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Isotopic and geochemical evidence for a heterogeneous mantle plume origin of the Virunga volcanics, Western rift, East African Rift system

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ABSTRACT

Virunga volcanics in the western rift of the East African Rift system (EARS) show silica-undersaturated, ultra-alkaline, alkalic-mafic compositions. The two active Virunga volcanoes, Nyiragongo and Nyamuragira, are 15 km apart. Nyiragongo shows unusual compositions not seen globally and has the lowest recorded viscosity among terrestrial magmas while Nyamuragira is unusually effusive. These volcanoes occur along the fringes of a topographic uplift within the EARS. We analyzed major, trace elements and Nd–Sr–Pb isotopes in Nyiragongo and Nyamuragira lavas including samples from 2002, 2003 and 2004 eruptions.

The youngest Nyiragongo lavas are ultrapotassic, ultrasodic and have low SiO_2 (36.6 wt.%). Nyamuragira lavas are basalts, basanites and tephrites, distinct from the foiditic Nyiragongo lavas. Both volcanic products show enrichment in light rare earths, large ion lithophile and high field strength elements. High chondrite-normalized Dy/Yb suggests residual garnet in the source(s). Nyiragongo and Nyamuragira lavas show high Nb/U similar to oceanic basalts while Ce/Pb ratios are unusually high in Nyiragongo. 2002 and 2003 Nyiragongo lavas show superchondritic Zr/Hf suggesting carbonate metasomatism in their source. Both these volcanics show low K/Rb (231–356) ratios suggesting phlogopite-melting in their mantle source. Residual garnet and phlogopite suggests melt derivation from depths between 80 and 150 km.

Nyiragongo lavas show bulk-earth like Nd–Sr isotopes. In Nd–Sr–Pb isotope space, both Nyiragongo and Nyamuragira show correlations similar to ocean island basalts with a strong affinity for EM II. Pb-isotopic variations in some parts of the 2002 Nyiragongo lava flow are caused by leaching of Pb by various degrees of fusion of granitic basement rocks found as xenoliths in the lava and within the volcanic cone. In a plot of $^{207}Pb/^{206}Pb$ versus $^{208}Pb/^{206}Pb$, however, the least contaminated lavas plot conspicuously in the field of Type I kimberlites suggesting a sub-lithospheric mantle origin. Also, the reported MORB-like He-isotopic composition of the Nyiragongo lavas are distinctly higher than those of the sub-continental lithospheric mantle. In summary, the Nyiragongo lavas are bulk silicate earth-like in Nd and Sr-isotopes, Group I kimberlite-like in their Pb-isotopes, and their high Ce/Pb, low SiO_2 rule out continental lithospheric sources, particularly in conjunction with all the isotopic data. Our combined geochemical data with available experimental petrological data on peridotitic compositions suggest that Nyiragongo lavas formed at greater depths by low degree partial melting of a garnet, clinopyroxene, and phlogopite-bearing carbonated mantle, while the Nyamuragira lavas are products of larger degree partial melting at comparatively shallower mantle depths with a recycled crustal component. We argue that simultaneous volcanism in adjacent Nyiragongo and Nyamuragira, with magmas originating from different depths requires the presence of a heterogeneous mantle plume beneath the Tanzanian craton. This plume caused chemically distinctive volcanic provinces around the Tanzanian craton, in the Western and Kenya Rift.

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1. Introduction

The East African Rift System (EARS) is a classic example of ongoing continental rifting and provides an excellent framework to investigate mafic, alkalic and ultrabasic magmatism in an extensional setting. The rift system extends over 2000 km from the Red Sea in the north to Mozambique in the south and traverses two regions of topographic uplift, the Ethiopian dome to the north and the Kenyan dome to the south, which are separated by the Turkana depression characterized by Quaternary volcanism, and the Anza graben representing a ~150 km wide zone of NW–SE trending extension (Fig. 1a).

Basaltic magmatism commenced ~45 Ma ago (George et al., 1998) in southern Ethiopia and has been reported along the entire length of

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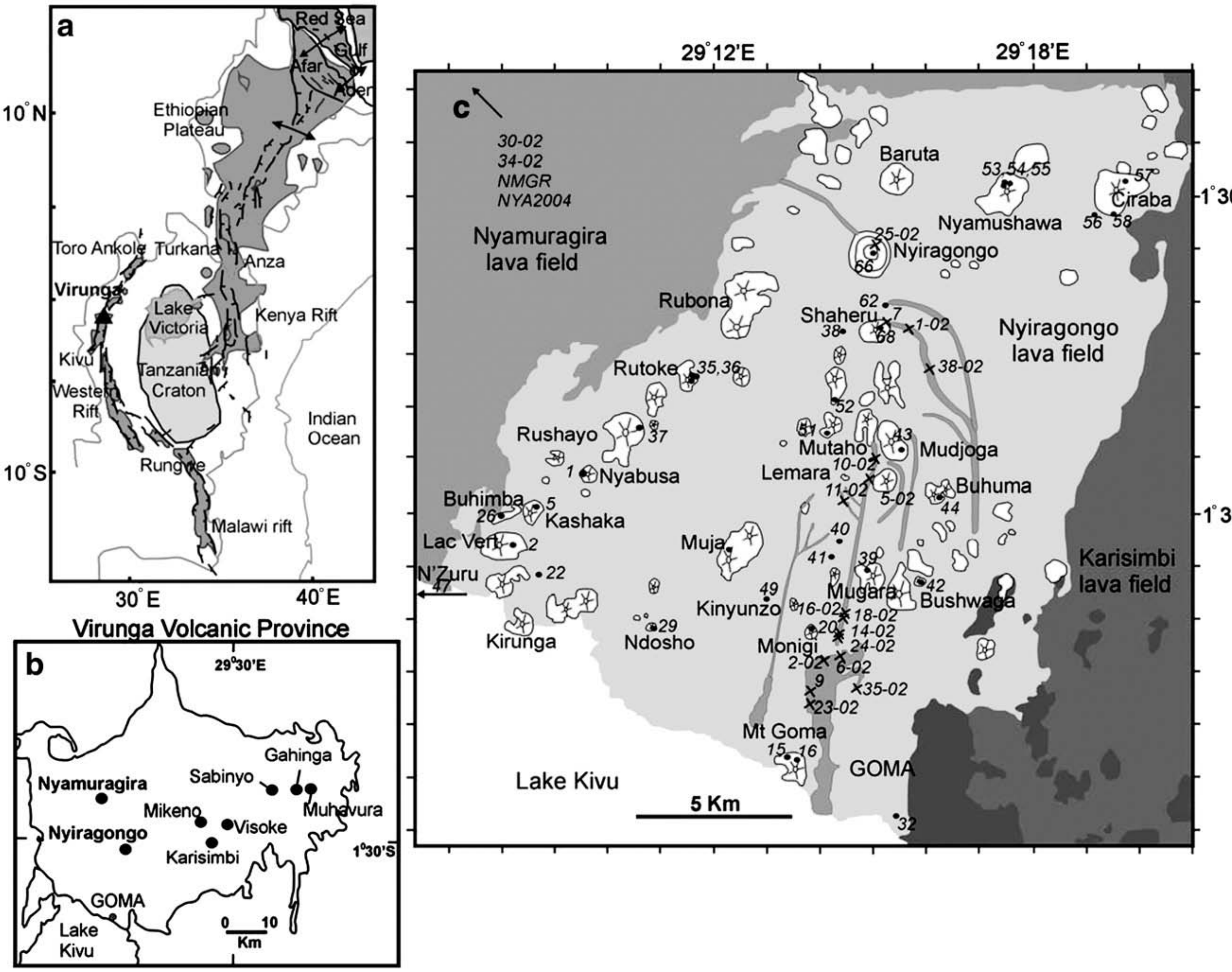

Fig. 1. (a) Simplified map showing the major structures of the East African Rift System (EARS), including the Ethiopian plateau to the north, the Tanzanian craton to the south and the Turkana depression between these two domal structures. South of Turkana, the EARS splits into the western rift and the Kenya rift running on either side of the Tanzanian craton. Prominent volcanic provinces around the Tanzanian craton include Virunga (filled triangle), Kivu, Toro Ankole, and Rungwe in the western rift, the Turkana basalts to the north and the Kenya rift basalts to the east. (b) The locations of the different volcanoes of the Virunga Volcanic Province (VVP) including the Nyiragongo and Nyamuragira volcanoes of the present study. (c) Geological map of the Nyiragongo volcanic complex showing several plugs and cones and the lava field. Also shown are the Nyamuragira and Karisimbi volcanic fields, the locations of samples from the 2002 lava flow (cross) and older lavas (filled circle), and the approximate location of the Nyamuragira samples.

the EARS (Fig. 1a) although there are clear geochemical distinctions between the volcanics to the north and the south (George et al., 1998; Rogers et al., 2000; Furman et al., 2004; Furman, 2007). Volcanism is more voluminous to the north where there is geochemical, petrological and geophysical evidence for the Afar mantle plume underlying the Ethiopian plateau (Ebinger et al., 1989; Hofmann et al., 1997) with flood basalt eruptions ~30 Ma ago in northern Ethiopia and Yemen (Schilling et al., 1992; Pik et al., 1999). Towards the south the EARS splits into two halves, the Kenyan rift in the east and the western rift to the west of Lake Victoria. The western rift extends from Lake Albert in the north to the southeast of the Lake Tanganyika in the south (Fig. 1a). GPS and earthquake slip vector data suggest the existence of a microplate between the Kenyan and western rifts (Calais et al., 2006) and at the core of this microplate is the 2.5–3.0 Ga old Tanzanian craton.

In this study, we will focus on the volcanics in the southern part of the EARS. Several volcanic provinces occur along the Kenya rift and the western rift. Among them, the Virunga Volcanic Province (VVP), located in the west rift of the EARS, is characterized by unusual silica-undersaturated, ultra-alkaline mafic volcanism that started erupting ~11 Ma ago with continuing activity today. The two currently active volcanoes of Virunga (Fig. 1b), Nyiragongo and Nyamuragira, are in a seismically active zone of the west rift at the periphery of the Archean Tanzanian craton. The Virunga volcanics, especially Nyiragongo, had received considerable attention for their unusual mineralogy and petrology (Holmes and Harwood, 1937; Sahama, 1957; Sahama, 1960; Sahama, 1973; Demant et al., 1994) unmatched by any other lava of this region as well as other global volcanics (Sahama, 1962; Rogers et al., 1998; Platz et al., 2004). The Nyiragongo volcano was also the focus of global attention in January 2002 when its spectacular eruption caused a humanitarian crisis as the lava rapidly flowed through the city of Goma before draining into Lake Kivu to the south. Thermal and rheological properties (Giordano et al., 2007) of this lava suggest the dry viscosity of the Nyiragongo lavas to be the lowest measured among terrestrial natural magmas. Nyamuragira, located only 15 km to the north of Nyiragongo, is compositionally less extreme but is one of the most effusive global volcanoes erupting several times in the past few decades.

We present a major, trace element and Nd–Sr–Pb isotopic study of the VVP, in particular the currently active Nyiragongo and Nyamuragira, including the 2002 Nyiragongo lava flows. The goal of the present study on spatially as well as temporally distributed lava samples from the western part of the VVP is to understand the petrogenesis of these unique ultra-alkaline and ultrabasic volcanics in the context of rift extension by using the above geochemical and isotopic tracers and by integrating available geochemical and geophysical information. In particular, we wish to investigate the nature of the magma source composition and possible contributions from the sub-continental lithospheric mantle, asthenospheric or sub-asthenospheric mantle as well as the role of fluids in generating these compositionally unique magmas.

2. Sample description and analytical methods

We analyzed 19 lava samples from the 2002 lava flow of Nyiragongo along a ~20 km stretch from the summit to Lake Kivu, one 2003 lava lake sample, and 41 samples from older cones, plugs and lava flows of this volcanic complex (Fig. 1c). Multiple samples were analyzed from the 2002 lava to check for possible flow differentiation effects, such as crystal settling and fractional crystallization, which affect trace element partitioning. In addition, we also analyzed 5 samples from Nyamuragira, including one 2004 lava, located only ~15 km northwest of Nyiragongo. Two quartzites from farther east possibly representing the basement of the Virunga volcanics were also analyzed.

The Nyiragongo lava has low viscosity, on account of its unusually low SiO_2 content, as discussed later. This lava is aphyric to microcrystalline, with a porphyritic texture that is characterized by small phenocrysts of melilite, kalsilite, leucite, Ti-augite and olivine in a fine-grained glassy groundmass. Common groundmass minerals that are petrographically discernible in the older lavas include kalsilite, nepheline and smaller amounts of leucite with minor clinopyroxene, olivine, perovskite, apatite and titanomagnetite (Sahama, 1973). The lavas of Nyiragongo are unique and to the best of our knowledge are unmatched by any other terrestrial occurrence. These lavas are strongly alkaline and silica-undersaturated, and in terms of normative mineralogy can be termed as melilitite, melilite nephelinite, pyroxene nephelinite, leucitite, and leucite nephelinite (Platz et al., 2004). These extreme normative compositions differ from all other volcanoes of the VVP including Nyamuragira (Sahama, 1962; Demant et al., 1994; Rogers et al., 1998; Platz et al., 2004).

The Nyamuragira volcanics comprise olivine basanites, tephrites, phonolitic tephrites and tephro-phonolites with phenocrysts of olivine, Ca-rich clinopyroxene, plagioclase, leucite, titanomagnetite, apatite, and nepheline. These rocks are also silica-undersaturated having 7.5–15.7% normative nepheline in spite of their higher SiO_2 (Aoki et al., 1985) contents than the Nyiragongo lavas.

All the whole rock samples of this study were powdered in an alumina Spex™ ball mill. Major elements (Table 1) were analyzed in a commercial laboratory, Actlabs, in Ontario, Canada. The samples underwent lithium metaborate/tetraborate fusion followed by measurement using an ICP-OES (optical emission spectrometry). Trace element concentrations (Table 2) were measured using a quadrupole ICP-MS (Thermo elemental X-7 series) at the University of Rochester. 25-mg of rock powder for each sample was treated with 2:1 HF/HNO_3 acid mixtures in sealed teflon beakers on a hotplate for 48 h. After evaporation to incipient dryness, the samples were twice treated with 1 ml of HNO_3 and dried down. Each sample was diluted to 100 ml of 2% HNO_3 solution (dilution factor 4000) with ~10 ppb internal standard of In, Cs, Re, and Bi. Table 2 shows the elemental concentrations of the samples obtained by using BCR-2 and BIR-2 (concentrations from USGS) as known standards. The concentrations of the various elements, other than the rare earth elements, are within 5% error, as estimated from repeated measurements of AGV-2 (andesite-USGS) and BHVO-2 (basalt-USGS) rock standards which were run as unknowns. The rare earth elements (REE) are more precisely determined to within 2% error. For Ba and the REEs, oxide correction was necessary. The sizes of the oxide and hydroxide interferences were estimated by analyzing single element solutions of Ba and the REEs. A typical measurement sequence involved blank, USGS standard, blank, sample, blank USGS standard. Lower limits of detection for the 25 elements reported in Table 2 are typically less than 5 ppb.

For Nd, Sr, Pb isotopic analyses, between 100 and 200 mg of powdered rock samples were digested in HF, HNO_3 and HCl acid mixtures. Nd and Sr-isotopes were measured with a VG Sector multi-collector TIMS using the procedures established for our laboratory at the University of Rochester (Basu et al., 1991). Pb isotopes were measured using the silica-gel technique by TIMS, also at the University of Rochester (Sharma et al., 1992). Filament temperatures during Pb-

Table 1
Major element compositions of selected lava samples

Sample #		SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	LOI	K_2O+Na_2O	K_2O/Na_2O	Mg#	K/Rb
NY1-02	Nyiragongo	36.6	19.9	12.4	0.27	3.9	11.4	5.5	5.1	2.6	1.4	−0.21	10.6	0.9	23.7	350
NY 2-02	2002 lava	38.7	15.5	13.2	0.28	4.0	12.1	5.6	5.3	2.8	1.5	−0.16	10.9	1.0	23.5	343
NY 5-02		38.9	15.0	13.3	0.29	4.1	12.2	6.0	5.6	2.8	1.4	−0.85	11.6	0.9	23.5	289
NY 6-02		38.6	14.9	13.1	0.29	4.1	12.2	5.7	5.5	2.8	1.4	−0.42	11.2	1.0	23.6	271
NY10-02		38.2	16.0	13.2	0.28	4.0	12.1	5.9	5.4	2.8	1.4	−0.78	11.3	0.9	23.5	258
NY11-02		38.1	15.7	13.1	0.28	4.0	12.0	5.5	5.2	2.8	1.4	0.28	10.7	1.0	23.5	262
NY14-02		38.4	15.6	13.1	0.28	4.0	12.0	5.8	5.6	2.8	1.4	−0.68	11.4	1.0	23.4	268
NY16-02		38.9	14.9	13.3	0.29	4.1	12.2	5.8	5.7	2.8	1.5	−0.23	11.5	1.0	23.4	276
NY18-02		39.0	14.8	13.4	0.29	4.1	12.3	6.0	5.7	2.8	1.5	−0.94	11.7	0.9	23.5	278
NY23-02		39.1	15.0	13.4	0.29	4.1	12.3	6.0	5.7	2.8	1.5	−0.90	11.7	1.0	23.4	329
NY24-02		38.9	15.4	13.3	0.29	4.1	12.2	5.9	5.4	2.8	1.5	−1.05	11.3	0.9	23.5	253
NY25-02		39.2	15.6	13.3	0.28	4.1	12.2	5.7	5.4	2.9	1.5	−0.57	11.2	1.0	23.6	333
NY26-02		39.1	15.0	13.3	0.28	4.2	12.2	5.6	5.3	2.9	1.5	−0.70	11.0	0.9	24.0	264
NY35-02		39.4	14.9	13.4	0.29	4.1	12.1	5.8	5.4	2.8	1.5	−0.72	11.3	0.9	23.2	289
NY38-02		39.5	14.9	13.4	0.29	4.1	12.1	5.7	5.6	2.8	1.5	−0.75	11.3	1.0	23.2	258
NY7		38.7	14.4	13.2	0.29	4.1	12.2	5.8	5.4	2.7	1.5	−1.16	11.2	0.9	23.6	311
NY9		38.1	14.2	13.1	0.29	4.1	12.2	5.8	5.4	2.7	1.4	−0.82	11.2	0.9	23.7	305
NY1	Nyiragongo	39.2	14.8	13.7	0.29	4.5	12.3	4.9	5.4	3.0	1.7	−0.12		1.1	24.7	338
NY2	Older lavas	39.2	10.6	12.6	0.22	10.4	16.5	2.6	2.6	3.2	1.3	0.68	5.1	1.0	45.2	356
NY5		40.6	10.7	12.3	0.20	13.2	13.6	2.1	2.4	2.9	0.8	0.17	4.5	1.1	51.8	330
NY15		38.6	14.6	13.9	0.28	4.6	12.5	5.6	2.5	3.2	1.7	1.49	8.1	0.4	25.0	231
NY16		38.8	15.0	14.2	0.29	4.2	12.0	5.1	4.6	3.1	1.7	0.24	9.7	0.9	22.8	303
NY20		39.8	14.6	13.2	0.29	4.1	12.1	5.7	5.3	2.7	1.5	−0.85	11.1	0.9	23.6	328
NY22		44.0	16.3	12.6	0.22	4.3	9.5	4.4	4.3	3.2	1.1	−0.23	8.6	1.0	25.5	289
NY26		39.4	15.3	13.4	0.27	4.3	12.1	4.8	4.6	3.1	1.7	0.14	9.5	1.0	24.4	318
NY29		39.1	15.1	13.3	0.29	4.1	12.0	4.9	4.9	3.0	1.6	−0.02	9.8	1.0	23.5	338
NY32		45.2	13.4	12.4	0.19	8.0	11.9	2.8	3.0	3.3	0.6	−0.52	5.8	1.1	39.3	328
NY34-02	Nyamuragira	45.2	9.8	10.8	0.16	14.9	13.0	1.5	1.6	2.4	0.3	−0.50	3.1	1.0	58.0	253

Table 2
Trace element compositions of the Nyiragongo and Nyamuragira volcanics and the basement rocks

2002 Nyiragongo lava

Sample	NY 1-02	NY 2-02	NY 5-02	NY 6-02	NY 10-02	NY 11-02	NY 14-02	NY 16-02	NY 18-02	NY 23-02	NY 24-02	NY 25-02	NY 26-02	NY 35-02	NY 38-02	NY 40-02	NY 42-02	NY 7	NY 9	2003 lava lake
Rb	121	129	160	167	175	166	173	173	170	144	176	136	167	156	178	110	171	145	146	139
Sr	2341	2481	2820	2913	2917	2824	2928	2995	2863	2901	2895	2446	2746	2775	2907	2359	115	2937	2936	2613
Y	33	34	46	51	54	50	54	51	51	45	52	35	52	46	52	30	0.60	42	42	42
Zr	269	284	327	332	337	328	339	342	331	340	334	310	330	326	333	301	39	331	331	327
Nb	243	256	327	324	318	314	327	337	321	330	321	319	309	323	324	295	25	351	350	298
Ba	2339	2436	3296	3172	3127	2682	3530	2934	3668	3445	3363	2200	2626	3034	3653	1985	1594	2398	2383	2305
La	158	167	212	230	235	227	241	227	231	207	229	182	229	217	232	150	0.94	199	198	177
Ce	321	330	479	505	462	477	484	494	487	482	478	373	463	477	487	335	4.8	365	362	361
Pr	32	33	44	47	47	46	49	46	47	43	46	37	46	44	47	30	0.17	39	39	36
Nd	108	112	146	159	153	156	164	157	157	143	155	125	155	146	159	100	0.68	129	128	123
Sm	15	16	21	22	22	22	22	23	21	20	21	18	22	22	22	14	0.10	19	19	18
Eu	4.1	4.3	5.5	6.0	5.6	5.9	5.8	6.1	5.5	5.2	5.7	4.8	5.7	5.6	5.8	3.6	0.17	5.3	5.3	5.2
Gd	9.4	10.5	12.7	13.5	14.5	13.8	13.9	14.5	13.0	13.6	13.0	11.9	13.1	13.1	13.4	8.8	0.21	13.3	13.3	13.2
Tb	1.25	1.34	1.73	1.94	1.92	1.82	1.94	1.90	1.81	1.87	1.81	1.49	1.88	1.77	1.85	1.13	0.04	1.66	1.66	1.69
Dy	6.6	6.9	9.0	9.8	9.9	10.3	10.9	10.0	11.6	10.1	10.7	7.0	10.6	9.4	10.8	6.1	0.29	8.3	8.2	8.2
Ho	1.15	1.19	1.70	1.81	1.80	1.91	2.0	1.90	2.1	1.85	2.1	1.29	1.92	1.78	2.0	1.10	0.06	1.51	1.49	1.49
Er	2.8	2.9	4.1	4.4	4.5	4.7	4.9	4.7	5.1	4.3	5.1	3.2	4.7	4.5	5.0	2.6	0.15	3.7	3.7	3.7
Tm	0.34	0.34	0.57	0.61	0.64	0.65	0.69	0.63	0.70	0.61	0.70	0.43	0.66	0.60	0.70	0.38	0.02	0.52	0.52	0.50
Yb	2.4	2.4	3.4	3.7	3.8	4.0	4.1	3.7	4.3	3.7	4.1	2.6	4.0	3.6	4.2	2.3	0.15	3.2	3.2	3.1
Lu	0.36	0.35	0.50	0.53	0.52	0.57	0.58	0.52	0.60	0.51	0.58	0.37	0.56	0.48	0.60	0.33	0.02	0.43	0.42	0.41
Hf	3.9	4.0	4.4	4.5	4.3	4.6	4.6	4.4	4.8	4.5	4.6	4.3	4.6	4.3	4.7	4.0	0.60	4.4	4.3	4.4
Ta	12.5	13.1	23	23	22	24	23	23	24	23	23	22	22	23	24	20	0.56	17.0	17.0	15.4
Pb	5.3	5.4	6.3	6.7	6.5	6.1	6.3	6.3	6.1	6.4	5.9	6.2	11.0	6.0	16.2	6.1	228	6.1	6.0	5.4
Th	19	20	28	30	32	30	33	30	32	27	31	22	30	28	32	18.8	0.37	24	24	23
U	8.3	8.6	10.3	10.1	9.8	10.5	10.3	10.7	10.7	9.5	10.8	10.2	9.3	10.0	10.7	8.5	6.4	9.8	9.8	9.1
La/Yb(N)	44	47	42	42	41	39	39	41	37	38	37	48	39	41	38	44		43	42	39
Dy/Yb(N)	1.79	1.88	1.73	1.74	1.69	1.69	1.72	1.74	1.76	1.80	1.68	1.77	1.74	1.72	1.69	1.72		1.70	1.70	1.74
U/Pb	1.59	1.58	1.64	1.51	1.51	1.74	1.64	1.70	1.77	1.48	1.84	1.64	0.85	1.67	0.66	1.40	0.03	1.62	1.64	1.68
Th/U	2.3	2.4	2.8	3.0	3.3	2.9	3.2	2.8	3.0	2.9	2.9	2.2	3.2	2.8	2.9	2.2	0.06	2.4	2.4	2.6
Ce/Pb	61	61	76	76	71	79	77	79	80	75	82	60	42	79	30	55	0.02	60	61	66
Nb/U	29	30	32	32	32	30	32	32	30	35	30	31	33	32	30	35		36	36	33
Zr/Hf	69	72	75	74	79	71	74	77	69	75	73	72	72	75	70	75		76	77	75

Older Nyiragongo lavas

Sample	Rumoka	NY 1	NY 2	NY 5	NY15	NY 16	NY 20	NY 22	NY 26	NY 29	NY 32	NY 35 A	NY 35 B	NY 36	NY 37	NY 38	NY 39	NY 40	NY 41	NY 42
Rb	92	133	60	60	89	125	135	123	121	121	75	79	68	68	118	139	99	129	139	89
Sr	872	2379	1776	1188	2099	2515	2740	1700	2298	2422	914	1539	1602	1595	2042	2649	1593	2209	2202	1116
Y	25	40	29	27	42	43	39	33	39	40	27	32	30	31	39	41	34	41	40	28
Zr	303	326	282	279	363	369	311	322	333	343	255	288	298	275	302	311	296	347	350	290
Nb	132	293	173	128	253	277	329	215	269	283	125	153	152	147	209	306	208	251	240	149
Ba	972	2107	1199	940	1862	2012	2214	1497	1714	1965	1033	1011	981	976	1009	2265	2013	1954	1938	1085
La	72	179	116	87	169	175	184	131	169	172	73	111	114	109	127	179	122	158	152	92
Ce	168	334	224	171	320	333	336	240	315	320	143	217	215	200	331	336	203	308	304	171
Pr	15	37	25	20	36	37	36	26	35	36	16	25	24	26	34	36	25	34	34	21
Nd	54	123	90	71	122	126	119	88	117	118	59	91	91	91	120	121	93	116	121	76
Sm	9.1	18.4	14.4	12.0	18.8	19.2	17.7	13.8	17.6	17.9	10.3	15.1	14.8	15.2	18.8	18.2	15.1	17.9	18.5	13.0
Eu	2.5	5.1	4.0	3.4	5.2	5.4	4.9	3.8	4.9	5.0	2.8	4.3	4.1	4.3	5.2	5.1	4.2	5.1	5.1	3.7
Gd	7.0	13.1	10.3	8.9	13.5	13.7	12.4	10.0	12.5	12.8	7.9	11.1	11.0	11.6	13.6	13.9	11.1	13.5	13.1	9.6
Tb	0.92	1.63	1.26	1.12	1.69	1.70	1.56	1.26	1.56	1.58	1.05	1.40	1.33	1.43	1.67	1.80	1.38	1.77	1.64	1.25
Dy	4.7	8.0	6.0	5.5	8.2	8.5	7.7	6.4	7.7	7.9	5.4	6.6	6.3	6.7	8.2	8.8	6.9	8.5	8.0	6.0
Ho	0.91	1.46	1.06	0.99	1.50	1.55	1.43	1.20	1.43	1.46	1.02	1.16	1.12	1.18	1.50	1.60	1.25	1.55	1.48	1.09
Er	2.3	3.6	2.5	2.4	3.7	3.8	3.5	3.0	3.5	3.6	2.5	2.7	2.6	2.8	3.6	4.0	3.1	3.8	3.7	2.6
Tm	0.33	0.49	0.33	0.32	0.51	0.52	0.49	0.42	0.49	0.50	0.36	0.36	0.34	0.37	0.50	0.52	0.42	0.51	0.50	0.35

Yb	2.1	3.0	2.0	2.0	3.1	3.2	3.0	2.7	3.0	3.1	2.2	2.1	2.1	2.2	3.1	3.2	2.6	3.1	3.1	2.1
Lu	0.29	0.41	0.27	0.26	0.42	0.43	0.41	0.37	0.40	0.41	0.30	0.29	0.27	0.30	0.41	0.44	0.35	0.43	0.42	0.28
Hf	5.7	4.6	5.5	5.9	5.4	5.2	4.1	5.8	4.9	5.0	6.0	5.6	6.2	5.7	5.7	4.3	7.0	5.1	5.2	6.1
Ta	6.2	14.5	9.1	6.7	13.0	14.2	16.5	10.7	14.1	14.7	6.6	8.9	8.8	9.1	13.1	17.3	12.6	14.3	14.7	9.8
Pb	9.7	5.8	3.8	4.7	6.5	5.2	6.0	9.6	5.6	4.9	5.9	5.3	3.4	6.1	5.3	5.6	4.7	4.4	4.9	2.9
Th	11.9	19.1	11.9	9.8	17.7	18.9	23	20	20	20	10.9	12.2	11.2	11.8	17.4	24	15.3	20	21	9.9
U	2.3	7.2	4.4	2.8	4.7	6.3	9.3	6.1	7.0	6.7	2.2	4.4	4.4	4.3	5.7	9.2	5.6	6.2	7.1	3.8
La/Sm(N)	23	40	39	30	37	37	41	33	38	38	22	35	37	33	28	38	32	35	33	29
Dy/Yb(N)	1.45	1.73	1.92	1.83	1.72	1.71	1.68	1.55	1.68	1.66	1.58	2.0	2.0	2.0	1.74	1.78	1.72	1.79	1.69	1.85
U/Pb	0.23	1.23	1.14	0.60	0.72	1.22	1.55	0.63	1.25	1.37	0.38	0.82	1.26	0.71	1.08	1.64	1.19	1.40	1.44	1.32
Th/U	5.3	2.7	2.7	3.5	3.8	3.0	2.4	3.2	2.8	3.0	4.9	2.8	2.6	2.7	3.1	2.6	2.7	3.2	2.9	2.6
Ce/Pb	17	57	59	36	50	64	56	25	56	65	24	41	62	33	63	60	43	69	62	59
Nb/U	58	41	40	46	54	44	35	35	38	42	56	35	35	34	37	33	37	40	34	39
Zr/Hf	53	70	51	47	67	71	75	55	68	69	43	52	48	48	53	72	42	68	68	47

Older Nyiragongo lavas

Sample	NY 43 A	NY 43 B	NY 44	NY 45	NY 46	NY 48	NY 49 A	NY 51	NY 52	NY 53	NY 54	NY 55	NY 56	NY 57	NY 58	NY 62	NY 63	NY 66	NY 68	MUJA
Rb	86	126	74	84	141	82	136	89	86	89	67	64	130	157	114	27	135	133	143	176
Sr	1432	1478	1046	976	2497	1650	2545	1910	1858	1504	1417	1318	2625	2512	1924	591	2350	2612	2976	1629
Y	29	33	29	27	44	33	44	28	37	33	34	29	41	37	34	17	35	40	42	31
Zr	312	320	282	267	280	334	356	290	286	308	315	315	316	320	341	142	299	311	321	307
Nb	160	157	145	131	303	189	282	172	183	156	142	133	273	250	229	49	230	293	309	206
Ba	1502	1484	1327	1130	1235	1128	2212	1052	1147	1049	1065	958	2108	1989	1608	377	2203	2234	2379	1515
La	100	101	82	79	185	132	179	126	139	116	109	102	164	172	133	43	171	174	176	120
Ce	178	186	151	149	353	335	347	272	284	211	206	199	329	349	263	84	340	349	362	215
Pr	22	23	20	18	44	26	39	30	30	24	25	21	35	33	28	10	33	36	36	24
Nd	79	82	74	66	145	95	128	94	105	90	91	80	116	116	92	37	113	117	119	81
Sm	13.0	13.9	12.7	11.2	21.3	15.0	19.3	13.6	16.5	14.8	14.9	13.1	17.7	17.7	14.3	6.7	17.0	17.7	18.2	12.5
Eu	3.6	3.9	3.5	3.2	5.8	4.3	5.5	4.0	4.7	4.2	4.3	3.7	5.0	4.9	4.1	2.0	4.7	5.0	5.2	3.6
Gd	9.4	10.4	9.2	8.3	14.5	10.9	14.1	9.2	12.1	10.9	11.5	9.6	13.5	12.3	10.6	5.2	11.9	13.3	13.8	9.4
Tb	1.20	1.34	1.19	1.07	1.80	1.38	1.78	1.19	1.51	1.39	1.43	1.23	1.74	1.55	1.38	0.70	1.48	1.73	1.80	1.20
Dy	6.0	6.6	5.9	5.3	8.8	6.8	8.7	6.1	7.3	6.8	7.0	6.0	8.3	7.6	6.7	3.5	7.3	8.3	8.7	5.9
Ho	1.09	1.22	1.08	0.97	1.61	1.23	1.60	1.18	1.32	1.24	1.26	1.09	1.53	1.40	1.24	0.66	1.35	1.52	1.59	1.11
Er	2.7	3.0	2.6	2.4	4.0	3.0	3.9	3.1	3.2	3.0	3.1	2.6	3.8	3.5	3.1	1.62	3.3	3.8	4.0	2.8
Tm	0.36	0.41	0.36	0.33	0.55	0.42	0.54	0.46	0.43	0.40	0.41	0.35	0.51	0.47	0.42	0.21	0.46	0.50	0.53	0.39
Yb	2.3	2.5	2.2	2.0	3.4	2.5	3.2	3.0	2.6	2.4	2.5	2.2	3.1	2.9	2.6	1.32	2.8	3.1	3.3	2.4
Lu	0.30	0.35	0.30	0.27	0.46	0.34	0.44	0.42	0.35	0.33	0.34	0.29	0.42	0.39	0.36	0.18	0.37	0.42	0.43	0.33
Hf	6.4	6.5	7.1	5.5	3.6	6.6	4.6	4.2	5.0	6.5	6.2	5.8	4.4	4.4	5.8	3.0	3.9	4.3	4.4	5.1
Ta	9.5	9.6	8.8	7.7	21	9.8	15.1	17.9	9.8	8.9	8.4	7.6	15.0	15.1	12.4	2.8	15.7	15.9	17.3	10.7
Pb	2.6	4.7	5.2	2.9	8.0	5.3	4.4	9.6	2.0	2.5	4.9	1.6	4.0	8.8	6.3	2.0	5.7	5.6	12.5	5.9
Th	12.4	13.4	10.9	9.8	31	14.6	21	30	16.5	11.9	11.8	10.1	20	19	20	4.7	21	23	25	19
U	4.1	3.8	3.4	3.2	14.7	5.2	6.4	12.0	5.7	4.3	3.9	3.7	7.2	7.0	6.0	1.26	8.7	8.8	9.6	5.5
La/Yb(N)	30	27	25	27	37	35	37	29	37	32	30	32	35	41	35	22	41	38	36	34
Dy/Yb(N)	1.73	1.71	1.74	1.75	1.70	1.75	1.75	1.34	1.85	1.81	1.84	1.82	1.73	1.74	1.66	1.71	1.69	1.74	1.72	1.59
U/Pb	1.60	0.80	0.66	1.13	1.84	0.97	1.47	1.26	2.87	1.72	0.80	2.36	1.81	0.80	0.95	0.63	1.51	1.56	0.76	0.92
Th/U	3.0	3.5	3.2	3.0	2.1	2.8	3.2	2.5	2.9	2.8	3.0	2.8	2.8	2.7	3.3	3.8	2.5	2.6	2.6	3.5
Ce/Pb	70	39	29	52	44	63	79	28	143	84	42	128	82	39	42	42	59	62	29	36
Nb/U	39	41	42	40	21	37	44	14	32	36	36	36	38	36	38	39	27	33	32	38
Zr/Hf	49	49	40	48	77	51	77	70	58	47	51	54	72	73	59	48	76	73	74	60

(continued in next page)

Table 2 (*continued*)

Basement				Nyamuragira			
Sample	NY 59	NY 61	NY 30-02	NY 34-02	NMGR	NYA 2004	NY 47
Rb	1.35	1.85	61	51	52	79	57
Sr	4.4	14.1	880	550	781	915	878
Y	0.16	6.6	26	23	32	33	26
Zr	15.2	38	266	176	289	279	210
Nb	0.38	2.3	105	63	110	104	80
Ba	2.8	8.5	921	593	1010	929	767
La	1.59	42	68	46	82	71	56
Ce	4.5	83	134	87	144	139	107
Pr	0.52	9.0	16.0	11.2	20	17.0	13.1
Nd	1.76	31	59	43	71	61	49
Sm	0.37	5.9	10.0	7.9	12.7	11.2	8.8
Eu	0.09	1.21	2.6	2.1	3.2	3.2	2.5
Gd	0.20	3.4	7.7	6.3	9.7	9.1	6.8
Tb	0.027	0.42	1.02	0.80	1.28	1.26	0.94
Dy	0.11	1.76	5.3	4.3	6.3	6.5	4.9
Ho	0.02	0.27	1.01	0.81	1.19	1.25	0.94
Er	0.029	0.60	2.5	2.1	2.9	3.2	2.4
Tm	0.004	0.083	0.35	0.30	0.43	0.45	0.34
Yb	0.037	0.55	2.2	1.9	2.6	2.8	2.1
Lu	0.004	0.08	0.30	0.25	0.36	0.39	0.29
Hf	0.42	1.06	5.9	4.3	6.3	6.5	4.7
Ta	0.024	0.16	7.3	4.7	8.0	5.9	4.3
Pb	0.55	1.59	6.9	3.8	6.6	6.9	5.2
Th	0.46	4.9	10.0	7.3	11.7	11.2	8.8
U	1.04	6.6	1.76	1.25	2.0	2.1	1.69
La/Yb(N)	29	53	21	17	21	17	18
Dy/Yb(N)	1.88	2.1	1.60	1.50	1.57	1.51	1.55
U/Pb	1.88	4.1	0.26	0.33	0.30	0.30	0.32
Th/U	0.44	0.74	5.7	5.8	5.9	5.4	5.2
Ce/Pb	8.2	52	19	23	22	20	20
Nb/U	0.36	0.36	60	50	56	51	48
Zr/Hf	36	36	45	41	46	43	44

Table 3
Nd, Sr, Pb isotopic composition of the Nyiragongo and Nyamuragira volcanics and the basement rocks

Sample #	$^{147}Sm/^{144}Nd$	$^{143}Nd/^{144}Nd(0)$[a]	εNd (0)[b]	$^{87}Rb/^{86}Sr$	$^{87}Sr/^{86}Sr$ (0)[c]	$^{206}Pb/^{204}Pb$[d]	$^{207}Pb/^{204}Pb$[d]	$^{208}Pb/^{204}Pb$[d]
Nyiragongo 2002 lava								
NY-1-02	0.086	0.512675	0.72	0.15	0.70469	19.67	15.67	39.64
NY-2-02	0.089	0.512667	0.57	0.15	0.704658	19.68	15.68	39.68
NY-5-02	0.089	0.512698	1.17	0.16	0.704674	19.68	15.68	39.65
NY-6-02	0.089	0.512687	0.96	0.16	0.704745	19.69	15.69	39.69
NY-10-02	0.093	0.512664	0.51	0.17	0.704692	19.69	15.68	39.68
NY-11-02	0.089	0.512645	0.14	0.17	0.704653	19.68	15.67	39.65
NY-14-02	0.085	0.512681	0.84	0.17	0.704675	19.69	15.68	39.67
NY-16-02	0.092	0.512709	1.38	0.16	0.704689	19.69	15.68	39.67
NY-18-02	0.083	0.512688	0.98	0.17	0.704757	19.71	15.70	39.76
NY-23-02	0.090	0.512628	−0.20	0.14	0.704608	19.66	15.66	39.60
NY-24-02	0.086	0.512694	1.09	0.17	0.704573	19.67	15.68	39.63
NY-25-02	0.093	0.512732	1.83	0.16	0.704674	19.66	15.66	39.60
NY-26-02	0.088	0.512685	0.92	0.17	0.704597	19.48	15.66	39.45
NY-35-02	0.094	0.512660	0.43	0.16	0.704694	19.69	15.70	39.74
NY-38-02	0.086	0.512667	0.57	0.17	0.704689	19.67	15.68	39.65
NY-40-02	0.087	0.512708	1.37	0.13	0.704618	19.70	15.71	39.72
NY-42-02		0.512123	−10.05	4.21	0.792726	19.83	15.77	39.85
NY-7	0.093			0.14	0.704746	19.67	15.66	39.59
NY-9	0.093	0.512688	0.98	0.14	0.704553	19.67	15.67	39.62
2003 lava lake	0.093			0.15		19.66	15.71	39.72
Nyiragongo older lavas								
Rumoka	0.106	0.512556	−1.60	0.30	0.705658	19.40	15.72	40.04
NY-1	0.095			0.16	0.704507	19.51	15.65	39.51
NY-2	0.101	0.512742	2.03	0.09	0.704344	19.30	15.60	39.41
NY-5	0.106	0.512689	0.99	0.14	0.704496	19.36	15.62	39.52
NY-15	0.097	0.512679	0.80	0.12	0.704406	19.45	15.64	39.40
NY-16	0.096	0.512714	1.48	0.14	0.704361	19.52	15.65	39.50
NY-20	0.094	0.512699	1.19	0.14	0.704602	19.67	15.66	39.61
NY-22	0.098			0.20	0.705111	19.73	15.74	39.88
NY-26	0.095	0.512757	2.32	0.15	0.70454	19.31	15.64	39.28
NY-29	0.096			0.14	0.704488	19.48	15.63	39.47
NY-32	0.110	0.512572	−1.29	0.23	0.705366	19.20	15.68	40.08
NY 35 B	0.102	0.512685	0.92	0.12	0.704373	19.37	15.61	39.46
NY 36	0.106	0.512674	0.70	0.12	0.704382	19.38	15.61	39.46
NY 37	0.099	0.512639	0.02	0.16	0.704431	19.49	15.65	39.43
NY 38	0.095	0.512644	0.12	0.15	0.704642	19.67	15.66	39.61
NY 39	0.103	0.512599	−0.76	0.18	0.704485	19.42	15.64	39.69
NY 40	0.097			0.16	0.70445	19.48	15.64	39.51
Nyiragongo older lavas								
NY 41	0.097	0.512671	0.64	0.18	0.704458	19.48	15.65	39.53
NY 42	0.108	0.512641	0.06	0.22	0.704524	19.30	15.62	39.57
NY 43 A	0.104	0.512583	−1.07	0.17	0.704864	19.39	15.66	39.89
NY 43 B	0.108	0.512661	0.45	0.24	0.704828	19.38	15.66	39.87
NY 44	0.108	0.512652	0.27	0.20	0.704737	19.53	15.70	39.95
NY 45	0.108	0.512637	−0.02	0.24	0.704551	19.46	15.65	39.81
NY 46	0.093	0.512744	2.07	0.16	0.704788	19.65	15.68	39.68
NY 48	0.100	0.512607	−0.60	0.14	0.704538	19.44	15.66	39.66
NY 49 A	0.096	0.512676	0.74	0.15	0.704477	19.54	15.66	39.53
NY 49 B					0.704622	19.55	15.67	39.59
NY 51	0.091			0.13	0.705289	19.39	15.67	39.71
NY 52	0.099	0.512734	1.87	0.13	0.704515	19.76	15.74	40.02
NY 53	0.104	0.512669	0.60	0.17	0.704518	19.40	15.66	39.68
NY 54	0.104	0.512697	1.15	0.13	0.704595	19.38	15.64	39.63
NY 55	0.104	0.512676	0.74	0.14	0.704376	19.38	15.64	39.62
NY 56	0.096			0.14		19.50	15.64	39.50
NY 57	0.096			0.18	0.704554	19.53	15.66	39.56
NY 58	0.098	0.512673	0.68	0.17	0.704699	19.52	15.67	39.66
NY 62	0.114	0.512758	2.34	0.13	0.704606	19.69	15.70	39.71
NY 63	0.095	0.512662	0.47	0.16	0.704587	19.67	15.66	39.60
NY 66	0.095	0.512737	1.93	0.14	0.704701	19.71	15.69	39.71
NY 67						19.70	15.70	39.72
NY 68	0.096	0.512723	1.66	0.14		19.69	15.71	39.75
NY-23E		0.512661	0.45		0.704373			
Basement								
NY 59	0.133			0.87	0.72422	71.36	19.12	48.43
NY 61	0.121	0.511705	−18.20	0.37	0.733914	176.02	25.38	49.35
Nyamuragira								
NY-30-02	0.107	0.512527	−2.17	0.20	0.705736	19.28	15.72	40.45
NY-34-02	0.117	0.512540	−1.91	0.26	0.705681	19.34	15.72	40.30
NMGR	0.113	0.512549	−1.74	0.19	0.706031	19.27	15.74	39.97
NYA 2004	0.113	0.512545	−1.74	0.24	0.706027	19.32	15.72	40.03
NY 47	0.113	0.512448	−3.71	0.18	0.70601	19.39	15.76	40.55

[a] Measured $^{143}Nd/^{144}Nd$ ratios were normalized to $^{146}Nd/^{144}Nd=0.7219$. Uncertainties for the measured $^{143}Nd/^{144}Nd$ were less than 3 in the 5th decimal place. La Jolla Nd-standard analyzed during the course of this study yielded $^{143}Nd/^{144}Nd=0.511852+24$ ($2\sigma_{mean}$) ($n=10$).

[b] $\varepsilon_{Nd(0)}$ were calculated using the present day bulk earth (CHUR) value of $^{143}Nd/^{144}Nd=0.512638$. The $\varepsilon_{Nd(0)}$ values represent the deviation of $^{143}Nd/^{144}Nd$ in parts per 10^4 from the present day CHUR value.

[c] Measured $^{87}Sr/^{86}Sr$ ratios were normalized to $^{86}Sr/^{88}Sr=0.1194$. Uncertainties for the measured $^{87}Sr/^{86}Sr$ were less than 4 in the 5th decimal place. The NBS-987 Sr standard analyzed during the course of this study yielded $^{87}Sr/^{86}Sr=0.710245+23$ ($2\sigma_{mean}$) ($n=8$).

[d] Pb isotopic data corrected for mass fractionation. Estimated errors are less than +0.05% per mass unit.

isotope ratio measurements were monitored continuously and raw ratios were calculated as weighted averages of the ratios measured at 1150 °C, 1200 °C and 1250 °C, respectively. The reported Pb-isotopic data are corrected for mass fractionation of 0.12±0.03% per amu based on replicate analyses of the NBS-981 Equal Atom Pb Standard measured in the same fashion. Our laboratory procedural blanks were less than 400 pg for Sr and less than 200 pg for both Nd and Pb. No blank correction was necessary for the isotope ratios measured. Analytical uncertainties in the isotope ratio measurements (Table 3) were less than 4 in the 5th decimal place for Sr, 3 in the 5th decimal place for Nd and $<+0.05\%$ per mass unit for Pb.

3. Results

3.1. Major element composition

Major element concentrations (Table 1) were determined for 27 selected samples from the Nyiragongo lava field including both 2002 and older lavas and one Nyamuragira sample. Nyiragongo volcanics,

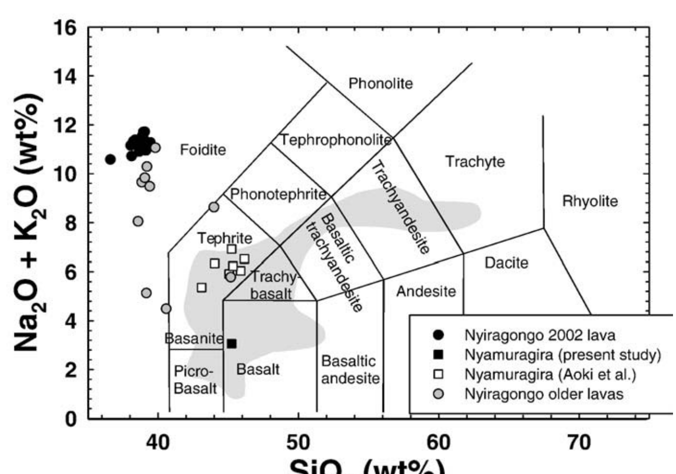

Fig. 2. Major element composition of the Nyiragongo and Nyamuragira volcanics represented in a Total Alkali Silica diagram (LeMaitre, 1984). The shaded region represents the composition of other volcanics from Virunga (Rogers et al., 1992, 1998), Kivu (Furman and Graham, 1999), Turkana (Furman et al., 2004), and Kenya rift (Rogers et al., 2000) located around the Tanzanian craton (Fig. 1a). The most silicic lavas in the shaded field are from Sabinyo in the VVP. The silica-undersaturated Nyiragongo lavas plot in the field of foidites with high alkali contents. The 2002 lavas (dark circles) do not show much variation in composition whereas the older Nyiragongo rocks (gray circles) show varying alkali contents. The Nyamuragira volcanics (squares, present study and from Aoki et al., 1985) are basaltic to tephritic in composition and overlap with the other volcanics of this region.

especially the 2002 lavas, are ultrapotassic with as high as 5.7 wt.% K_2O. Na_2O contents are also very high ranging up to 6.0 wt.%. Nyiragongo volcanics are silica-undersaturated with SiO_2 as low as 36.6 wt.%. In the total alkali versus silica diagram (LeMaitre, 1984), these rocks plot in the foidite field (Fig. 2), outside the field of commonly occurring volcanic rocks. Compared to the 2002 lava, the older lavas of Nyiragongo show greater variability in bulk rock chemistry, especially in their alkali content (Fig. 2). Mg-numbers (Mg# = 100 * Mg/Mg + Fe) are as low as 22.8 for the Nyiragongo lavas.

The Nyamuragira sample (NY-34-02) has lower alkali (K_2O = 1.6 wt.%, Na_2O = 1.5 wt.%), higher silica (45.2 wt.%), and higher Mg# (58) compared to the Nyiragongo volcanics, and this analysis (Table 1) is consistent with reported analyses of multiple Nyamuragira lavas (Aoki et al., 1985) that range from basalts, basanites to tephrites (Fig. 2).

3.2. Trace element composition

The Nyiragongo and Nyamuragira volcanics show uniform, sub-parallel, light rare earth element (LREE) enriched patterns (Fig. 3a–c). The REE concentrations (Table 2) of the Nyiragongo volcanics are unusually high with La as high as 197 ppm (100–1000x chondrite). Nyamuragira volcanics also show high La (average 65 ppm). The highest REE concentrations and the most fractionated LREE (average La_N/Yb_N = 41.0) are observed in the 2002 Nyiragongo lavas, while the Nyamuragira lavas show much lower REE concentrations and are less fractionated (average La_N/Yb_N = 19.2) compared to the Nyiragongo lavas. Average Dy_N/Yb_N is also high for these volcanics (Table 2), although the heavy REE patterns for the Nyiragongo and Nyamuragira volcanics are relatively less steep (Fig. 3a–c) compared with the LREE fractionation.

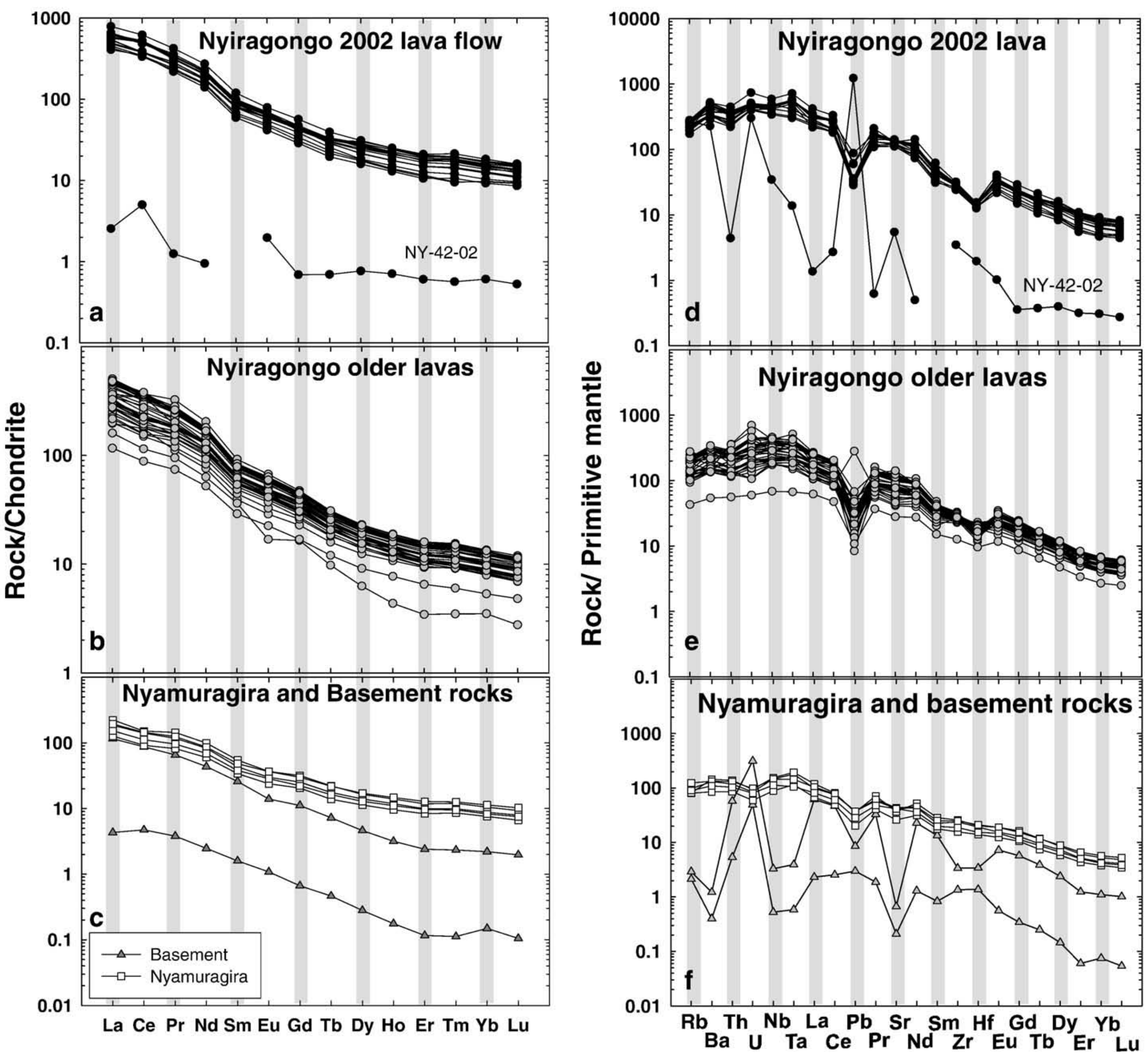

Fig. 3. Chondrite normalized rare earth element patterns (a–c) and primitive mantle-normalized multiple trace element concentration patterns (d–f) of the 2002 and older lava flows of Nyiragongo, the Nyamuragira volcanics as well as the two basement rocks. None of the lavas show depletions in Nb and Ta. The sillimanite-bearing 2002 lava, NY-42-02, shows much lower trace element concentrations, prominent enrichments in Pb and Sr and depletion in Th. The trace element patterns of the lavas are very different from the basement rocks implying lack of continental crustal contamination.

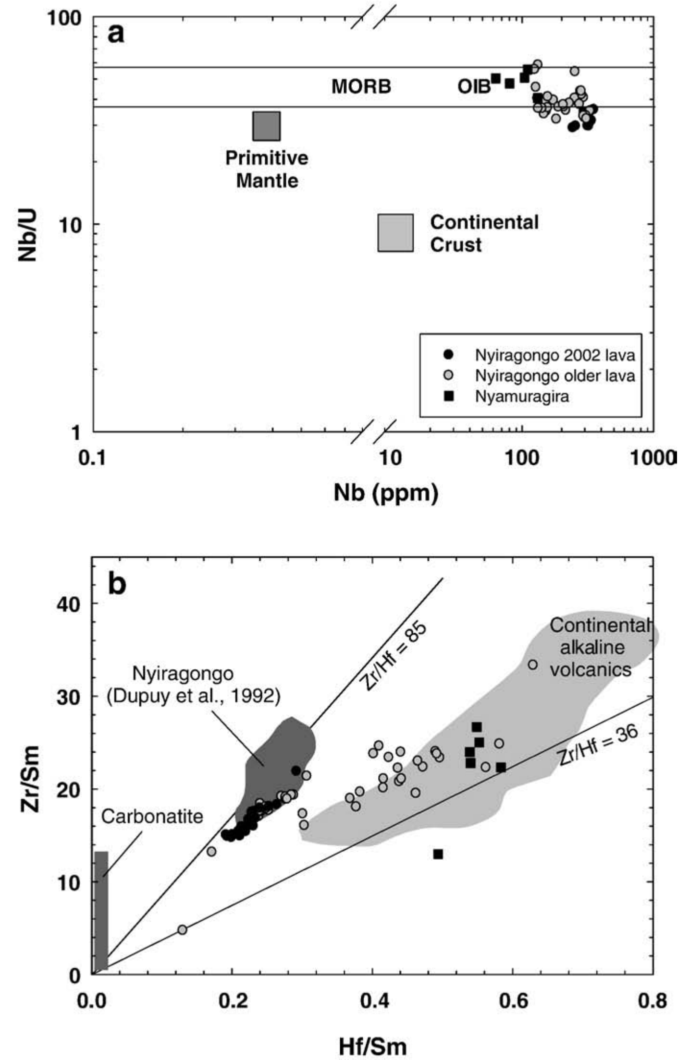

Fig. 4. (a) A plot of Nb/U versus Nb (Hofmann et al., 1986). Both the Nyiragongo and Nyamuragira volcanics show mantle-like Nb/U that is much higher than continental crustal values suggesting the absence of granitic crustal component in the magma source or as a contaminant. (b) Zr/Sm versus Hf/Sm (Dupuy et al., 1992) in the Nyiragongo and Nyamuragira lavas. Nyiragongo volcanics show very high Zr/Hf that is much higher than those of the Nyamuragira rocks. Zr/Hf ratios of Nyiragongo are among the highest observed in both continental and oceanic alkali basalts. High Zr/Hf indicates presence of carbonates in the mantle source of Nyiragongo.

The primitive mantle-normalized (Sun and McDonough, 1989) "spider" plots for a wide range of compatible and incompatible elements are shown in Fig. 3(d–f). The high concentrations of the large ion lithophile elements (LILE) and the high field strength elements (HFSE) without anomalous Nb and Ta are noteworthy in these plots (Fig. 3d–f). The Nyiragongo volcanics show prominent depletion in Pb relative to the adjoining REEs and a less prominent depletion in Hf (Fig. 3d, e). In contrast, the Nyamuragira volcanics (Fig. 3f) do not show any depletion in Hf although a small negative Pb-anomaly is also observed for these rocks.

The 2002 lava sample, NY-42-02, is clearly different from the others with low REE, enrichments in Ce and Eu (Fig. 3a), prominent enrichments in Pb and Sr and depletion in Th (Fig. 3d). This sample has abundant sillimanite crystals unlike any other lava sample of this study. Such sillimanite-bearing rocks occur as isolated patches within the 2002 lava flow. Two quartzite samples (NY-59, 61), possibly fragments of the basement beneath Virunga, have much lower REE concentrations (Fig. 3c), with prominent Nb, Ta depletions (Fig. 3f).

Nyiragongo volcanics show high Ce/Pb ratios (average 64 for the 2002 flow, 53 for the older lavas) (Table 2) that are much higher than average continental crustal rocks and mantle-derived oceanic basalts (Hofmann et al., 1986; Sims and DePaolo, 1997). In contrast, the Nyamuragira volcanics show lower Ce/Pb values with an average of 21 similar to oceanic volcanic rocks. Nb/U ratios for most of the Nyiragongo volcanics are similar to mantle values of 47 + 10 (Hofmann et al., 1986; Sims and DePaolo, 1997) as observed in oceanic basalts (Fig. 4a). However, along with a few older lavas of Nyiragongo, the Nyamuragira volcanics show higher-than mantle Nb/U (48–60).

The Nyiragongo volcanics are characterized by high U/Pb (highest 1.7), much higher than Nyamuragira (highest 0.3). Th/U is higher in Nyamuragira (average 5.7) compared to both the Nyiragongo 2002 flow (average 2.62) and the older lavas (average 2.97). K/Rb ratios (Table 1) of the Nyiragongo (231–356) and Nyamuragira (253) lavas are low. A characteristic feature of the Nyiragongo volcanics is their superchondritic (>36.6 + 2.9) (Jochum et al., 1986) Zr/Hf, as high as 79 for the 2002 lava and 77 for the older flows that are distinctly higher than those of the Nyamuragira volcanics (average Zr/Hf = 44) (Fig. 4b). REE/HFSE e.g. Eu/Ti is also high (~4.0) especially in the 2002 Nyiragongo lava. The latter relationship is consistent with the general observations on the fluid-mobile character of the lighter REE compared with the less mobile HFSE in magmatic fluids.

3.3. Nd, Sr, Pb isotopic composition

Nd-isotopic compositions of the Nyiragongo volcanics do not vary appreciably (Fig. 5a) and cluster around bulk earth values whereas the Nyamuragira lavas show more enriched compositions with comparatively radiogenic Sr and unradiogenic Nd. The sillimanite-bearing 2002 lava sample, NY-42-02, has more radiogenic Sr and unradiogenic Nd (Table 3) unlike any other Nyiragongo lava sample and plots well off scale in Fig. 5. Measured $^{87}Sr/^{86}Sr$ ratios for the quartzite basement rocks, are 0.72422 and 0.73391. Present day $\varepsilon_{Nd(0)}$ for one of these quartzites (NY-61) is −18.2 (Table 3).

Pb isotopic compositions of the samples of this study are shown in Table 3. The Nyiragongo lavas, especially the 2002 samples, show a broader range of $^{207}Pb/^{204}Pb$ for a small range in $^{206}Pb/^{204}Pb$, resulting in a vertical trend in Fig. 5b. This variation in $^{207}Pb/^{204}Pb$, observed in samples from a single lava flow of Nyiragongo is noteworthy. The sillimanite-bearing lava sample, NY-42-02, with distinct trace element concentration patterns including enriched U and Pb concentrations (Fig. 3d) has the highest $^{207}Pb/^{204}Pb$ (15.769) among all the Nyiragongo samples (Fig. 5b). Overall, the Pb-isotopic compositions of the Nyiragongo volcanics, both old and young, overlap with those of ocean island basalts (OIB) (Fig. 5b). The Nyamuragira volcanics have distinctly higher $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ but lower $^{206}Pb/^{204}Pb$ ratios compared to Nyiragongo and overlap with the compositions of other Virunga volcanics (e.g. Rogers et al., 1992, 1998) (Fig. 5b). The Nyamuragira volcanics also show higher $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ compared to the Nyiragongo volcanics, among which the 2002 lava samples show relatively lower values than the older lavas (Fig. 6). The basement rocks (NY-59, 61) have very radiogenic present day Pb-isotopic composition as shown in Table 3 and they plot far outside the field shown in Fig. 5b.

4. Discussion

Despite their close proximity, the Nyiragongo and Nyamuragira volcanics show distinctly different geochemical and isotopic compositions as documented above. It is evident that these two volcanoes have very different pedigrees, in terms of their source compositions and/or depth of origin, which will be discussed in the following sections by comparing other volcanoes from the VVP and those from around the Tanzanian craton, particularly the Kenya rift (Fig. 1a).

The low-viscosity Nyiragongo lavas have extreme silica-undersaturated compositions (Fig. 2) that are not seen in any other terrestrial silicic

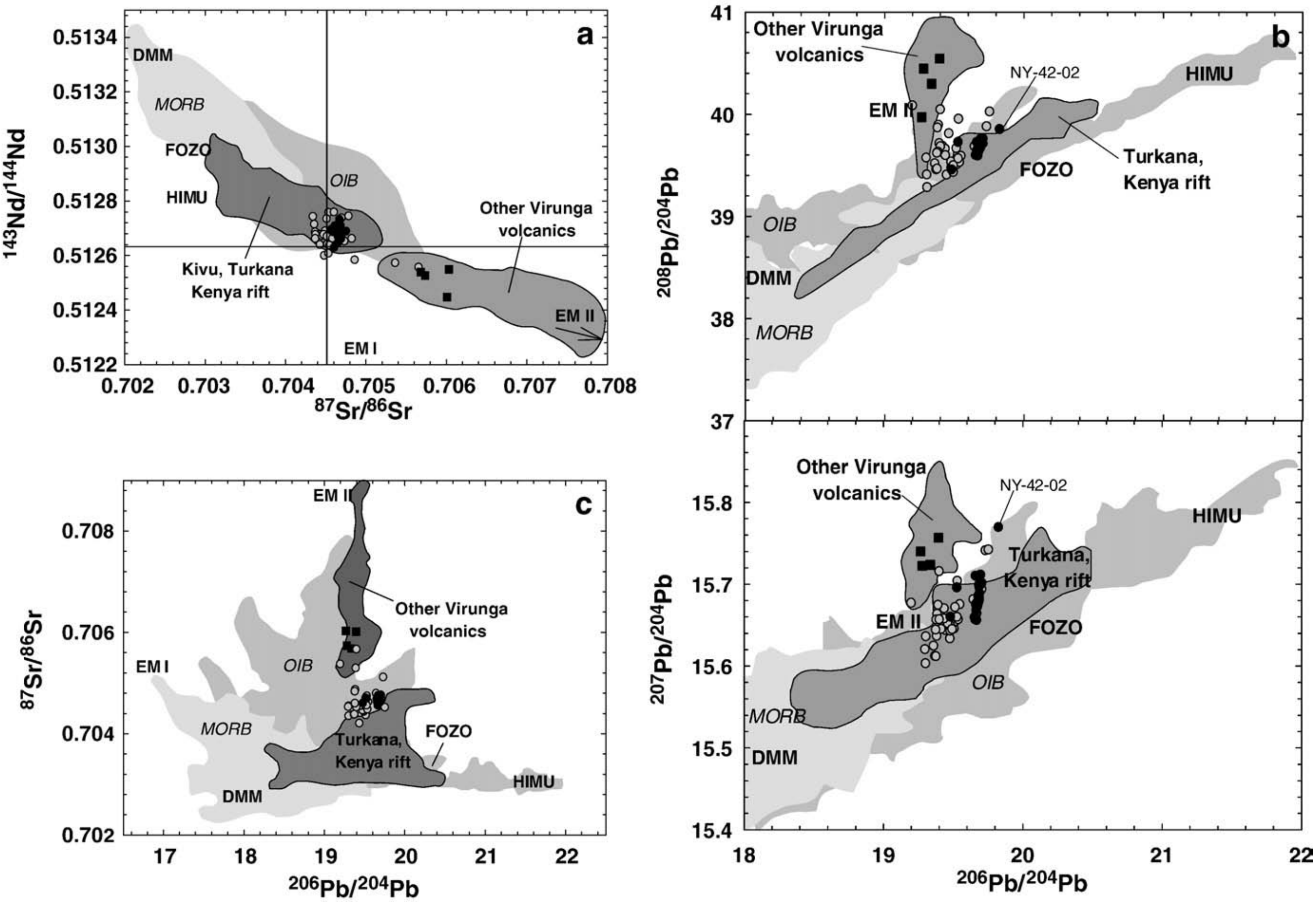

Fig. 5. Nd–Sr–Pb isotopic composition of the Nyiragongo (circles) and Nyamuragira (filled squares) volcanics of the present study. Also shown are the domains of oceanic basalts, mantle reservoirs, other Virunga volcanics (Vollmer and Norry, 1983a; Rogers et al., 1992, 1998) as well as the Kivu (Furman and Graham, 1999), Turkana (Furman et al., 2004) and Kenya rift (Rogers et al., 2000) volcanics of EARS located around the Tanzanian craton. (a) Nyiragongo volcanics show bulk-earth-like Nd–Sr isotopic compositions whereas the Nyamuragira volcanics and a few older Nyiragongo lavas overlap with the compositions of other Virunga volcanics showing more radiogenic Sr and less radiogenic Nd. The sillimanite bearing lava sample, NY-42-02 and the basement rock samples show very different compositions with more radiogenic Sr and less radiogenic Nd (Table 3) and plot outside the field of this diagram. (b) Pb isotopic compositions of the volcanics of this study. The 2002 Nyiragongo lava plots in a narrow slightly vertical trend in the $^{207}Pb/^{204}Pb$–$^{206}Pb/^{204}Pb$ plot. The sillimanite-bearing sample, NY-42-02 shows the highest $^{207}Pb/^{204}Pb$. The older Nyiragongo samples have slightly lower $^{206}Pb/^{204}Pb$ compared to the 2002 lavas, and show greater variations in $^{207}Pb/^{204}Pb$. (c) A plot of $^{87}Sr/^{86}Sr$ versus $^{206}Pb/^{204}Pb$ of the Nyiragongo and Nyamuragira volcanics of the present study. The contribution of the EM II component to the source of the Virunga lavas, including Nyamuragira and a few Nyiragongo samples may be indicated from this diagram (see text for discussion).

volcanics. Another distinctive chemical feature is the high K_2O (5.5 wt.%) (Fig. 2) and unusually high Na_2O, as high as 6 wt.%. While the 2002 lavas do not show appreciable variation in alkalis, the older lavas of Nyiragongo show variable bulk rock chemistry, plotting generally at lower total alkalis but grading into the 2002 lava composition. All these lavas are dominated by feldspathoidal mineralogy. The compositional variability in the older Nyiragongo lavas is not due to variable contribution of the feldspathoidal minerals since these lavas are very fine-grained with micro-phenocrysts in a glassy matrix and large quantities of rock samples were crushed to obtain a compositionally representative powder for analyses. This observation suggests that the older lavas may have evolved from or may have similar genetic relations with lava compositions of the 2002 flow. It is interesting to note that the bulk rock compositions of the 2002 Nyiragongo lava are close to those of the 1977 and 1982 eruptions (Santo et al., 2002).

The unusual chemical compositions of these highly potassic and sodic ($K_2O/Na_2O \sim 1$) lavas are difficult to explain by derivation from a normal peridotitic mantle. Although phlogopite is often suggested as a source of high K in mantle derived lavas, the ultimate source and mechanism of K-metasomatism in the mantle is enigmatic. The highly sodic composition of the Nyiragongo volcanics suggest the presence of sodic-pyroxene, jadeite-rich diopside (omphacitic) and/or perovskite in their mantle source, which are the Na-bearing mineral phases commonly known for greater mantle depths.

Nyamuragira lavas show major elemental compositions similar to other volcanics from Virunga and those from Kivu, Rungwe, Kenya rift, and Turkana (Fig. 2) (Vollmer and Norry, 1983a; Rogers et al., 1992; Furman, 1995; Paslick et al., 1996; Rogers et al., 1998; Furman and Graham, 1999; Rogers et al., 2000; Furman et al., 2004; Furman et al., 2006; Furman, 2007). These contemporaneous volcanics, in structural proximity to the Tanzanian craton (Fig. 1a), show much lower alkali contents and range from basalts and picro-basalts to basaltic trachyandesites (Fig. 2). The only exceptions are the Sabinyo volcanics from Virunga (Fig. 1b), which are silica-saturated and show evidence of lower crustal contamination (Holmes and Harwood, 1937; Vollmer and Norry, 1983a; Rogers et al., 1998) in contrast to the other above mentioned-volcanics, none of which show evidence of crustal contamination.

Experimental petrological data suggest that relatively high degrees of partial melting (20–30%) of a phlogopite–clinopyroxenite mantle source with minor amounts of titanomagnetite, sphene and apatite can produce compositions of highly potassic mafic lavas with K_2O in the range of 3.07–5.05 wt.% and SiO_2 varying between 35 and 39.2 wt.% (Lloyd et al., 1985). However, these high degree experimental melts have Mg# ranging from 59 to 62, which is much higher than those observed in Nyiragongo (average 29.4, Table 1). Even the rare olivine-bearing Nyiragongo lava samples also show low Mg# indicating that olivine-fractionation is not the cause of the low MgO content in these lavas.

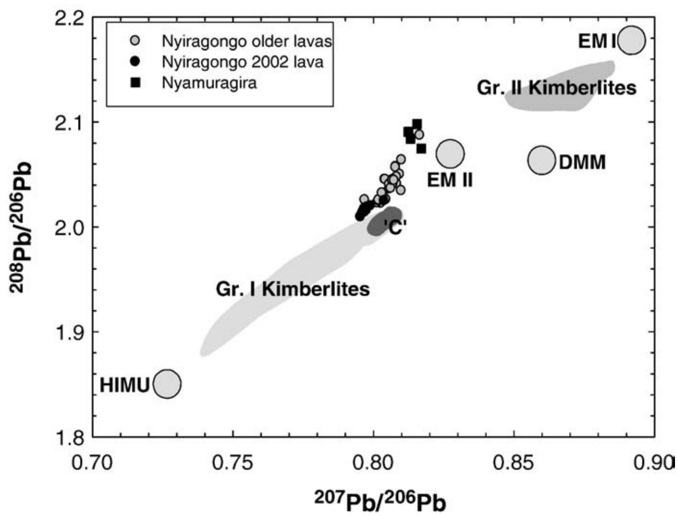

Fig. 6. $^{208}Pb/^{206}Pb$ versus $^{207}Pb/^{206}Pb$ plot of the samples of the present study. The mantle reservoirs EM I, EM II, DMM and HIMU are also plotted as the four end-members of the mantle tetrahedron after Hart et al. (1992). Also shown are the fields of Group I and Group II kimberlites (Collerson et al., in preparation) and a common component 'C' as derived from the Pacific, Atlantic and Indian MORB data by Hanan and Graham (1996). The 2002 Nyiragongo lavas are similar to the Group I kimberlite field and all samples considered together, the Virunga volcanics, particularly Nyamuragira, of this study show a trend close to the EM II component. Note the correspondence of the 2002 Nyiragongo lavas with the field for 'C' (see text for details).

Large degree melting of a mantle source cannot produce the fractionated La_N/Yb_N of the Nyiragongo lavas (Fig. 3), unless this mantle source is already metasomatized with the observed highly fractionated LREE pattern. Experimental petrological data for the magmatic evolution of highly potassic lava are sparse and not available for silica-undersaturated lavas like Nyiragongo, which are equally enriched in K and Na. The thermodynamic properties of multi-component systems involving nepheline–kalsilite–leucite were developed only recently to predict crystallization sequences, involving nepheline–kalsilite solutions (Sack and Ghiorso, 1998).

Based on the major elemental compositions of these volcanics, a bimodal character emerges between the foiditic Nyiragongo lavas and the Virunga basanites and their derivative lavas. This bimodality is also observed in the Sr, Nd and Pb isotopic compositions of these volcanics (Fig. 5) where the Nyiragongo lavas form a cluster while the other Virunga lavas show distinct linear trends suggesting assimilation and fractional crystallization processes (AFC). These observations suggest that the foidite lavas of Nyiragongo are of an independent origin than the basanites of Virunga.

The above conclusion is in direct contrast with recent inferences of Platz et al. (2004), who suggested a common line of liquid descent by fractional crystallization for Nyiragongo and the associated Virunga lavas from a single parental liquid. This inference of Platz et al. (2004) in our opinion, is improbable, mainly because crystallizing the undersaturated phenocrystal phases, such as Mg-rich olivine, melilites and feldspathoids, in a common basanitic or nephelinitic parental magma, cannot yield any residual liquid that is lower in silica and simultaneously enriched in the alkalis, as observed for the Nyiragongo lavas. Thus, the Nyiragongo lava compositions such as those reported for the 2002 eruption, must represent a "primary" mantle-derived liquid that is unique among terrestrial silicic lavas.

4.1. Issue of crustal contamination

The Virunga volcanics are underlain by Precambrian granites and sediments of the Karagwe–Ankolean system which were deposited ~2100 Ma ago. These sediments were deformed and metamorphosed during the Kibaran Orogeny (1300–800 Ma), but were not affected by the later Pan-African events (700–450 Ma) (Cahen, 1970). The extreme enrichment of the LREEs in the Nyiragongo volcanics (100–1000x chondrite), especially in the 2002 lava, precludes any significant continental crustal contamination since average continental crust is characterized by much lower LREE concentrations and distinctly different REE patterns (Taylor and McLennan, 1985). This conclusion is also supported by the very low SiO_2 (~36.6 wt.%), and very high concentrations of other incompatible trace elements such as Sr (~2000 ppm), Ba (~2200 ppm), Zr (~300 ppm), in the Nyiragongo lavas. Lack of any negative Nb and Ta anomaly (Fig. 3d) along with higher than mantle Ce/Pb (Table 2) and mantle-like Nb/U (Fig. 4a), which are much higher than continental crustal values, also suggests the absence of any granitic crustal component in the magma source of Nyiragongo. Trace element concentrations are also high in the Nyamuragira volcanics (e.g. average Sr=781 ppm; Ba=863 ppm), although lower compared to Nyiragongo. The lack of any depletion in Nb and Ta as well as high Ce/Pb and Nb/U also rules out any significant contribution of continental crust in the genesis of the Nyamuragira volcanics.

The sillimanite-bearing 2002 Nyiragongo lava sample NY-42-02, however, has considerably different trace element compositions (Fig. 3a,d) with generally lower concentrations, prominent depletion in Th and enrichments in Sr and Pb. When considered together with its irregular occurrence as pockets in the lava flow, unusual sillimanite-bearing mineralogy, unradiogenic Nd, radiogenic Sr and highly radiogenic Pb-isotopic compositions, as discussed in the following sections, it is clear that this lava sample crystallized sillimanite by local crustal assimilation, possibly when the hot lava erupted on the surface. In this context it is important to note the observations of Sahama (1978) regarding crustal xenoliths found either as ejected blocks or as inclusions in the Nyiragongo lavas representing samples of the underlying basement rocks, which are almost invariably granitic in composition, in their various stages of fusion. These fused granite xenoliths were found by Sahama to be common in the second intracraterial lava platform located 180 m below the first lava platform from the top of the Nyiragongo crater. In addition, Sahama observed that when the granite xenoliths locally dissolved almost entirely in the lava, the sillimanite masses remained as the last remnants of the fused granite (Sahama, 1978). We consider the NY-42-02 sample to be one such last remnant of a fused granite.

4.2. Depth of melting

K/Rb ratios of the Nyiragongo and Nyamuragira volcanics are low (231–356, Table 1) and similar to those reported by Bell and Doyle (1971) for several East African Rift volcanics including Nyiragongo. K/Rb ratio of phlogopite is usually less than 250 but ranges from 40 to 400 (Beswick, 1976; Basu, 1978). In contrast, K/Rb ratio of amphiboles and melts derived from amphibole-bearing sources is much higher, usually greater than 1100 (Hart and Aldrich, 1967; Basu and Murthy, 1977; Basu, 1978). Hence, low K/Rb in Nyiragongo and Nyamuragira rules out melting of an amphibole-bearing source and is consistent with the melting of a phlogopite-bearing source assemblage. The presence of phlogopite in the magma source has important implications for the depth of origin of the magma (Green and Falloon, 1998) (Fig. 7). Phlogopite is a stable phase in the upper mantle up to depths of ~150 km (Modreski and Boettcher, 1970; Foley, 1993) with temperature being the ultimate limiting factor for the stability of phlogopite in the mantle (e.g. Class and Goldstein, 1997). However, fluorine-rich phlogopites are stable at much higher temperatures (~1260^{0}C) than normal phlogopite (Dooley and Patiño Douce, 1996; Motoyoshi and Hensen, 2001). The possible occurrence of fluorine-rich phlogopite in the source of the Nyiragongo lavas is supported by the high fluorine-content in gas vents and groundwater around the Nyiragongo volcanic complex (Vaselli et al., 2007).

Average Dy_N/Yb_N values for the Nyiragongo and Nyamuragira volcanics vary from ~1.74 in Nyiragongo to 1.54 in Nyamuragira (Table 2). These high Dy_N/Yb_N ratios as well as the highly fractionated LREE

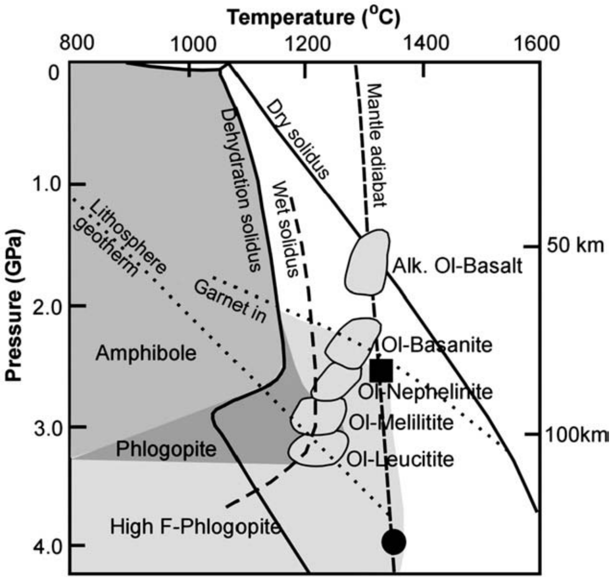

Fig. 7. Pressure versus Temperature diagram modified after Green and Falloon (1998) showing stability fields of amphibole, phlogopite and F-rich phlogopite in a peridotitic upper mantle composition. Simultaneous, compositionally different volcanism in the spatially adjacent Nyiragongo and Nyamuragira volcanoes can be explained by magmas being derived from greater depths in Nyiragongo (filled circle) compared to Nyamuragira (filled square).

relative to HREE suggest low degree partial melting with residual garnet. The comparatively lower Dy_N/Yb_N in the Nyamuragira lavas could be due to larger degree melting of the mantle source. Other geochemical proxies for the presence of residual garnet include Th/U, which is low in garnet (Beattie, 1993; LaTourrette et al., 1993) and hence, partial melts with residual garnet are expected to have high Th/U ratios, much greater than the present-day depleted mantle (DM) value of 2.6 (Kramers and Tolstikhin, 1997). Th/U ratios vary from 2.2 to 3.2 for the younger lavas, from 2.1 to 5.3 for the older Nyiragongo lavas and from 5.2 to 5.9 for Nyamuragira, which is consistent with the presence of residual garnet in the source of these rocks. These observations may suggest an eclogite or a garnet peridotite or both in the mantle source of these lavas. Since garnet is not a stable phase in the mantle at depths shallower than 80 km, presence of residual garnet in the source implies origin from depths greater than 80 km for these lavas. Thus, the required presence of both phlogopite and garnet in the source of the Nyiragongo and Nyamuragira lavas suggests that the depth of melting must be between approximately 80 and 150 km.

4.3. Role of CO_2 and metasomatism of the source

Fumaroles in and around the Nyiragongo summit crater have very high $CO_2/^3He$ ratios, about 10 times higher than those observed in MORB (Tedesco et al., 2007). This high ratio is due to the greater solubility of CO_2 in ultrabasic lavas, like Nyiragongo, compared to more silica saturated basalts although the solubility of He maybe comparable in feldspathoid-bearing ultrabasic lavas and basalts. Ground-based UV and IR spectroscopic measurements of volatile flux from the Nyiragongo volcano (Sawyer et al., 2008) from May, 2005 to June, 2006 have confirmed that Nyiragongo is a strong source of CO_2 to the atmosphere compared to other passively degassing volcanoes on Earth. In addition, these measurements show little temporal variations in proportions of CO_2, SO_2 and CO during a 24 month observation period. Similarly, the amount of CO_2 currently emanating from Nyamuragira, if sustainable over a year, would be comparable to the amount of CO_2 presently emanating from global mid-oceanic ridge volcanism in a year (Tedesco et al., 2007). What is the source of the high CO_2 in these lavas that is not observed in other volcanoes of the Virunga province? A limited number of Nyiragongo lava samples were analyzed for CO_2 by Sahama (1978) that allow potentially high $CaCO_3$ (5.7–11.9 wt.%) in the groundmass of these lavas. Such high calcite content results from magmatic enrichment in the carbonate which was also confirmed by the presence of modal calcite in the groundmass of some Nyiragongo lavas as noted by Sahama (1978). A few of our Nyiragongo samples show as high as 1.5% LOI (Table 1) which could also represent micro-crystalline carbonate given that no other hydrous mineral phases were recognized.

A diagnostic feature of the Nyiragongo volcanics is their superchondritic Zr/Hf (Jochum et al., 1986), as high as 78.6, that are much higher than those of the Nyamuragira lavas (average 43.6) or other global continental and oceanic alkali basalts (Dupuy et al., 1992) (Fig. 4b). Zr and Hf are both incompatible HFSE with nearly identical ionic radii and charge. During partial melting of the mantle, these elements are not expected to fractionate from one another. Experimental studies indicate that in garnet and clinopyroxene, the distribution coefficient of Zr is less than that of Hf ($D_{Zr}<D_{Hf}$) (Hamilton et al., 1989). However, 1–10% partial melting of a garnet–clinopyroxene bearing mantle source assemblage with primitive Zr/Hf of 36 can result in Zr/Hf values ranging from 40 to 48 (Dupuy et al., 1992) which are similar to the ratios observed in Nyamuragira but are much lower than those in Nyiragongo. It is noteworthy that carbonatites from continents as well as from oceanic settings have highly fractionated Zr/Hf ratios (>70) (Gerlach et al., 1988; Nelson et al., 1988; Andrade et al., 2002). Experimental studies indicate that in a carbonate–silicate melt pair, $D_{Zr}<D_{Hf}$ in the silicate melt (Hamilton et al., 1989). Hence, the carbonate melt in equilibrium with the silicate melt has higher concentrations of Zr relative to Hf resulting in high Zr/Hf ratios in the carbonatites. The influence of carbonate-rich melts in the peridotitic mantle source can explain the high Zr/Hf ratios observed in Nyiragongo lavas. High Zr/Hf in carbonate-metasomatized peridotite xenoliths from the Olmani cinder cone in northern Tanzania have been similarly attributed to such processes (Rudnick et al., 1993). The Nyiragongo volcanics, in particular the 2002 lavas show high Eu/Ti as high as 4.1. Similar observations have also been made by Platz et al. (2004) on these lavas. High Eu/Ti is also a characteristic signature of carbonate metasomatism (Rudnick et al., 1993; Furman and Graham, 1999). U–Th–Ra series disequilibria data on the older Nyiragongo lavas (Williams and Gill, 1992) also suggest metasomatism in the source of these lavas.

What is the source of carbonates in the mantle? Experimental petrological studies have shown that carbonate minerals remain stable in anhydrous or slightly hydrous carbonated eclogites to temperatures >1100 °C at 5 GPa and >1200 °C at higher pressures of 9 GPa (Dasgupta et al., 2004). Thus, carbonate minerals are expected to survive deep subduction considering even the hottest subduction geotherm, higher than the continental geotherm (Dasgupta et al., 2004). Near-solidus partial melts of carbonated eclogites are also extremely enriched in Na_2O, which could explain the unusually high Na-content of Nyiragongo. Therefore, it is possible that a phlogopite-bearing carbonate-metasomatized eclogite–peridotite source assemblage is a viable source for the Nyiragongo volcanics that could explain the high Zr/Hf and high alkali content ($Na_2O=K_2O=6$ wt.%) in these rocks. However, contribution from the subducted eclogitic component, which would have higher SiO_2 than the silica-undersaturated Nyiragongo volcanics, must have been minimal during melting to generate the Nyiragongo lavas. In addition, this above scenario must take into account the Nd, Sr and Pb isotopic constraints that do not allow any recycled crustal component in the source of the Nyiragongo lavas, as discussed below.

4.4. Nd–Sr–Pb and He-isotopic constraints

Nd–Sr isotopic compositions of the Nyiragongo volcanics, especially the 2002 lava, cluster around the "bulk earth" values (Fig. 5a). Most of the geochemical data of older Nyiragongo lava of this study and the 1972 crater-lake sample (Vollmer and Norry, 1983a) also overlap with the

2002 flow. Thus on a first order of approximation, the Nyiragongo crater lava has remained uniform isotopically, and perhaps chemically, over the last 30 years.

The Nyamuragira samples, along with a couple of older Nyiragongo lavas plot distinctly below and to the right of bulk Earth values, and overlap with those of other Virunga volcanics from Muhavura, Gahinga (Rogers et al., 1998) and Karisimbi (Rogers et al., 1992) (Fig. 5a). The silica-saturated Sabinyo volcanics (Vollmer and Norry, 1983a; Rogers et al., 1998), also from Virunga (Fig. 1b), show the most radiogenic Sr-isotopic compositions among the Virunga lavas (Fig. 5a) and have been interpreted as derivatives of mantle-derived melts mixing with a lower crustal component (Rogers et al., 1998). However, no crustal component has been envisaged in the genesis of any of the other Virunga volcanics based on combined trace element and isotopic characteristics (Rogers et al., 1998). As discussed in the earlier sections, the Nyamuragira and Nyiragongo lavas also do not show any sign of continental crustal contamination in their major and trace element characteristics. We argue that the position of the Nyamuragira and other Virunga volcanics in the so-called enriched field in the Nd–Sr isotopic space, in the lower right hand quadrant of Fig. 5a, indicates contributions from an enriched mantle, such as an EM II-like (Zindler and Hart, 1986; Workman et al., 2004) source. The only Nyiragongo sample showing isotopic signatures of crustal contamination is NY-42-02 (Table 3) consistent with its distinctive and unique sillimanite-bearing mineralogy and trace element characteristics (Fig. 3a, d). It is interesting to note that Nd–Sr isotopic composition of the Nyiragongo volcanics is similar to those of global ocean island basalts (Fig. 5a). Unlike most other Virunga volcanics, lavas from the Kivu region (Furman and Graham, 1999), the Turkana depression (Furman et al., 2004) and those from the Kenya rift (Rogers et al., 2000) (Fig. 1a) show more radiogenic Nd and less radiogenic Sr, and overlap with the domains of global MORB and OIB.

What does the bulk earth-like Nd, Sr isotopic composition of the Nyiragongo lavas imply? Are these indicative of a primitive component in the source of these rocks or do they represent a mixing between EM II and depleted MORB mantle? Bulk earth-like Nd, Sr isotopic compositions are also observed in xenoliths derived from some portions of sub-continental lithospheric mantle (SCLM) (Erlank et al., 1987; Menzies et al., 1987). However, the role of the SCLM can be ruled out based on He-isotopic constraints. Along the EARS, a high ^{3}He component (R/Ra ~ 20) is restricted only to the north, consistent with the occurrence of the Afar plume (Pik et al., 2006). Towards the south, the volcanics are characterized by much lower $^3He/^4He$ (R/Ra ~ 7 ± 2). Olivines and pyroxenes from Nyiragongo have $^3He/^4He$ ranging from 7.1 to 8.5 (R/Ra) (Pik et al., 2006) that overlap with the He-isotopic composition of MORB. Global peridotites, representing the SCLM, have a well defined average $^3He/^4He$ of 6.1 ± 0.9 (R/Ra) (Gautheron and Moreira, 2002), which is distinctly lower than the $^3He/^4He$ found in MORB as well as those observed in Nyiragongo (Pik et al., 2006), indicating a sub-lithospheric origin for the Nyiragongo lavas. Other He-isotopic data (Porcelli et al., 1986) from mantle xenoliths (both mineral separates and whole-rock), including those from kimberlites in Eastern Africa, comply mostly with the recent data of Gautheron and Moreira (2002) except for a few less radiogenic samples. It is important to note here that Gautheron and Moreira (2002) analyzed only olivine separates (low U, Th) by sample crushing to avoid contamination of diffused He from the matrix unlike whole-rock analysis of Porcelli et al. (1986) which might be less accurate.

SCLM as a mantle reservoir is isolated from the convecting mantle and is sampled as mantle xenoliths hosted by alkali basalts, kimberlites and related lavas. The SCLM is enriched in incompatible trace elements compared to the primitive mantle but is not a significant reservoir of these elements (McDonough, 1990). There is also a regional variation in the trace element composition of the SCLM (Saunders et al., 1992; Bedini and Bodinier, 1999; Gregoire et al., 2003; Jourdan et al., 2007). Differences also exist between sheared mantle xenoliths, which show relatively higher trace element concentrations than granular xenoliths. Sheared xenoliths from the East African rift show pronounced negative Nb, Ta anomalies (Bedini and Bodinier, 1999) whereas most SCLM-derived xenoliths do not show any Nb, Ta anomalies (McDonough, 1990). Clinopyroxenes from ultramafic xenoliths in the Kaapvaal craton also show negative Nb, Ta anomalies (Gregoire et al., 2003). Overall trace element patterns of the SCLM are comparable to ocean island basalts although the absolute abundances of these elements are much lower. SCLM components are envisaged by some as the end-member EM II component in ocean island basalts (Jackson et al., 2007).

Overall trace element abundances in the Nyiragongo lavas are ~ 100 times higher than those observed in mantle xenoliths derived from the SCLM. Although, it might be argued that low degree partial melting of a metasomatized SCLM source can produce the high trace element concentrations in the Nyiragongo lavas, it cannot explain why Nyiragongo lavas have distinctly higher Ce/Pb (average 63 for the 2002 lavas, Table 2) compared to mantle xenoliths e.g. from the Kaapvaal craton (average 12.6) (Gregoire et al., 2003) Moreover, we cannot generate the undersaturated Nyiragongo lavas with as low as 36.6 wt.% SiO_2 by partially melting the SCLM which typically has 40–45 wt.% SiO_2 (McDonough, 1990; Gregoire et al., 2003). In addition Primitive Mantle normalized Th/Nb ratios of the Nyiragongo volcanics are less than unity indicating the lack of a SCLM component in their source, which shows Th/Nb greater than one (Saunders et al., 1992; Gregoire et al., 2003).

While the role of an SCLM can be ruled out from the above arguments involving He-isotopes and some major and trace elements, some questions can still be raised concerning the role of a plume-underplated SCLM source as a component in the Nyiragongo lavas. However, the Nd and Sr-isotopic constraints of Nyiragongo lavas as presented above would require such an underplating to be a very recent event, consistent with our suggestion of the involvement of a mantle plume in Virunga volcanism as discussed in a later section.

It may be argued that the source of the strong alkali enrichment in Nyiragongo lavas (Na_2O = 6.0 wt.% and K_2O = 5.7 wt.%, Table 1) are from a recycled continental crustal component as in SCLM. However, the various isotope systematics, the high trace element concentrations and elemental ratios, such as Nb/U, Ce/Pb, Zr/Sm and Hf/Sm of the Nyiragongo lavas do not allow the involvement of any recycled continental crustal component or recycled eclogitized basalts, due to the low SiO_2, in the source of these volcanics. Therefore, "pristine mantle minerals" like jadeite ($NaAlSi_2O_6$)-rich clinopyroxene, perovskite and K-rich phlogopite and/or hollandite are the likely candidates for the observed alkali enrichment in the Nyiragongo lava source.

The Pb-isotopic compositions of the Virunga lavas are more complex (Fig. 5b). Nyamuragira lavas show $^{207}Pb/^{204}Pb$ higher than oceanic basalts and overlap with those of other Virunga volcanics from Bifumbira, Sabinyo, Karisimbi etc. (Rogers et al., 1992, 1998). Nyiragongo lavas overlap with ocean island basalts and can be interpreted by mixtures of several mantle reservoir sources including EM II, DMM, HIMU and/or FOZO (Zindler and Hart, 1986). However, like the other Virunga volcanics mentioned above, the 2002 Nyiragongo lava plots in a narrow but discernible vertical trend in $^{207}Pb/^{204}Pb$ ratios when plotted against $^{206}Pb/^{204}Pb$ (Fig. 5b), and is almost identical with the Nyiragongo 1972 Crater Lake lava data (Vollmer and Norry, 1983b). The older Nyiragongo samples have slightly lower $^{206}Pb/^{204}Pb$ compared to the 2002 lavas, and show greater variations in $^{207}Pb/^{204}Pb$ (Fig. 5b).

The very high $^{207}Pb/^{204}Pb$ observed in the Nyamuragira volcanics suggests the role of a component with a time-integrated high U/Pb in the source of these lavas, possibly of Archean age. Evidence for the presence of subducted Archean oceanic crust below the African craton has been presented based on the oxygen isotopic composition of eclogite xenoliths from the Roberts Victor kimberlite pipe (Garlick et al., 1971; Ongley et al., 1987). A similar Archean subducted oceanic crustal component arguably may have been incorporated into the source of the Nyamuragira volcanics of the present study.

A different argument has been proposed by Rogers et al. (1992) for the Karisimbi volcanics of Virunga, which along with the Bufumbira and Sabinyo volcanics also show high $^{207}Pb/^{204}Pb$ (Fig. 5b) (Rogers et al., 1998; Vollmer and Norry, 1983a). According to these authors, the Archean component could not have been the upper continental crust as none of the volcanic rocks of their study show physical signs of continental crustal contamination or on the basis of their major and trace element compositions as discussed earlier. The same argument may hold true for the other Virunga volcanics except for the silica-enriched end-member Sabinyo (Rogers et al., 1998).

Why would the Pb-isotopic composition vary in the single 2002 Nyiragongo lava flow if the high $^{207}Pb/^{204}Pb$ is a source characteristic? In the case of the Nyiragongo lavas, we argue that the Pb-isotopic variability is possibly a surficial feature. Variable degrees of fusion of granitic basement rocks, as discussed in section 4.1, in which Pb (and Sr) was leached out in the hot lava locally, caused the observed variation in the Pb-isotopic ratios (Fig. 5b). Pb-isotopes were affected by this local process because of the fluid-mobile nature of Pb and as expected, Nd and Sr isotopes or other trace element patterns still preserved the mantle signature indicating that the effects of this fusion were not pervasive enough to resolvably affect the Nd and Sr isotopic compositions or other trace element concentrations. The sillimanite-bearing sample NY-42-02 is clearly the most affected by granite fusion. This sample not only has the highest $^{207}Pb/^{204}Pb$ among the Nyiragongo lavas, it also shows radiogenic Sr, unradiogenic Nd, and crust-like trace element patterns, which is not seen in other Nyiragongo lavas with high $^{207}Pb/^{204}Pb$.

Most of the Nyiragongo lavas, except a few older samples, show $^{208}Pb/^{204}Pb$ similar to those observed in OIB. Higher $^{208}Pb/^{204}Pb$ (Fig. 5b) is observed in some of the older Nyiragongo lavas, the Nyamuragira volcanics and in other Virunga volcanics and requires time-integrated high Th/Pb in the source. Note that the lavas with the highest $^{208}Pb/^{204}Pb$ show higher $^{207}Pb/^{204}Pb$, suggesting an ancient component in their source and radiogenic Sr and less radiogenic Nd suggesting contribution from an enriched mantle EM II-like source. This high $^{208}Pb/^{204}Pb$ in Nyamuragira could be due to an old phlogopite-bearing mantle source, with high Th/U ratios (Williams et al., 1992). The occurrence of phlogopite in the mantle source of these volcanics is consistent with the low K/Rb ratios both in the Nyamuragira and Nyiragongo lavas (Table 1). However, the relatively lower $^{208}Pb/^{204}Pb$ in Nyiragongo suggests that the phlogopite in the source of Nyiragongo formed relatively recently, possibly shortly before partial melting.

The contribution of an EM II component in the source of the Virunga lavas, especially Nyamuragira and the older Nyiragongo lavas, is more visible in a plot of $^{87}Sr/^{86}Sr$ versus $^{206}Pb/^{204}Pb$ (Fig. 5c) with possible contributions from DMM, HIMU and/or FOZO (Zindler and Hart, 1986). The standard model for the origin of the EM II reservoir involves recycling and long-term storage of a subducted oceanic crust along with terrigenous sediments (Weaver, 1991; Zindler and Hart, 1986). The recent discovery (Jackson et al., 2007) of enriched Sr and Nd isotopic signatures in dredged samples from Samoa also suggests the presence of recycled continental crustal sediments in the EM II reservoir. Alternatively, it has been proposed that this enriched mantle end-member was formed by ancient subduction and long-term storage of a metasomatized oceanic lithosphere without requiring a terrigenous sediment component (Workman et al., 2004). The contribution of an ancient oceanic lithosphere in the source of some of the Virunga volcanics, and possibly in other volcanics from the EARS, is in agreement with oxygen isotopic composition of eclogite xenoliths from the Roberts Victor kimberlite pipe of the Kapvaal craton in Southern Africa that indicated such ancient subduction beneath the craton (Garlick et al., 1971; Ongley et al., 1987). The presence of such an ancient oceanic lithosphere would also explain the observed high $^{207}Pb/^{204}Pb$ and the absence of continental crustal signatures (e.g. Nb, Ta anomaly) in the Nyamuragira and other Virunga volcanics showing similar radiogenic Pb-isotopic compositions (Fig. 5b).

We have plotted the Pb-isotopic ratios of the Nyiragongo and Nyamuragira lavas as discussed above and reported in Table 2 in Fig. 6 as $^{207}Pb/^{206}Pb$ in the abscissa and $^{208}Pb/^{206}Pb$ in the ordinate. We note the similarity of the 2002 Nyiragongo lava data with part of the Group I kimberlite field (Collerson et al., in preparation), and the data array defined by our analyzed samples of the Virunga volcanics showing a trend close to the EM II component. In particular, the Nyamuragira lavas plot close to this enriched reservoir. Note the correspondence of the 2002 Nyiragongo lavas with the field for "C," interpreted to be a common mantle source region for ocean island basalts sampled by mantle plumes (Hanan and Graham, 1996). This component "C" is similar to 'focal zone' (FOZO) of Hart et al. (1992), the high $^{3}He/^{4}He$ reservoir that are common to many hot spot mantle sources with similar Pb, Nd and Sr isotopic signatures. Thus it is remarkable that the Nyiragongo lavas are similar to oceanic lavas of supposedly a lower mantle plume origin. We note,

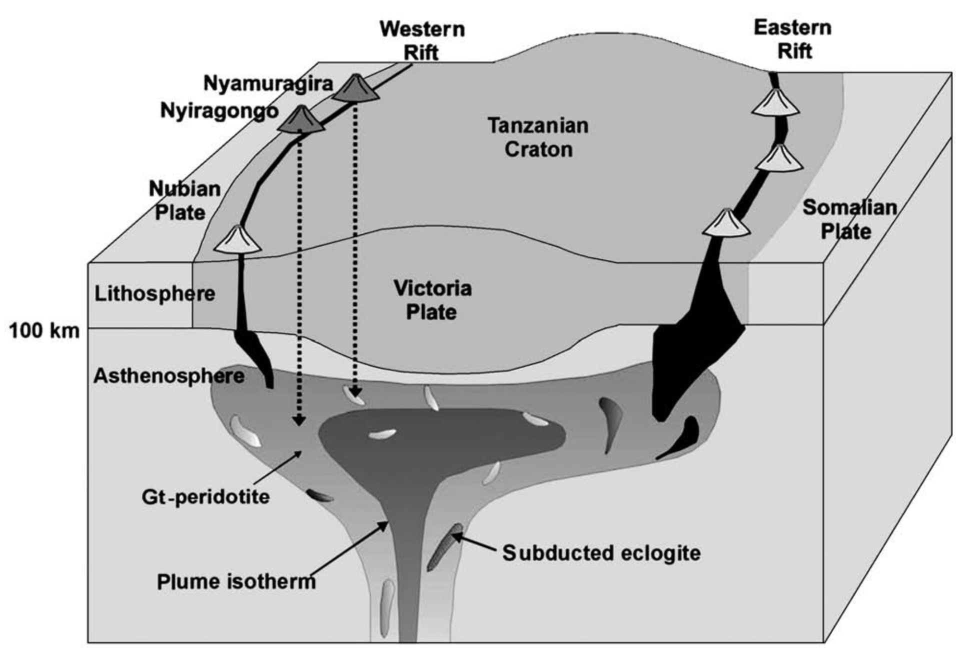

Fig. 8. Diagram showing the proposed model for the genesis of the Virunga volcanics in the west rift. Based on our geochemical data we propose that volcanism in the spatially adjacent Nyiragongo and Nyamuragira volcanoes is due to partial melting of different parts of a heterogeneous mantle plume at different depths beneath the Tanzanian craton. Relatively low degrees of melting of this heterogeneous mantle plume could explain the less voluminous volcanism in the western rift compared to the Kenya rift to the east.

however, that the Nyiragongo lavas are compositionally different from 'C' and 'FOZO' components with respect to their bulk-Earth like Sr and Nd-isotopic characteristics.

4.5. Summary of geochemical data and implications for a plume source

Any model attempting to explain the origin of the Nyiragongo lavas must take into account the silica-undersaturated, highly sodic and potassic nature of these lavas. Experimental studies suggest that silica-poor ultra-potassic lavas can be generated in an oxidizing environment with high CO_2 (Foley et al., 1986). In this context it is important to note the high groundmass $CaCO_3$ content (5.7–11.9 wt.%) in some of the Nyiragongo lavas (Sahama, 1978) and their high Zr/Hf ratios. Recent discovery (Brenker et al., 2007) of inclusions of olivine, walstromite-structured $CaSiO_3$ and Si–$CaTiO_3$ within calcite found in diamonds confirm ultradeep origin of these inclusions and along with other studies (Dasgupta et al., 2004) support ultradeep mantle carbonates at depths greater than 670 km.

Experimental petrological evidence suggests (Green and Falloon, 1998) that leucite and melilite bearing lavas, similar to those seen in Nyiragongo, are generated at greater depths compared to basanites, such as those found in other Virunga lavas including Nyamuragira (Fig. 7). Based on this evidence, we suggest that simultaneous, compositionally distinct volcanism in the spatially adjacent Nyiragongo and Nyamuragira volcanoes, with magmas being derived from greater depths in Nyiragongo compared to Nyamuragira is due to the melting of different parts of a heterogeneous mantle plume at different depths beneath the Tanzanian craton (Fig. 8). The highly sodic and potassic compositions of the Nyiragongo lavas deserve some extraordinary considerations. The sources of high sodium and potassium are usually interpreted to be the result of metasomatism via subduction-introduced fluids in the magma source. However, as discussed above the Nd, Sr and Pb-isotopes of the Nyiragongo lavas as well as their highly concentrated trace element contents and patterns cannot be explained by any recycled materials. These constraints necessitate some pristine mineralogy, possibly in the deep lower mantle where magnesite, Mg–perovskite, Ca–perovskite, CaTiSi–perovskites and hollandite are stable and consideration of low degree partial melts from these phases (e.g. Collerson et al., in preparation) may be invoked to generate the entire spectrum of trace element concentration patterns (Fig. 3) as well as the unique major oxide composition (Fig. 2, Table 1) of the Nyiragongo lavas.

This suggestion is consistent with the Nd–Sr–Pb isotopic compositions of the west rift lavas, which overlap with those of global ocean island basalts that are products of plume volcanism inferred from their geochemical composition and associated geodynamical evidence. Moreover, Nd–Sr isotopic compositions of Nyiragongo are clearly bulk-earth like which suggests a primitive component in the source of these lavas or could represent a mixing between EM II and a depleted MORB mantle; both of these two possibilities require the contribution of a plume component in these lavas. Nd–Sr–Pb isotopic compositions of Nyamuragira lavas (Fig. 5) suggest significant contributions from an EM II mantle reservoir.

4.6. Geophysical constrains on the sub-surface structure of the Western rift

It has been suggested (Ebinger and Furman, 2003; Furman et al., 2004) that the Virunga Volcanic Province is within a large area of uplift associated with anomalously hot upwelling asthenosphere. Weeraratne et al. (2003) used seismic tomographic images to infer a deep thermal perturbation up to depths of 600 km reflecting a plume stem beneath the Tanzanian craton, centered at 4°S and 34°E. However, it remains unclear whether this plume is related to the African superswell to the south (Nyblade and Robinson, 1994). Geophysical data from the western rift of the EARS are sparse. One such study involves inversion of teleseismic data (Nolet and Mueller, 1982) from 60 to 70 km north of Nyiragongo and Nyamuragira of the present study area (Fig. 1a). Based on this study, the thickness of the crust in this region has been estimated to be ~35 km. This thin high-velocity lid overlies a zone of low P and S wave velocities, which extend to depths of 140 km. A strong reflector at depths of 140 km marks the sharp transition to a high-velocity material. Beneath the eastern rift, the low velocity region extends at least up to depths of 160 km, beyond which its existence cannot be ascertained because of limited resolution. It has been suggested (Nolet and Mueller, 1982) that a slice of a mushroom-shaped diapiric intrusion has stalled below the western branch and is cooling while being exposed to the surface in the eastern branch. The key issue here is really the small-scale topography along the lithosphere–asthenosphere boundary, which cannot be resolved by the available seismic data. It is possible that the lithosphere–asthenosphere boundary beneath the east and west rift is highly variable. Alternatively, if this low velocity zone is at a constant depth throughout the Tanzanian craton, it is our suggestion that the degree and depth of melting of this heterogeneous plume is variable as shown in Fig. 8.

The above interpretation is consistent with our plume-model for the origin of the Virunga volcanics proposed here on the basis of geochemical and isotopic data presented in this study (Fig. 8). Relatively low degree melting of this compositionally heterogeneous mantle plume could explain the less voluminous volcanism in the western rift compared to the Kenya rift in the east. Future seismic studies with better resolution are required in the western rift to further validate our model.

5. Conclusions

Major and trace element as well as Nd–Sr–Pb isotopic compositions of the Nyiragongo volcanics are distinctly different from Nyamuragira, which is located only ~15 km north of Nyiragongo. These silica-undersaturated, alkali-rich Nyiragongo lavas are also geochemically distinct from the other volcanoes of the Virunga volcanic province in the western rift of the East African Rift system.

Major and trace element compositions of the Nyiragongo and Nyamuragira rocks suggest that they were formed by partial melting of a carbonate-rich, high-F phlogopite-bearing source in the presence of residual garnet but in the absence of amphibole. Trace element composition of these lavas also precludes any continental crustal contamination and suggests sub-lithospheric as well as sub-asthenospheric origin. The highly sodic and potassic compositions of the Nyiragongo lavas cannot be explained by subduction-introduced fluids and require partial melting of a source with pristine mineralogy, possibly in the deep lower mantle.

Carbonates are more important in the source of the Nyiragongo lavas compared to the Nyamuragira volcanics. This indicates that carbonate metasomatism of the mantle source was not uniform and the scale of source heterogeneity is on the order of a few kilometers.

Nd, Sr, Pb isotopic compositions of the Nyiragongo lavas overlap with those of global ocean island basalts. The bulk earth-like Nd–Sr isotopic compositions of the Nyiragongo lavas suggest a primitive component in the source of these rocks and the Pb-isotopes are similar to Type I kimberlites and Cook–Austral island basalts, for example, all requiring a plume component in the source. The Nyamuragira lavas show greater contribution from an EM II type mantle reservoir.

He-isotopic composition of the Nyiragongo volcanics are MORB-like and are distinctly higher than those characteristic of the sub-continental lithospheric mantle indicating a sub-lithospheric source for these lavas. This is in agreement with the high Ce/Pb and low SiO_2 (relative to SCLM-derived xenoliths) of Nyiragongo.

The Pb-isotopic compositions of the Nyiragongo lavas, especially high $^{207}Pb/^{204}Pb$ in the same lava flow from the 2002 eruption can be explained by Pb leaching out of the granitic xenoliths found in different degrees of fusion by the erupting lava. Even higher $^{207}Pb/^{204}Pb$ in Nyamuragira suggests the presence of an ancient subducted component in the source of these lavas. High $^{208}Pb/^{204}Pb$ ratios, are derived by melting of phlogopite, with time integrated high Th/Pb, in the mantle source.

Combined major, trace element and isotopic composition of these volcanics suggest that the Nyiragongo lavas were derived from comparatively greater depths by very low degree partial melting of a phlogopite-bearing carbonated mantle source assemblage. The Nyamuragira lavas are products of larger degree partial melting of this source from comparatively shallower depths.

Simultaneous volcanism in the spatially adjacent Nyiragongo and Nyamuragira volcanoes, with magmas being derived from greater depths in Nyiragongo compared to Nyamuragira, is due to partial melting of different parts of a heterogeneous mantle plume at different depths beneath the Tanzanian craton. We also suggest that the MORB-like He-isotopes in the Nyiragongo lavas are a result of interaction of this plume with the ambient asthenosphere. Relatively low degrees of melting of this heterogeneous mantle plume could explain the less voluminous volcanism in the western rift compared to the Kenya rift to the east.

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