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Carbonatite Magmatism and Plume Activity: Implications from the Nd, Pb and Sr Isotope Systematics of Oldoinyo Lengai

New Nd (0.51261–0.51268), Pb ($^{206}\text{Pb}/^{204}\text{Pb}$: 19.24–19.26), and Sr (0.70437–0.70446) isotopic compositions from ten natrocarbonatite lavas, collected in June 1993 from Oldoinyo Lengai, the only known active carbonatite volcano, are relatively uniform, and are similar to data from the 1960 and 1988 flows. Three of the samples contain silicate spheroids, one of which has Nd and Sr isotopic ratios similar to host natrocarbonatite, consistent with an origin by liquid immiscibility or the mixing of melts with similar isotopic compositions. Pb isotope data for two samples of trona are inconsistent with its involvement in the genesis of natrocarbonatite. New Pb isotope data from silicate volcanic and plutonic blocks (ijolite, nephelinite, phonolite, syenite) from Oldoinyo Lengai are highly variable ($^{206}\text{Pb}/^{204}\text{Pb}$, 17.75–19.34; $^{207}\text{Pb}/^{204}\text{Pb}$, 15.41–15.67; $^{208}\text{Pb}/^{204}\text{Pb}$, 37.79–39.67), and define near-linear arrays in Pb–Pb diagrams. The isotopic data for the silicate rocks from Oldoinyo Lengai are best explained by invoking discrete partial melting events which generate undersaturated alkaline silicate magmas with distinct isotopic ratios. Pb isotope ratios from most ijolites and phonolites are predominantly lower and more variable than from the natrocarbonatites, and are attributed to interaction between silicate melts involving HIMU and EMI source components and an additional component, such as lower-crustal granulites, DMM or PREMA (prevalent mantle). Variations in Nd, Pb and Sr isotope ratios from Oldoinyo Lengai, among the largest yet documented from a single volcano, are attributed to mantle source heterogeneity involving mainly the mixing of HIMU and EMI mantle components. Based on the new isotopic data from Oldoinyo Lengai and data from other East African carbonatites, and mantle xenoliths, we propose a two-stage model in an attempt to explain the isotope

variations shown by carbonatites in this area. The model involves (1) the release of metasomatizing agents with HIMU-like signatures from upwelling mantle ('plume') source, which in turn metasomatize the sub-continental (old, isotopically enriched, EMI-like) lithosphere, and (2) variable degrees and discrete partial melting of the resulting heterogeneous, metasomatized lithosphere.

KEY WORDS: carbonatite; isotopes; Oldoinyo Lengai; mantle plumes

INTRODUCTION

Nd, Pb and Sr isotope data from carbonatites have now been shown to be effective in monitoring the nature and the temporal evolution of the sub-continental mantle (e.g. Bell & Blenkinsop, 1987a, 1987b; Nelson *et al.*, 1988; Tilton & Bell, 1994). In addition, recent studies of mantle xenoliths and high-pressure experiments have demonstrated that carbonatite melts may be important metasomatic agents within the mantle (e.g. Dalton & Wood, 1993; Rudnick *et al.*, 1993). With their unusual chemistries (Woolley & Kempe, 1989), extremely low viscosities (Dawson *et al.*, 1990; Pinkerton *et al.*, 1995) and low dihedral wetting angles, carbonatite melts have all the properties needed to bring about profound chemical changes in the upper mantle. Some of the major issues surrounding carbonatite genesis are: (1) whether carbonatites are primary or secondary magmas; and (2) whether the silicate rocks associated with carbonatites represent conjugate liquids

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resulting from immiscibility, or are the products of interaction between carbonatitic melts and mantle or crustal wall rocks. The origin of carbonatite melts remains a topic for debate, with some workers favouring magmatic differentiation, including immiscible separation from a carbonated, parental silicate magma (Kjarsgaard & Hamilton, 1989; Church & Jones, 1995; Jones *et al.*, 1995; Kjarsgaard *et al.*, 1995), whereas others advocate a primary mantle melt (Wallace & Green, 1988; Dalton & Wood, 1993). On the basis of phase equilibrium studies, Wallace & Green (1988) first proposed the idea of a 'primary' sodic, dolomitic carbonatite magma in equilibrium with an amphibole lherzolite assemblage. Although the carbonatite melt was considered sodic, it contained only 5.5 wt% Na₂O. However, closed system fractionation of the primary melt could generate natrocarbonatites. Recent experiments by Sweeney *et al.* (1995) failed to duplicate these findings and showed that if dolomite, olivine and spinel crystallized from such a liquid to produce natrocarbonatite, then the primary melt would have to be more silicic, less FeO rich, and have a greater K/Na ratio than the primary liquid of Wallace & Green (1988). Further experimental work by Dalton & Wood (1993) demonstrated that it is, however, possible to produce parental melts similar to natrocarbonatites by low-pressure (<15 kbar) wall-rock reaction between a primary calciocarbonate melt and fertile lherzolite.

As the only known active carbonatitic volcano, Oldoinyo Lengai plays a key role in unravelling the difficulties involved in understanding the evolution and genesis of carbonatitic magmas. Although a great deal of Nd, Pb and Sr isotope data is available from carbonatites from several continents, there are only a handful of case studies where carbonatites and their associated silicate rocks have been investigated involving all three isotope systems (e.g. Chilwa Island, Simonetti & Bell, 1994b). Samples from Oldoinyo Lengai provide an opportunity to establish a complete Nd–Pb–Sr isotope database for both carbonate and silicate rocks.

Oldoinyo Lengai

Situated within the eastern branch of the East African Rift Valley (Fig. 1) in northern Tanzania, the 2200 m cone of Oldoinyo Lengai is the youngest of a series of Neogene–Quaternary nephelinite–phonolite–carbonatite volcanoes. The volcano consists mainly of nephelinitic and phonolitic ashes, tuffs and agglomerates with minor flows of nephelinite and natrocarbonatite; plutonic blocks occur within the pyroclastics and are mainly cumulates, including

ijolite (roughly equal amounts of clinopyroxene and nepheline), jacupirangite (mainly clinopyroxene with some nepheline), nephelinite syenite and pyroxenite. Although in the literature the most recent eruptions of natrocarbonatite from Oldoinyo Lengai have been highlighted [see review by Bell & Keller (1995)], there is evidence for earlier periods of carbonatitic activity (Dawson, 1962).

The natrocarbonatite lavas from Oldoinyo Lengai are chemically different from all other known carbonatites, whether plutonic or volcanic, in containing large amounts of alkalis (up to 33 wt% Na₂O and 5 wt% K₂O). Other chemical characteristics unique to natrocarbonatites include: (1) extremely high Ba contents (>10 000 p.p.m.) and Ba/Sr > 1.0; (2) Nb < 110 p.p.m. and Zr < 5 p.p.m.; (3) Mo and W abundances > 90 p.p.m.; (4) Th/U ~ 1.0; (5) LREE concentrations > 1000 times chondritic (Keller & Spettel, 1995; Simonetti *et al.*, 1996b). The unusual chemical composition of natrocarbonatite has been explained by crystal fractionation of a sövite parent magma (Gittins, 1989), immiscible separation from a carbonated peralkaline nephelinite (Kjarsgaard *et al.*, 1995), and interaction between a silicate magma and Na-rich evaporite products such as trona or saline brine (Milton, 1968; Eugster, 1970). Natrocarbonatites are now considered to represent the products of protracted magmatic fractionation rarely encountered in nature (Bell & Keller, 1995; Kjarsgaard *et al.*, 1995).

A considerable amount of Nd and Sr isotope data has been obtained in recent years from the natrocarbonatites from Oldoinyo Lengai, and to a lesser extent from related silicate rocks. Among the findings are: (1) the C, Nd, O and Sr isotope ratios are consistent with a mantle origin for the natrocarbonatites (Deines, 1989; Bell & Blenkinsop, 1989; Keller & Hoefs, 1995); (2) the Nd and Sr isotope compositions of the natrocarbonatites fall close to the intersection of bulk Earth and CHUR on an Nd–Sr isotope diagram (Bell & Blenkinsop, 1987a; Keller & Krafft, 1990; Simonetti *et al.*, 1995a); (3) Nd and Sr isotope compositions of associated silicate flows and plutonic blocks are extremely variable (Bell & Dawson, 1995a), spanning almost two-thirds of the East African Carbonatite Line (Bell & Blenkinsop, 1987a), a line that mimics the 'LoNd' array of Hart *et al.* (1986) based on data from oceanic island basalts; (4) most lavas from Oldoinyo Lengai appear to have been produced by discrete melting events involving the mixing of the same two end-member source components (Bell & Dawson, 1995a); (5) the Pb isotope compositions from the natrocarbonatites fall close to or slightly above the NHRL in both ²⁰⁸Pb/²⁰⁴Pb vs ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁷Pb/²⁰⁴Pb vs

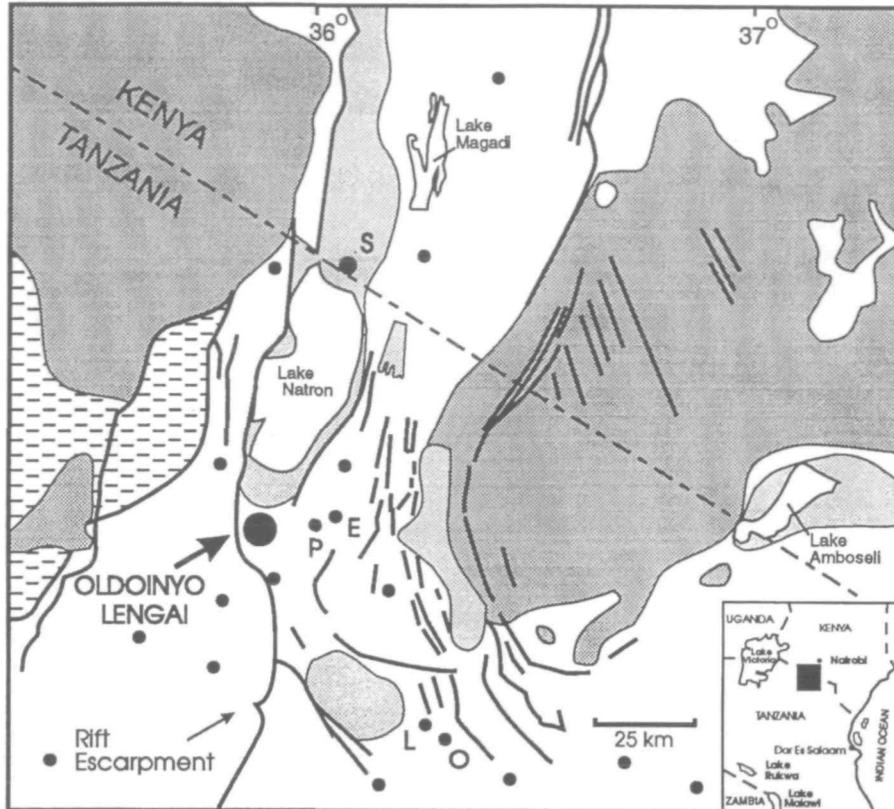


Fig. 1. Regional geology of East Africa showing part of the Tanzanian Shield, the East African Rift Valley System and Neogene-Quaternary lavas. Also shown is the distribution of some Tertiary and Recent eruptive centres marked by the black dots. S, Shombole; E, Eledoi; L, Lashaine; O, Olmani; P, Pello. Dark grey area, basement; light grey unit, modern alluvium and/or detritus; dashed pattern, Neogene tuffs; white area, Neogene lavas [modified after Dawson (1992)].

$^{206}\text{Pb}/^{204}\text{Pb}$ plots (Williams *et al.*, 1986; Dawson *et al.*, 1995; Simonetti *et al.*, 1995a); (6) U-Th isotope disequilibrium studies suggest that segregation of the natrocarbonatite and eruption took place over an interval of 20–81 years (Williams *et al.*, 1986; Pyle, 1995).

A suite of ten natrocarbonatite samples collected in June 1993 was donated to us by C. Shradly. Included among these are samples from the Chaos Crags flow, a highly viscous spheroid-bearing natrocarbonatite flow with a viscosity similar to that of a rhyolite (Dawson *et al.*, 1994). The spheroids, described in detail by Dawson *et al.* (1994), Church & Jones (1995) and Dawson *et al.* (1996), are <2 mm in size, and most are porphyritic containing phenocrysts of nepheline, schorlomite-Ti-andradite, garnet, hedenbergitic pyroxene and wollastonite, along with rare titanite and melilite. Many spheroids contain cores of single euhedral phenocrysts. The phenocrysts are set in a fine-grained matrix consisting of much smaller grains of the same minerals, plus pyrrhotite, perovskite, Ti-magnetite, melilite and peralkaline silicate glass. The mineralogy of the

silicate spheroids is essentially that of a wollastonite nephelinite but they also contain minor, but ubiquitous, rounded or ovoid globules of natrocarbonatite.

The BD suite of samples, collected by J. B. Dawson, contains both volcanic and plutonic magmatic blocks and metasomatized rocks. The volcanic rocks are highly evolved olivine-free nephelinites and phonolites that represent samples of a gradational series, the detailed petrography, mineral chemistry and bulk chemistry of which has been given by Donaldson *et al.* (1987). In general terms, the lavas are porphyritic, and consist of nepheline, aegirine-augite, Ti-andradite and in the phonolites, sanidine phenocrysts are set in a matrix of nepheline and aegirine microphenocrysts, apatite, pyrrhotite, wollastonite, glass (commonly palagonitized) and perovskite or titanite. The magmatic plutonic rocks, ranging from jacupirangite to ijolite, show cumulate textures and consist of varying proportions of clinopyroxene, nepheline, titanomagnetite, apatite, Ti-andradite, perovskite and wollastonite. One of the plutonic blocks is a eucolite-bearing nepheline

syenite (BD872), containing aegirine-augite, nepheline, K-feldspar and trace amounts of apatite and titanite.

The major and trace element data for both the natrocarbonatite and silicate samples, taken from Donaldson *et al.* (1987) and Simonetti *et al.* (1996b), are shown in Tables 1 and 2. The aims of our study are to: (1) compare the Sr, Nd and Pb isotope compositions of the 1993 natrocarbonatite flows with those from the 1960 and 1988 eruptions, (2) evaluate the relationship of silicate minerals from the spheroids in the 1993 flows to the enclosing natrocarbonatite, (3) provide new Pb isotopic data from older, associated silicate lavas and blocks, (4) place additional constraints on rejecting the trona hypothesis as a way to provide the high Na₂O contents of the natrocarbonatites, and (5) explain the large variation in Nd, Pb and Sr isotope ratios, particularly for the silicate rocks from Oldoinyo Lengai.

ANALYTICAL METHODS

Analytical procedures for Nd and Sr are similar to those described by Bell & Blenkinsop (1987b). All of the whole-rock silicate samples (~50 mg) were dissolved using HNO₃ and HF in sealed Savillex beakers for at least 24 h. The separation of silicate material from spheroid-bearing natrocarbonatite lava (OL-6) was achieved by acid leaching using cold 2.5 N HCl. Carbonate-free clinopyroxene and garnet crystals were then hand-picked from the silicate residue and ultrasonically washed several times in ultrapure water. The pyroxene-garnet separate (~150 mg) was then subjected to sequential leachings of 2 h each using hot 2.5 N HCl. The grains were again washed with ultrapure water between successive leachings. The sequential leachings were carried out to ensure that all carbonate-held Sr was removed. Sample dissolution was identical to that used for the whole-rock silicate samples. Lead was separated from other elements in HBr and HCl using a two-column anion ion-exchange technique (Simonetti *et al.*, 1995b). No leaching was carried out on the whole-rock, silicate samples so that results from this study may be compared with those for nephelinites from Mount Elgon and Napak. A few samples of nephelinite from Napak, however, were leached (Simonetti & Bell, 1994a) and the data show that the isotope signatures for the whole rocks and associated leachates were similar (within analytical error). Neodymium and Sr were analysed using a double Re-filament technique, and all isotope ratios were measured (in static mode) using Finnigan-MAT 261 multicollector mass spectrometers at Carleton University, Ottawa, and the

Max-Planck-Institut für Chemie at Mainz. The Pb samples were run on a single Re filament using silica gel and phosphoric acid.

Nd-Sr-Pb isotope data

Nd-Sr isotope data

The new Nd and Sr isotopic data from the natrocarbonatites, listed in Table 3 and plotted in Fig. 2, are extremely uniform, clustering close to the intersection of CHUR and bulk Earth on an Nd-Sr isotope plot, and similar to previously published data for the 1960 and 1988 flows (Keller & Krafft, 1990; Bell & Dawson, 1995a). Despite the fact that the trace element geochemistry of natrocarbonatites differs significantly from that of all other carbonatites (Keller & Spettel, 1995; Simonetti *et al.*, 1995a, 1996b), their Nd and Sr isotope signatures correlate with isotope data from calcicarbonatites from other parts of East Africa. The new Nd and Sr isotope data fall close to the East African Carbonatite Line (EACL—Bell & Blenkinsop, 1987a), a linear array that mimics the 'LoNd' array of Hart *et al.* (1986). The data points which form the EACL do not cluster into groups but form a continuum. Also shown in Fig. 2 are the data from Homa Bay, the most EMI-like end-member for the EACL. Nd and Sr isotope data from mantle xenoliths from cinder and tuff cones (Eledoi, Lashaine, Olmani, Pello Hill—Cohen *et al.*, 1984; Rudnick *et al.*, 1993) proximal to Oldoinyo Lengai and the field representing isotope ratios for carbonatite from the Canary Islands (Hoernle & Tilton, 1991), a supposed-plume-related, 'oceanic' carbonatite, are shown for comparison in Fig. 3. The large variation shown by the data includes analyses from a variety of mantle xenoliths, supposedly metasomatized by carbonatitic magmas (Rudnick *et al.*, 1993), and an amphibole-phlogopite vein from a spinel lherzolite (Cohen *et al.*, 1984). Most of the isotope data in Fig. 3 plot close to the EACL, and proximal to the HIMU end-member composition, including five analyses from three young (<40 Ma) Ugandan carbonatites, Bukusu, Napak and Sukulu (Bell & Blenkinsop, 1987a; Nelson *et al.*, 1988; Simonetti & Bell, 1994a).

Previously published Nd and Sr isotope data from the Oldoinyo Lengai silicate flows and plutonic blocks (Bell & Dawson, 1995a), define a linear array in Fig. 2 with the nephelinites forming two distinct groups, one in the enriched quadrant corresponding to the wollastonite-bearing nephelinites (Group II) and the other (including three combeite-bearing lavas) in the depleted quadrant (Group I). The linear array defined by the data for the silicate flows, almost identical in slope to the EACL, does not

Table 1: Major and trace element data* from Oldoinyo Lengai 1993 natrocarbonatite lavas

Sample:	OL-1	OL-1(2)	OL-2	OL-5	OL-6	OL-7	OL-8	OL-9	OL-10	OL-11
<i>Major elements (wt %)</i>										
Fe ₂ O ₃	0.27	0.23	0.27	1.34	1.49	0.10	0.25	0.92	0.22	0.18
CaO	13.43	13.01	13.43	15.95	16.25	13.01	13.15	15.25	13.85	13.99
SrO	1.32	1.30	1.34	1.22	1.24	1.30	1.30	1.37	1.28	1.31
BaO	1.93	1.84	1.76	0.94	0.91	1.90	1.88	1.14	1.51	1.60
Na ₂ O	30.87	30.33	30.73	27.50	26.97	31.14	31.41	29.12	30.33	31.27
<i>Trace elements (p.p.m.)</i>										
Au (p.p.b.)	20	15	14	12	19	17	33	16	18	19
Ag	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
As	20	19	21	12	13	19	21	16	16	19
Ba	17300	16500	15800	8390	8180	17000	16800	10200	13500	14300
Br	120	110	110	46	42	120	120	53	97	100
Co	1.6	1.4	1.6	2.4	2.9	1.5	1.5	1.5	1.1	1.2
Cr	1.3	1.6	<0.5	2.6	1.8	<0.5	1.0	0.7	<0.5	<0.5
Cs	7.8	7.4	7.1	3.4	3.2	7.4	7.6	4.2	5.9	6.3
Hf	<0.2	<0.2	<0.2	1.4	1.2	<0.2	<0.2	1.0	<0.2	<0.2
Hg	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Ir	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Mo	176	165	154	73	69	174	175	70	133	136
Ni	<50	<50	<50	<50	<50	<50	<50	<50	<50	<50
Rb	235	233	221	134	134	239	233	160	199	208
Sb	3.0	2.9	2.8	1.4	1.3	2.9	3.0	1.7	2.4	2.6
Se	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Sr	11200	11000	11300	10300	10500	11000	11000	11600	10800	11100
Ta	<0.3	<0.3	<0.3	<0.3	0.5	<0.3	<0.3	0.3	<0.3	<0.3
W	113	112	106	41	47	114	116	56	94	97
Zn	64	69	58	184	198	39	36	190	56	38
Sc	0.1	0.1	0.1	0.2	0.3	0.1	0.1	0.2	0.1	0.1
Th	2.7	2.8	2.7	11.1	11.4	1.3	1.3	8.1	2.6	0.5
U	2.3	2.8	3.2	10.5	10.7	1.5	1.6	9.5	2.2	<0.1
La	347	336	328	480	487	312	310	531	294	284
Ce	313	306	296	552	565	272	272	575	285	251
Nd	58	60	56	122	128	48	50	117	59	47
Sm	4.45	4.42	4.41	9.85	10.20	3.76	3.78	9.03	4.58	3.31
Eu	1.10	1.13	1.09	2.42	2.54	0.95	0.93	2.18	1.10	0.77
Tb	0.2	0.3	0.3	0.6	0.7	0.2	0.2	0.4	<0.1	0.2
Yb	0.23	0.15	0.19	0.70	0.81	0.07	0.10	0.51	0.21	0.07
Lu	0.03	0.02	0.02	0.10	0.10	0.01	0.02	0.05	0.03	<0.01

*Data from Simonetti *et al.* (1996b). Data obtained by INAA at Activation Laboratories (Ancaster, Ontario). Samples were encapsulated in polyethylene vials and irradiated at a thermal neutron flux of $7 \times 10^{12} \text{ ncm}^{-2}\text{s}^{-1}$ together with CANMET MGR-1 standard and flux wires. After a 7-day decay, the samples were counted for 2000 s on a high-purity Ge detector with 15% efficiency and a resolution of 1.68 keV for the 1332 ⁶⁰Co photopeak. After a further decay of 24 h, the samples were re-counted. Four days after the first count the samples were re-counted for 5000 s. All data were calculated by using the flux monitors with the standard present used solely as a check on accuracy.

Table 2: Major and trace element data* for silicate rocks from Oldoinyo Lengai

Sample:	BD119	BD120	BD81	BD64	BD54	BD126	BD121	BD67	BD50	BD29	BD74
Type:	N-1	N-1	N-2	N-2	N-2	N-2	P	P	P	P	P
<i>Major elements (wt %)</i>											
SiO ₂	43.07	43.48	44.74	46.47	46.83	46.12	50.92	49.36	52.56	51.58	52.92
TiO ₂	0.99	0.97	1.88	0.99	1.21	0.91	1.19	0.77	0.94	0.78	0.90
Al ₂ O ₃	13.96	13.26	14.63	17.05	16.62	18.33	18.59	18.22	19.30	18.33	19.83
Fe ₂ O ₃	5.80	6.92	7.21	5.93	6.86	5.70	4.45	4.10	3.44	4.22	3.66
FeO	2.69	2.47	1.88	0.85	0.82	1.33	1.78	0.74	1.92	1.33	1.78
MnO	0.34	0.38	0.23	0.22	0.22	0.22	0.18	0.18	0.17	0.19	0.17
MgO	1.01	0.66	2.02	0.88	1.00	0.52	0.96	0.53	0.59	0.67	0.66
CaO	11.08	8.40	8.70	8.96	5.61	5.00	4.69	4.43	3.05	3.86	2.88
Na ₂ O	9.70	7.48	8.06	9.29	7.21	8.76	9.10	8.54	10.19	8.68	10.21
K ₂ O	4.90	5.43	4.31	5.17	5.88	5.06	4.78	5.59	4.85	5.17	4.65
P ₂ O ₅	0.66	0.39	0.63	0.27	0.43	0.45	0.38	0.35	0.18	0.35	0.18
SO ₃	0.28	0.57	0.09	0.14	0.10	0.08	0.07	0.03	0.15	0.13	0.05
H ₂ O ⁺	1.20	4.18	3.52	2.85	3.56	3.16	1.48	4.19	1.66	3.58	1.48
CO ₂	2.95	4.17	1.29	2.15	2.57	2.53	0.45	2.64	0.14	1.10	0.09
Total	98.63	98.76	99.19	99.20	98.92	98.17	99.02	99.67	99.14	99.97	99.46
<i>Trace elements (p.p.m.)</i>											
V	211	166	230	130	189	173	98	76	80	86	77
Cr	<5	<10	5	<5	<5	<5	<5	<5	<10	6	<5
Mn	2877	2802	1833	1793	1765	1705	1428	1564	1255	1452	1334
Ni	14	<10	14	10	11	9	11	11	<10	9	9
Cu	47	40	61	17	31	22	17	12	15	15	14
Zn	274	324	159	168	189	189	133	177	150	163	160
Rb	119	155	109	116	147	108	110	158	101	113	110
Sr	2288	3442	1646	2424	1926	1947	2080	2354	1655	1274	1438
Y	48	35	30	40	38	39	32	36	31	27	28
Zr	869	831	398	516	520	621	554	579	555	221	640
Nb	324	303	127	140	144	125	164	169	160	195	183
Ba	1720	2723	1468	1605	1457	1645	1622	2085	1653	1280	1448
Pb	43	51	17	26	33	22	17	48	41	32	34
Li	29	66	28	36	56	44	17	53	31	39	22
Mo	0	4	6	1	8	0	0	0	4	0	0
Cl	1600	1800	400	1450	400	200	1550	550	1800	300	1850
F	1740	1740	1980	1880	1800	1940	940	1260	1180	880	740
U†	10	9	2	5	4	2.5	7	7	6	2	7
Th†	20	19	9	n.d.	10.5	19	17.5	29.5	n.d.	32	32.5

Type: N-1, nephelinite (Group 1); N-2, nephelinite (Group 2); P, phonolite.

*Data from Donaldson *et al.* (1987).

†Data from Dawson & Gale (1970).

extend as far as the HIMU end-member. The Group I nephelinites have isotope ratios that overlap some of the plutonic rocks; mainly ijolite and all three combeite-bearing nephelinites belong to this depleted group. Relative to the EACL, most of the silicate rocks from Oldoinyo Lengai have higher

¹⁴³Nd/¹⁴⁴Nd ratios for a given ⁸⁷Sr/⁸⁶Sr ratio, but this divergence can be incorporated within the limits of the analytical uncertainty of the line (Bell & Dawson, 1995a).

The new data from the 1993 natrocarbonatite flows do not fall on any of the extreme positions of

Table 3: Isotopic data from June 1993 natrocarbonatites and trona

Sample	$^{87}\text{Sr}/^{86}\text{Sr}$	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$
<i>Carbonatites</i>					
OL-1	0.70441	0.51265	19.24	15.62	39.34
OL-1(2)	0.70437	0.51268	19.24	15.60	39.30
OL-2	0.70439	0.51268	19.26	15.63	39.38
OL-5	0.70442	0.51264	19.25	15.62	39.36
OL-6	0.70441	0.51262	19.25	15.62	39.35
OL-7	0.70439	0.51268	19.26	15.63	39.39
OL-8	0.70441	0.51268	19.24	15.61	39.31
OL-9	0.70446	0.51266	19.24	15.62	39.34
OL-10	0.70440	0.51261	19.24	15.61	39.33
OL-11	0.70440	0.51281	19.25	15.62	39.35
<i>Silicate spheroids</i>					
OL-6 (Py + Garnet)	0.70438	0.51266			
Leachate-1	0.70432				
Leachate-2	0.70431				
<i>Trona</i>					
BD1527	0.70462		18.62	15.45	36.33
BD1533	0.70560	0.51269	18.41	15.62	38.25

Nd and Sr isotope ratios normalized to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$ and $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. NBS 987 standard is 0.710256 ± 0.000014 ($n = 34$); La Jolla Nd standard is 0.511854 ± 0.00001 ($n = 25$); BCR-1 $^{143}\text{Nd}/^{144}\text{Nd}$ is 0.51266 ± 0.00002 . Uncertainties are given at the 2σ level. Reproducibility of Pb ratios is 0.1% at the 2σ level. An average fractionation factor of 0.1% per mass unit was applied to all measured ratios based on analyses of NBS 982.

the linear array (Fig. 2) and the uniform isotope ratios of the natrocarbonatites contrast markedly to the isotopic variability of the associated silicate lavas. This is unlike the isotopic data from Shombole, in which the carbonatites cover the complete range of Nd and Sr compositions similar to those of the associated nephelinites (Bell & Peterson, 1991). Because of the variable Nd and Sr isotope compositions of the nephelinites, Bell & Dawson (1995a) considered that the isotopic signatures of the 1960 and 1988 carbonatites may be one of many that could have existed in carbonatite magmas throughout the eruptive history of Oldoinyo Lengai, particularly as at least four phases of carbonatitic activity have been documented for the volcano (Dawson, 1962).

Pb isotope data

The new Pb data are listed in Tables 3 and 4. Also included in Table 4 are the Nd and Sr isotope compositions for the same silicate samples taken from Bell & Dawson (1995a). In both Pb isotope ratio diagrams the data form broad linear arrays (Figs 4 and 5). In the $^{207}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ diagram,

the slope for the data from Oldoinyo Lengai is 0.110 ± 0.007 (2σ), which is similar to the oceanic regression line. In both of the Pb isotope ratio diagrams the data also fall sub-parallel to the Northern Hemisphere Reference Line as defined by Hart (1984), and also fall close to the 'LoNd' array based on data from Walvis, San Felix, Comores, Northeast Seamounts, St Helena and Tubuai (Hart *et al.*, 1986). The Pb isotope data from Oldoinyo Lengai range between 17.75–19.34 for the $^{206}\text{Pb}/^{204}\text{Pb}$ ratios and 37.79–39.57 for the $^{208}\text{Pb}/^{204}\text{Pb}$ ratios, and are closer to the isotopic composition of EMI rather than HIMU. In general, the silicate lavas are less radiogenic than the natrocarbonatites, especially the ratios from the phonolites and some of the ijolites (Figs 4 and 5). In contrast to the two distinct groupings for the nephelinites based on the Nd and Sr isotope data, the Pb data are similar for both groups. Also shown for comparison in Figs 6 and 7 are data from mantle xenoliths from East Africa (Cohen *et al.*, 1984) and for the Ugandan carbonatites (Nelson *et al.*, 1988; Simonetti & Bell, 1994a). All of the Pb isotope data from the mantle xenoliths plot between HIMU and EMI in the Pb isotope ratio diagram, and within the broad array defined

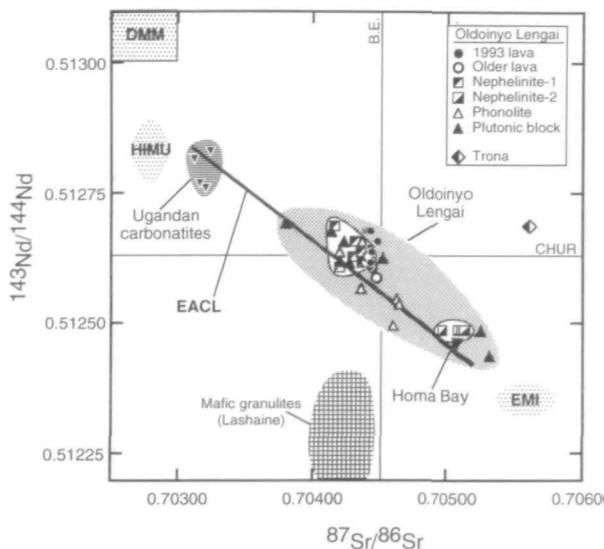


Fig. 2. Nd-Sr isotope plot showing the data from Oldoinyo Lengai. EACL from Bell & Blenkinsop (1987a). DMM, HIMU and EMI are from Hart (1988). $^{143}\text{Nd}/^{144}\text{Nd}$ value for CHUR is 0.512638, and $^{87}\text{Sr}/^{86}\text{Sr}$ for bulk Earth (B.E.) is 0.7045. Data for Ugandan carbonatites from Bell & Blenkinsop (1987a), Nelson *et al.* (1988) and Simonetti & Bell (1994a). Hatched field marks mafic granulite data from Lashaine (Cohen *et al.*, 1984). One granulite has ratios that lie below and to the left of the enclosed field.

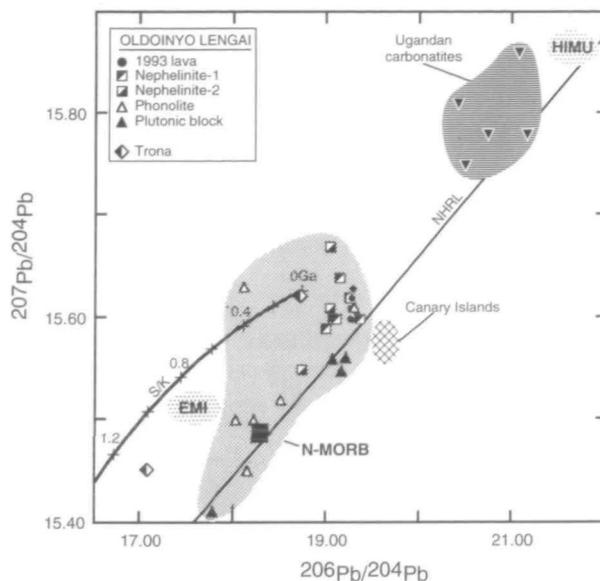


Fig. 4. $^{207}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ plot. S/K, Stacey-Kramers growth curve (Stacey & Kramers, 1975). Crosses along S/K curve are at 200 Ma intervals. Data for carbonatites from Uganda (Nelson *et al.*, 1988; Simonetti & Bell, 1994a) and Canary Islands (Hoernle & Tilton, 1991). HIMU, EMI and N-MORB mantle components are from Hart (1988). NHRL, Northern Hemisphere Reference Line (Hart, 1984).

by the data from Oldoinyo Lengai, excluding the phonolite analyses.

The Pb data from Oldoinyo Lengai fall close to an array defined by other East African carbonatites

(Grünenfelder *et al.*, 1986), but are the least radiogenic, by far, of any that have been analysed from young carbonatites world-wide (<200 Ma; Tilton & Bell, 1994), with the one exception of the Jacupiranga carbonatite from Brazil (Huang *et al.*,

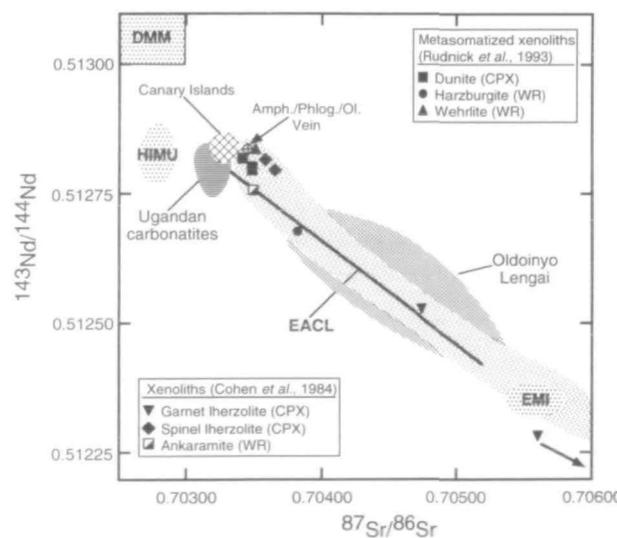


Fig. 3. Comparison Nd-Sr isotope plot with data for mantle xenoliths from East African extrusive centres. Mantle xenolith data from Cohen *et al.* (1984) and Rudnick *et al.* (1993). Data from the Canary Islands from Hoernle & Tilton (1991). Other features as in Fig. 2.

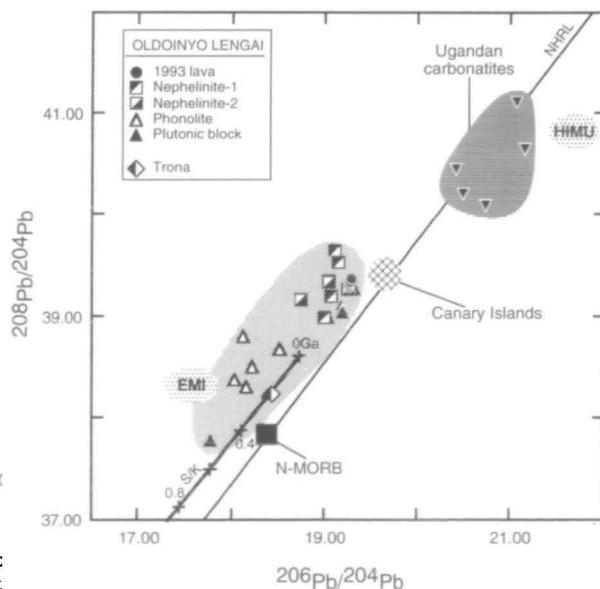


Fig. 5. $^{208}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ plot. Trona sample BD1527 has been omitted from the diagram. References as in Fig. 4.

Table 4: Isotope data for silicate rocks from Oldoinyo Lengai

Sample	Mode of occurrence	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{87}\text{Sr}/^{88}\text{Sr}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$
<i>Ijolites</i>						
BD33	B	0.51263	0.70439	19.15	15.58	39.14
BD35	B	0.51268	0.70412	18.33	15.47	38.26
BD45	B	0.51263	0.70450	19.02	15.59	39.03
BD49	B	0.51249	0.70522	18.61	15.55	39.07
BD52	B	0.51262	0.70425	17.75	15.41	37.79
BD122	B	0.51266	0.70421	19.16	15.56	39.08
BD343	B	0.51262	0.70441	19.13	15.55	39.08
<i>Nephelinites—Group I</i>						
BD66	L	0.51261	0.70418	19.34	15.60	39.26
BD120	L	0.51263	0.70428	19.05	15.60	39.22
HOL-6	L	0.51266	0.70429	19.08	15.60	39.67
HOL-10	L	0.51266	0.70429	18.97	15.59	39.01
HOL-14	L	0.51269	0.70414	19.22	15.62	39.29
HOL-16	L	0.51269	0.70414	19.12	15.64	39.55
<i>Nephelinites—Group II</i>						
BD54	B	0.51249	0.70612	19.02	15.61	39.37
BD64	L	0.51249	0.70507	18.71	15.55	39.18
BD126	B	0.51249	0.70495	19.02	15.67	39.36
<i>Phonolites</i>						
BD29	B	0.51257	0.70434	18.50	15.52	38.68
BD50	B	0.51255	0.70460	18.11	15.63	38.81
BD67	B	0.51250	0.70458	18.21	15.50	38.51
BD74	B	0.51254	0.70462	18.02	15.50	38.38
BD91	B	0.51264	0.70418	19.29	15.61	39.29
BD121	B	0.51266	0.70435	18.14	15.45	38.31
<i>Nepheline syenite</i>						
BD872	B	0.51262	0.70418	19.03	15.56	39.00

See Table 3 footnote. Nd and Sr ratios from Bell & Dawson (1995a). Mode: B, block; L, flow (Donaldson *et al.*, 1987; Bell & Dawson, 1995a).

1995). The East African carbonatite array (Grünenfelder *et al.*, 1986) in a $^{207}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ plot is almost identical to the slope of the oceanic regression line (Tatsumoto, 1978), and was interpreted by Grünenfelder *et al.* (1986) to be the result of mixing of two mantle components, one a large ion lithophile element (LILE) depleted source and the other a metasomatic fluid with a high $^{206}\text{Pb}/^{204}\text{Pb}$ ratio. With the exception of one sample, previous Pb isotope data for older Oldoinyo Lengai natrocarbo-

natites (Dawson *et al.*, 1995) are similar to those reported here.

INTERPRETATION OF THE ISOTOPE DATA

Natrocarbonatite—the result of liquid immiscibility?

Table 3 shows the Nd and Sr isotopic data for

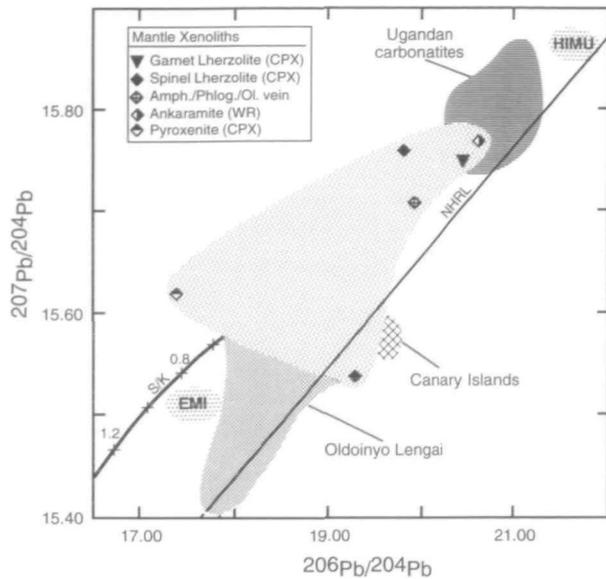


Fig. 6. $^{207}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ diagram for mantle xenoliths (Cohen *et al.*, 1984).

silicate grains (a mixture of garnet and clinopyroxene) from spheroids within natrocarbonatite sample OL-6 from the Chaos Crags flow. The formation of these spheroid-bearing flows was attributed by Dawson *et al.* (1994, 1996) to the mingling of a nyerereite–gregoryite crystal mush (remnant of an older, fractionated carbonatite magma), and a newly formed, carbonatitic ijolitic magma, which formed immiscible silicate spheroids

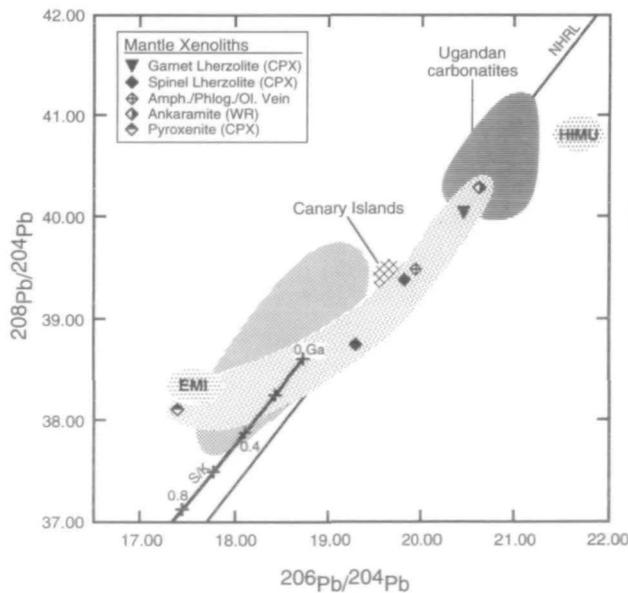


Fig. 7. $^{208}\text{Pb}/^{204}\text{Pb}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ diagram for mantle xenoliths (Cohen *et al.*, 1984).

within the crystal mush. In contrast, Church & Jones (1995) considered these spheroids as conjugate silicate liquids formed by liquid immiscibility after extensive fractionation of a carbonate-rich silicate magma. The Nd (0.51266) and Sr (0.70438) isotope ratios for the silicate minerals from spheroids are within analytical uncertainty of those from the host lava (OL-6), the Group I nephelinites, as well as the other 1993 natrocarbonatites (Fig. 8). The Sr isotopic ratios of the leachates are marginally lower than the isotopic composition of the natrocarbonatites (Table 1), suggesting that the leachable material is not in equilibrium with the host natrocarbonatite. The similar Nd and Sr isotopic ratios of the garnet–pyroxene separate and the natrocarbonatite may support the liquid immiscibility hypothesis, although the data do not rule out magma mixing as proposed by Dawson *et al.* (1994), provided that the ijolitic magma had the same isotope composition as the natrocarbonatite lavas.

The question remains as to whether natrocarbonatites are primary or derivative melts. Based on the Nd–Sr–Pb isotopic data from Oldoinyo Lengai the conclusion might be drawn that natrocarbonatites are derived directly from a relatively undifferentiated mantle source with Nd and Sr close to bulk Earth and CHUR. It is then possible to attribute the isotopic variations in the silicate melts from Oldoinyo Lengai to interaction between a primary natrocarbonatitic melt and an inhomogeneous

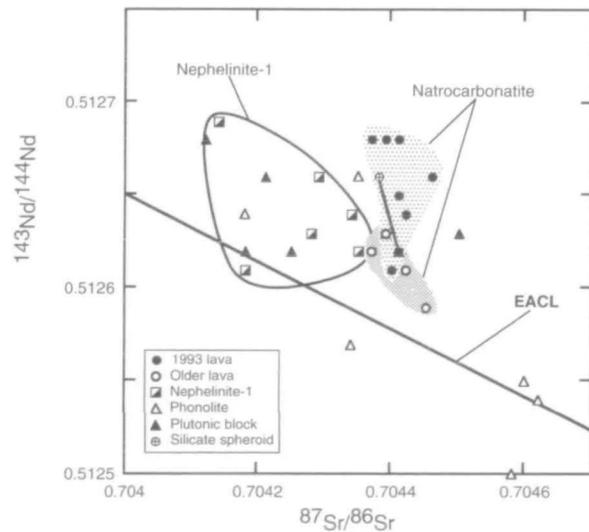


Fig. 8. $^{143}\text{Nd}/^{144}\text{Nd}$ vs $^{87}\text{Sr}/^{86}\text{Sr}$ diagram showing data from the 1993 and older natrocarbonatites. Tie-line joins data for mineral phases from silicate spheroid and host natrocarbonatite. Shown for comparison are data from various silicate rocks. Data from the Group I nephelinites are outlined. EACL from Bell & Blenkinsop (1987a).

mantle source made up of two isotopically distinct components. However, a further constraint imposed by the isotopic data is that the primary natrocarbonatite melt has also been generated by mixing the same two end-members because the natrocarbonatite data lie along the linear trend defined by the Nd and Sr isotopic data from the silicate rocks, and the EACL. A simpler model might involve the production of a series of silicate rocks using two mantle end-members, which under appropriate conditions of pressure and temperature can fractionate to generate a series of conjugate carbonatitic liquids.

Previous attempts to model the petrogenesis of the silicate lavas from Oldoinyo Lengai invoked liquid immiscibility between natrocarbonatite and peralkaline nephelinites such as wollastonite nephelinites or combeite nephelinites (Kjarsgaard & Hamilton, 1989; Peterson, 1989a, 1989b; Keller & Krafft, 1990; Kjarsgaard *et al.*, 1995; Dawson *et al.*, 1996). Bell & Dawson (1995a), in their earlier work on the silicate lavas, divided the nephelinites into two groups, a more depleted group that has Nd and Sr isotope ratios close to undifferentiated mantle (Group I), and a second group (Group II) that has Nd–Sr isotope compositions much closer to EMI. The Group I nephelinites contain all of the combeite nephelinite samples, and are isotopically similar in Sr and Nd to all of the natrocarbonatites so far measured from Oldoinyo Lengai. Although not unequivocal, this is certainly consistent with a genetic relationship between peralkaline lavas and natrocarbonatite, including models that involve immiscible separation. Although there are differences in Al_2O_3 , CaO, SiO_2 and FeO contents between the two groups, their highly fractionated nature (low Cr, Ni and Mg contents) makes it difficult to use the chemical characteristics of the nephelinites to identify their mantle sources.

Natrocarbonatite—the role of trona

During an evaluation of the trona model of Milton (1968, 1989), in which trona ($\text{Na}_2\text{CO}_3 \cdot \text{NaHCO}_3 \cdot 2\text{H}_2\text{O}$), an evaporite found in some of the lakes within the Rift Valley system and in Lake Magadi, which lies close to Oldoinyo Lengai, is either remobilized or taken into solution and a sodium carbonate exsolved, Bell & Dawson (1995b) argued that it seemed unnecessary to increase the Na content of a silicate melt by involving trona because nephelinitic melts are already rich in alkalis. In addition, they pointed out that the Sr, Nd, C and O isotope compositions of the natrocarbonatites are consistent with a mantle origin, and that marked differences do exist between

the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of natrocarbonatite and trona. Concern, however, was expressed by Bell & Dawson (1995b) that the isotopic ratios of the natrocarbonatites could be dominated by the isotopic signatures of the silicate melt, particularly if most of the Sr and Nd from the silicate melt were partitioned into the carbonatitic liquid during exsolution. This assumes that natrocarbonatites are formed by immiscible separation from a carbonated, peralkaline, silicate magma.

The Nd, Pb and Sr isotope compositions of two trona samples (BD1527, BD1533) are given in Table 3 and some of the data are shown in Figs 2, 4 and 5. Sample BD1527, from Lake Natron, is composed of 80% NaCl and 20% trona, whereas sample BD1533, from Lake Magadi, consists of 100% trona (Bell *et al.*, 1973). Compared with the Nd and Sr isotope data for the natrocarbonatites (Fig. 2), the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for sample BD1533 is significantly higher, whereas the $^{143}\text{Nd}/^{144}\text{Nd}$ ratio is similar. The Pb isotope compositions are unlike those of the natrocarbonatite (see Figs 4 and 5), plotting to significantly lower $^{206}\text{Pb}/^{204}\text{Pb}$ values, although the $^{207}\text{Pb}/^{204}\text{Pb}$ values are similar. Figure 5 does not include data from sample BD1527, which has a $^{207}\text{Pb}/^{206}\text{Pb}$ ratio like EMI but a much lower $^{206}\text{Pb}/^{204}\text{Pb}$ ratio. Remobilization of trona by a parental nephelinite magma from Oldoinyo Lengai (Milton, 1968) could not produce the more radiogenic Pb isotope values of the natrocarbonatites. These new data support the previous conclusions of Bell & Dawson (1995b) against the involvement of trona in the genesis of natrocarbonatite.

Silicate rocks and discrete partial melting

Based on available Nd and Sr data, Bell & Dawson (1995a) concluded that at least two mantle source components (HIMU and EMI) contributed to the formation of the silicate melts, and this is supported by the new Pb data for the silicate rocks from Oldoinyo Lengai presented here. The silicate rocks cannot be the products of simple differentiation from a single parent melt under closed-system conditions, or by mixing between a primary natrocarbonatite melt and a single mantle source. The fact that the isotope ratios are dissimilar but mostly coherent (i.e. lie along the same regression line) is consistent with the model of Bell & Dawson (1995a) involving generation of discrete, small-volume, mantle melts by the mixing of the same two end-members with HIMU and EMI isotope compositions. Isotopic data and major element variations of diopside phenocrysts from primitive nephelinite lavas (mg -number > 0.65) from other East African volcanic centres such as

Napak (Simonetti & Bell, 1994a; Simonetti *et al.*, 1996a) and Mount Elgon (Simonetti & Bell, 1995), led to similar conclusions involving the mixing of isotopically distinct mantle-derived melts. On the basis of the limited data set it seems unlikely that upper-crustal material was involved in the derivation of the phonolites, as no clear trends emerged in plots of isotope ratios or elemental abundances such as K, Si and Al.

In plots of $^{87}\text{Sr}/^{86}\text{Sr}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 9), and $^{143}\text{Nd}/^{144}\text{Nd}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 10) the data from the silicate rocks show scatter, requiring an additional component to HIMU and EMI to explain the distribution of the data. Donaldson *et al.* (1987) concluded that the nephelinites and phonolites could not be related by crystal fractionation of the phenocryst phases, and argued that some other process such as palagonitization or selective interaction with wall rock must have been involved. Variation in isotopic compositions of phonolites from Shombole volcano, 80 km north of Oldoinyo Lengai, was attributed to interaction between the magmas and lower-crustal granulites (Bell & Peterson, 1991). Similar interpretations were also made by Schleicher *et al.* (1990) to explain the isotope compositions of phonolites associated with the Kaiserstuhl carbonatite, Germany, and by Simonetti & Bell (1994b) for nepheline syenites from the Chilwa Island carbonatite complex, Malawi.

Two significant observations that emerge from the Pb–Pb isotope ratio diagrams (Figs 4 and 5) are: (1) none of the data plot to the right of the natrocarbonatite analyses, and (2) most ratios approximate a linear array similar to the oceanic regression line. Mixing between lower-crustal granulites and a silicate melt would be difficult to detect in Nd vs Sr isotope ratio plots (Fig. 2) given that the high abundances of Nd and Sr commonly observed in alkaline silicate rocks would buffer their initial isotope ratios inherited from the mantle source. Lower-crustal contamination is also difficult to distinguish from lithospheric mantle contamination on the basis of Pb isotopes given that granulites (e.g. Lashaine tuff cone, Cohen *et al.*, 1984) are similar in isotopic composition to the EMI mantle component. Of the silicate rocks from Oldoinyo Lengai, the greatest scatter of data is shown by the ijolites and phonolites (see Figs 4, 5, 9 and 10), a feature reported for other carbonatite-alkalic complexes (e.g. Shombole, Bell & Peterson, 1991; Ivaara, Kramm, 1994; Napak, Simonetti & Bell, 1994a). Bell & Peterson (1991) attributed the displacement to lower $^{143}\text{Nd}/^{144}\text{Nd}$ ratios for the phonolites from Shombole to interaction with lower-crustal granulites, a mechanism which might have happened at Oldoinyo Lengai.

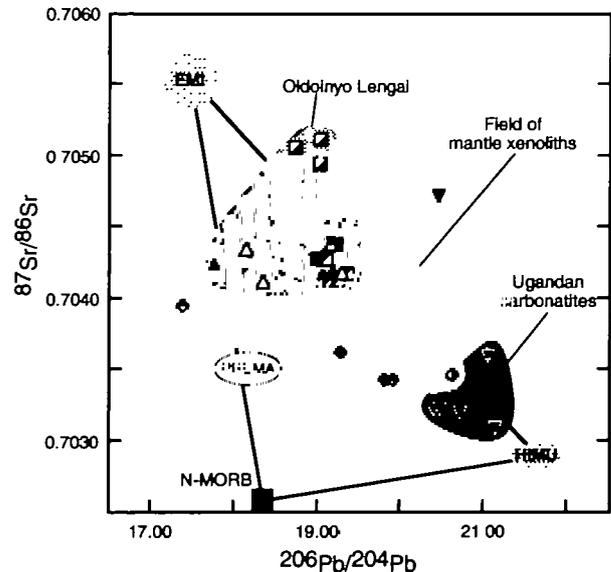


Fig. 9. $^{87}\text{Sr}/^{86}\text{Sr}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ for Oldoinyo Lengai samples and Ugandan carbonatites. Also shown is the field for mantle xenoliths (Cohen *et al.*, 1984). HIMU, EMI, N-MORB and PREMA mantle components from Zindler & Hart (1986) and Hart (1988). Symbols as in Figs 2 and 3.

An alternative explanation to contamination by lower-crustal granulites might be the involvement of yet a third mantle component. One possibility might be PREMA (prevalent mantle, Zindler & Hart, 1986) and the other might be DMM (depleted

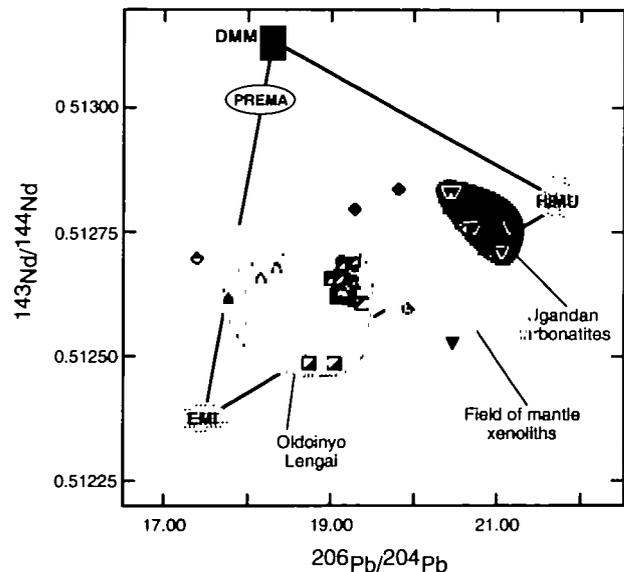


Fig. 10. $^{143}\text{Nd}/^{144}\text{Nd}$ vs $^{206}\text{Pb}/^{204}\text{Pb}$ for Oldoinyo Lengai samples and Ugandan carbonatites. Also shown is the field for mantle xenoliths (Cohen *et al.*, 1984). HIMU, EMI, N-MORB and PREMA mantle components from Zindler & Hart (1986) and Hart (1988). Symbols as in Figs 2 and 3.

MORB mantle), both of which are consistent with the distribution of most of the data points shown in Figs 4, 5, 9 and 10). Stein & Hoffmann (1994) have proposed that plume heads represent an average isotopic composition of the plume source plus material entrained from the overlying mantle and that their composition may be close to that of the mean mantle, i.e. PREMA. On the other hand, basaltic lavas in the Huri Hills in northern Kenya suggest the involvement of a possible DMM component (Class *et al.*, 1994).

EAST AFRICAN CARBONATITES— INTERACTION BETWEEN A HIMU PLUME AND LITHOSPHERE

The role of mantle plumes in carbonatite genesis, both direct and indirect, has been previously proposed, e.g. for the Cape Verdes (Gerlach *et al.*, 1988), the Canary Islands (Hoernle & Tilton, 1991), Brazil (Toyoda *et al.*, 1994; Huang *et al.*, 1995) and India (Simonetti *et al.*, 1995b). HIMU signatures have been attributed to plume activity by Gerlach *et al.* (1988), Hoernle & Tilton (1991) and Wilson *et al.* (1995). Plume activity is also supported by the spatial relationship of some carbonatites to flood basalts, including those in India associated with Deccan volcanism, those in Brazil associated with Parana volcanism, and those in Canada associated with Keeweenawan volcanism. Several problems arise, however, in relating carbonatite-alkaline complexes directly to plumes. The first involves the absence of carbonatites in truly oceanic environments (although carbonatites have been reported from the Cape Verde and Canary Islands, some of these islands are in transition zones between oceanic and continental crust and are therefore not truly oceanic). The depth of generation of carbonatite and associated nephelinite parental melts is a second, more serious problem, because phase equilibria evidence suggests that primary magmas associated with typical carbonatites are nephelinitic or melilititic in composition, probably representing derivation or segregation at 20–30 kbar pressures consistent with lithospheric depths within the mantle (Brey & Green, 1975, 1977; Egger, 1974, 1978, 1989; see also review by Edgar, 1987). Carbonatite melts with Na as the dominant alkali are also in equilibrium with phlogopite lherzolite at ~25 kbar (Sweeney *et al.*, 1995). Such experimental work is consistent with the restriction of most carbonatites to continental areas, although CO₂-rich melts might

play a role in producing variations in major and trace element data from oceanic island lavas (Hémond *et al.*, 1994). Yet another problem is the repetition of carbonatitic activity over thousands of millions of years (Woolley, 1989) in certain areas such as south and west Greenland, and the Kapuskasing Structural Zone, Canada.

In the Cape Verdes, magmas derived from sources with HIMU signatures are enriched in incompatible trace elements, and are both very alkaline and silica-undersaturated, features attributed to small degrees of partial melting at the plume margins under conditions dominated by CO₂ (Gerlach *et al.*, 1988). Elsewhere, Pb and Sr isotopic signatures of some carbonatites from Brazil are similar to EMI, and such a signature has been attributed either directly to an EM plume such as the Tristan da Cunha hotspot, or to derivation from enriched sub-continental lithospheric mantle (Toyoda *et al.*, 1994).

The isotopic variations shown in oceanic basalts led Zindler & Hart (1986) to propose five end-member components—DMM, HIMU, EMI, EMII and PREMA. HIMU has been interpreted as (1) ancient altered, subducted oceanic crust (Chase, 1981; White & Hofmann, 1982), (2) mixtures of subducted oceanic crust plus sediment (Chauvel *et al.*, 1992), (3) mantle that has lost Pb to the core (Vollmer, 1977), and (4) metasomatized mantle, either where fluid has removed elements from the mantle (Zindler & Hart, 1986), or where CO₂-rich fluids have enriched the mantle in U and Rb, and to a lesser extent in REE (Nakamura & Tatsumoto, 1988). Interpretations of EMI signatures involve: (1) recycled lower continental crust or lithosphere (Hawkesworth *et al.*, 1984; Nakamura & Tatsumoto, 1988; Gerlach *et al.*, 1988; Menzies, 1989), (2) mantle metasomatized by CO₂-rich fluids with low U/Pb and high Rb/Sr ratios (Zindler & Hart, 1986), and (3) mixtures of HIMU and ancient pelagic sediments (Weaver, 1991).

Initially, Hart *et al.* (1986) sited HIMU and EMI in the sub-continental lithosphere and argued that HIMU must be the result of metasomatic activity, with HIMU and EMI being complementary parts of the same metasomatic process. On the basis of Nd and Sr isotope ratios from metasomatized (pargasite-bearing) lherzolite nodules and host basanite lavas from the Ataq diatreme (Yemen), Menzies & Murthy (1980a) proposed that the pre-metasomatic, sub-continental mantle source for the basanite lavas contained an enriched isotopic composition (⁸⁷Sr/⁸⁶Sr = 0.705–0.706 and ¹⁴³Nd/¹⁴⁴Nd = 0.51240–0.51260) similar to that for the EMI mantle component and to those reported from mantle xenoliths

from kimberlites (Menzies & Murthy, 1980b). In addition, Hawkesworth *et al.* (1986) suggested that the Dupal signature in the South Atlantic (Walvis Ridge, Tristan da Cunha) originated in the sub-continental lithosphere that was thermally remobilized and metasomatized before the breakup of Gondwana.

If EMI is a slightly modified bulk Earth component it clearly needs to be stored out of the upper-mantle circulation for a long period of time (Hart, 1988). On the basis of the isotope systematics of carbonatites we suggest that a principal storage area for EMI might be the sub-continental lithosphere. On the basis of geochemical data for Neogene–Quaternary primitive alkali basalts, and basanites from the Ross Sea Rift, Antarctica, Rochell *et al.* (1995) suggested a stratified mantle below the Ross Sea Rift in the order (from deep to shallow): MORB, HIMU, and EMI sources. Rochell *et al.* (1995) have further suggested that the EMI reservoir resides in the mantle lithosphere, and that the HIMU-type component may be related to an active plume head, or may represent part of a ‘fossilized’ mantle plume attached to the base of the lithosphere beneath the Ross Sea area. To explain the large isotopic variation shown by Oldoinyo Lengai and other East African carbonatites we propose a model that involves mixing between two mantle components, one related to plume activity with a HIMU signature, and the other sub-continental lithosphere with isotopic characteristics close to EMI.

Although it is perhaps impossible to recognize plumes solely on the basis of geochemical characteristics, the geophysical evidence and magma types for mantle upwelling in East Africa are convincing. Evidence in favour of plume-driven rifting from the Kenya rift segment includes the long-wavelength gravity and topographic anomalies, the volume of eruptive rocks, the relative timing of rifting and magmatism, the distribution of physical anomalies, and the pattern of tectonomagmatic evolution of the rift (Macdonald *et al.*, 1994). Additional evidence for mantle upwelling in the Gregory Rift includes voluminous outpourings of plateau phonolites (25 000–50 000 km³), three-dimensional geophysical data (Henry *et al.*, 1990; Davis, 1991; Green *et al.*, 1991), crustal uplift (Kenya Dome), and a marked zonal arrangement of alkaline rocks (kimberlite diatremes, carbonatitic complexes and alkali basaltic centres; Le Bas, 1989). Figure 11 shows the approximate surface expression of the Kenya Dome, an area undergoing crustal uplift (Smith, 1994). Oldoinyo Lengai, along with Shombole, lies close to the southernmost limit of the Kenya Dome. It is interesting to note that the carbonatite–nephelinite

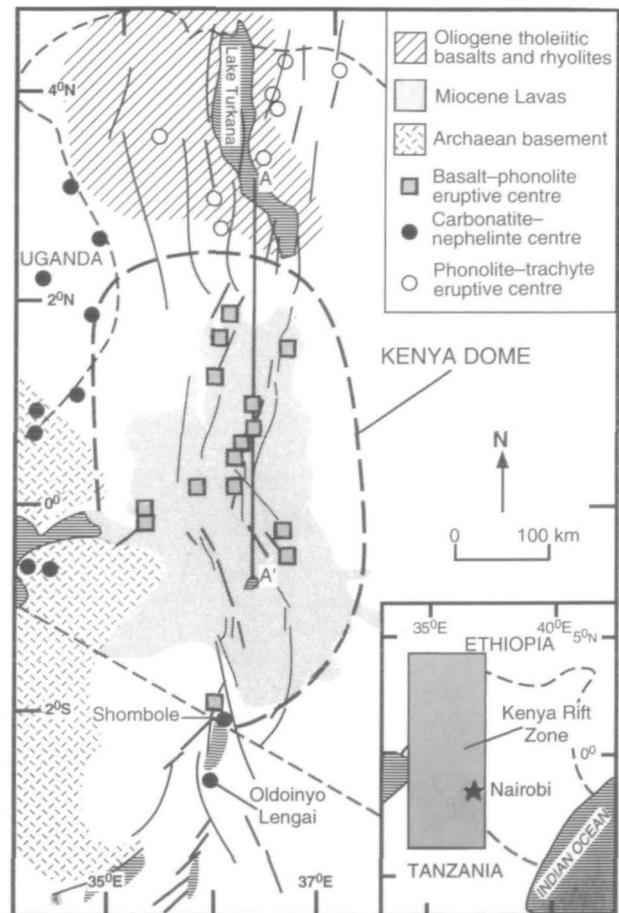


Fig. 11. Map showing surface expression of the Kenya Dome and the distribution of volcanism and rifting (continuous lines) within the Kenya Rift Zone [modified after Smith (1994)]. Carbonatite–nephelinite centres are located marginal to the present-day location of mantle upwelling; basalt–phonolite centres are located within the Kenya Dome. Oldoinyo Lengai lies close to the southernmost margin of the dome. Line AA' shows location of axial profile KRISP 90 line (Mechie *et al.*, 1994).

centres lie at the margins of the dome, whereas the basalt–phonolite centres lie within the central area. On the basis of this evidence, deep mantle upwellings or major convective overturns can be linked to continental rifting and alkaline magmatic activity (Smith, 1994).

High-pressure experiments have shown that alkaline melts, such as nephelinites and alkali basalts, are not in equilibrium with an anhydrous lherzolitic mantle source (Bultitude & Green, 1968, 1971; Merrill & Wyllie, 1975; Allen *et al.*, 1975) but can be derived from an amphibole peridotite at 20–25 kbar (e.g. Olafsson & Eggler, 1983; Eggler, 1989) or volatile-bearing (CO₂-dominated) mantle at ~30 kbar pressure (Eggler, 1974, 1978, 1989; Brey & Green, 1975, 1977; Edgar, 1987; Wyllie, 1989). A

volatile-rich mantle, therefore, is considered important in deriving nephelinitic and associated carbonatitic melts.

We consider, therefore, the derivation of nephelinitic and carbonatitic melts from East Africa to be a two-stage process, the first involving generation of fluids or melts with HIMU signatures derived from an upwelling mantle (thermal cell or plume, Fig. 12). Metasomatism of the sub-continental lithosphere below East Africa, therefore, can be attributed to plume activity, involving release of volatiles and/or melts from the plume. The second stage of our model involves low-degree partial melting of the metasomatized sub-continental lithosphere, an event that probably occurs soon after metasomatism of the lithosphere. Incorporation of volatiles and melts into the EMI-like lithosphere, with concomitant lowering of the mantle solidus, is followed by low degrees of partial melting that can then generate either carbonated silicate melts (which can form conjugate silicate and carbonatite liquids via liquid immiscibility), or primary carbonatite magmas. Such melts would contain variable proportions of the HIMU and EMI components, and perhaps additional material from the upwelling plume (PREMA? or DMM?). Wilson *et al.* (1995)

have also argued that low-degree, CO₂-H₂O-enriched partial melts from an ascending mantle plume may freeze at the base of the lithosphere creating a heterogeneous carbonate-phlogopite-lherzolite layer. The timing of the metasomatism from the plume relative to the production of carbonatite-nephelinite melts from the lithosphere still remains uncertain. On the basis of U/Pb ages of accessory minerals from metasomatized mantle xenoliths in kimberlites (Kinny & Dawson, 1992) and initial ¹⁴³Nd/¹⁴⁴Nd ratios from carbonatites of different age from Canada (Bell & Blenkinsop, 1987b), it appears that metasomatic activity can be contemporaneous with melt formation. We suggest, therefore, that derivation of young (<30 Ma) East African carbonatites can be attributed to recent plume-mantle interaction, as also proposed by Smith (1994). An alternative explanation is to suggest that the isotopic variation shown by the East African carbonatites can be attributed to a heterogeneous plume source containing both HIMU and EMI mantle signatures. We view this latter model, however, as an ad hoc explanation which would remain difficult to prove or disprove.

The considerable range and linear array of isotopic compositions for East African carbonatites have been previously attributed to mixing between mantle and metasomatic veins by Meen *et al.* (1989), but in their model a depleted mantle is invaded by metasomatic incursions of alkaline silicate melts with low U/Pb, Th/Pb and Sm/Nd ratios, significantly lower than bulk Earth, and with Rb/Sr ratios similar to bulk Earth, the reverse of what we propose here.

Chemical and isotopic data from mantle xenoliths and basaltic lavas from East Africa are consistent with a plume-lithosphere interaction model. Isotopic data for mantle xenoliths from the nearby tuff and cinder cones of Eledoi, Lashaine, Pello Hill (Cohen *et al.*, 1984) and Olmani (Rudnick *et al.*, 1993), shown in Fig. 3, approximate the EACL. One xenolith documented by Bell & Dawson (1995a) from Oldoinyo Lengai (not shown in Fig. 3), a nepheline-veined mica pyroxenite, is exceptional in terms of its ⁸⁷Sr/⁸⁶Sr (0.7086) and ¹⁴³Nd/¹⁴⁴Nd (0.51174) ratios. These high ratios may simply reflect an earlier period of metasomatic activity that affected the lithosphere. The peridotite xenoliths from Olmani are considered to be the result of interaction between depleted peridotite and carbonatitic melts (Rudnick *et al.*, 1993), and those from Eledoi and Pello Hill appear to be non-equilibrium mineral assemblages (Cohen *et al.*, 1984). Although the database is not large, these xenoliths could be samples of relic metasomatized lithosphere capable, on melting, of generating the parental melts for car-

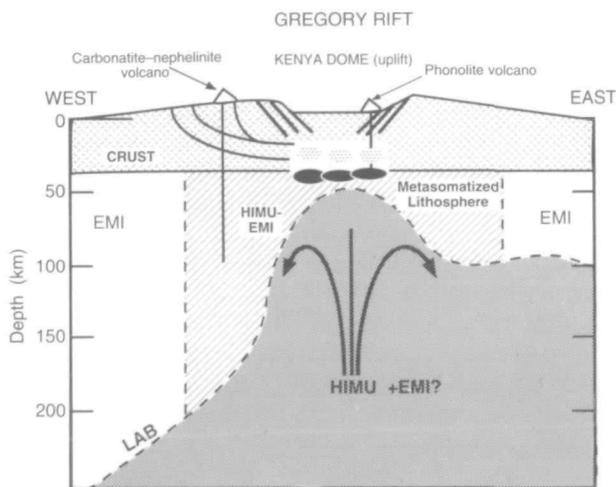


Fig. 12. Schematic east-west cross-section across the Kenya Rift Zone [modified after Smith (1994)]. Shown is the location of the mantle upwelling (thermal cell or plume) and consequent area of crustal uplift (Kenya Dome). Diagram illustrates our preferred two-stage model to explain the large isotope variations shown by East African carbonatites. The model consists of a HIMU-like 'plume' releasing (degassing?) volatiles and/or melts that invade the overlying EMI-like lithosphere. This process results in metasomatized lithosphere characterized by heterogeneous isotopic signatures that lie between HIMU and EMI. Seismic evidence suggests that directly above the mantle upwelling, large-volume mafic melts underplate the continental crust, and are responsible for the generation of the basalt-phonolite centres. LAB, lithosphere-asthenosphere boundary.

bonatites. The covariation between Nd, Pb and Sr isotope compositions and certain trace element ratios (e.g. Ba/Th, K/La, Sr/Nd) for Pliocene–Quaternary basaltic lavas from the nearby Huri Hills in northern Kenya, have been attributed to binary mixing between a plume with HIMU affinities and either lithospheric mantle or a second plume component (Class *et al.*, 1994). Compared with the Nd and Sr isotopic data from other African carbonatites, which contain more HIMU-like signatures (e.g. Bukusu, Napak, Tororo, and Sukulu), those from Oldoinyo Lengai lie closer to EMI, probably reflecting a greater lithospheric component. The greater interaction with lithospheric mantle may be attributed to the location of Oldoinyo Lengai above a zone of thicker lithosphere relative to that found beneath the Kenya Dome, which is supported by seismic data obtained from a north–south axial profile (KRISP 90 line; Mechie *et al.*, 1994). The results show that lithospheric thicknesses range from 20 km in the north (beneath Lake Turkana) to 35 km in the south (beneath Lake Naivasha, Fig. 11).

Our proposed model may have some broader implications. Isotopic data from young (<0.2 Ga) carbonatites from five continents closely fit a model corresponding to the mixing of EMI and HIMU mantle components (Tilton & Bell, 1994). If we assume that the HIMU signatures are associated with fluids or melts generated by large-scale mantle perturbations, such as major convective overturns and plume activity, then the excellent correlation between the temporal distribution of carbonatites and major orogenic cycles for the past 3.0 billion years (Woolley & Kempe, 1989; Veizer *et al.*, 1993) adds support to our interpretation.

CONCLUSIONS

In all of the isotope ratio diagrams, the natrocarbonatite data fall close to or on a straight line that joins estimates of the isotopic compositions of HIMU and EMI (Hart, 1988). The Nd, Pb and Sr isotope compositions of the 1960, 1988 and 1993 natrocarbonatites are extremely uniform. Their clustering close to present-day values for bulk Earth and CHUR requires mixing of the same two end-members in roughly the same proportions. That the Sr and Nd isotope ratios from other East African carbonatites are also consistent with HIMU and EMI mixing illustrates that both components are contiguous and underlie a large segment of East Africa.

In our preferred model, the Nd, Pb and Sr isotope variation defined by young (<30 Ma) East African carbonatites may be attributed to interaction between a HIMU-like 'plume (thermal cell)' and

EMI-like lithosphere. Evidence for mantle upwelling in the Gregory Rift includes voluminous outpourings of plateau phonolites, three-dimensional geophysical data, a marked zonal arrangement of alkaline rocks, and crustal uplift (Kenya Dome). Oldoinyo Lengai lies close to the southern margin of the present-day location of the mantle upwelling, which is situated beneath the surface expression of the Kenya Dome. Volatiles and/or melts from a HIMU-like plume metasomatize the overlying lithosphere and provide a mechanism for generating nephelinitic and associated carbonatitic melts at pressures of 20–30 kbar. In spite of the limited database, the isotope and chemical data for carbonatite-metasomatized mantle xenoliths from nearby tuff and cinder cones (Tanzania) are consistent with plume (HIMU)–lithosphere (EMI) interaction.

Complex histories for the associated silicate magmas from Oldoinyo Lengai are evident from the Pb isotope signatures. Not only are these silicate rocks the product of discrete partial melting events with variable HIMU–EMI contributions, but most ijolites and phonolites have isotope ratios that require an additional component, such as lower-crustal granulites, PREMA or DMM, to be involved in their origin.

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