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Chemical effects of deuteric alteration in some Kenyan trachyte lavas

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ABSTRACT. The Kenyan trachyte volcanoes Kilombe and Londiani show some unusual chemical features. Analyses of elements normally regarded as residual in salic peralkaline rocks do not show the linear covariance characteristic of these elements in other East African volcanoes. The scatter from expected trends is greatest for Y and the lanthanides, less for Nb and Rb, and not apparent for Zr. The lavas also show variable loss of Na, P, and Fe. It is suggested that this chemical deviation is the result of a deuteric event during the later stages of crystallization of the lavas. The flows showing the chemical deviations, in particular negative Ce anomalies, also tend to have their ferroaugite phenocrysts deuterically altered to a mineraloid closely resembling iddingsite.

During the crystallization of these trachyte lavas an H_2O-, Cl-, and F-rich vapour separates from the liquid. This absorbs some elements, particularly Na, P, Fe, Y, and the lanthanides from the silicate liquid as water-soluble or volatile halides. Some of the absorbed material is redeposited in other parts of the flow and the rest is lost altogether as the vapour escapes from the surface.

LONDIANI and Kilombe (30° 35′ E., 0° 07′ S., and 35° 50′ E., 0° 05′ S.) (fig. 1) are trachytic caldera volcanoes situated, 12 km apart, on the western flank of the Kenya Rift Valley (McCall, 1964; Jennings, 1971; Jones, 1975). They have been dated at about 3 Ma and 2 Ma respectively (Jones, 1975). Kilombe consists entirely of trachyte lavas; Londiani of trachyte lavas with a few late alkali rhyolite flows. Early flows on both volcanoes are aphyric whereas later flows are progressively more porphyritic. They overlie the Saos Mugearite, a series of alkali basalts, hawaiites, and mugearites.

Petrography of the trachytes. The trachytes of Londiani and Kilombe are indistinguishable in thin section. Feldspar phenocrysts, which are larger and more abundant in the upper part of the succession, are sanidine in the early flows, with anorthoclase forming up to 35% of the later flows. The groundmass alkali feldspar has a pronounced fluidal texture and often a bimodal grain-size distribution.

Pale yellow-green mildly pleochroic ferroaugite phenocrysts up to 1 mm long are present in about half of the sections examined and vary in abundance with the feldspar phenocrysts, though never forming more than 1% of the rock.

Occasional microphenocrysts of pale-green olivine and an ore mineral are both mantled by reaction rims of aenigmatite. The groundmass mafic minerals are aenigmatite (15-20%), katophorite with rims transitional to arfvedsonite and aegirine. There is usually about twice as much pyroxene as amphibole and together they form 15-20% of the rock, but amphibole occasionally predominates. Interstitial pale yellow-brown, very fine-grained highly birefringent material, probably an aggregate of hydrous minerals formed by alteration of original glass, comprises 5-20% of the rock.

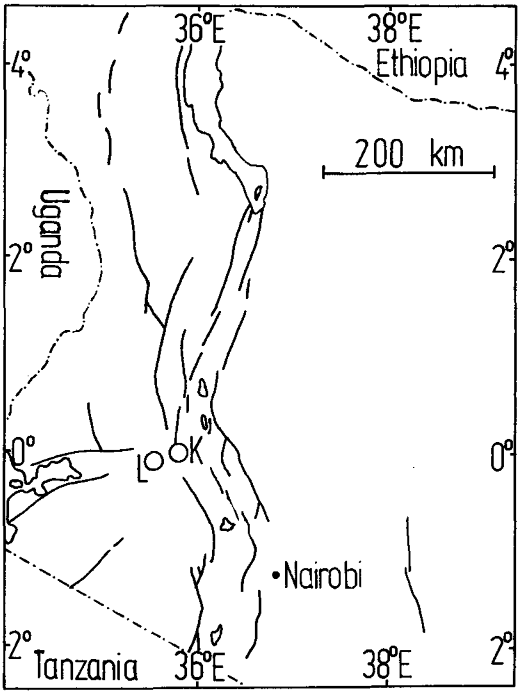

FIG. 1. Index map of Kenya, showing location of Kilombe, K, and Londiani, L.

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Quartz is sometimes present as pools of equidimensional grains in the groundmass and as occasional vermicular intergrowths with feldspar but constitutes up to 25% of the alkali rhyolites.

TABLE I. *Representative analyses of Londiani and Kilombe trachytes*

Wt %	14/183	14/254	14/325	14/236	14/382	14/323
SiO_2	60.43	61.90	61.20	61.84	67.80	60.16
TiO_2	0.88	0.58	0.76	0.65	0.39	0.56
Al_2O_3	17.36	14.46	16.00	15.02	14.01	13.89
Fe_2O_3	3.78	4.43	6.05	4.75	4.05	5.67
FeO	1.90	4.11	1.54	3.30	1.99	3.73
MnO	0.15	0.30	0.16	0.24	0.11	0.38
MgO	0.64	0.41	0.12	0.29	0.10	0.43
CaO	1.98	1.06	0.64	0.59	0.32	0.99
Na_2O	6.84	6.22	7.01	6.54	4.94	6.94
K_2O	5.18	5.01	5.43	5.08	4.88	4.01
H_2O^+	0.58	1.07	0.95	1.21	1.42	1.55
P_2O_5	0.24	0.15	0.08	0.15	0.02	0.18
	99.96	99.70	99.94	99.66	100.03	99.29
ppm						
Ba	1435	198	380	99	46	74
Ce	207	526	306	317	44	510
La	237	600	804	429	27	294
Nb	113	341	180	251	292	365
Nd	131	399	447	262	26	184
Pr	30	100	102	61	9	38
Rb	77	192	115	151	182	198
Sr	268	18	22	6	4	4
Y	87	141	129	132	38	116
Zr	332	1072	455	640	1274	935
Percentage norms						
	(14/183	14/254	14/325	14/236	14/382	14/323)
q	—	4.13	0.56	3.55	19.61	0.66
or	30.61	29.61	32.09	30.02	28.84	28.42
ab	53.05	46.49	52.07	48.98	41.80	44.67
an	1.37	—	—	—	1.46	—
ne	2.61	—	—	—	—	—
ac	—	5.42	6.39	5.61	—	12.38
di	3.44	3.71	0.65	1.68	—	3.24
hy	—	4.13	—	2.96	0.25	4.87
ol	—	—	—	—	—	—
il	1.67	1.10	1.44	1.23	0.74	1.06
mt	4.06	3.71	3.28	4.08	5.65	2.01
hm	0.98	—	1.58	—	0.16	—
ap	0.56	0.35	0.19	0.35	0.05	0.42
cc	—	—	—	—	—	—
H_2O	0.58	1.07	0.95	1.21	1.42	1.55
wo	1.03	—	0.76	—	—	—
co	—	—	—	—	0.07	—

I4/183	Anorthoclasephyric trachyte with fresh ferroaugite phenocrysts, Kilombe.
I4/254	Sanidinephyric trachyte with partly altered ferroaugite phenocrysts, Kilombe.
I4/325	Sanidinephyric trachyte without ferroaugite phenocrysts, Londiani.
I4/236	Sparsely sanidinephyric trachyte without ferroaugite phenocrysts, Londiani.
I4/382	Sanidinephyric pantelleritic trachyte with completely altered ferroaugite phenocrysts, Londiani.
I4/323	Aphyric trachyte glass, Londiani.

Representative analyses of samples from Londiani and Kilombe are given in Table I. They are usually peralkaline and slightly oversaturated. Similar analyses have been published for other Kenyan trachyte volcanoes: Paka (Sceal and Weaver, 1971), Menengai (McCall, 1967), Silali (McCall and Hornung, 1972), Ribkwo and Nasaken (Weaver *et al.*, 1972).

Geochemical patterns in the lavas. Recent studies have shown that the elements Zr, Rb, Y, Nb, and the lanthanides behave coherently in the lavas of several East African alkaline volcanoes (Barberi *et al.*, 1975; Gibson, 1972; Sceal and Weaver, 1971; Weaver *et al.*, 1972). Plots of any pair of these elements approximate to straight lines passing through the origin. The concentrations of the elements increase with the degree of differentiation. This is thought to show that these elements are strongly partitioned into the liquid phase during fractional crystallization because they are not readily accommodated in the structures of any of the early-crystallizing minerals (Baker, 1978).

Six specimens of Saos Mugearite, nine of Kilombe Trachyte, and nineteen of Londiani Trachyte were analysed for Rb, Sr, Y, Zr, Nb, Ba, La, Ce, Pr, and Nd on a Phillips PW 1212 automatic XRF spectrometer at Bedford College, University of London. Precisions are ±1% of the amount present above the 100 ppm level for Nb, Rb, Sr, and Zr and ±2% for Ba, Ce, and La (Weaver *et al.*, 1972). Plots of Nb *v.* Zr and Ce *v.* Zr for the three formations are presented in fig. 2. The Nb/Zr plot shows the straight line for the two volcanoes as expected. In this paper such a plot will be said to show a normal pattern. The Ce/Zr plot shows a marked scatter. This sort of pattern will be called here an anomalous pattern. On the Nb/Zr plot the Kilombe rocks and the majority of the Londiani rocks form close but distinct lines. This is as expected since it is known that different Kenyan volcanoes show different Nb/Zr ratios (Weaver *et al.*, 1972). The Saos Mugearite forms a line at an intermediate slope. Most of the Londiani trachytes lie on a line but a few fall below it. Those points which lie below the line for Nb/Zr are even further below on the Ce/Zr plot. If Zr has been enriched in these rocks then the Ce/Zr plot implies much greater enrichment than is indicated by the Nb/Zr plot. This difficulty is avoided by assuming that Zr is unaltered and that Nb and Ce are both depleted in some rocks, Ce to a greater extent than Nb. The latter interpretation is followed here.

The Rb/Zr plot shows some scatter but for Y and the lanthanides there is little trace of a linear relationship. This scattering only affects the trachytes and is more pronounced for Londiani than for Kilombe. In the Saos Mugearite all these

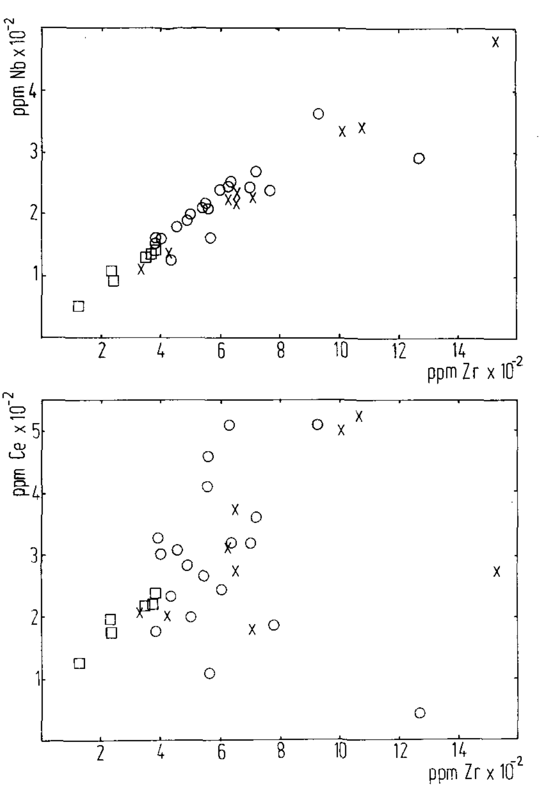

FIG. 2. Nb/Zr and Ce/Zr plots for the Londiani Trachyte, Kilombe Trachyte and Saos Mugearite. ○ Londiani Trachyte, × Kilombe Trachyte, □ Saos Mugearite.

elements behave coherently with Zr. The degree of scattering for the different elements is shown by their correlation coefficients with Zr:

	Nb	Rb	Y	La	Ce	Pr	Nd
Kilombe	0.996	0.968	0.449	0.479	0.459	0.510	0.482
Londiani	0.805	0.735	−0.269	−0.460	−0.117	−0.485	−0.515

In the Londiano Trachyte, and to a lesser extent in the Kilombe Trachyte, these elements do not behave in the coherent manner characteristic of other East African salic volcanoes. This effect is least for Nb and Zr since the Nb/Zr diagram is the least anomalous.

On the Nb/Zr and Rb/Zr plots the trends for the two volcanoes are close to the extension of the trend of the Saos Mugearite. It would be reasonable to expect the plots of the other elements against Zr also to be close to the Saos Mugearite trend. In that case by extending the trend of the Saos Mugearite on each plot and finding the point on that trend which corresponds to the Zr content of a specimen, we can find a 'predicted value' for the concentration of each element. This predicted value maybe compared with the measured value and the ratio measured value/predicted value = m/p can be used as a measure of the degree of chemical deviation of a specimen for a particular element. When this ratio is close to unity then the specimen is normal; if it is greater, the specimen has an excess of that element, and when less it is depleted. Since the assumption that the normal trace-element plots of the trachytes would be close to those of the Saos Mugearite is only approximate, then this ratio can only be a very approximate measure of the chemical deviation.

Major-element analyses of the lavas showed fairly smooth trends for all these elements except for Na, Fe, and P. Na shows a trend of falling concentrations with Zr while total Fe increases. There are less data for P but it appears to have a nearly linear positive correlation with Zr. The phenocryst phases in the lavas are alkali feldspar and minor ferroaugite. The major element trends probably reflect evolution dominated by crystallization of alkali feldspar more sodic than the magma. The crystallization of ferroaugite was inadequate to prevent Fe enrichment and there was no crystallization of apatite. However, there is a strong tendency for particular samples to be depleted in all three elements, and for these samples to be the ones which show irregularities in the trace elements.

Timing of the deuteric event. Possible explanations for the irregular trace-element patterns are: leaching due to weathering; mobilization during metamorphism; deuteric effects on extrusion and crystallization; and contamination of the trachyte magma by reaction with wall rock or another magma.

Weathering is unlikely to be the cause because the two volcanoes are only slightly eroded; apparently fresh rocks were collected in the field and signs of weathering were not seen in thin section. Metamorphism cannot have taken place because the volcanoes have not been buried under younger deposits. Y and the lanthanides are generally considered immobile, and Rb very mobile, during these secondary processes (Pearce, 1975), while in this case Y and the lanthanides show considerable, and Rb only moderate, chemical deviation.

Strong evidence that the chemical scatter is a consequence of a deuteric event is provided by a study of the alteration of the ferroaugite phenocrysts. These are often completely fresh but in about half of the sections examined they show varying degrees of alteration to a red-brown mineraloid. This is not yet well characterized and here it will be merely referred to as a phyllosilicate. It has a high birefringence, small to moderate $2V_\alpha$ and shows straight extinction parallel to the single good cleavage. It is slightly pleochroic, the colour being more pronounced parallel to the cleavage which is itself parallel to the (100) faces of the original pyroxene. X-ray diffraction studies show that it has a prominent interlayer spacing of 15.3 Å, falling to 14.6 Å after heating for two days at 110 °C (Weaver, 1973). There are three reasons for believing that the alteration of ferroaugite to the phyllosilicate takes place during the crystallization of the lava: phenocrysts sometimes show phyllosilicate mantling fresh ferroaugite cores; phyllosilicate forms small pyroxene-shaped prisms in the groundmass of some sections; and aegirine similar to that forming poikilitic patches in the groundmass forms rims around both the small prisms and the phenocrysts pseudomorphed by the phyllosilicate. In fig. 3, Nb, Rb, Y, La, Ce, Na_2O, P_2O_5, and total FeO are plotted against Zr for the Londiani trachyte,

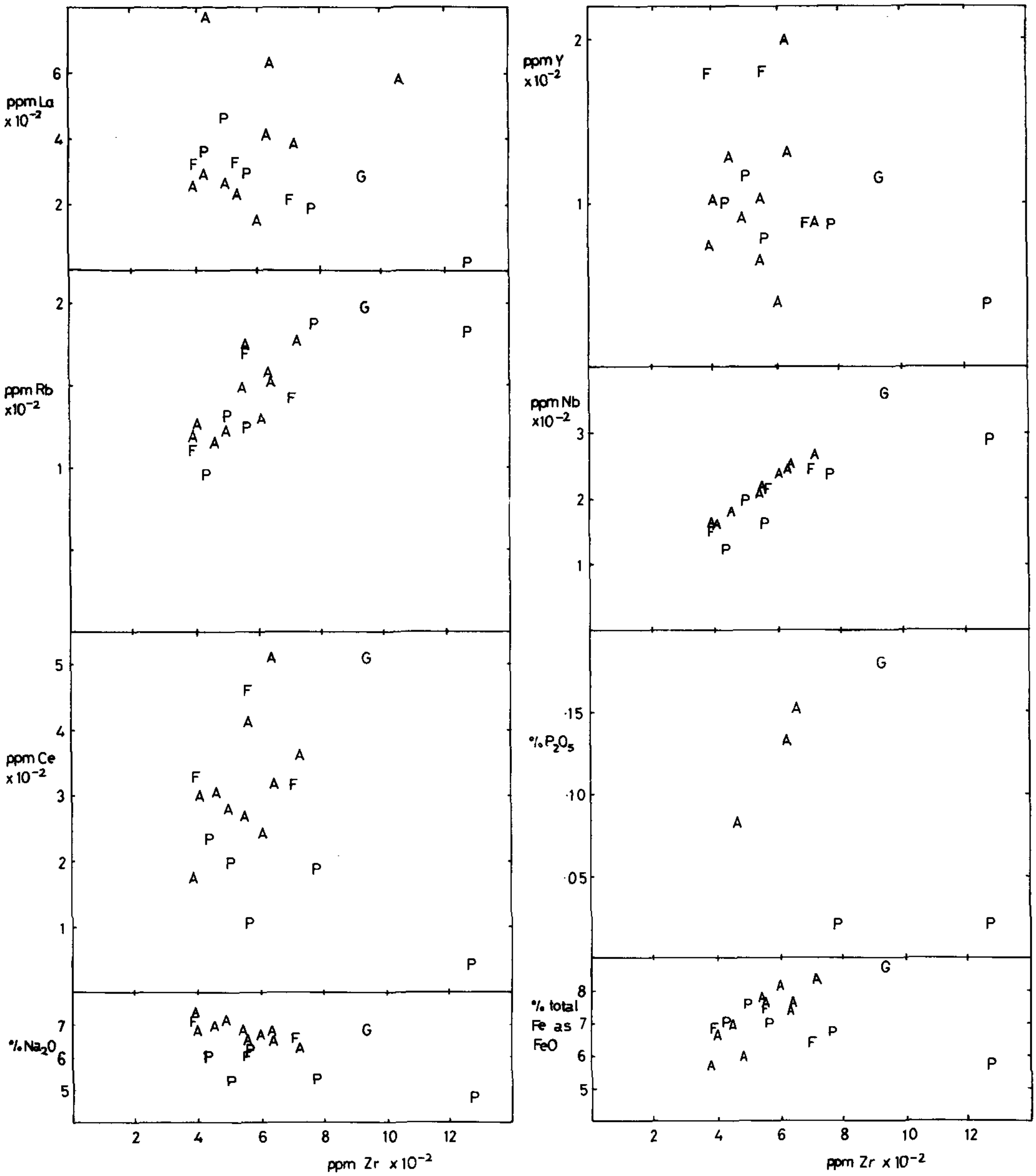

FIG. 3. La, Rb, Ce, Na_2O, Y, Nb, P_2O_5, and total Fe *v.* Zr for the Londiani Trachyte. F, trachyte with ferroaugite phenocrysts. P, trachyte with phyllosilicate pseudomorphs after ferroaugite. A, trachyte without ferroaugite phenocrysts. G, glassy trachyte 14/323.

the specimens being distinguished according to whether they contain unaltered pyroxene phenocrysts, phyllosilicate pseudomorphs, or neither. These plots show that the phyllosilicate tends to occur in rocks which have lost Nb, Ce, P, and Na while unaltered ferroaugite is found in undepleted rocks. There is no clear correlation between the alteration and loss or gain of La, Y, Rb or Fe, but that between the presence of the phyllosilicate and the anomalous trace-element patterns is well brought out in fig. 4, a plot of the chondrite-normalized concentrations of the first four lanthanide elements for those specimens which contain either pseudomorphs or fresh ferroaugite phenocrysts. Londiani and Kilombe rocks are shown separately. Londiani phyllosilicate-bearing rocks have substantial negative Ce anomalies while ferroaugite-bearing rocks have little or no such anomaly. The same is true for Kilombe although the distinction is not so marked. It seems that the Ce anomalies are produced under the same circumstances as the phyllosilicate, during the crystallization of the flows.

Further evidence for the disturbance taking place during the crystallization of the flows is provided by the analysis of specimen 14/323 which is from a glass boulder on Londiani, and is the only glassy specimen analysed. On all the trace-element plots it has close to its predicted trace-element concentrations. It has higher Na, P, and total Fe than any other Londiani trachyte of similar Zr content (fig. 3) and the concentrations of these elements lie on the extension of the normal trends. Its lanthanide pattern shows no Ce anomaly. It has clearly not undergone the same chemical scattering process as many of the crystallized rocks.

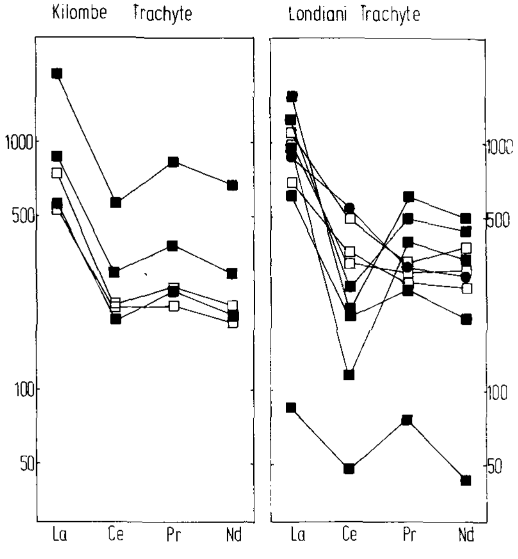

FIG. 4. Chondrite-normalized rare-earth element plots for the Londiani Trachyte and Kilombe Trachyte. □ trachyte with ferroaugite phenocrysts. ■ trachyte with phyllosilicate pseudomorphs after ferroaugite phenocrysts. ● glassy trachyte 14/323.

Evidence for mixing of the trachyte magma with a distinct magma type is shown by the existence of blobs of benmoreite in certain flows on Londiani (Jones, 1979*a*) but there is no indication in these flows for complete absorption of the benmoreite by the trachyte to produce a single homogeneous liquid. Contamination of the trachyte by another magma would produce a scatter on one side of the predicted trend only, whereas some elements show both enrichment and depletion in different samples. Contamination by another magma lying on approximately the same trend as the trachytes (such as the benmoreite) would not produce a scatter away from the predicted trend. The same arguments apply to reaction of the trachyte with wall rock, so contamination is unlikely to contribute to the geochemical anomalies.

Discussion. Loss of Na during the crystallization of peralkaline silicic rocks is well established in the literature (Baker and Henage, 1977; Macdonald and Bailey, 1973; Noble, 1965, 1970; Romano, 1969). This is generally attributed to its expulsion in a Na-halide-rich vapour (Baker and Henage, 1977). The loss of Fe from peralkaline liquids is less well documented (Baker and Henage, 1977; Gibson, 1972) but seems to be correlated with Na loss and may also be due to removal in a halide-rich fluid since $FeCl_3$ is a volatile compound. Loss of P on crystallization has not previously been reported but could also be due to expulsion with the halogens since P forms several volatile halides (Heslop and Robinson, 1967).

Gains and losses of Y and the lanthanides similar to those described here have been reported for other Kenyan salic peralkaline rocks by Baker and Henage (1977) although their data do not show Ce anomalies. It is known that lanthanide elements form mobile halide complexes (Herman, 1970), and expulsion in a halogen-rich vapour has been used by Weaver *et al.* (1972) to explain anomalous lanthanide concentrations in some specimens from Eburru, Kenya. Of all the lanthanides, Ce is the most readily oxidized to the M^{4+} ion (Heslop and Robinson, 1967). The Ce anomalies can therefore be explained by the oxidation of Ce to Ce^{4+} in which state its chemical behaviour is different from that of the other lanthanides which remain as M^{3+} ions.

The formation of the phyllosilicate from ferroaugite is very similar to the conversion of olivine into iddingsite and probably occurs under the same conditions. Iddingsitization is believed to be caused by an oxidizing hydrous fluid which separates from the silicate liquid and escapes from the flow (Gay and Le Maitre, 1961; Wilshire, 1958). It seems that the chemical and mineralogical effects discussed in this paper may all be explained by invoking halogen-rich hydrous fluids separating from the lavas. There is no direct evidence that these trachytes have lost halogens on crystallization, but Noble *et al.* (1967) state that in general silicic volcanic rocks lose an average of half of their original F and four-fifths of the Cl. The glassy specimen 14/323 has a much lower major element total than the other trachytes; this difference could be accounted for by about 0.7% of halogens which are not present in the crystallized rocks. The very vesicular flow tops on many of the trachytes point to the loss of a large amount of volatiles after eruption. Johnson (1969) has demonstrated that loss of up to 0.25% of Cl from certain trachyphonolite flows on Suswa volcano, Kenya.

Bailey and Macdonald (1975) found F up to 7800 ppm and Cl up to 4290 ppm in a series of pantelleritic trachyte and pantellerite obsidians from Eburru volcano, Kenya. Their data show correlation coefficients with Zr of 0.994 for F and 0.876 for Cl suggesting that the halogens behave as incompatible elements in peralkaline liquids.

It is thus reasonable to suggest that the Londiani and Kilombe trachyte lavas may have had up to 1% of halogens on eruption and subsequently lost most of it during crystallization. These halogens could have redistributed some of the other elements in the flows in the form of volatile halides. If the halogen content increases with Zr, this would explain the most anomalous rocks being the more Zr-rich ones. The halogens are present in sufficient quantity to combine with the P and trace elements but probably not with the Na and Fe in the more anomalous rocks; the most anomalous rock, 14/382, would require 5% F or 9% Cl to alter it from its predicted composition. Other volatile species must also contribute to the mobility, in particular H_2O and also probably CO_2, since CO_2 is the main gas escaping from fumaroles in the Kenya Rift Valley now (McCall, 1967), and it has been shown to be more important as a volatile than its concentration in lavas suggests (Price and Bailey, 1980).

The gradual increase in the proportion of phenocrysts in the lavas, the existence of calderas on each volcano, and the presence of syenite enclaves in post-caldera lavas and tuffs shows that the trachyte magma occupied magma chambers under the volcanoes. Examination of these syenites (Jones, 1979*b*) has shown that at stages of fractionation more extreme than represented by any whole-rock composition among the lavas, Zr and Rb do enter the crystallizing minerals and a residual volatile-rich liquid may separate to crystallize as a distinct rock type enriched in Na, P, Fe, Nb, Y, and the lanthanides, especially La and Ce. The crystallization of a trachyte lava is likely to be very similar to the crystallization of syenite magma in a high-level magma chamber and the liquid left when most of a flow has crystallized will be similar to the late-stage syenites. This enables the course of crystallization of the trachyte flows to be reconstructed.

As a trachyte lava erupts it usually contains phenocrysts of alkali feldspar and ferroaugite and sometimes microphenocrysts of olivine and ore minerals.

Most of the period of solidification of the groundmass is dominated by the crystallization of feldspar laths and ferroaugite prisms. During this period Nb, Y, and the lanthanides remain in the liquid while Zr and Rb initially partition into the liquid but become increasingly incorporated in the crystallizing minerals. Na, P, and Fe slowly concentrate in the liquid because of their presence in excess of the requirements of the solid phases. H_2O, Cl, and F are partitioned into the liquid but when the flow is about half or three-quarters crystallized these volatiles begin to separate as a distinct fluid. This fluid is strongly oxidizing and tends to oxidize the Fe in the lava to Fe^{3+} and the Ce to Ce^{4+}, at the same time converting the ferroaugite phenocrysts to the phyllosilicate. The volatile fluid incorporates Na, P, Fe, Y, lanthanides, and to a lesser extent Nb from the enclosing silicate liquid in the form of water-soluble or volatile halides. Some of this material is redeposited in other parts of the flow while the rest is removed as the volatiles escape at the surface. The solidification of the flow is completed by the crystallization of mossy aegirine, aenigmatite, and alkali amphibole from the remaining volatile-saturated silicate liquid with aegirine also mantling the phyllosilicate. The last of the liquid solidifies either as glass which is later altered to the yellow-brown very fine grained crystal aggregate or crystallized directly as this material.

These volatile effects in trachytes show that the composition of the crystalline rock may be quite different from that of the lava during its eruption. Analyses of trachytes may therefore be very misleading for petrogenetic studies. Fortunately the very obvious alteration of the ferroaugite phenocrysts, even in hand specimen, indicates rocks in which alteration by volatile loss may have taken place.

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