite and halloysite, and simultaneously iron oxide, as goethite or hematite, is concentrated in the oxidized parts of the weathering zone (Loughnan, 1969). The absence of kaolinite at Killundine is particularly interesting and was also noted by Godard *et al.* (1961) for interbasaltic beds in the same region.

Hematite, like kaolinite and gibbsite, is often regarded as representative of fairly advanced weathering and its coexistence with montmorillonite, which forms early in weathering sequences, may seem anomalous. However, hematite has in fact been recorded as forming at an earlier stage of weathering than kaolinite (Nilsen and Kerr, 1978).

Conclusions

It would seem that the fresh basalt (which has an unusually high CEC of 10 meq/100 g due to the presence of a swelling chlorite-type mineral) has weathered in a localized pocket to an intimate mixture of dioctahedral interstratified montmorillonite-illite with some hematite and a little anatase. This process involved the relative accumulation of Al, Fe, and Ti and depletion (in increasing order) of Si, Mg, Ca, and Na. Much of the K released on weathering was retained in the formation of the illite component of the interstratified mineral. These processes would probably have occurred during the same period as the formation of the Antrim lateritic bauxites but, as it appears that desilication and weathering were not so intense in north-west Scotland, the products have not reached such an advanced state in the weathering sequence.

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