[2]

Possible origin of K-rich volcanic rocks from Virunga, East Africa, by metasomatism of continental crustal material: Pb, Nd and Sr isotopic evidence

R. Vollmer [1] and M.J. Norry [2]

[1] *Department of Earth Sciences, The University, Leeds LS2 9JT (U.K.)*
[2] *Department of Geology, Leicester University, Leicester LE1 7RH (U.K.)*

The isotopic compositions of Sr, Nd and Pb together with the abundances of Rb, Sr, U and Pb have been determined for mafic and felsic potassic alkaline rocks from the young Virunga volcanic field in the western branch of the East African rift system. $^{87}Sr/^{86}Sr$ varies from 0.7055 to 0.7082 in the mafic rocks and from 0.7073 to 0.7103 in the felsic rocks. The latter all come from one volcano, Sabinyo. Sabinyo rocks have negative ϵ_{Nd} values of $\epsilon_{Nd} = -10$. Nd and Sr isotopic variations in the basic potassic rocks are correlated and plot between Sabinyo and previously reported [1] compositions ($\epsilon_{Nd} = +2.5$; $^{87}Sr/^{86}Sr \approx 0.7047$) for Nyiragongo nephelinites. The Pb isotopic compositions for Sabinyo rocks are nearly uniform and average $^{206}Pb/^{204}Pb \approx 19.4$, $^{207}Pb/^{204}Pb = 15.79$–15.84, $^{208}Pb/^{204}Pb \approx 41.2$. The basic potassic rocks have similar $^{206}Pb/^{204}Pb$ values but range in $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ from the Sabinyo values to less radiogenic compositions.

Excellent correlations of $^{87}Sr/^{86}Sr$ with Rb/Sr, 1/Sr and $^{207}Pb/^{206}Pb$ for Sabinyo rocks suggest these to be members of a hybrid magma series. However, the nearly uniform Pb compositions for this series points to radiogenic growth of ^{87}Sr in the magma source region following an event which homogenized the isotopic compositions but not Rb/Sr. The Rb-Sr age derived from the erupted Sabinyo isochron-mixing line is consistent with the ~ 500 Myr Pb-Pb age from Nyiragongo [1], which suggests that this event affected all Virunga magma sources. The event can again be traced in the Pb-Pb, Pb-Sr and Nd-Sr isotopic correlations for all Virunga rocks, including Nyiragongo, when allowances are made for radiogenic growth subsequent to this mixing or incomplete homogenization event. Inferred parent/daughter element fractionations point to a metasomatic event during which a mantle fluid invaded two lithospheric reservoirs: a $+ \epsilon_{Nd}$ reservoir sampled by the Nyiragongo nephelinites and suggested to be the subcontinental mantle and a $- \epsilon_{Nd}$ reservoir sampled by the mafic and felsic potasssic volcanism. Whether this $- \epsilon_{Nd}$ reservoir is the crust, continental crustal material in the mantle or anomalous mantle cannot be decided from the data. The simplest answer, that this reservoir is the continental crust, seems to be at variance with experimental evidence suggesting a subcrustal origin for basic potassic magmas. Partial melting of the ancient metasomatised lithospheric domains and ensuing volcanism seems to be entirely a response to decompression and rising geotherms during rifting and thinning of the lithosphere.

1. Introduction

The genesis of basic potassic alkaline volcanic rocks is still extremely controversial. There seems to be a tendency in more recent studies (e.g. [2–6]) to ascribe the extreme abundances of K, Rb, Sr, Ba, La and other large ionic lithophile (LIL) ele-

ments in these rocks to their concentration, by fluids, in the sources of the magmas prior to partial melting, a process referred to as mantle metasomatism [7]. Experimental evidence has been interpreted to suggest a mantle depth for the magma source regions (e.g. [8,9]). Yet the role of the continental crust—if at all involved—is far

from clear: A chemical component has been traced in the Italian occurrence which has a negative ϵ_{Nd} and high $^{87}Sr/^{86}Sr$ isotopic signature. These isotopic ratios indicate derivation of this component from a reservoir which, since ancient times, has been enriched in light rare earth and other LIL elements. Is the negative ϵ_{Nd} component a mantle fluid derived from an ancient enriched mantle as Hawkesworth and Vollmer [2] propose? Or is the enriched reservoir the continental crust? For example, Turi and Taylor [10] and Taylor et al. [11] suggest large-scale contamination of mantle-derived magmas by continental crustal material, while Holm and Munksgaard [4] favour subduction of continental sediments, followed by release of fluids which then metasomatise the overlying mantle.

It seems that answers have to come from comparative studies of potassic volcanics from geodynamically less controversial regions than the Mediterranean. One such occurrence is the Late Pliocene to recent [12,13] Virunga volcanic field in the western branch of the East African rift system (Fig. 1).

The Virunga volcanics are underlain by sediments of the Karagwe-Ankolean System which were deposited ~ 2100 Myr ago and deformed

and metamorphosed during the Kibaran orogeny (1300–800 Myr), but apparently were not here affected by Pan-African events (700–450 Myr) [14].

The majority of the Virunga volcanic rocks are silica-deficient, leucite-bearing potassic rocks [15,16]. The two compositional extremes at Virunga are represented by lavas from the volcanoes of Sabinyo and Nyiragongo. Sabinyo lavas form a shoshonite to hypersthen-latite series and are the only silica-saturated to oversaturated rocks at Virunga [15]. The melilite-nephelinites from Nyiragongo, with ~ 39% SiO_2, are the silica-undersaturated extreme and are richer in sodium than potassium [17].

In the first of two articles dealing with the Sr, Nd and Pb isotope systematics of the Virunga volcanic field we concentrated on the Nyiragongo volcano [1]. Here we report isotope data for the Sabinyo series and for basic potassic rocks from, mainly, the Bufumbira field, the eastern region of Virunga. The majority of the samples used in this study have been described by Holmes and Harwood [15] (see Table 1).

2. Results

Rb-Sr. Sr isotopic compositions are plotted against Rb/Sr and 1/Sr in Fig. 2. $^{87}Sr/^{86}Sr$ is positively correlated with Rb/Sr and 1/Sr when all Virunga data are considered, a feature already observed by Bell and Powell [16]. But each of the three rock groups, the Nyiragongo nephelinites, the basic potassic rocks and the Sabinyo series, show peculiar features in both diagrams: The nephelinites are tightly clustered; no correlations are observed for the basic potassic rocks on their own in either of the two diagrams, although the scatter seems to be somewhat less in the diagram $^{87}Sr/^{86}Sr$ versus Rb/Sr; the Sabinyo series stands out through excellent correlations in both diagrams and through $^{87}Sr/^{86}Sr$ and Rb/Sr variations which exceed half the total observed range and comprise solely the higher values.

Sr-Nd. In Fig. 3 Sr and Nd isotopic compositions for Virunga are compared with isotopic variations
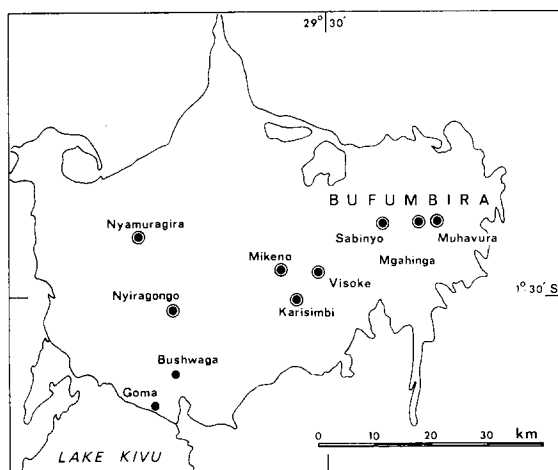

Fig. 1. The Virunga volcanic field within the western branch of the East African rift system. The map shows the eight major volcanoes and two small cones (Bushwaga, Goma) which are related to Nyiragongo [1]. In addition, numerous smaller vents occur at Bufumbira, the eastern region of Virunga. The age of volcanism is Late Pliocene to historic [12,13].

TABLE 1

Rock type, key for petrographic description, partial chemical analyses, and Sr and Nd isotopic compositions for potassic rocks from Virunga, East Africa

Sample rock type, locality	Description [a]	SiO_2 [b] (wt. %)	K_2O [b] (wt. %)	Rb (ppm)	Sr (ppm)	$^{87}Sr/^{86}Sr$ [c]	$^{143}Nd/^{144}Nd$ [d]
Bufumbira							
C1904 kivite B	374,700		3.19	104	890	0.70690 ± 5	
C1909 kivite M, Muhavura	371,697			164 [e]	1117	0.70695 ± 2	
C1910 kivite B, Muhavura	372,697			164 [e]	1231	0.70681 ± 3	
C1912 shosh. absarokite	491,698			117 [e]	939	0.70817 ± 4	
C1918 shosh. absarok., Mgahinga	471,688	48.82	4.09	129 [e]	806	0.70752 ± 4	
C1960 kivite B	299			127 [e]	1059	0.70690 ± 3	
C1961 kivite B	299			87 [e]	885	0.70671 ± 3	
C2007 murambite	432,642			62 [e]	612	0.70688 ± 2	
C2017 shosh. absarokite	169			93 [e]	1070	0.70739 ± 4	
C2022 ugandite	219,703			117 [e]	839	0.70596 ± 6	
C2765 ol-leucitite	255,643			126 [e]	1245	0.70628 ± 4	
C2794 shosh. absarokite	494,701	50.05	4.46	165	1109	0.70756 ± 5	0.51240 ± 2
C2799 banakite	528,626	52.94	5.36	171	1381	0.70720 ± 6	
C2800 murambite	421,646			95 [e]	770	0.70663 ± 4	
C2821 ugandite	205,694			99 [e]	1117	0.70658 ± 3	0.51244 ± 3
C2824 absarokite	454,713			109 [e]	1034	0.70625 ± 4	
C2840 kivite B	315,634	44.41	4.24	119 [e]	1152	0.70621 ± 6	0.51246 ± 3
C2865 banakite	524,737			141 [e]	1326	0.70726 ± 4	0.51234 ± 3
C2881 shosh. absarokite	516,729			95 [e]	851	0.70729 ± 3	
C2883 kivite B–ol-leucitite	331,730			90 [e]	816	0.70655 ± 4	
C2897 ugandite–murambite	429,655		2.83	98	862	0.70595 ± 4	
C2909 mikenite, Mikeno	279,692		5.96	165	1550	0.70546 ± 3	0.51259 ± 2
C2911 mikenite, Mikeno	276,692	47.68	6.45	153	1757	0.70572 ± 4	
C3046 ol-leucitite	253,703			130	1079	0.70591 ± 3	0.51252 ± 4
Bufumbira: Sabinyo volcano							
C2888 hypersthene-latite	561,736		4.80	207	655	0.70954 ± 3	0.51211 ± 3
C2889 shoshonite	560,736			179	784	0.70859 ± 4	0.51213 ± 4
C2893 ol-shoshonite	554,736			149 [e]	1109 [e]	0.70771 ± 3	
C2894 hypersthene-latite	563,736		5.05	203	604	0.70977 ± 4	
C2895 ol-shoshonite	557,736			152	1027	0.70727 ± 4	
CV258 latite		57.40	4.60	167	876	0.70789 ± 8	
CV690 ol-shoshonite			4.74	138	1243	0.70746 ± 4	
CV693 latite			4.88	206	527	0.71030 ± 3	
Nyamuragira							
C3032 kivite M	349,723					0.70577 ± 3	
Southwest Virunga							
C3021 mel-nephelinite, Bushwaga	267,636			136	2866	0.70463 ± 5	0.51275 ± 5 [f]
C3022 mel-nephelin., Nyiragongo	268,712	38.47	5.41	138	3643	0.70479 ± 5	0.51277 ± 2 [f]
C9872 lc-nephelinite, Goma crater				110	2421	0.70457 ± 5	0.51276 ± 2 [f]
1972 mel-nephelin., Nyiragongo crater-lake		39.10	5.45			0.70465 ± 4	0.51276 ± 3 [f]

[a] Single numbers refer to paragraphs in Combe and Simmons [35], double numbers to paragraphs in Holmes and Harwood [15].

[b] Major elements from Holmes and Harwood [15] and Bell and Doyle [34].

[c] Normalized to $^{86}Sr/^{88}Sr = 0.1194$ and relative to an E + A standard value of 0.70800.

[d] Normalized to $^{146}Nd/^{144}Nd = 0.7219$; BCR-1: $^{143}Nd/^{144}Nd = 0.51266 \pm 2$.

[e] Isotopic dilution analyses. Estimated uncertainty: Sr: ±0.5%; Rb: ±2%. The remaining Rb and Sr determinations are XRF data from Bell and Powell [16].

[f] From Vollmer and Norry [1]. Note that, by mistake, an erroneous correction was applied to the Nd data.
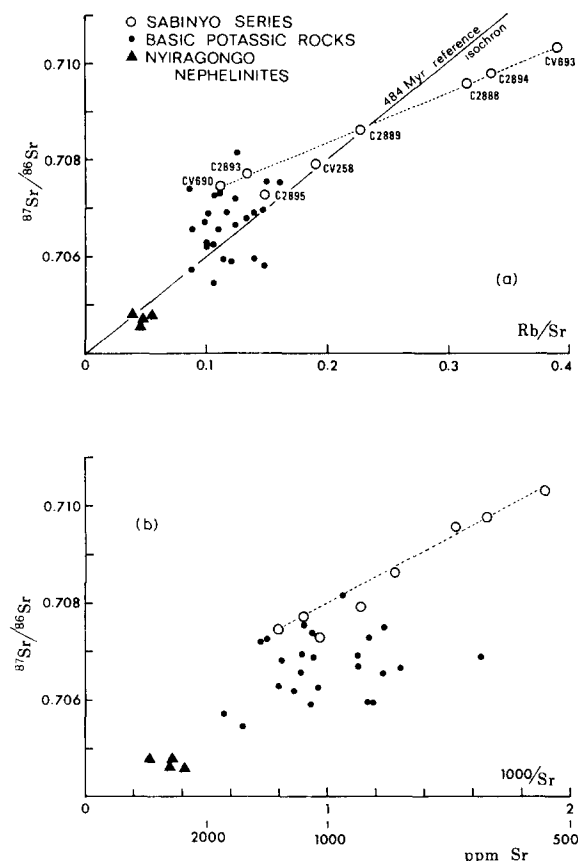
Fig. 2. $^{87}Sr/^{86}Sr$ versus Rb/Sr (a) and 1/Sr (b) for Virunga volcanic rocks. Both diagrams allow to distinguish three rock groups at Virunga: Nyiragongo nephelinites are tightly clustered; no clear correlations are observed for the basic potassic rocks; the Sabinyo series comprises exclusively the higher $^{87}Sr/^{86}Sr$ values and shows excellent correlations in both diagrams (dotted line). These strongly suggest the Sabinyo series to be a hybrid magma series, but the slope $\Delta^{87}Sr/^{86}Sr/\Delta Rb/Sr$ is interpreted—for reasons discussed in the text—to date also a source event. The good correlations further indicate that feldspar fractionation after mixing has not been of any significance for this magma series. The 484-Myr reference isochron in Fig. 2a is derived from whole-rock Pb-Pb data for Nyiragongo [1].
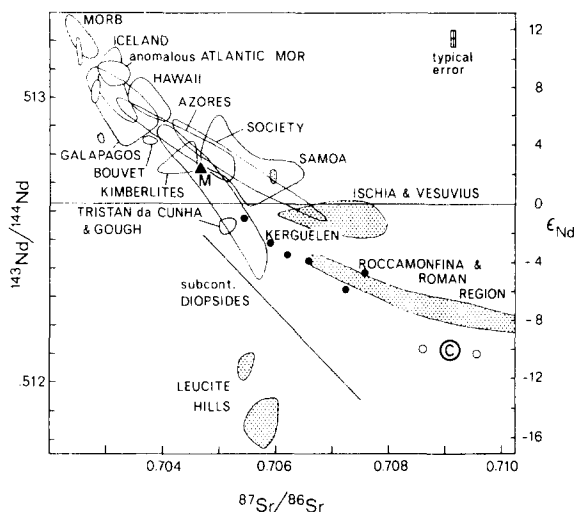

Fig. 3. Comparison of Nd and Sr isotopic variations for Virunga with oceanic basalts, basic continental rocks (kimberlites and basic potassic rocks: shaded) and mantle-derived diopsides. Symbols as in Fig. 2. Isotopic variations for Sabinyo and most of the Virunga basic potassic rocks are outside the field for oceanic basalts but would approximately coincide with the range of mantle diopside compositions prior to the inferred 500-Myr Rb enrichment. C and M denote end-member compositions (transposed to present-day values) for this event. See White and Hofmann [29] for data sources. Additional data—diopsides: Menzies and Murthy [30]; kimberlites: Kramers et al. [31]; Italian potassic rocks: Hawkesworth and Vollmer [2]; Leucite Hills: Vollmer et al. [32].

observed for oceanic basalts and basic continental rocks. The isotopic ratios are well correlated and cover a large range from $\epsilon_{Nd} = +2.5$ for Nyiragongo to $\epsilon_{Nd} = -10$ for the Sabinyo series. The basic potassic rocks have intermediate isotopic characteristics. Sabinyo rocks and the majority of the basic potassic rock fall outside the fields for oceanic basalts. Sabinyo magmas in particular are

derived from an ancient LIL-element-enriched source with an isotopic signature typical of upper continental crust, but this does not necessarily preclude a mantle origin as the isotopic range for subcontinental diopsides demonstrates.

Pb. The Virunga Pb isotopic compositions (Table 2) are plotted in fig. 1 of Vollmer and Norry [1] and are compared with oceanic basalts in Fig. 4. Pb isotopic variations in Nyiragongo nephelinites are the largest ever observed for young volcanic rocks. The Bufumbira volcanics (including Sabinyo) are characterized by minor variations in $^{206}Pb/^{204}Pb$ but relatively large variations in $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$. Such variations are again unique; similar trends have not been observed before for any oceanic island group or continental volcanic field (Fig. 4). Sabinyo Pb is almost homogeneous and isotopic variations ex-

TABLE 2

Pb isotopic composition and U, Pb concentrations of potassic rocks from Virunga, East Africa

Sample	$\dfrac{^{208}Pb}{^{206}Pb}$	$\dfrac{^{207}Pb}{^{206}Pb}$	$\dfrac{^{206}Pb}{^{204}Pb}$	$\dfrac{^{207}Pb}{^{204}Pb}$	$\dfrac{^{208}Pb}{^{204}Pb}$	Pb (ppm)	U (ppm)	$\dfrac{^{238}U}{^{204}Pb}$
Bufumbira								
C1904	2.0888 ± 4	0.81089 ± 12	19.420 ± 9	15.748	40.565	6.99	2.34	22.2
C1909	2.0957 ± 2	0.81432 ± 6	19.392 ± 7	15.791	40.641	11.5		
C1910	2.0956 ± 8	0.81439 ± 16	19.394 ± 17	15.794	40.642	11.69	4.27	24.2
C1912	2.0952 ± 4	0.81398 ± 11	19.429 ± 24	15.815	40.708			
C1918	2.0936 ± 9	0.81095 ± 20	19.482 ± 14	15.799	40.786			
	2.0936 ± 3	0.81105 ± 10	19.458 ± 10	15.781	40.737			
C1960	2.1033 ± 2	0.81273 ± 7	19.425 ± 10	15.787	40.858		2.31	
C1961	2.1045 ± 4	0.81298 ± 13	19.381 ± 11	15.756	40.788	7.81	2.17	18.7
C2007	2.1150 ± 15	0.81668 ± 31	19.327 ± 30	15.784	40.878	5.34	1.57	19.5
C2017	2.1187 ± 3	0.81615 ± 3	19.367 ± 12	15.806	41.032			
C2022	2.0979 ± 7	0.81180 ± 22	19.347 ± 11	15.706	40.589			
C2765	2.0967 ± 2	0.81263 ± 6	19.374 ± 23	15.744	40.622			
C2794	2.1060 ± 2	0.81357 ± 4	19.402 ± 16	15.785	40.861	12.02	3.83	21.2
	2.1050 ± 3	0.81304 ± 9	19.417 ± 7	15.786	40.871			
C2799	2.1128 ± 9	0.81601 ± 20	19.334 ± 11	15.777	40.849	18.5	5.15	18.4
	2.1100 ± 9	0.81562 ± 20	19.316 ± 14	15.755	40.758			
C2800	2.0993 ± 3	0.81195 ± 9	19.382 ± 18	15.738	40.690			
C2821	2.0920 ± 5	0.81163 ± 15	19.449 ± 15	15.785	40.686	9.39	2.99	21.1
C2824	2.1062 ± 9	0.81406 ± 20	19.408 ± 24	15.799	40.877			
C2840	2.1088 ± 2	0.81063 ± 6	19.418 ± 15	15.741	40.948	9.30	2.88	20.6
C2865	2.1187 ± 3	0.81486 ± 8	19.384 ± 13	15.795	41.068	13.15	4.78	24.2
C2881	2.1154 ± 3	0.82084 ± 9	19.222 ± 17	15.778	40.662	7.30	2.01	18.2
C2883	2.1106 ± 8	0.81416 ± 18	19.386 ± 22	15.783	40.915			
C2897	2.0828 ± 8	0.81101 ± 18	19.409 ± 30	15.741	40.426			
	2.0805 ± 2	0.81045 ± 7	19.408 ± 15	15.729	40.377			
C2909	2.0763 ± 5	0.81062 ± 9	19.396 ± 12	15.723	40.272	4.53	4.18	60.4
C2911	2.0810 ± 2	0.81282 ± 6	19.409 ± 11	15.776	40.390			
C3046	2.0983 ± 2	0.81197 ± 6	19.370 ± 18	15.728	40.643			
Bufumbira: Sabinyo								
C2888	2.1257 ± 4	0.81792 ± 12	19.360 ± 8	15.835	41.154	36.5	6.07	11.1
C2889	2.1263 ± 13	0.81653 ± 26	19.405 ± 18	15.845	41.260			
C2893	2.1189 ± 2	0.81282 ± 6	19.422 ± 15	15.787	41.152			
C2894	2.1247 ± 5	0.81849 ± 11	19.338 ± 18	15.828	41.088			
C2895	2.1189 ± 2	0.81291 ± 7	19.435 ± 22	15.798	41.180			
	2.1238 ± 9	0.81402 ± 16	19.462 ± 22	15.843	41.333			
CV258	2.1159 ± 2	0.81462 ± 7	19.391 ± 11	15.797	41.030	22.0		
CV690	2.1205 ± 7	0.81324 ± 11	19.436 ± 24	15.806	41.213			
Nyamuragira								
C3032	2.0940 ± 4	0.82228 ± 9	19.101 ± 27	15.706	39.997			

See Vollmer and Norry [1] for analytical details.

ceed the analytical uncertainty significantly only in the more precise $^{207}Pb/^{206}Pb$ ratios (Fig. 5). In both diagrams of Fig. 4 Sabinyo Pb isotopic compositions constitute the one extreme of the Bufumbira trends. A correlation is observed between the $^{208}Pb/^{204}Pb$ ratio (and, less distinctly, the $^{207}Pb/^{204}Pb$ ratio) and major element compositions exemplified by the modal feldspar content [1, fig. 1]. Sabinyo Pb has an isotopic signature which falls far outside the fields for any
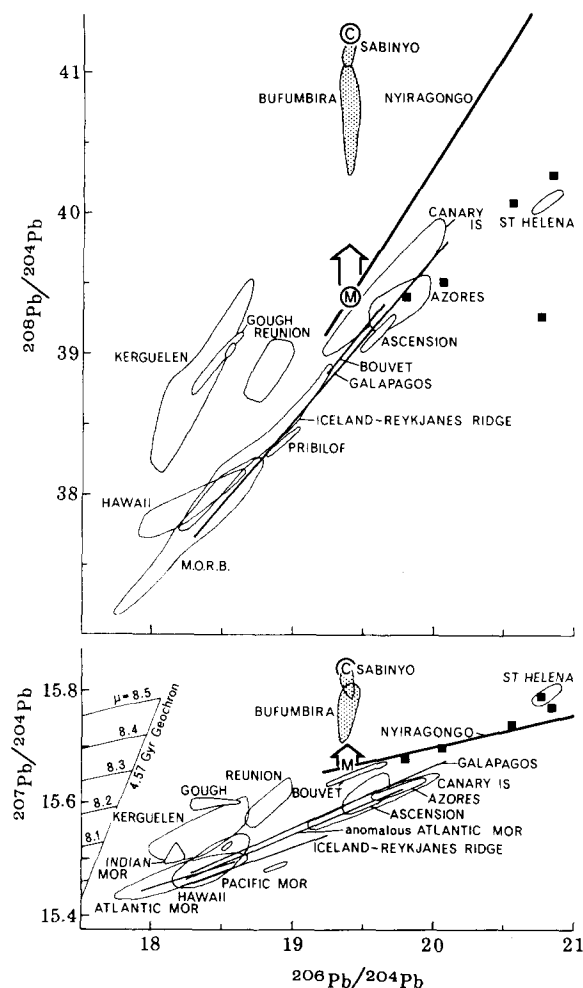
Fig. 4. Comparison of Pb isotopic compositions for Virunga volcanic rocks with oceanic basalts and Ugandan carbonatites (■). The Nyiragongo nephelinites inherited a 484 ± 28 Myr (2σ), Th/U = 5.0 isochron from their source region dating a metasomatic Th and U mantle enrichment [1]. The unique trends of the Bufumbira volcanics are caused by a binary mixing event and are thought to be relict features of the same ~ 500-Myr metasomatic event dated at Nyiragongo. *C* and *M* denote end-member compositions transposed to present-day values. The recent Sabinyo mixing (see Fig. 2) is not discernible in these diagrams as both end-members have nearly identical Pb isotopic compositions. See Sun [33] for data sources. Additional data—carbonatites: Lancelot and Allègre [25]; Nyiragongo: Vollmer and Norry [1].

known mantle compositions; such Pb compositions are usually regarded as typical for upper continental crust [18,19].

Sr-Pb. $^{87}Sr/^{86}Sr$ ratios are plotted against two different Pb isotopic ratios in fig. 5. A well-defined hyperbolic correlation is observed in the diagram $^{87}Sr/^{86}Sr$ versus $^{208}Pb/^{204}Pb$ but only an obscure correlation in the diagram $^{87}Sr/^{86}Sr$ versus $^{207}Pb/^{206}Pb$. But note the excellent correlation for the Sabinyo series in the latter diagram.

3. Discussion

We will start the discussion with Sabinyo. It will become clear later that the key to the understanding of the basic potassic rocks lies with the silica-saturated to -oversaturated rocks of this volcano. Also, a recent mixing event superimposes upon an ancient event here, thus complicating the isotope systematics. Both points make it necessary to devote a seemingly disproportionate space to the discussion of Sabinyo.

3.1. Sabinyo

To our knowledge, similar excellent correlations as shown by the Sabinyo series in the diagrams Figs. 2 and 5b have not been observed before for any young volcanic rocks. They point to the generation of this series by a recent two-component mixing. We tentatively suggest the series to be a hybrid magma series formed by mixing of a shoshonite and a latite primary magma, although this conclusion needs to be confirmed by using a range of compatible to incompatible elements [20]. Mixing of sources seems to be unlikely; Sr may not be an incompatible element for the low-Sr (felsic) end-member and variations in the degree of partial melting would probably result in a much larger scatter than is observed. Feldspar fractionation would strongly partition Sr and, if subsequent to mixing, would effect scatter in both the diagrams (Fig. 2) involving this element. This could be the cause for the small scatter among the shoshonite samples (CV690, C2893, C2895), although we tend not to think so because (a) despite feldspar being a dominant phenocryst phase [15], the other Sabinyo samples could not have fractionated feldspar, and (b) the scatter among these three samples is repeated also in the $^{87}Sr/^{86}Sr$
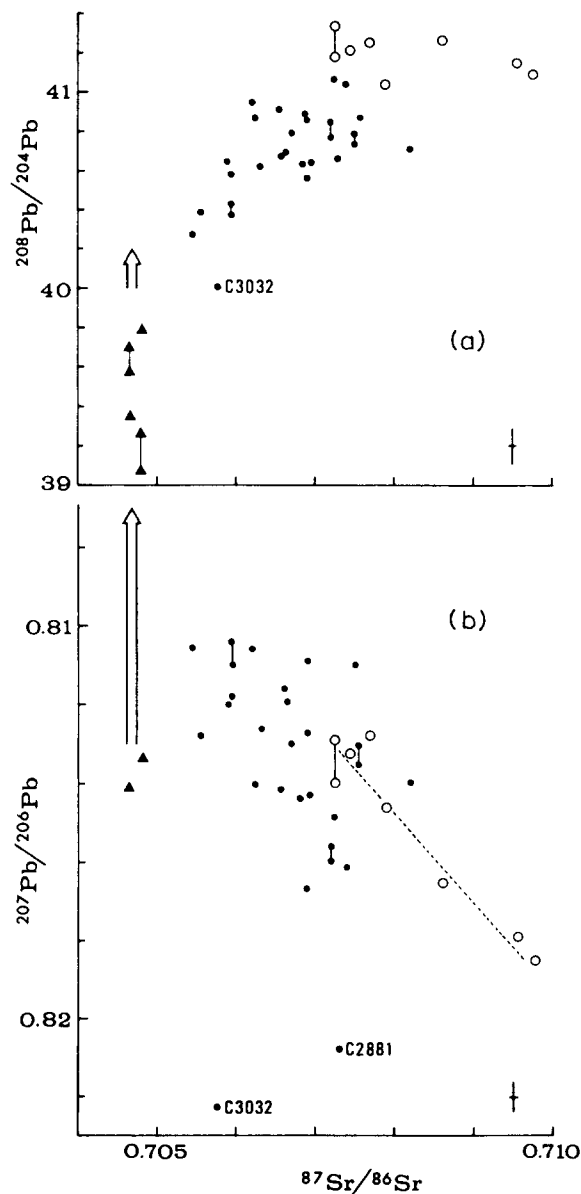
versus $^{207}Pb/^{206}Pb$ diagram (Fig. 5b) which instead suggest that one or two of these samples represent not hybrid but primary magma batches whose isotopic and trace element imprint reflects slightly differing source characteristics.

We do not consider variable contamination of a magma a viable hypothesis to account for this mixing relation. We understand here under *contamination* the incorporation, into a magma, of xenolithic material (which could be a solid, liquid or ions at the time of incorporation) from a lithological unit which clearly differs from the magma source itself [21]. One may also regard a hybrid magma series having as end-members a mantle- and a crustal-derived magma as a mantle-derived magma contaminated by a crustal melt, although we would tend not to do so, if both primary magmas are of equal importance.

The strongest argument against contamination is provided by the nearly identical Pb isotopic composition of all Sabinyo samples (Table 2 and Fig. 4). Crustal material shows typically very large Pb isotopic variations [18,19] and it would seem to require an exceptional coincidence if a contaminant were to have a similar isotopic signature to the magma. It could be argued that the contaminant was almost lead free or, alternatively, had a very much higher Pb concentration than the magma and therefore would either not effect, or would dominate the Pb isotopic composition of a mixture. This argument can, however, be confidently rejected (see Fig. 5b and the discussion next section).

Fig. 5. $^{87}Sr/^{86}Sr$ versus $^{208}Pb/^{204}Pb$ (a) and $^{207}Pb/^{206}Pb$ (b). Symbols as in Fig. 2. The arrows point towards the highly radiogenic Pb compositions from Nyiragongo. The length of the arrows are equal to the radiogenic growth in $^{208}Pb/^{204}Pb$ and $^{207}Pb/^{206}Pb$ for a 100-Myr period with $\mu = 10$ and $\kappa = 4$ and indicate variations in the Pb isotopic ratios which are to be expected for 20% source heterogeneities in Th/Pb and U/Pb since the 500-Myr source enrichment event The relative effect of radiogenic growth is much stronger in diagram (b), therefore the information gained from both diagrams is not identical: The data show a well-defined hyperbolic correlation in diagram (a) but only an obscure correlation in diagram (b). These differences suggest that not end-member heterogeneity but differential radiogenic growth between source domains subsequent to mixing is responsible for the scatter: mixing has not been a recent event. If average μ for Bufumbira sources has been about 10 then source heterogeneities in μ are $\leq 15\%$. Samples C2881 and C3032 are exceptional in representing source domains with a significantly lower μ. (b) The well-defined correlation shown by the Sabinyo series supports the Rb-Sr evidence (Fig. 2) for a hybrid origin of this series by *recent* mixing of two primary magmas. We estimate from the curvature and the Sr concentrations in the magmas the Pb concentration differences in the two Sabinyo primary magmas to be within a factor of two and conclude from this that the very similar Pb isotopic ratios for both primary magmas cannot be due to Pb domination by one end-member.

3.2. Sabinyo: Sr-Pb isotopic paradox

A combination of nearly constant Pb isotopic ratios with large Sr isotopic variations is a conspicuous feature of Sabinyo. The Tuscan Igneous Province is the only other known volcanic area with similar Pb and Sr isotope relations [22] and here also silicic and basic potassic alkaline volcanism are associated. Some possible answers to such an unusual situation are:

(1) Some time in the past an igneous or metasomatic event homogenised the Sabinyo source for the Sr and Pb isotopic compositions and U/Pb ratios but not for Rb/Sr. The observed Sr isotopic variations are a result of subsequent radiogenic growth.

(2) A recent event homogenized the volatile element Pb in its isotopic composition but not the perhaps less mobile refractory element Sr.

(3) The Pb isotopic composition throughout the mixing series is dominated by a Pb-rich end-member. This model can be tested for Sabinyo by using combined Sr and Pb isotope relations (Fig. 5b). Sabinyo samples fall on an almost straight line (more generally, variable degrees of mixing would result in hyperbolic curves in this diagram [20]) supporting the Rb-Sr evidence for mixing. However, calculated end-member Pb concentrations are similar, therefore we do not consider domination of the Pb isotopic composition in the mixture by one end-member to be a viable explanation for the Sabinyo Sr-Pb isotopic paradox.

The data from Sabinyo do not allow a choice between models (1) and (2). However, we will show that the Pb, Sr and Nd isotopic evidence from Virunga is consistent with growth of radiogenic ^{87}Sr subsequent to the 484 Myr event recorded from Nyiragongo. The model implies that the slope of the ^{87}Sr/^{86}Sr-Rb/Sr correlation for Sabinyo—despite being a result of recent magma mixing—dates the (ancient) homogenization event, subject of course to Rb/Sr fractionation during partial melting. The apparent Rb-Sr age is ~ 250 Myr, considerably less than the 484-Myr Pb-Pb age from Nyiragongo, however, a too young apparent age is to be expected if feldspar has been a residual phase. This is likely for the less Sr-rich latite end-member (the high ^{87}Sr/^{86}Sr end-mem-

ber in Fig. 2). The Rb-Sr systematics of Sabinyo are therefore consistent with and support a ~ 500-Myr source event for the Virunga volcanic field.

3.3. The 500-Myr event: Pb isotopic evidence

The extreme Pb isotope variations at Nyiragongo have made it possible to date this event precisely [1]. In contrast, the Pb isotope systematics of the mafic and felsic potassic rocks do not constrain the age of this event. But as homogenization has been incomplete, relicts allow inferences of pre-event features.

The Pb isotope characteristics of the Bufumbira volcanics are unique (Fig. 4 and [1, fig. 1]). Such trends, characterized by minor variations in ^{206}Pb/^{204}Pb but relative large variations in ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb cannot be reproduced by multi-stage evolution models [23]. These Pb compositions are mixtures of Pb which have evolved in unrelated reservoirs isolated from each other for a billion years or more. We emphasize that the Bufumbira Pb isotope trends do not reasonably allow any other interpretation: it is by far the simplest explanation. Probably only two isotopically distinct components were involved. Both components (C and M in Fig. 4) were fairly similar in ^{206}Pb/^{204}Pb but quite different in ^{207}Pb/^{204}Pb and ^{208}Pb/^{204}Pb.

The Sr-Pb isotopic relations (Fig. 5) make it unlikely that this mixing has been a recent event. Certainly the simplest answer is to equate the Bufumbira mixing event with the event recorded by the Rb-Sr systematics and the Pb-Pb isochron from Nyiragongo. The low-^{207}Pb/^{204}Pb, low-^{208}Pb/^{204}Pb end-member (M) is then likely to be represented by the Nyiragongo nephelinites. Because the Sabinyo series represents the other end-member (C in Figs. 3, 4), comprehension of the geochemistry of Sabinyo is decisive for an understanding of the origin of the basic potassic volcanics.

3.4. Parent-daughter element fractionation during the 500-Myr event

The reasonable fit of all Virunga Rb-Sr data to the reference isochron in Fig. 2a suggests that the

high Rb/Sr ratios are neither a result of extremely small degrees of melting nor are they due to extensive fractional crystallization; they seem, largely, to reflect source variations. This enables us to use them, together with observed isotopic ratios, to make some inferences about parent/daughter element fractionation during the 500-Myr event and about the isotopic characteristics of the source reservoirs prior to this event.

Rb/Sr. All rocks in the sequence nephelinites–mafic and felsic potassic rocks have high abundances of Rb, Sr and are strongly enriched in the light rare earth elements (LREE). It seems that Rb/Sr increased for *all* Virunga sources (due to Rb gain rather than Sr loss) and that this increase was least for the Nyiragongo source and highest for the Sabinyo sources. Rb/Sr source variations are restricted by the observed $^{87}Sr/^{86}Sr$ variations to < 30% for Nyiragongo but range from ~ 0.1 to ~ 0.3 for Sabinyo. The $^{87}Sr/^{86}Sr$ source ratios 500 Myr ago must have been fairly low, about 0.704 for the Nyiragongo nephelinites, 0.704–0.706 for the basic potassic rocks and ~0.706 for the Sabinyo
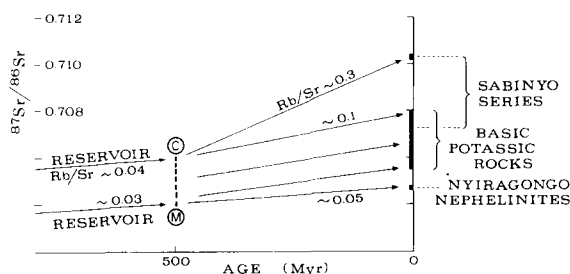

Fig. 6. Model for growth of radiogenic Sr with time in lithospheric reservoirs and domains sampled by the Virunga volcanism, showing the effects of metasomatism by a mantle-derived fluid 500 Myr ago. Prior to metasomatism, both lithospheric reservoirs *C* and *M* followed fairly similar Sr isotopic evolution curves with Rb/Sr of ~ 0.04 and ~ 0.03 resulting in only a small separation of the Sr isotopic compositions between both reservoirs at 500 Myr (*C*: $^{87}Sr/^{86}Sr$ ~ 0.706; *M*: ~ 0.704). Metasomatism, basically a mixing between a fluid with the isotopic composition of reservoir *M* and a solid (*C*), generated a range of domains with intermediate Sr isotopic compositions (broken line). All sampled domains gained Rb in preference over Sr but this caused strong Rb/Sr heterogeneities only between the two domains represented by the two Sabinyo primary magmas.

series (Fig. 2a). Fig. 6 summarizes the Rb-Sr evolution.

Sm/Nd. Observed Sm/Nd ratios are fairly constant (Sm/Nd_N ~ 0.5 [24]) throughout the whole sequence but ϵ_{Nd} varies from + 2.5 (Nyiragongo) to − 10 (Sabinyo) with intermediate values for the basic potassic rocks (Fig. 3). The observed Sm/Nd ratios suggest fairly uniform source Sm/Nd ratios: the large variations in ϵ_{Nd} are likely to be a pre-metasomatic relic. Prior to the metasomatic event, the Nyiragongo source had been LREE depleted while the sources for the Sabinyo magmas had already been enriched in LREE. It follows that metasomatism caused a general enrichment in LREE relative to HREE; LREE/HREE (Nd/Sm) and Rb/Sr behaved coherently. But in contrast to Rb/Sr, increase in LREE was highest for the Nyiragongo source and least pronounced for Sabinyo sources.

U/Pb (μ). Fractionation of μ was highly erratic for the Nyiragongo source, causing in some source domains very large increase in μ by U gain [1]. By contrast, the two Sabinyo primary magma sources which responded so strongly in the Rb-Sr system, differ in μ by only about 20%. Fractionation of μ within source domains for basic potassic rocks followed the Sabinyo rather than Nyiragongo pattern. The least radiogenic Pb from Nyiragongo, which probably has not had a history in an anomalous high-μ environment (judging from Figs. 4 and 5b, μ has been perhaps even slightly reduced by the 500-Myr event), plots within the fields for African carbonatites [25] and with a similar relative ^{207}Pb enrichment which seems to be an upper mantle feature for the whole region encompassing Central and South Africa, the South Atlantic (Gough, Bouvet) and the western Indian Ocean (Réunion, Kerguelen) (Fig. 4). Sabinyo Pb has an isotopic signature generally regarded as typical for upper continental crust [19]; the isotopic ratios fall far outside the fields for any known mantle compositions and must have done so already prior to the 500-Myr event (Fig. 4).

These fractionation patterns can be understood if a trace-element-rich fluid migrates through the source rocks and leaches, exchanges, or deposits

mobile elements according to their solubility. Major fractionation will occur when one or several of these elements are fixed in a newly formed metasomatic mineral. In the absence of any such mineral the fluid is expected to homogenize the solid for a particular element or group of elements. The physical and chemical conditions controlling the solubility of Rb, Sr, U, Th, Pb and the REE seem to have been drastically different for the Nyiragongo and Sabinyo sources with the sources for basic potassic rocks occupying an intermediate position.

Petrographic evidence for metasomatism beneath the western branch of the East African rift system has been presented by Holmes and Harwood [15], Lloyd and Bailey [26] and Lloyd [27].

3.5. Crustal metasomatism?

One of the end-members (M in Figs. 3, 4 and 6) of the 500-Myr metasomatic event, the mantle fluid, is isotopically indistinguishable from the pre-metasomatic upper mantle. No isotopic relics of either the fluid or the pre-metasomatic mantle are observed. This suggests that the invading fluid had been in equilibrium with the solid and had isotopically homogenized it.

Because of large Nd and Pb isotopic differences, the other end-member reservoir (C in Figs. 3, 4, 6) characterized by low ϵ_{Nd} and high $^{208}Pb/^{204}Pb$ values and represented by Sabinyo, must have been isolated from the Nyiragongo upper mantle reservoir for 10^9 years or more, certainly pre-dating metasomatism. Although chemically isolated, both source reservoirs were mechanically coupled for, at least, the last 500 Myr. Is it possible to identify these reservoirs among known Earth reservoirs?

The base of the lithosphere is commonly perceived as a thermal and mechanical boundary with ductile flow of the asthenosphere relative to the lithosphere. The requirement of being coupled for 500 Myr confines both reservoirs to the lithosphere. The two obvious units of the continental lithosphere with which the two Virunga reservoirs could correspond are the lithospheric mantle and the crust: The Sr, Nd and Pb isotopic composi-

tions of the Nyiragongo magmas and their nephelinitic chemistry are compatible with an upper mantle origin. Detailed mineralogical and chemical modeling of the Sabinyo source has to await further work but the fairly high silica content of the felsic Sabinyo magmas and their Pb and Nd isotopic signature suggest derivation from continental crustal material.

When basing a crustal or mantle origin of a magma on isotopic arguments one has to be aware that isotopic signatures alone do not give any information about the physical source relations and a melt derived from a crustal block tectonically depressed into the mantle (or derived from recycled crustal material in the mantle) may be isotopically indistinguishable from a truly crustal anatectic melt. Therefore the choice whether the Sabinyo source lies within the crust, within crustal material subducted or depressed into the mantle or within an ancient enriched mantle region coexisting in close proximity with ancient depleted lithospheric upper mantle cannot be made from the present data. We are facing again the same difficulty already encountered in the interpretation of the Italian potassic volcanism [2].

Certainly the simplest answer would be to assume a crustal source. But as the two components of which the chemistry of the basic potassic rocks is made up of are then perceived as being the mantle fluid and the pre-metasomatic crust, it follows that the source depth for these magmas would be constrained to ~ 35 km [28] by the thickness of the non-metasomatised crust in East Africa. This appears to be at variance with experimental evidence suggesting a much greater depth for these magmas (e.g. [8,9]). Nevertheless, comparison with the isotope systematics of the K-rich volcanic rocks from Italy lends strong support to this solution [21].

Should it be the correct answer then inferences can be made about present isotopic signatures in the lower crust and the depth of magma sources (Fig. 7). The basic potassic magmas range in their isotopic signatures from near mantle compositions to crustal compositions. As metasomatism will establish isotopic gradients in the lower crust, isotopic variations will correspond to variations in the height of the magma source above the crust-
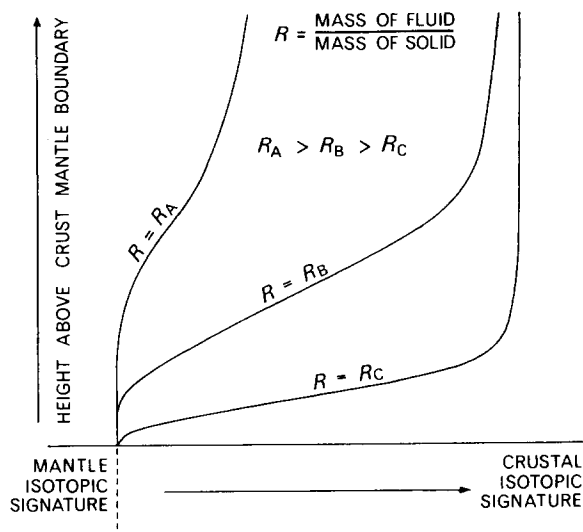
Fig. 7. Crustal metasomatism by a mantle fluid: schematic isotopic depth profiles for three different fluid/solid mass ratios. Isotopic gradients in the metasomatised lower crust are a function of the height above the crust-mantle boundary. These schematics are generally valid for any metasomatising fluid crossing an interface between two solids and are not confined to the special case of a crust/mantle boundary shown here. (From Vollmer, in preparation.)

mantle boundary. Acidic magmas from the Sabinyo volcano are nearest in their isotopic imprints to the crustal end-member compositions or coincide with them and are probably drawn from a higher crustal level than any of the basic potassic magmas.

It is worth mentioning that the present data do not give any evidence for surviving pre-metasomatic heterogeneities within the $-\epsilon_{Nd}$ reservoir. If it is the crust, then the lower crust immediately above the crust-mantle boundary had, before being metasomatised, the same average isotopic signature as the Sabinyo source. There is, in particular, no evidence for an ancient U-depleted lower crust. This suggests that other mobile elements were also not depleted, perhaps an essential condition in the evolution of basic high-K magmas for which extreme high abundances of alkali metals and other LIL elements are typical.

No isotopic evidence is found for newly injected asthenospheric material initiating partial

melting. We suggest that partial melting of the ancient metasomatised lithospheric domains leading to the volcanism is entirely a response to decompression and rising geotherms during rifting and thinning of the lithosphere.

4. Conclusions

The Pb, Nd and Sr isotope systematics of the young Virunga volcanic field are dominated by an ancient homogenization or mixing event affecting the source regions of the magmas. The age of this event could be dated to 484 ± 28 Myr from a Pb-Pb isochron which the Nyiragongo magmas inherited from their sources [1]. The Rb-Sr systematics of the Sabinyo volcano are consistent with and support this age when allowance is made for Rb/Sr fractionation during partial melting.

Parent/daughter element fractionation inferred for this ancient event point to a metasomatic event during which a mantle fluid invaded and metasomatised the subcontinental mantle (represented by Nyiragongo nephelinites and characterised by positive ϵ_{Nd} values) and another lithospheric reservoir with a negative ϵ_{Nd} signature. No pre-metasomatic isotopic mantle relicts are observed, suggesting isotopic homogenisation and equilibration of the fluid with the upper mantle material.

The metasomatic model implies that all basic high-K magmas and the acidic Sabinyo magmas come from the metasomatised $-\epsilon_{Nd}$ reservoir. Metasomatism established isotopic gradients in this reservoir. If it is assumed that $a + \epsilon_{Nd}$ fluid migrated *upwards*, then the ϵ_{Nd} imprint of the magma is directly related to the height of the magma source above the boundary separating the $+\epsilon_{Nd}$ and $-\epsilon_{Nd}$ reservoirs. Feldspar-free basic potassic magmas then come from a greater depth than the feldspar-bearing variety and the acidic Sabinyo magmas will be drawn from a higher level than any of the basic magmas.

Whether the $-\epsilon_{Nd}$ reservoir is the crust, continental crustal material subducted or depressed into the mantle or highly anomalous enriched mantle cannot be decided from the data. The simplest answer, that this reservoir is the crust, seems to be in contradiction to experimental evi-

dence suggesting a subcrustal origin for basic high-K magmas.

Isotopic variations not attributable to the metasomatic event itself can be accounted for by subsequent radiogenic growth. At Sabinyo, a young mixing event superimposes the ancient event. It suggests the Sabinyo shoshonite-latite series to be a hybrid magma series. No feldspar fractionation is observed at Sabinyo and fractional crystallization could only have been of minor importance for all other samples. Contamination of magmas during their ascent could be excluded for Sabinyo magmas and did not determine the isotopic characteristics of any of the other sampled magmas.

Volcanism does not seem to be initiated by material transport from the asthenosphere but entirely a response to rifting of the lithosphere.

Acknowledgements

References

1 R. Vollmer and M.J. Norry, Unusual isotopic variations in Nyiragongo nephelinites, Nature 301, 141, 1983.

2 C.J. Hawkesworth and R. Vollmer, Crustal contamination versus enriched mantle: ^{143}Nd/^{144}Nd and ^{87}Sr/^{86}Sr evidence from the Italian volcanics, Contrib. Mineral. Petrol. 69, 151, 1979.

3 L. Civetta, F. Innocenti, P. Manetti, A. Peccerillo and G. Poli, Geochemical characteristics of potassic volcanics from Mts. Ernici (southern Latium, Italy), Contrib. Mineral. Petrol. 78, 37, 1981.

4 P.M. Holm and N.C. Munksgaard, Evidence for mantle metasomatism: an oxygen and strontium isotope study of the Vulsinian district, Central Italy, Earth Planet. Sci. Lett. 60, 376, 1982.

5 M.F. Roden, Origin of coexisting minette and ultramafic breccia, Navajo volcanic field, Contrib. Mineral. Petrol. 77, 195, 1981.

6 N.W. Rogers, S.W. Bachinski, P. Henderson and S.J. Parry, Origin of potash-rich basic lamprophyres: trace element data from Arizona minettes, Earth Planet. Sci. Lett. 57, 305, 1982.

7 D.K. Bailey, Mantle metasomatism—continuing chemical change within the Earth, Nature, 296, 525, 1982.

8 M. Barton and D.L. Hamilton, The melting relationships of a madupite from the Leucite Hills, Wyoming, to 30 kb., Contrib. Mineral. Petrol. 69, 133, 1979.

9 A.D. Edgar, E. Condliffe, R.L. Barnett and R.J. Shirran, An experimental study of an olivin ugandite magma and mechanisms for the formation of its K-enriched derivatives, J. Petrol. 21, 475, 1980.

10 B. Turi and H.P. Taylor, Oxygen isotope studies of potassic volcanic rocks of the Roman Province, Central Italy, Contrib. Mineral. Petrol. 55, 1, 1976.

11 H.P. Taylor, B. Giannetti and B. Turi, Oxygen isotope geochemistry of the potassic igneous rocks from the Roccamonfina volcano, Roman comagmatic region, Italy, Earth Planet. Sci. Lett. 46, 81, 1979.

12 G.P. Bagdasaryan, V.I. Gerasimovskiy, A.I. Polyakov and R.K.H. Gukasyan, Age of volcanic rocks in the rift zones in East Africa, Geochem. Int. 10, 66, 1973; transl. from Geokhimiya S.S.S.R. 1, 84, 1973.

13 P. Guibert, M. Delaloye and J. Hunziker, Contribution à l'étude géologique du volcan Mikeno, chaîne des Virunga (République du Zaïre), C.R. Soc. Phys. Hist. Nat. Genève, N.S. 10, 57, 1975.

14 L. Cahen, Igneous activity and mineralisation episodes in the evolution of the Kibaride and Katangide orogenic belts of Central Africa, in: African Magmatism and Tectonics, T.N. Clifford and I.G. Gass, eds., p. 97, Oliver and Boyd, Edinburgh, 1970.

15 A. Holmes and H.F. Harwood, The volcanic area of Bufumbira: the petrology of the volcanic area of Bufumbira, Mem. Geol. Surv. Uganda 3, Part II, 300 pp., 1937.

16 K. Bell and J.L. Powell, Strontium isotopic studies of alkalic rocks: the potassium-rich lavas of the Birunga and Toro-Ankole regions, East and Central equatorial Africa, J. Petrol. 10, 536, 1969.

17 Th.G. Sahama, The Nyiragongo main cone, Ann. Mus. R. Afrique Centrale, Tervuren (Belgique), Sér. 8°, Sci. Géol. 81, 1–88, 1978.

18 B.R. Doe, Lead Isotopes, 137 pp., Springer, Berlin, 1970.

19 B.R. Doe and R.E. Zartman, Plumbotectonics, I. The Phanerozoic, in: Geochemistry of Hydrothermal Ore Deposits, H.L. Barnes, ed., 2nd ed., pp. 22–70, John Wiley and Sons, New York, N.Y., 1979.

20 C.H. Langmuir, R.D. Vocke, G.N. Hanson and S.R. Hart, A general mixing equation: applied to the petrogenesis of basalts from Iceland and Reykjanes Ridge, Earth Planet. Sci. Lett. 37, 380, 1977.

21 R. Vollmer, Comparative Sr, Nd and Pb isotope geochemistry of potassic alkaline volcanic rocks from Virunga and Italy: petrogenetic and geodynamic inferences, Earth Planet. Sci. Lett. (submitted).

22 R. Vollmer, Isotopic evidence for genetic relations between acid and alkaline rocks in Italy, Contrib. Mineral. Petrol. 60, 109, 1977.

23 N.H. Gale and A.E. Mussett, Episodic uranium-lead models and the interpretation of variations in the isotopic composition of lead in rocks, Rev. Geophys. Space Phys. 11, 37, 1973.

24 R.H. Mitchell and K. Bell, Rare earth element geochemistry of potassic lavas from the Birunga and Toro-Ankole regions of Uganda, Africa, Contrib. Mineral. Petrol. 58, 293, 1976.

25 J.R. Lancelot and C.J. Allègre, Origin of carbonatitic magma in the light of the Pb-U-Th isotope system, Earth Planet. Sci. Lett. 22, 233, 1974.

26 F.E. Lloyd and D.K. Bailey, Light element metasomatism of the continental mantle: the evidence and consequences, Phys. Chem. Earth 9, 389, 1975.

27 F.E. Lloyd, Upper mantle metasomatism beneath a continental rift: clinopyroxenes in alkali mafic lavas and nodules from South West Uganda, Min. Mag. 44, 315, 1981.

28 J. Wohlenberg, Geophysikalische Aspekte der ostafrikanischen Grabenzonen, Geol. Jahrb. E4, 1-84, 1975.

29 W.M. White and A.W. Hofmann, Sr and Nd isotope geochemistry of basalts and mantle evolution, Nature 296, 821, 1982.

30 M. Menzies and V.R. Murthy, Enriched mantle: Nd and Sr isotopes in diopsides from kimberlite nodules, Nature 283, 634, 1980.

31 J.D. Kramers, C.B. Smith, N.P. Lock, R.S. Harmon and F.R. Boyd, Can kimberlites be generated from an ordinary mantle?, Nature 291, 53, 1981.

32 R. Vollmer, P. Ogden and J.G. Schilling, Nd isotope geochemistry of Leucite Hills ultra-potassic volcanism, EOS Trans. Am. Geophys. Union 62, 431, 1981.

33 S.-S. Sun, Lead isotopic study of young volcanic rocks from mid-ocean ridges, ocean islands and island arcs, Philos. Trans. R. Soc. London, Ser. A, 297, 409, 1980.

34 K. Bell and R.J. Doyle, K-Rb relationships in some continental alkaline rocks associated with the East African rift valley system, Geochim. Cosmochim. Acta 35, 903, 1971.

35 A.D. Combe and W.C. Simmons, The volcanic area of Bufumbira, Mem. Geol. Surv. Uganda 3, Part I, 1-150, 1933.