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Differentiation of natrocarbonatite magma at Oldoinyo Lengai volcano, Tanzania

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ABSTRACT

Natrocarbonatite magma, erupted as lava flows in the Tanzanian volcano Oldoinyo Lengai in June and November of 1988, has evolved chemically since its formation. The June and November flows of 1988 display increasing Cl, F, Ba, K, Mg and Mn, concomitantly with Na, Ca and P depletion. Furthermore, the June magma, at the time of eruption, had higher Cl, F, Ba and K contents and lower Ca than the November magma and evolved to higher levels of Cl, F, Ba and K content and lower Ca, Na and P. The mineralogy of the lavas reflects these trends. Crystallization of fluorite and halite–sylvite solid solution, usually as a symplectic intergrowth, occurs when Cl and F concentrations reach the critical value necessary to stabilize both minerals and explains why neither occurs as a phenocryst phase. Natrocarbonatite magma has undergone considerable and rapid magmatic evolution, probably in small and separate magma chambers. Two minerals, nyerereite and gregoryite, have dominated the crystallization history of natrocarbonatite magma, and many lavas are phenocryst-rich. However, because most of the lavas are composed principally of these two minerals, crystal accumulation has not greatly changed their composition and, consequently, we suggest that the bulk composition of the lavas closely approximates that of the parental magma.

KEYWORDS: natrocarbonatite, Oldoinyo Lengai, magmatic evolution.

Introduction

NATROCARBONATITE magma at Oldoinyo Lengai volcano in Tanzania displays substantial chemical evolution in flows erupted over a duration of only a few days, and significant differences between flows erupted in June and November of 1988. This evolution is displayed by whole-rock chemical analyses and by mineralogical relationships.

Natrocarbonatite mineralogy and petrography has been well described (Dawson 1962, Dawson *et al.*, 1995; Peterson, 1990; Keller and Krafft, 1990) but it is only recently that there has been available a sufficient number of whole-rock chemical analyses to allow a more detailed study of their compositional evolution. The rocks are composed principally of the two carbonate minerals nyerereite $(Na,K)_2Ca(CO_3)_2$ and gregoryite $(Na,Ca,K)_2CO_3$. Porphyritic lavas consist largely of phenocrysts of both minerals in a matrix of symplectically intergrown fluorite and $(\text{halite-sylvite})_{SS}$. Aphanitic lavas consist of nyerereite and gregoryite as microphenocrysts in a symplectite of fluorite and sylvite, together with a nyerereite-like mineral and 'phase X' (Dawson *et al.*, 1996). In addition, Mitchell (1997) has described, from a flow erupted on October 17th 1995, a variant in which rounded areas (probably sections through globules) of gregoryite and sylvite are contained within a symplectite of nyerereite and gregoryite in which no fluorite is present. The aphanitic varieties occur as 'squeeze-ups' or driblets that have escaped from cracks in the surface of porphyritic flows, and also as vent-erupted lavas.

Compositional evolution of natrocarbonatite magma

The origin of natrocarbonatite magma has been debated extensively (Freestone and Hamilton, 1980; Kjarsgaard and Hamilton, 1989; Kjarsgaard, Hamilton and Peterson, 1995; Dawson *et al.*, 1996; Church and Jones, 1995) but there has been very little discussion of how the magma has evolved since its formation. It is this magmatic evolution that we propose to examine.

Analyses of aphyric and porphyritic natrocarbonatite lavas erupted in June and November 1988 (Table 1) are taken from Keller and Krafft (1990) and Dawson *et al.* (1995). It seems clear that the aphyric lavas represent a later phase of magmatic evolution than do the porphyritic types and so their compositions can safely be taken as indicating the direction of any evolutionary trends. In Figs 1–6 the variation of several elements in 16 lava flows erupted in June and November 1988 is examined. Distinct trends are present and, while those for June and November overlap, they are also distinguishable as representing separate lines of descent. Our discussion is limited to these two groups for several reasons. Compositions of rocks from other eruptions fall along the same trends, with rare exceptions, and do not change the conclusions that emerge from the more limited data set. Furthermore, the eruptive dates of these other flows are either not known, or there are not enough of them to establish any trend in a particular eruptive phase of the volcano. The discussion is further limited to the behaviour of Cl, F, Ba, Ca, Na, K, Mn, Mg and P. Similar trends are observable for some trace elements (such as Rb) but the elements chosen are sufficient to establish the character of magmatic evolution and its general direction.

From Figs 1–6 we may conclude that:

1. Elements that are increasingly concentrated during evolution of the magma are Cl, F, Ba, K, Mg and Mn, with the possibility of very late Na enrichment.
2. Elements whose concentrations steadily decrease during evolution of the magma are Na, Ca and P, with the possibility that the Na decrease is reversed at a very late stage.

Both of these trends are consistent with the conclusions of Dawson *et al.* (1995) "... that K_2O, BaO, the halogens and Rb are concentrated preferentially within the matrix." Differences between the data for June and November 1988

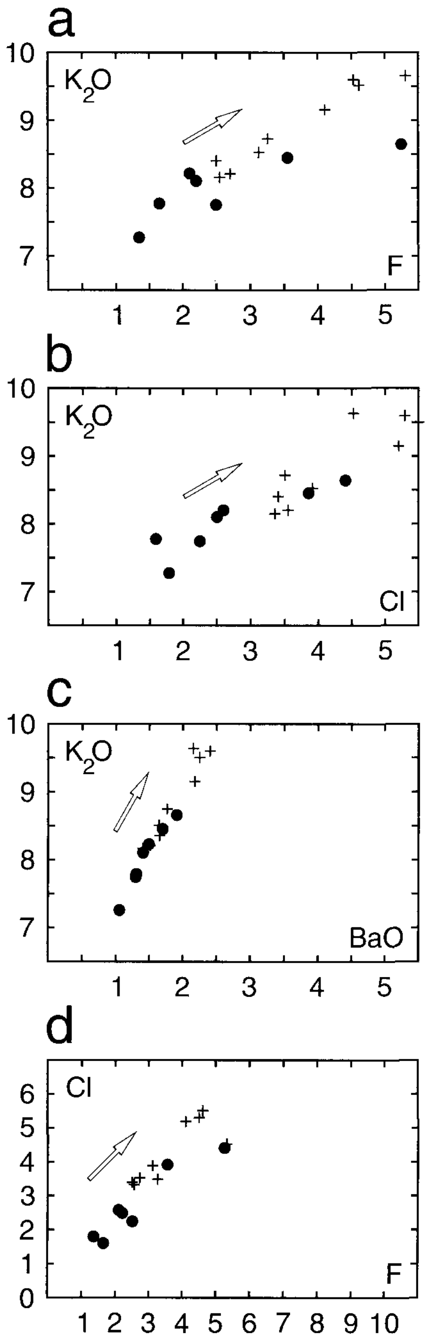

FIG. 1. Variation of elements (in wt.%) for Oldoinyo Lengai natrocarbonatite lavas. Symbols: crosses – June 1988 flows; solid circles – November 1988 flows. (*a*) K_2O *vs* F; (*b*) K_2O *vs* Cl; (*c*) K_2O *vs* BaO; (*d*) Cl *vs* F. Arrows indicate direction of magmatic differentiation.

TABLE 1. Chemical composition of Oldoinyo Lengai natrocarbonatite lavas erupted in June and November 1988

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
SiO_2	0.23	0.16	0.19	0.13	0.12	0.24	0.21	0.15	0.16	0.29	0.14	0.17	0.19	0.22	0.23	0.18
TiO_2	0.02	0.02	0.02	0.02	0.02	0.03	0.02	0.03	0.02	0.02	0.01	<0.01	0.01	0.01	0.02	<0.01
Al_2O_3	b.d.	b.d.	b.d.	b.d.	b.d.	b.d.	b.d.	b.d.	b.d.	0.10	0.10	0.10	0.10	0.12	0.10	0.10
Fe_2O_3	0.30	0.28	0.39	0.38	0.21	0.47	0.48	0.67	0.59	0.50	0	0.13	0.20	0.21	0.36	0.21
FeO										0.24	0.10	0.06	0.14	0.08	0.10	0.12
MnO	0.38	0.38	0.51	0.56	0.34	0.59	0.60	0.99	0.89	0.61	0.24	0.24	0.42	0.31	0.38	0.45
MgO	0.31	0.38	0.41	0.39	0.32	0.57	0.52	0.55	0.57	0.42	0.46	0.27	0.28	0.27	0.26	0.22
CaO	14.08	14.02	13.48	13.55	14.11	12.33	12.86	12.25	12.24	13.36	13.04	14.15	13.81	13.52	14.18	14.63
SrO	1.41	1.42	1.42	1.41	1.42	1.43	1.43	1.42	1.43	1.74	1.79	1.68	1.69	1.70	1.69	1.63
BaO	1.39	1.66	1.63	1.77	1.52	2.41	2.17	2.25	2.16	1.68	1.92	1.30	1.39	1.47	1.29	1.05
Na_2O	32.33	32.22	32.46	31.42	31.80	31.08	30.42	31.03	30.82	31.06	32.58	32.03	32.18	31.92	32.45	32.72
K_2O	8.16	8.38	8.52	8.73	8.21	9.61	9.14	9.50	9.64	8.44	8.64	7.75	8.11	8.22	7.78	7.27
P_2O_5	0.87	0.85	0.80	0.75	0.79	0.73	0.75	0.59	0.58	0.73	0.63	0.91	0.86	0.86	0.99	1.02
CO_2	31.19	31.55	30.36	30.41	30.99	27.40	28.12	27.39	27.10	32.7	29.2	35.0	34.7	34.3	35.0	34.3
Cl	3.35	3.40	3.90	3.50	3.55	5.30	5.18	5.50	5.30	3.85	4.40	2.25	2.50	2.60	1.60	1.80
F	2.55	2.50	3.12	3.26	2.69	4.53	4.10	4.58	4.51	3.55	5.25	2.50	2.20	2.10	1.65	1.35
SO_3	4.43	3.72	4.63	4.80	4.18	4.92	5.58	5.24	5.77	3.12	3.07	2.92	3.17	3.10	2.88	3.18
H_2O+	0.41	0.56	0.41	0.24	0.47	0.43	0.44	0.51	0.38	0.05	0.23	0.03	0.03	0.0	0.18	0.22
-O = F,Cl	−1.83	−1.82	−2.19	−2.16	−1.92	−3.11	−2.90	−3.13	−3.09	−2.35	−2.03	−1.55	−1.48	−1.46	−1.05	−0.97
Total	99.58	99.68	100.06	99.16	99.82	98.96	99.12	99.52	99.07	100.12	99.97	99.93	100.50	99.51	100.29	99.48

b.d. indicates that the element, if present, is in an amount less than the limit of detection
1–9 Keller and Krafft 1990, Table 2, page 636. June 1988. Total iron expressed as Fe_2O_3.
10–16 Dawson *et al.*, 1995, Table 7, page 58. November 1988.
In analysis 11 the value of 0 for Fe_2O_3 is quoted as printed in Dawson *et al.*, 1995.

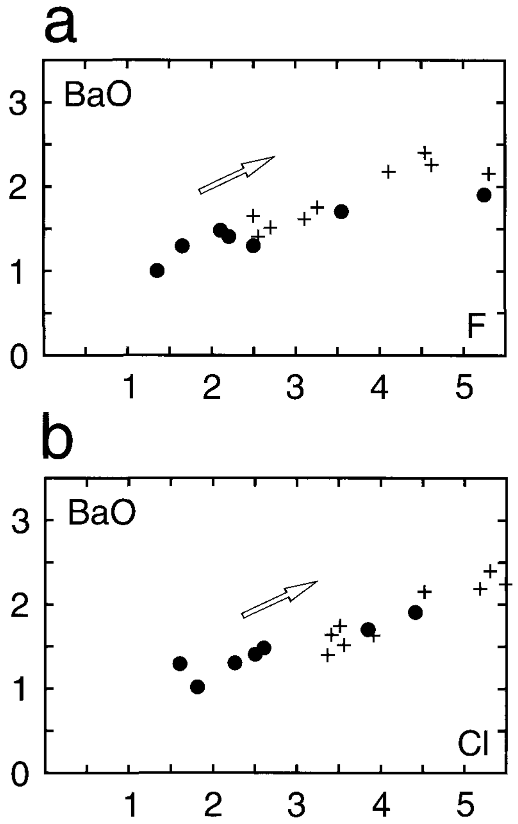

FIG. 2. Variation if elements (in wt.%) for Oldoinyo Lengai natrocarbonatite lavas. Symbols: crosses – June 1988 flows; solid circles – November 1988 flows. (*a*) BaO *vs* F; (*b*) BaO *vs* Cl. Arrows indicate direction of magmatic differentiation.

suggest that the magma erupted in June had a higher content of Cl, F (with one exception), Ba and K and a slightly lower Ca content than the November magma. The June magma also evolved to higher contents of Cl, F, Ba and K and lower contents of Ca, Na and P than are found in the November lavas.

Mineralogical behaviour that reflects magmatic evolution

Accessory quantities of K-Fe sulphide minerals occur in many of the natrocarbonatite lavas and have been reported by Dawson *et al.* (1995) and Mitchell (1997). We have reported elsewhere (Jago and Gittins, 1999) tiny grains of alabandite

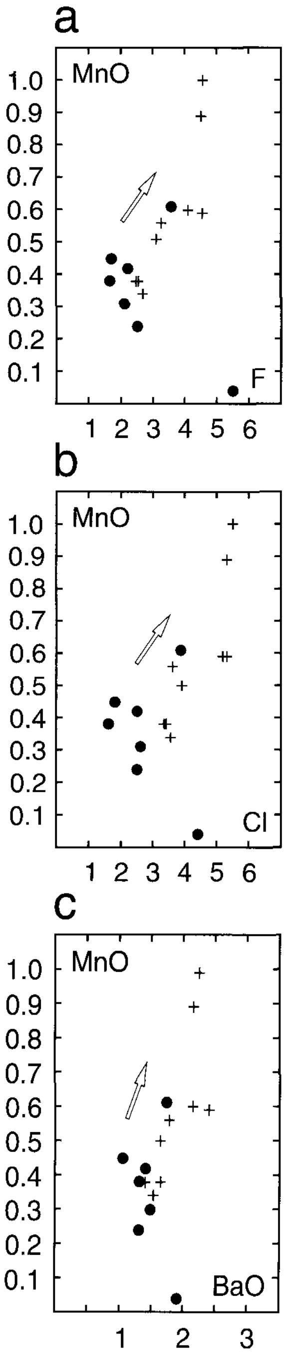

FIG. 3. Variation of elements (in wt.%) for Oldoinyo Lengai natrocarbonatite lavas. Symbols: crosses – June 1988 flows; solid circles – November 1988 flows. (*a*) MnO *vs* F; (*b*) MnO *vs* Cl; (*c*) MnO *vs* BaO. Arrows indicate direction of magmatic differentiation.

(MnS) with thin rims of a Mn- and F-bearing K-Fe sulphide that is probably a type of rasvumite. We interpret these rims as the product of reaction of alabandite with natrocarbonatite magma. The most likely cause of such a reaction is an increase in the K content as crystallization proceeded, and Fig. 1 clearly shows that K enrichment does occur.

Other mineralogical behaviour reflects the whole rock compositional trends. For example, Mn enrichment is consistent with the presence, in accessory amounts, of; (*a*) manganiferous magnetite (MnO = 14.35–15.34 wt.%; Jago, 1991; and MnO 10.3 wt.%; Dawson *et al*., 1995)) which is a magnetite–jacobsite solid solution, (*b*) manganiferous monticellite (Peterson, 1990; Church and Jones, 1995), and (*c*) a manganiferous silicate that is probably spurrite or tilleyite (Dawson *et al.*, 1995).

Fluorite is restricted to the matrix of the lavas as symplectic intergrowths, generally with sylvite; it does not occur as phenocrysts. Jago and Gittins (1991) have shown that F is extremely soluble in liquids throughout the system Na_2CO_3–$CaCO_3$–F and that crystallization of fluorite does not begin until the concentration of F reaches 5–8 wt.%. Such concentrations are reached only in the liquid that is interstitial to phenocrysts, and in the aphyric 'squeeze-ups' and driblets; the fact that the aphyric lavas are richer in fluorite than the porphyritic type is consistent with this experimental finding, and with progressive enrichment of the magma in F.

Although the behaviour of Cl has not been studied experimentally it is probably similar to that of F. Sylvite, like fluorite, does not occur as phenocrysts but is found only in the quenched matrix. Since sylvite crystallization does not occur until the magma reaches a highly evolved stage, K concentration is able to continue increasing throughout most of the crystallization history, as is displayed by Fig. 1. When crystallization of sylvite does begin there will be a rapid removal of K from the magma. An indication of this effect is the development of distinctly more sodic rims on many phenocrysts of gregoryite. Dawson *et al.* (1996) cite a groundmass gregoryite grain with 50.9 wt.% Na_2O and Peterson (1990) gives examples of core compositions with 43.38 wt.% Na_2O and rim compositions with 46.00 wt.% Na_2O. It was Peterson who first suggested that this is due to late crystallization of sylvite actively depleting the magma in K. It appears that Cl has a greater affinity for K than does gregoryite.

We have also noted that as the magma becomes enriched in Cl, F, Ba, K, Mg and Mn it is concomitantly depleted in Na, Ca and P. The most likely cause of this depletion is the continuous co-crystallization of nyerereite and gregoryite, both of which contain considerable quantities of both Ca and Na (nyerereite averages 24 wt.% CaO and 24 wt.% Na_2O; gregoryite averages 9 wt.% CaO and 43 wt.% Na_2O; Peterson, 1990) and so they are very effective in removing Ca and Na from the magma. They also contain only minor to trace amounts of F and Cl so are equally effective at concentrating both elements in the interstitial liquid. This concentration continues until fluorite and sylvite are stabilized as liquidus phases, and then the concentration of both will decrease markedly. Gregoryite also averages 2 wt.% P_2O_5 (Peterson, 1990) and so is responsible for almost all of the P depletion, given the abundance of gregoryite in natrocarbonatite. Apatite does not crystallize readily in natrocarbonatite liquid (Jago, 1991); consequently it is a rare mineral and has little effect on P control.

In all of the June lava analyses, and in four of the six November analyses, wt.% Cl > F (Cl/F = 1.0–1.4) and the probable course of Cl and F behaviour can be predicted by the mineralogy of the October 17th 1995 flow that has been described by Mitchell (1997). As noted earlier in this paper, it has circular patches, that appear to be sections of globules. These consist of gregoryite and sylvite and are encased in a symplectic intergrowth (quenched liquid) of nyerereite and gregoryite. Most importantly, there is no fluorite in either the globules or the surrounding matrix. Mitchell interprets this texture as the result of two immiscible liquids, one of which is Cl- rich and F-poor, and the other contains no significant Cl or F.

Such an explanation is consistent with progressive removal of F from the magma until its concentration is such that fluorite is no longer a liquidus phase, while Cl concentration remains high enough for sylvite to continue crystallizing.

A further effect of F concentration being followed by its depletion is the absence of villiaumite (NaF) in natrocarbonatite lava, despite the fact that the system Na_2CO_3–$CaCO_3$–F (Jago, 1991; Jago and Gittins, 1991) contains a primary phase field of villiaumite. The most probable explanation is that crystallization of fluorite eventually reduces the concentration of F to a level that is too low to allow for villiaumite being a liquidus phase. At this stage Na combines with Cl

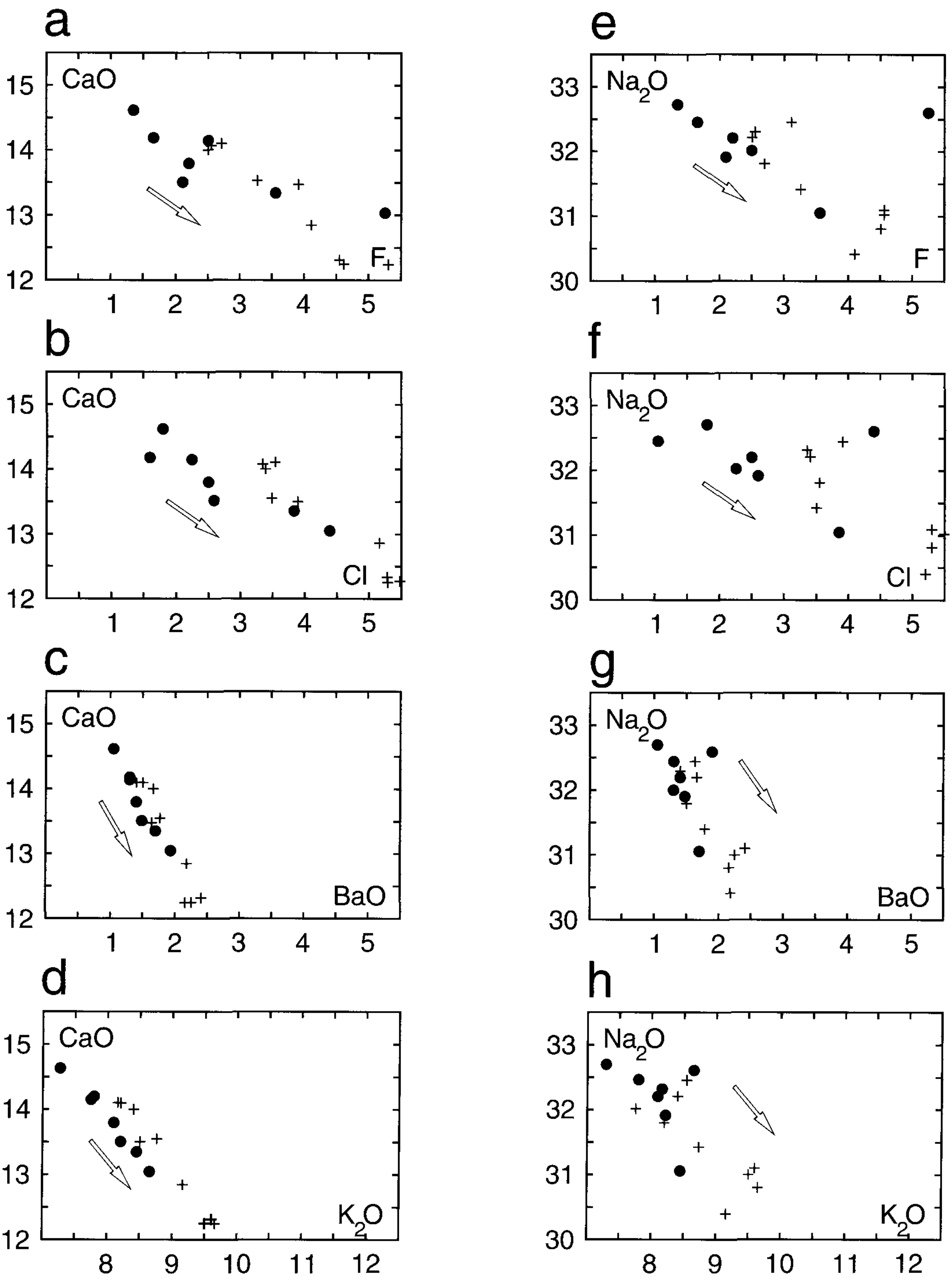

FIG. 4. Variation of elements (in wt.%) for Oldoinyo Lengai natrocarbonatite lavas. Symbols: crosses – June 1988 flows; solid circles – November 1988 flows. (*a*) CaO *vs* F; (*b*) CaO *vs* Cl; (*c*) CaO *vs* BaO; (*d*) CaO *vs* K_2O; (*e*) Na_2O *vs* F; (*f*) Na_2O *vs* Cl; (*g*) Na_2O *vs* BaO; (*h*) Na_2O *vs* K_2O. Arrows indicate direction of magmatic differentiation.

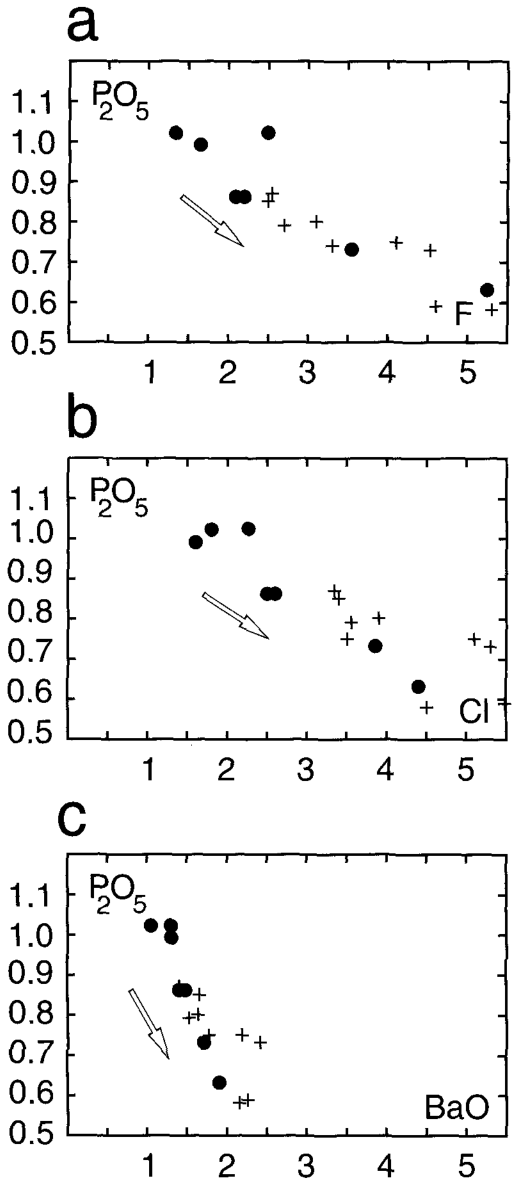

FIG. 5. variation of elements (in wt.%) for Oldoinyo Lengai natrocarbonatite lavas. Symbols: crosses – June 1988 flows; solid crosses – November 1988 flows. (*a*) P_2O_5 *vs* F; (*b*) P_2O_5 *vs* Cl; (*c*) P_2O_5 *vs* BaO; Arrows indicate direction of magmatic differentiation.

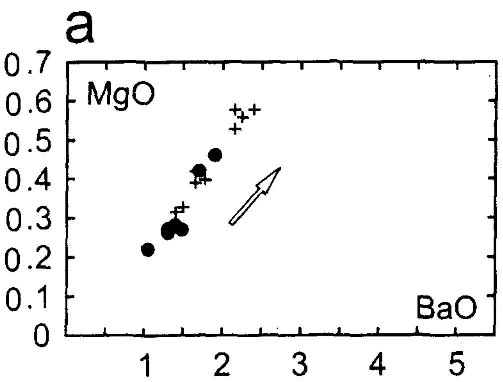

FIG. 6. Variation of MgO *vs* BaO (in wt.%) for Oldoinyo Lengai natrocarbonatite lavas. Symbols: crosses – June 1988 flows; solid circles – November 1988 flows. Arrow indicates direction of magmatic differentiation.

as (halite-sylvite)$_{SS}$ rather than forming villiaumite. Dawson *et al.* (1996) report halite contents of 25–31 mol. % and Mitchell (1997) reports a range of 26–36 mol.%. Increasing halite content might be associated with a very late-stage reversal in the trend of Na depletion. Each of Fig. 4(*e*–*h*), which display the depletion of Na that accompanies increasing concentration of F and Cl, has three points at the most evolved end of the trends in which Na depletion sharply reverses itself. This is probably a genuine trend that manifests itself in the most evolved, aphyric rocks, although we cannot rule out analytical error.

Although reduced F concentration seems to prevent the crystallization of villiaumite, Mitchell (1997) reports tiny grains of a Na-K-Mg-Ba fluoride mineral thought to be a neighborite–K-neighborite solid solution that is intimately intergrown with the principal minerals of the lava. It appears, therefore, that there is still sufficient F in the magma at this late stage in its evolution to allow some fluoride minerals to crystallize. This mineral is particularly interesting for its high Mg and Ba contents (MgO 15–17 wt.%; BaO 8–15 wt.%) in light of the fact that both elements increase steadily as the natrocarbonatite magma evolves (Fig. 6), no doubt reaching the concentration necessary to stabilize the mineral as a liquidus phase only in the late stages of this evolution. A further indication of the concentration of Mg and Ba in the later stages of magmatic evolution is the presence of sellaite (MgF_2), and of 'Phase X' (Dawson *et al.*, 1995) which is a possible carbonate mineral (MgO 0.8–3.0 wt.%; BaO 18–44 wt.%). Mg enrichment is the opposite of what is commonly encountered in silicate magmas, where Mg is progressively replaced by Fe^{2+}. But, as we have already seen, natrocarbonatite magma crystallization is dominated by nyerereite and gregoryite, neither of which contains more than minor Mg. The result must be a progressive increase in Mg concentration.

The directions of magmatic evolution described here also make it impossible for natrocarbonatite magma to be considered as a parental magma from which calcitic and dolomitic carbonatites might be derived, as was proposed by Le Bas (1977) and still occasionally referred to. Declining Na, Ca and P and increasing K, Cl and F are entirely the wrong direction to produce calcite and dolomite and the Na-enrichment trends so common in carbonatite amphiboles. Furthermore, since P declines from an already low level, it is impossible for it to become concentrated to the levels reached in apatite-bearing carbonatites

Status of aphyric and porphyritic natrocarbonatites

How closely does natrocarbonatite lava represent the composition of the parental magma from which it crystallized? Some believe that crystal accumulation has been extensive, that "The liquids erupted cannot represent primary natrocarbonatite magma", and that the aphyric liquid is a more primitive composition to which crystals of nyerereite and gregoryite have been added, either by disruption of an early-formed cumulate or by flotation accumulation (Mitchell, 1997). We are inclined to disagree. It seems to us that the aphyric lavas, with their very high contents of F, Cl and SO_3, must represent a later stage of differentiation than do the porphyritic varieties. By our interpretation the aphyric lavas are representative of the interstitial liquid that has been squeezed from the porphyritic varieties.

We have previously emphasized that it is nyerereite and gregoryite which effectively control the course of magmatic evolution in all but the very last stages. We caution against assuming that "the very high modal abundance of phenocrysts of gregoryite and nyerereite indicates that crystal accumulation has occurred prior to eruption" and, consequently, that "the bulk composition of the lavas cannot represent the composition of their parental magma" (Mitchell, 1997). We accept that crystal accumulation might well have occurred but, because of the dominance and continuous co-crystallization of nyerereite and gregoryite, their accumulation changes the bulk composition of natrocarbonatite very little. We, therefore, suggest that the parental magma composition is little different from that of the lavas, with the exception of the very small-volume aphyric driblets and squeeze-ups.

The ratio of nyerereite:gregoryite appears to have remained constant throughout most of the crystallization history. Evidence of this is provided by the behaviour of Sr, which resides principally in nyerereite and gregoryite. Nyerereite has an average SrO content of 2.24 wt.%, whereas gregoryite has one of 0.8 wt.% (Peterson, 1990). Although there are some Sr-rich minerals, they are present in only accessory quantities. Yet, the SrO content of the lavas is remarkably constant at 1.42 wt.% (standard deviation 0.1) for June 1988, and 1.70 (standard deviation 0.05) for November 1988. This is in striking contrast to all other major elements, which display sympathetic variation as illustrated in Figs. 1–6. Given the ratio of 2.8:1 for SrO (nyerereite)/SrO (gregoryite), any substantial variation in the proportions of co-crystallizing nyerereite and gregoryite would have a strong effect on the SrO content of the co-existing natrocarbonatite magma. Yet, there is no significant difference between the SrO content of aphyric and porphyritic natrocarbonatite (see Table 1) and so we may reasonably conclude that nyerereite and gregoryite have co-crystallized in roughly constant proportion. This is very suggestive of crystallization on a nyerereite–gregoryite cotectic such as exists in the synthetic systems Na_2CO_3–$CaCO_3$–CaF_2 and Na_2CO_3–$CaCO_3$–F (Jago, 1991; Jago and Gittins, 1991).

The exact composition of the parental natrocarbonatite magma will have to await more detailed experimental study but we incline to the view that it will plot in, or at the edge of, the primary phase field of nyerereite in these systems.

We agree with the commonly held view that natrocarbonatite magma is highly evolved but we stress that fractionation has caused even greater evolution, and that the aphyric compositions represent the most evolved compositions rather than being "the actual liquid present in the magma chamber underlying the crater floor" (Mitchell, 1997) to which cumulate nyerereite and gregoryite has been added.

Conclusions

There is general agreement that natrocarbonatite magma at Oldoinyo Lengai, whatever its origin, is highly evolved and some of the directions outlined here have been pointed out by Dawson, Keller and Peterson in various publications. We suggest, however, that this evolution is more extensive than has previously been noted. Progressive differentiation affects an already evolved magma and drives it to the very highly evolved aphyric types.

Although it is common to assume that phenocryst-rich rocks are the result of crystal accumulation, and so do not represent the composition of the liquid from which they crystallized, we suggest that natrocarbonatites are a special case in which phenocryst-rich lavas really are very close in composition to the parental magma.

We emphasize that the analyses employed in this study are of lavas that erupted over a period of only a couple of days and so we know very little about the trend that would emerge from a suite collected over a much longer period of continuous eruptive activity. It is tempting to speculate, however, that fractionation may be occurring in isolated, relative small magma reservoirs. Indeed, Pyle *et al.* (1995) have estimated that some of the individual reservoirs in the crater area have radii as small as 5–10 m.

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We are indebted to Dr B.C. Kjarsgaard for pointing out an ambiguity in our use of the term 'parental magma' in this paper. We have used it solely in the sense of the natrocarbonatite magma from which the series of variously differentiated natrocarbonatite lavas crystallized. We do not intend to imply that this magma is in any way a primitive magma and have scrupulously avoided committing ourselves to fractionation or to liquid immiscibility origins for the magma that we have here referred to as 'parental'.