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Béatrice Luais. Temporal changes in Nd isotopic composition of Piton de la Fournaise magmatism (Réunion Island, Indian Ocean). *Geochemistry, Geophysics, Geosystems*, 2004, 5, 10.1029/2002GC000502 . insu-03619226

HAL Id: insu-03619226

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Submitted on 25 Mar 2022

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Temporal changes in Nd isotopic composition of Piton de la Fournaise magmatism (Réunion Island, Indian Ocean)

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[1] A Nd isotopic study of twelve historical samples and fifty-seven core samples covering the last 530 ka of volcanic activity of the Piton de la Fournaise volcano of Réunion Island was undertaken, with additional new Sr isotopic analyses of historical samples. The core samples were obtained from the cliffs of the deep (ca.1000 m) canyons of the Remparts and Langevin Rivers. Four types of rocks, namely basalts (MgO = 5–9 wt.%), high-Mg basalts (MgO = 10–20 wt.%), plagioclase-rich basalts, and differentiated samples (SiO₂ = 48–53 wt.%) occur randomly throughout the lava sections. The overall variations in Nd isotopic ratios (0.512751 to 0.512902) are small, but significant. The range of Nd isotopic variations is larger than previously described in the literature, and extends toward lower values. Nd isotopic ratios increase through time and are broadly negatively correlated with the alkalinity index. The older alkaline Remparts samples (527–290 ka) (average $\epsilon_{Nd} = +3.26$) exhibit lower ¹⁴³Nd/¹⁴⁴Nd ratios than the younger tholeiitic Langevin samples (70–40 ka) (average $\epsilon_{Nd} = +4.17$) and the historical lavas (1901–1956) (average $\epsilon_{Nd} = +4.74$). Comparison of ¹⁴³Nd/¹⁴⁴Nd ratios with available ⁸⁷Sr/⁸⁶Sr ratios on the same set of samples (Remparts and Langevin series [Albarède *et al.*, 1997]; historical series, this study) shows no correlation between Sr and Nd isotopes. It is suggested that the high Nd isotopic ratios of historical tholeiitic samples would be representative of the isotopic composition of the Réunion plume. The increase in Nd isotopic ratios over 530 ka reflects the effect of decreasing contribution of an enriched component to the Réunion plume through time. This enriched component would occur as isolated blobs in the mantle source.

Components: 7737 words, 7 figures, 2 tables.

Keywords: Reunion Island; Nd isotopes; Indian Ocean; plume composition.

Index Terms: 1040 Geochemistry: Isotopic composition/chemistry; 1010 Geochemistry: Chemical evolution; 1025 Geochemistry: Composition of the mantle.

Received 20 December 2002; **Revised** 13 October 2003; **Accepted** 17 October 2003; **Published** 28 January 2004.

Luais, B. (2004), Temporal changes in Nd isotopic composition of Piton de la Fournaise magmatism (Réunion Island, Indian Ocean), *Geochem. Geophys. Geosyst.*, 5, Q01008, doi:10.1029/2002GC000502.

1. Introduction

[2] Temporal isotopic variations in oceanic island volcanic series have been attributed to variable interactions between a deep mantle plume and the

surrounding shallow depleted upper mantle. High-resolution isotopic evolution through time in volcanic series might thus shed light on plume dynamics as well as on the composition of the



plume, and on the heterogeneity of the deep mantle (e.g., Sr and Nd isotopic variations in Hawaiian volcanism [Chen *et al.*, 1991; Kennedy *et al.*, 1991; Lassiter *et al.*, 1996; Hauri *et al.*, 1996]). Specifically, these authors have emphasized the temporal relationship between the degree of alkalinity and the variation in Sr, Nd, and Pb isotopic ratios and conclude that alkali/tholeiitic character is not a source feature.

[3] Detailed geochemical study of Piton de la Fournaise volcano (Réunion Island) has shown an inverse evolution compared to Mauna Kea in Hawaii [Lassiter *et al.*, 1996] with older Remparts samples being more alkaline than the younger historical tholeiitic lavas, without any correlation with Sr isotopic variations [Albarède *et al.*, 1997]. The limited Sr-Nd [Newsom *et al.*, 1986; Fisk *et al.*, 1988] and He-Sr [Graham *et al.*, 1990] isotopic data as well as a detailed investigation of ³He/⁴He ratios [Hanyu *et al.*, 2001] have led to the conclusion that Piton de la Fournaise lavas originated from an isotopically homogeneous source. The aims of this study are (1) to define a high-resolution Nd isotopic record of 0.5 Ma of volcanic activity at Piton de la Fournaise, which would be potentially less sensitive to possible seawater alteration than Sr isotopes, (2) to relate the possible Nd isotopic variations to the alkaline character of the samples, and thus (3) to define mantle source characteristics beneath Piton de la Fournaise volcano. The geochemistry of the samples discussed in this paper was studied by Albarède *et al.* [1997].

2. Geological Setting and Previous Work

[4] Réunion island, located in the Indian Ocean to the East of Madagascar (Figure 1), is the youngest volcanic island generated by a major hot spot whose track extends to Mauritius, the Mascarene Plateau and finally to the Deccan Traps erupted 65 Ma ago. Volcanic activity began at least 2.1 Ma ago at Piton des Neiges volcano located to the North [McDougall, 1971], which was active until 12 ka ago [Gillot and Nativel, 1982; Deniel *et al.*, 1992]. To the South, the oldest known activity of Piton de la Fournaise is dated at 527 ka [Gillot *et*

al., 1990]. Both volcanoes were thus concurrently active for about 0.5 Ma. Erosion has carved abrupt and deep canyons exposing about 1000 m sections of the lava pile, thus providing rather complete time coverage of Piton de la Fournaise activity.

[5] Piton de la Fournaise volcano has been studied by a number of authors and the geological and petrological background is summarized by Bachèlery and Mairine [1990]. Albarède and Tamagnan [1988] and more recently, Albarède *et al.* [1997] studied the geochemical evolution of the volcano in detail and developed a model for the genesis of Piton de la Fournaise on the basis of a high-resolution stratigraphic sampling. Outcrop locations are shown in Figure 1. Samples from the oldest series have been cored in the Rivière des Remparts cliff for which the oldest rocks give a K-Ar age of 527 ka [Gillot *et al.*, 1990]. The lavas cored at 1390 m give an age of 395 ka (P. Gillot, unpublished data, 1991) whereas the top of the cliff is estimated by extrapolation to be 290 ka [Gillot *et al.*, 1990]. These samples consist mainly of basalts and hawaiites. A few plagioclase-rich flows occur mainly at the base of the series at 700–900 m elevation whereas differentiated rocks can be found in the interval from 1150 to 1200 m. Above 1300 m, only basalts with a few picrites occur. Younger samples have been cored from the Langevin cliff. Their lava flow ages range from 70 ka at the base to 40 ka at the top (P. Gillot, unpublished data, 1991). Chemical compositions evolve through time from alkaline (Remparts samples) to tholeiitic (Langevin samples) and range from low Mg basalts to Mg-rich lavas (MgO > 10%) and picrites (MgO > 20%). In these two sequences Albarède *et al.* [1997] have defined four rock types on the basis of their SiO₂ contents (44.5 to 53 wt%), FeO/MgO ratios (0.5 to 3.5), or alkalinity index (see Table 1 for definition): (1) Steady State Basalts (SSB), which are characterized by a narrow range of MgO (5–9%) and other compatible element contents. Their low Ni, Cr and MgO contents suggest that they are not primitive basalts. This results from fractionation of cpx at intermediate pressure (8–10 Kbar) and olivine at lower pressure (4–8 Kbar). Their small range of geochemical variation is attributed to buffering

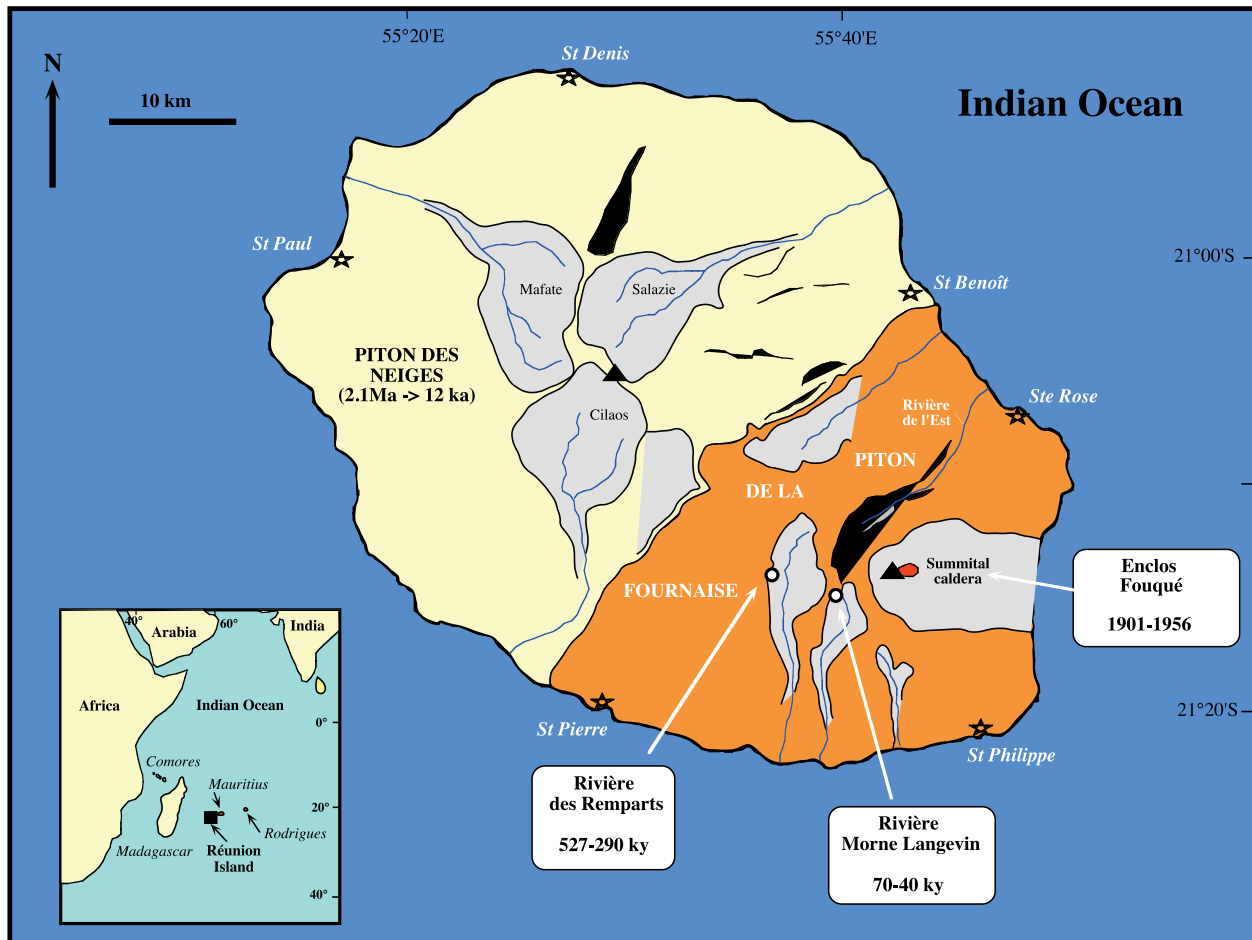


Figure 1. Schematic map of Réunion Island and its position in the Indian Ocean. Locations of Remparts, Langevin and the historical Grand Brûlé (Enclos Fouqué) series are indicated.

of these compatible elements by olivine \pm clinopyroxene-rich cumulates, (2) picrites with high MgO contents (MgO > 20%; FeO/MgO < 0.5–1.4; lowest SiO₂ = 44.5–45.8%) resulting from the entrainment of older olivine \pm clinopyroxene cumulates by SSB melts, (3) differentiated samples with the highest SiO₂ contents (48–53%), and alkalinity index (Alk = 1.6 to 2.5), FeO/MgO = 3–3.4. (4) plagioclase-rich basalts (SiO₂ = 46–47%, Alk = 0–1) with up to 50% plagioclase and occasional plagioclase+clinopyroxene crystal clusters. They are thought to represent dismantled gabbroic fragments [Albarède *et al.*, 1997].

[6] Most of the Holocene eruptive activity took place within the Enclos Fouqué caldera with occasional events occurring outside the caldera. These historical lavas consisting of basalts and picrites are described in Albarède *et al.* [1997].

[7] The present study is focused on the Nd isotope signature of the two old Remparts and Langevin stratigraphical lava sequences, and the historical lavas with ages ranging from 1901 to 1956. Additional Sr isotopic data have been obtained for the historical samples.

3. Analytical Techniques

[8] Analytical techniques and errors associated with the major and trace element data of all samples and with the Sr isotopic ratios of Remparts and Langevin samples discussed in the following sections are given in Albarède *et al.* [1997].

[9] Nd isotopic analyses have been performed on the same samples and the same chemical digestion and column separations as Sr isotopic analyses published in Albarède *et al.* [1997].



Table 1. Nd and Sr Isotopic Compositions, and Alkalinity Index of Remparts, Langevin and Historical Samples From Piton de la Fournaise Volcano

Samples	z^a	$^{143}\text{Nd}/^{144}\text{Nd}$	$\pm 2\sigma$	ϵ_{Nd}^b	$\pm 2\sigma$	$^{87}\text{Sr}/^{86}\text{Sr}^c$	$\pm 2\sigma$	Alkalinity Index ^d
REMPARTS								
RP 01	1690	0.512793	8	3.03	0.16	0.704179	15	0.266
		0.512779	8	2.75	0.16			
RP 02	1670	0.512778	11	2.73	0.21	0.704122	18	-0.146
		0.512819	9	3.52	0.18			
RP 03	1630	0.512808	9	3.31	0.18	0.704136	17	1.095
		0.512850	9	4.13	0.18			
RP 04	1605	0.512823	10	3.61	0.20	0.704083	15	0.843
RP 05	1590	0.512796	9	3.09	0.18	0.704148	20	0.321
		0.512796	10	3.08	0.20			
RP 06	1550	0.512820	8	3.54	0.16	0.704148	14	0.501
		0.512824	14	3.64	0.28			
RP 07	1540	0.512786	6	2.89	0.12	0.704092	17	0.662
		0.512799	8	3.13	0.16			
RP 09	1390	0.512821	10	3.57	0.20	0.704137	17	0.620
RP 10	1370	0.512791	7	2.98	0.14	0.704118	20	0.576
		0.512817	12	3.48	0.24			
RP 11	1345	0.512776	13	2.69	0.26	0.704114	15	0.417
		0.512810	7	3.36	0.14			
RP 12	1330	0.512777	9	2.71	0.18	0.704053	18	1.181
		0.512817	9	3.48	0.18			
RP 13	1280	0.512801	7	3.17	0.14	0.704145	15	1.883
		0.512830	6	3.75	0.12			
RP 14	1270	0.512803	7	3.22	0.14	0.704061	20	1.693
		0.512811	8	3.38	0.16			
RP 15	1240	0.512820	5	3.54	0.10	0.704084	15	2.254
		0.512829	7	3.72	0.14			
RP 16	1200	0.512807	7	3.29	0.14	0.704149	15	0.716
		0.512814	7	3.44	0.14			
RP 17	1190	0.512824	7	3.62	0.14	0.704056	18	1.948
RP 18	1170	0.512756	9	2.29	0.18	0.704156	14	1.026
RP 19	1145	0.512823	5	3.60	0.10	0.704050	23	1.899
		0.512818	16	3.50	0.32			
RP 20	1110	0.512810	9	3.36	0.18	0.704104	20	0.120
RP 21	1080	0.512763	9	2.44	0.18	0.704115	17	0.305
		0.512780	8	2.76	0.16			
RP 23	990	0.512772	9	2.62	0.18	0.704063	18	1.464
		0.512806	7	3.27	0.14			
RP 24	910	0.512835	7	3.83	0.14	0.704049	18	0.557
RP 25	890	0.512813	7	3.40	0.14	0.704034	18	0.670
RP 26	850	0.512825	8	3.64	0.16	0.704093	20	1.396
		0.512825	7	3.64	0.14			
RP 27	810	0.512787	7	2.91	0.21	0.704074	18	0.739
		0.512824	14	3.62	0.28			
RP 28	790	0.512823	6	3.60	0.12	0.704086	20	1.766
RP 29	760	0.512816	9	3.46	0.18	0.704129	20	0.941
RP 30	750	0.512812	9	3.40	0.18	0.704123	13	1.237
RP 31	740	0.512806	8	3.27	0.16	0.704117	21	0.229
RP 32	720	0.512751	10	2.21	0.20	0.704065	21	0.900
		0.512791	11	2.98	0.22			
Average		0.512805		3.26		0.704103		0.936
$\pm 2\sigma$ unweighted		0.000043		0.83		0.000077		1.26



Table 1. (continued)

Samples	z ^a	¹⁴³ Nd/ ¹⁴⁴ Nd	±2σ	ε _{Nd} ^b	±2σ	⁸⁷ Sr/ ⁸⁶ Sr ^c	±2σ	Alkalinity Index ^d
LANGEVIN								
LGV 01	2300	0.512843	11	4.00	0.22	0.704118	15	-0.034
LGV 02	2280	0.512827	11	3.68	0.22	0.704161	20	-0.061
		0.512859	11	4.31	0.22			
LGV 03	2260	0.512833	10	3.80	0.20	0.704138	20	0.398
		0.512838	10	3.90	0.20			
LGV 04	2240	0.512840	10	3.93	0.20	0.704101	15	0.663
		0.512871	10	4.54	0.20			
LGV 06	2200	0.512840	11	3.94	0.22	0.704104	14	0.282
		0.512874	8	4.61	0.16			
LGV 07	2190	0.512845	8	4.05	0.16	0.704103	21	0.219
		0.512888	11	4.88	0.22			
LGV 09	2160	0.512840	9	3.93	0.18	0.704065	14	0.442
		0.512901	8	5.12	0.16			
LGV 12	2100	0.512838	8	3.90	0.16	0.704090	13	-0.229
		0.512890	9	4.91	0.18			
LGV 13	2080	0.512828	11	3.71	0.22	0.704090	17	-0.214
LGV 14	2060	0.512848	9	4.09	0.18	0.704211	14	-0.188
LGV 15	2040	0.512851	12	4.15	0.24	0.704058	14	-0.005
		0.512876	8	4.64	0.16			
LGV 16	1990	0.512878	8	4.67	0.16	0.704209	15	0.200
		0.512820	7	3.54	0.14			
		0.512825	10	3.65	0.20			
LGV 17	1985	0.512823	10	3.61	0.20	0.704227	17	0.196
		0.512843	11	3.99	0.22			
LGV 18	1980	0.512812	6	3.39	0.12	0.704208	18	0.262
		0.512819	9	3.52	0.18			
		0.512847	9	4.08	0.18			
LGV 20	1960	0.512819	7	3.53	0.14	0.704126	17	-0.181
LGV 21	1930	0.512830	7	3.74	0.14	0.704119	13	0.371
LGV 22	1910	0.512864	8	4.41	0.16	0.704102	17	0.507
		0.512866	11	4.45	0.22			
LGV 23	1905	0.512835	7	3.84	0.14	0.704163	18	0.032
		0.512838	11	3.91	0.22			
LGV 25	1870	0.512868	8	4.48	0.16	0.704166	14	0.020
		0.512829	8	3.72	0.16			
LGV 26	1850	0.512899	8	5.09	0.16	0.704042	15	-0.556
		0.512888	11	4.88	0.22			
LGV 27	1830	0.512876	9	4.64	0.18	0.704047	15	-0.246
LGV 28	1820	0.512887	8	4.87	0.16	0.704091	15	0.075
		0.512849	10	4.11	0.20			
LGV 29	1815	0.512876	10	4.63	0.20	0.704106	17	0.086
Average		0.512852		4.17		0.704124		0.089
±2σ unweighted		0.000049		0.96		0.000107		0.578

^az, elevation.

^bε_{Nd} is calculated with present-day ¹⁴³Nd/¹⁴⁴Nd_{CHUR} = 0.512638 based on ¹⁴³Nd/¹⁴⁴Nd = 0.7219 [Jacobsen and Wasserburg, 1980].

^cSr isotopic compositions of samples from Remparts and Langevin series from Albarède *et al.* [1997].

^dAlkalinity index data from Albarède *et al.* [1997]. Alkalinity index is defined as excess of Na₂O + K₂O: Alk.I. = Na₂O + K₂O - 0.37 (SiO₂ - 39).

Core samples were cleaned in acetone and deionized water before crushing in an agate mortar. The entire Sr and Nd chemical processing was carried out at the ENS-Lyon. About 50–80 mg of rock powder were digested in screw top Savillex Teflon vials in HF-HNO₃-HClO₄ mixtures and evaporated

to dryness. Three successive re-dissolutions and evaporations were then performed in concentrated HNO₃, HCl 6M, and finally in HCl 4M before loading on AGV50W-X12 cation-exchange resin. LREE+MREE were collected with HCl 6M after elution of the Ca + Sr and Ba fractions.



Table 2. Nd and Sr Isotopic Compositions and Alkalinity Index of Historical Samples From Piton de la Fournaise Volcano

Samples	Age(AD)	¹⁴³ Nd/ ¹⁴⁴ Nd	±2σ	ε _{Nd} ^a	±2σ	⁸⁷ Sr/ ⁸⁶ Sr	±2σ	Alkalinity Index ^b
Historical samples								
S1901	1901	0.512879	13	4.70	0.26	0.704104	28	-0.071
		0.512896	11	5.04	0.22			
R1201	1905	0.512875	13	4.62	0.26	0.704134	33	0.086
Q1301	1915	0.512876	13	4.64	0.26	0.704072	32	-0.043
		0.512890	9	4.92	0.18			
T2101	1927	0.512888	7	4.87	0.14	0.704169	24	-0.091
T1602	1931	0.512881	17	4.74	0.34	0.704111	15	-0.153
		0.512902	18	5.16	0.36			
P1601	1938	0.512895	7	5.02	0.14	0.704116	20	-0.157
P1401	1942	0.512863	10	4.39	0.20	0.704138	19	-0.248
M1701	1945	0.512898	14	5.07	0.28	0.704069	32	-0.103
O1401	1948	0.512883	15	4.77	0.30	0.704163	12	-0.063
		0.512872	19	4.57	0.38			
T1601	1952	0.512870	13	4.52	0.26	0.704110	10	0.072
T1903	1953	0.512880	10	4.72	0.20	0.704138	17	0.017
O1402	1956	0.512843	13	4.00	0.26			-0.119
Average		0.512881		4.74		0.704120		-0.073
±2σ unweighted		0.000030		0.58		0.000065		0.170

^a ε_{Nd} is calculated with present-day ¹⁴³Nd/¹⁴⁴Nd_{CHUR} = 0.512638 based on ¹⁴³Nd/¹⁴⁴Nd = 0.7219 [Jacobsen and Wasserburg, 1980].

^b Alkalinity index data from Albarède *et al.* [1997]. Alkalinity index is defined as excess of Na₂O + K₂O: Alk.I. = Na₂O + K₂O - 0.37 (SiO₂ - 39).

[10] No subsequent separation of Nd from other LREE and MREE was performed. Nd isotopic ratios using the Plasma 54 of VG Elemental (ENS-Lyon) are accurately corrected for Sm isobaric interferences [Luais *et al.*, 1997]. Moreover, Ce hydrides are negligible and in contrast with thermal ionization measurements, there is no competition for ionization between neighboring REEs. About 70 ng of Nd can be analyzed with an internal precision better than 10 ppm in 18 minutes. These values are exponentially corrected for mass fractionation using ¹⁴⁶Nd/¹⁴⁵Nd = 2.071943 [Wasserburg *et al.*, 1981]. The unweighted average of four ¹⁴³Nd/¹⁴⁴Nd measurements of the La Jolla international standard over the period of analysis is 0.511844 ± 22 (two standard deviations) whereas the external reproducibility of our in-house JMC Nd standard is ±0.000032 (ε_{Nd} = +0.6) (two standard deviations, n = 77).

[11] Sr isotopic ratios of historical samples were analyzed at the CRPG (Nancy) using a Finigann Mat 262 multicollector mass spectrometer. The unweighted average of ⁸⁷Sr/⁸⁶Sr measurements of the NBS 987 international standard over the period of analysis is 0.710189 ± 32 (2σ) (n = 6).

This average is within error the value of NBS 987 determined during the measurement of Remparts and Langevin samples [Albarède *et al.*, 1997], and thus no correction for the standard values has been applied to the samples.

4. Results

4.1. Nd Isotopes

[12] The Nd isotopic ratios are reported in Tables 1 and 2 and plotted against FeO/MgO ratios in Figure 2. The overall range of variation of Nd isotopic ratios is small, with 0.512751 < ¹⁴³Nd/¹⁴⁴Nd < 0.512902 (ε_{Nd}: +2.21 to +5.16). However, these variations are outside the analytical uncertainty and thus meaningful. Within each of these three series, the various rock types, defined on the basis of FeO/MgO ratios, exhibit similar Nd isotopic values, except for some plagioclase-rich basalts. The old Remparts samples exhibit low Nd isotopic ratios (¹⁴³Nd/¹⁴⁴Nd = 0.512751 to 0.512850; average = 0.512805, ε_{Nd} = +3.26). The least radiogenic Nd values ever measured on Réunion samples are observed in some plagioclase-rich lavas. Langevin and historical samples have higher Nd isotopic

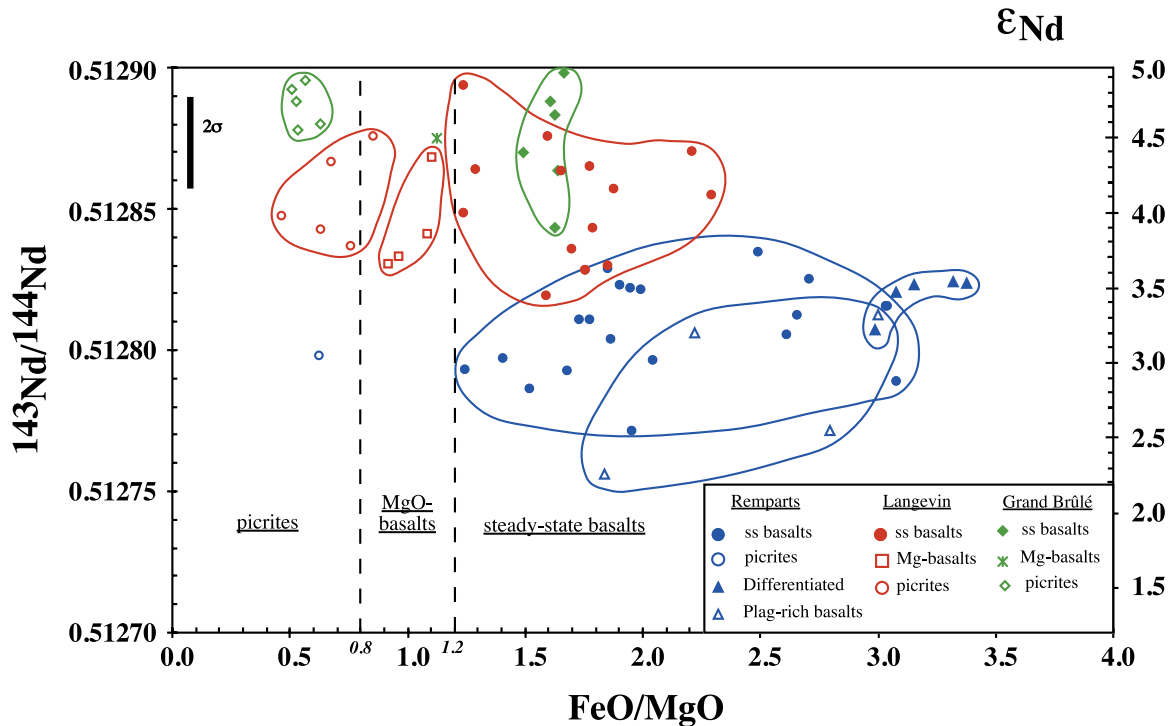


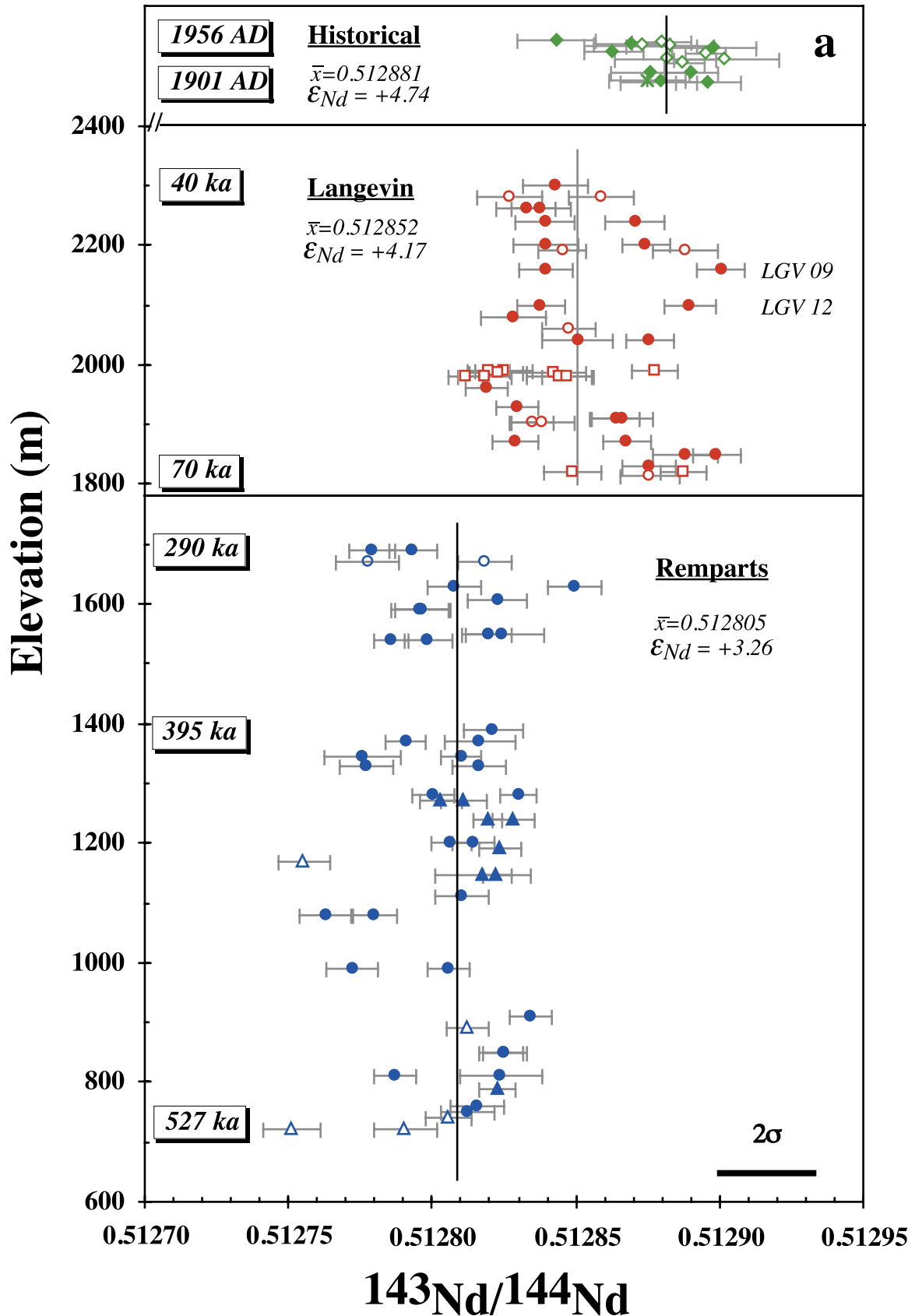
Figure 2. $^{143}\text{Nd}/^{144}\text{Nd}$ versus FeO/MgO diagram for samples from Remparts (in blue), Langevin (in red) and Grand Brûlé (historical, in green) series of Piton de la Fournaise volcano. Symbols as indicated in the inset. Picrites with $\text{MgO} > 20\%$ are characterized by low FeO/MgO ratios. $^{143}\text{Nd}/^{144}\text{Nd}$ ratios correspond to the average of replicate analyses for each sample (see Tables 1 and 2).

ratios, with $^{143}\text{Nd}/^{144}\text{Nd} = 0.512812$ to 0.512901 ; average = 0.512852 ($\epsilon_{\text{Nd}} = +4.17$), and $^{143}\text{Nd}/^{144}\text{Nd} = 0.512843$ – 0.512902 , average 0.512881 ($\epsilon_{\text{Nd}} = +4.74$), respectively. The Langevin series covers the range defined by *Newsom et al.* [1986], and *Fisk et al.* [1988] for Piton de la Fournaise based on a limited sampling, and by *Fisk et al.* [1988] and *Deniel* [1990] for Piton des Neiges.

[13] A temporal increase of $^{143}\text{Nd}/^{144}\text{Nd}$ ratios is clearly evident in Figure 3a. This corresponds to a sharp increase from the Remparts to the Langevin and historical lavas, that is especially well demon-

strated for picrites. Within the Remparts series, plagioclase-rich basalts have on average lower Nd isotopic ratios ($^{143}\text{Nd}/^{144}\text{Nd} = 0.512782 \pm 25$) than the neighboring SSB samples. Differentiated samples and picrites exhibit $^{143}\text{Nd}/^{144}\text{Nd}$ ratios similar to those of the SSB. The Langevin series shows two main segments with distinct $^{143}\text{Nd}/^{144}\text{Nd}$ evolution patterns. First, $^{143}\text{Nd}/^{144}\text{Nd}$ ratios decrease from 0.512899 at the base of the series at 1850 m to 0.512812 – 0.512847 at 1960–1980 m (from samples LGV29 to LGV16). These latter values are comparable to the average value of the Remparts samples. Second, in the upper part of

Figure 3. $^{143}\text{Nd}/^{144}\text{Nd}$ and alkalinity index versus stratigraphy/time for samples from Piton de la Fournaise lavas. Altitude (in meters) represents time spans from 527 ka to 40 ka to present. The older Remparts series ranges in age from 527 to 290 ka and the younger Langevin series from 70 ka to 40 ka, with a gap in sampling of 200 ka between the two series. Ages are derived from K-Ar data [Gillot et al., 1990, and P. Gillot unpublished data, 1991]. Symbols as in Figure 2. (a) $^{143}\text{Nd}/^{144}\text{Nd}$ ratios versus elevation (relative time). Error bars for individual samples represent two-sigma internal precision. The 2σ error bar represents the unweighted reproducibility of ± 0.000032 . Duplicate analyses have been performed on some samples, and except for samples LGV12, LGV9, $^{143}\text{Nd}/^{144}\text{Nd}$ values well agree within reproducibility (Tables 1 and 2). (b) Alkalinity Index (Alk.I.) versus elevation (relative time). Data from *Albarède et al.* [1997]. Positive spikes in Alk.I. ($\text{Alk.I.} > 1.5$) are associated with the differentiated lavas. Samples evolve from bottom to top of the sequences from alkaline to tholeiitic affinity.



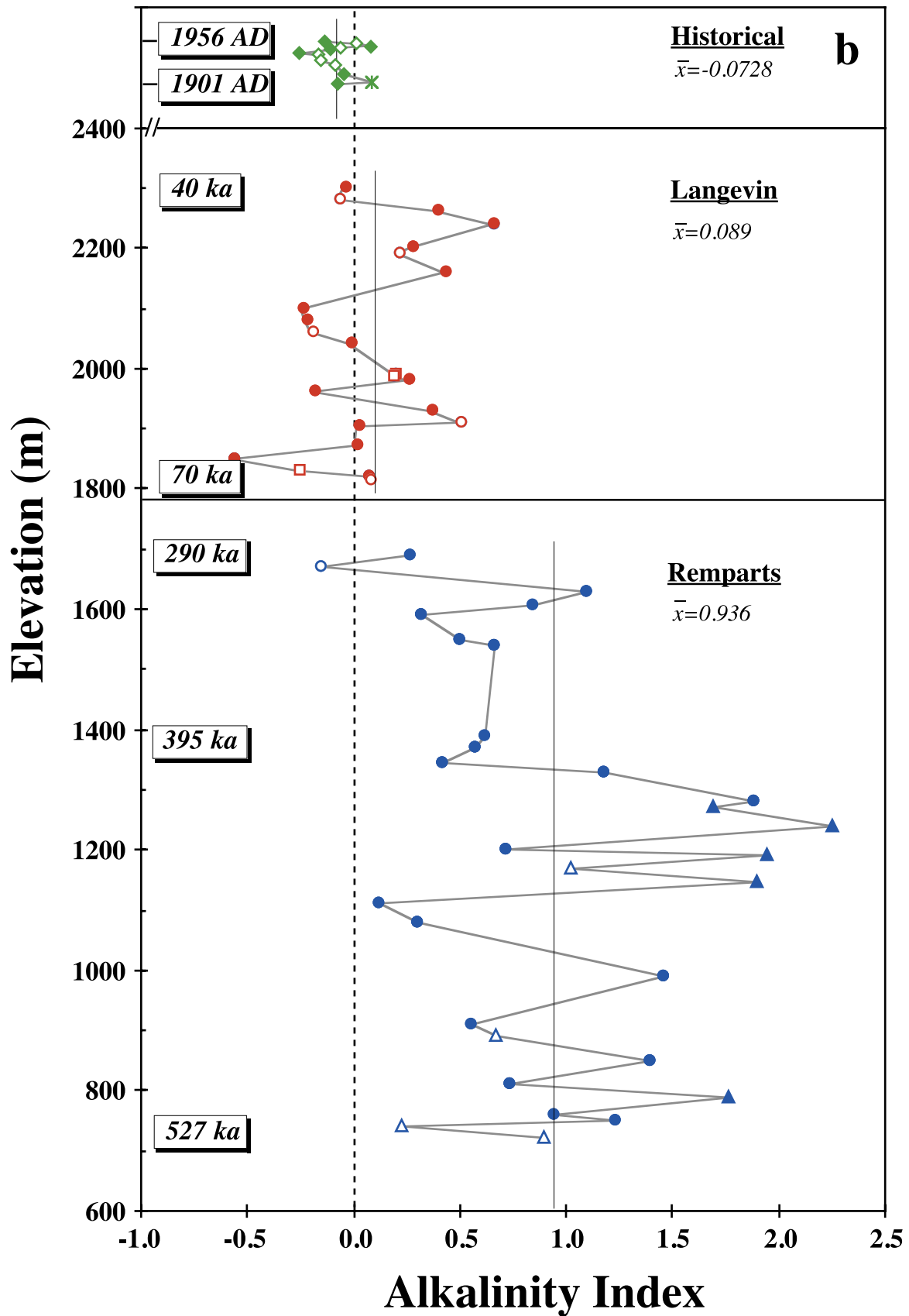


Figure 3. (continued)

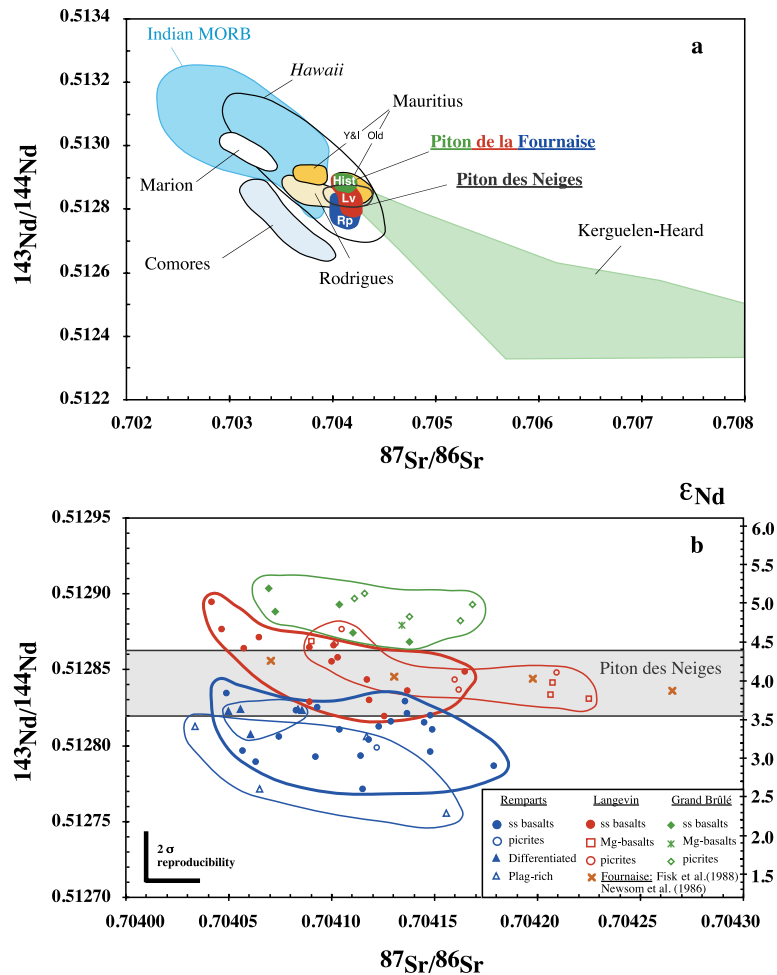


Figure 4. (a) Comparison of $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of Piton de la Fournaise samples (Rp: Remparts, Lg: Langevin, Hist: historical samples) with OIBs from Indian Ocean (Kerguelen [Dosso and Murthy, 1980; Storey et al., 1988], Heard [Barling et al., 1994], Comores [Deniel, 1998], Mauritius [Hamelin et al., 1986; Mahoney et al., 1989; Peng and Mahoney, 1995], Rodrigues [Baxter et al., 1985]), and Hawaii [Chen and Frey, 1985; Stille et al., 1986; Chen et al., 1991; Lassiter et al., 1996]. Indian Ocean MORB data from Hamelin and Allègre [1985], Hamelin et al. [1986], Le Roex et al. [1983], Ito et al. [1987], Mahoney et al. [1989, 1992], Schiano et al. [1997], Rehkämper and Hofmann [1997]. For Mauritius, distinct fields for Young and Intermediate series (Y&I) and Older series (Old) are shown. (b) $^{143}\text{Nd}/^{144}\text{Nd}$ versus $^{87}\text{Sr}/^{86}\text{Sr}$ diagram for Remparts, Langevin and historical lavas. Average data for $^{143}\text{Nd}/^{144}\text{Nd}$ ratios. Same symbols as in Figure 2. Error bars are two-sigma reproducibility. Remparts and Langevin $^{87}\text{Sr}/^{86}\text{Sr}$ data are from Albarède et al. [1997]. For comparison are also indicated data from Newsom et al. [1986] and Fisk et al. [1988] for Piton de la Fournaise lavas. The grey field corresponds to Piton des Neiges lavas [Fisk et al., 1988; Deniel, 1990].

the section, $^{143}\text{Nd}/^{144}\text{Nd}$ ratios remain constant, with an average value of 0.512854, up to the top of the sequence at 2300 m. Picrites and MgO-rich basalts in the Langevin series have also Nd isotopic ratios similar to those of SSB.

[14] $^{143}\text{Nd}/^{144}\text{Nd}$ ratios and alkalinity index are broadly negatively correlated through time: Nd isotopic values increase (Figure 3a) whereas alkalinity index decrease from Remparts (average

%alk. = 0.936) to Langevin (%alk. = 0.0886) and historical samples (%alk. = -0.0728) (Figure 3b).

4.2. Nd and Sr Isotopic Variations

[15] In addition to the previously published Sr isotopic data for the Remparts and Langevin series [Albarède et al., 1997], new Sr isotopic data for the historical samples are reported in Table 2.



[16] Figures 4a and 4b show that, like Nd isotopes, Sr isotopes exhibit a restricted range of variation, from 0.70403 to 0.70423 (Tables 1 and 2) [Albarède *et al.*, 1997] compared to those of other OIBs from the Indian Ocean and Hawaii. Data for Piton de la Fournaise and Piton des Neiges from Newsom *et al.* [1986], Fisk *et al.* [1988] and Deniel [1990] show a range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios extending to slightly lower (0.70397) and higher (0.704298) Sr isotopic values. Historical samples exhibit a smaller range of Sr isotopic variation (from 0.70407 to 0.70417). Within rock types, variations of Nd isotopic ratios are smaller than those of Sr isotopes. Although the high $^{87}\text{Sr}/^{86}\text{Sr}$ end-members of the Langevin (0.70420–0.70423) and historical (0.70416–0.70417) fields are represented by some picrites and Mg-rich basalts, a few other samples of the same type (LGV07, LGV28–29) display lower $^{87}\text{Sr}/^{86}\text{Sr}$ (0.70409–0.70410), overlapping those of the SSB. The differentiated samples from the Remparts series with their narrow range of $^{143}\text{Nd}/^{144}\text{Nd}$ (0.512803–0.512829), and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.70405–0.70409) lie at the high Nd-low Sr isotopic ratio side of the Remparts series field. On the other hand, in the Sr and Nd isotopic diagram (Figure 4b), the plagioclase-rich basalts (except RP 31) define a lower field covering nearly the entire Sr range defined for the Remparts series (0.70403–0.70418), with slightly less radiogenic Nd ratios.

[17] Variations in Sr and Nd isotopic ratios are not correlated. Only $^{143}\text{Nd}/^{144}\text{Nd}$ isotopic ratios can distinguish Remparts, Langevin and historical samples. On a larger scale in Figure 4a, it can be precised that only the older series of Mauritius show similar Nd and Sr isotopic ratios as the historical and some of the Langevin samples, in agreement with He isotopes [Hanyu *et al.*, 2001].

5. Discussion

5.1. Extent of Isotopic Heterogeneity

[18] Both the now extinct Piton des Neiges and the active Piton de la Fournaise of Réunion island were thought to have sources with homogeneous Sr, Nd and He isotopic compositions over the past 2 Ma [e.g., Newsom *et al.*, 1986; Fisk *et al.*, 1988; Graham *et al.*, 1990; Deniel, 1990; Hanyu *et al.*,

2001], whereas Oversby [1972] and Deniel [1990] suggest some Pb isotopic heterogeneities in the mantle source of Piton des Neiges. Previous work on the Piton de la Fournaise [Newsom *et al.*, 1986; Fisk *et al.*, 1988; Graham *et al.*, 1990] was based mainly on historical, and relatively young lavas which are geochronologically equivalent to Langevin samples. Only two picrites older than 290Ka (same age as the youngest Remparts samples), have been analyzed for He and Sr isotopes [Graham *et al.*, 1990]. No Nd isotope ratios are given for those samples. Therefore the assumption of isotopic homogeneity is biased by undersampling. Core sampling of high cliffs provided access to a more thorough distribution of samples through time. When the entire data set, including the data from this study, is considered, Sr and Nd isotopic ratios of Piton de la Fournaise show significant, albeit small, variations over the past 570 ka.

[19] The range of isotopic variations observed in Réunion samples is small compared to those of other ocean island basalts such as Marion, Rodrigues, Mauritius, Kerguelen-Heard, Comores, or Hawaii (Figure 4a). However, unlike Réunion Island, some of these other OIB fields are defined on the basis of several islands. Detailed examinations of core samples from individual volcanoes such as in Hawaii (Mauna Loa [Hauri *et al.*, 1996], Mauna Kea [Lassiter *et al.*, 1996]) revealed Sr and Nd isotopic variations similar in magnitude to those observed in Réunion island lavas; occurring over time spans (over 50 ka for Mauna Loa and 200 ka for Mauna Kea) similar to that of Réunion.

5.2. Role of Fractional Crystallization and Mineral Accumulation

[20] Although variations in Sr and Nd isotopic ratios are significant, variability in trace elements is small among the SSB samples in the historical, Langevin and Remparts series [Albarède *et al.*, 1997]. Meaningful trace element/isotopic ratio trends are restricted to a few picrites, differentiated and plagioclase-rich basaltic samples (Figures 5a and 5b).

[21] Fractionation of differentiated samples in the Remparts series has been extensively studied and

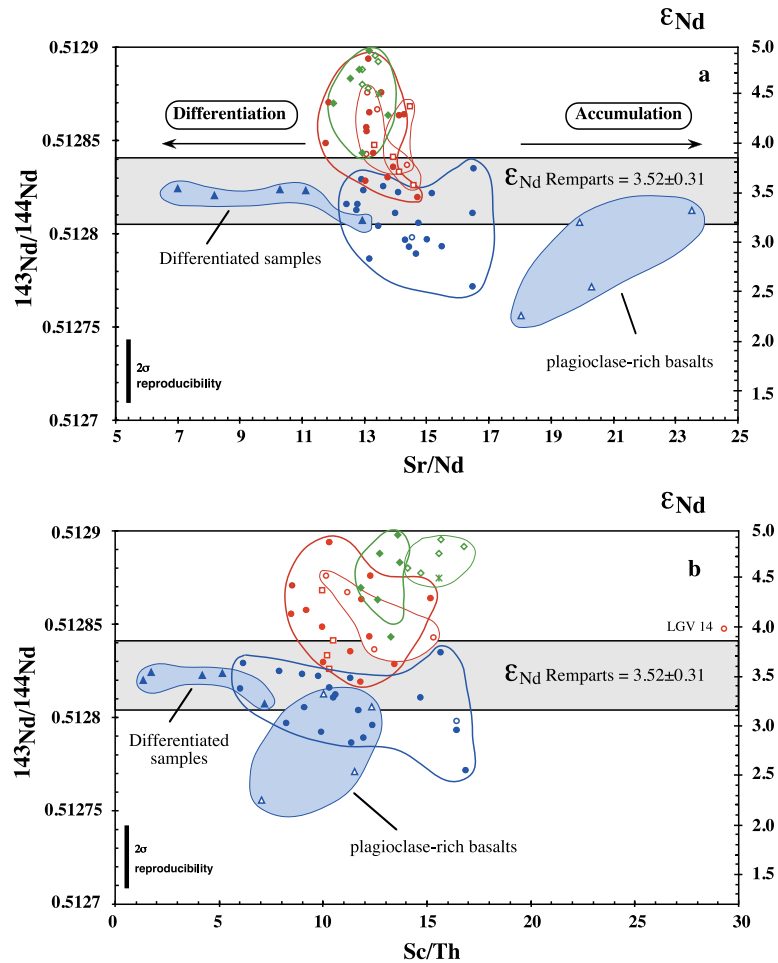


Figure 5. Sr and Nd isotope variations versus trace element contents and ratios: (a) $^{143}\text{Nd}/^{144}\text{Nd}$ versus Sr/Nd, (b) $^{143}\text{Nd}/^{144}\text{Nd}$ versus Sc/Th. Symbols as in Figure 2. The grey field represents the ϵ_{Nd} value of 3.52 ± 0.31 of the Remparts source. The high Sc/Th ratio of the sample LGV14 results from low Th contents. Trace element data from *Albarède et al.* [1997].

modeled by *Albarède et al.* [1997] on the basis of trace elements. *Albarède et al.* [1997] demonstrate that fractionation of differentiated lavas from basalts leaves a gabbroic cumulate of mainly clinopyroxene and plagioclase. They also conclude that because of the high Alk Index ($\text{Alk} > 1.5$) of these samples this early alkaline stage in the volcanic activity taps the rim of the hot spot conduit, at the thermal boundary with the surrounding mantle lithosphere. In order to clarify this model, and considering that Sr is strongly compatible in plagioclase ($K_d \approx 1.6$ [*Dostal et al.*, 1983; *Phinney and Morrison*, 1990]), and Sc in pyroxene ($K_d = 1.3$ [*Hart and Dunn*, 1993]), we have reported in Figures 5a and 5b the variations of Sr/Nd and Sc/Th ratios (compatible/incompatible

element ratios: “compatible” element here means element incorporated into the fractionated phases) and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios in all rock types. The differentiated samples from the Remparts display the lowest Sr/Nd (7–13) and Sc/Th (<8) values compared to the SSB, in agreement with cpx and plagioclase crystallization, and constant Nd isotopic values (average $^{143}\text{Nd}/^{144}\text{Nd} = 0.512819 \pm 8$; $\epsilon_{\text{Nd}} = 3.52 \pm 0.31$ (2σ)) similar to those of SSB. This demonstrates that the fractionation of differentiated samples from basalts occurs without any contamination by the mantle lithosphere, assuming that the latter has Nd isotopic signatures of Indian MORB ($^{143}\text{Nd}/^{144}\text{Nd} > 0.51276$ [*Frey et al.*, 1980; *Dupré and Allègre*, 1983; *Hart*, 1984; *Hamelin and Allègre*, 1985; *Ito et al.*, 1987]). The average ϵ_{Nd} of



3.52 ± 0.31 (2σ) thus characterize the Remparts lava source. The relation between alkalinity and the dynamic of the hot spot will be discussed in the following section.

[22] In contrast, plagioclase-rich basalts exhibit higher Sr/Nd than the SSB, which indicates plagioclase accumulation (Figure 5a). The Sr/Nd ratios of these samples are positively correlated with $^{143}\text{Nd}/^{144}\text{Nd}$. Assuming the ϵ_{Nd} value of +3.52 for the Remparts lavas, the plagioclase-rich basalts trend from a high Sr/Nd - high $^{143}\text{Nd}/^{144}\text{Nd}$ end-member toward an enriched component characterized by low Sr/Nd and $^{143}\text{Nd}/^{144}\text{Nd}$, and high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (the sample with low $^{143}\text{Nd}/^{144}\text{Nd}$ has high $^{87}\text{Sr}/^{86}\text{Sr}$, Figure 4). This trend results from the combined effects of fractional crystallization and mixing with this enriched component.

5.3. Temporal Relationship Between Alkalinity and Nd Isotopic Composition in Steady State Basalts (SSB)

[23] SSB from all series are characterized by an increase with time in $^{143}\text{Nd}/^{144}\text{Nd}$ for a similar range of Sr isotopes. $^{143}\text{Nd}/^{144}\text{Nd}$ evolution is inversely correlated with that of alkalinity index, from alkali-less radiogenic Nd Remparts samples to tholeiitic-more radiogenic Nd historical samples (Figures 3a and 3b).

[24] The fact that decreasing $^{143}\text{Nd}/^{144}\text{Nd}$ ratios are correlated both with increasing age and with increasing alkalinity (Figures 3a and 3b) raises again the question of the origin of the alkalinity variation. The negative correlation between alkalinity and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios precludes clinopyroxene fractionation [Albarède *et al.*, 1997] as the sole process controlling alkalinity. The possibility that alkalinity variations represent variable extents of melting [Chen *et al.*, 1991] can be tested using incompatible element ratios with different degrees of incompatibility as discussed by Hofmann and Jochum [1996]. In contrast with the very low ratios of Hawaiian samples ($\text{Th}/\text{Ba} = 0.001\text{--}0.01$, $\text{Th}/\text{La} = 0.062\text{--}0.077$) [Hofmann and Jochum, 1996], indicative of Th depletion of the Hawaiian source, Th/La and Th/Ba ratios of the Réunion samples ($\text{Th}/\text{Ba} = 0.012\text{--}0.02$, $\text{Th}/\text{La} =$

$0.08\text{--}0.15$) for all series are close to the primitive mantle values [Zindler and Hart, 1986]. The important observation is that whereas Th/Ba ratios are similar in all series and correlate positively with Th/La within series, Th/La ratios decrease through time from the Remparts series to the Langevin and historical series (Figure 6a), with increasing $^{143}\text{Nd}/^{144}\text{Nd}$ (Figure 6b), which precludes any difference in degree of partial melting to explain variations in alkalinity. In addition, highly incompatible element ratios indicative of source composition, such as Nb/La , Zr/Hf , are constant within errors for alkaline and tholeiitic series through time ($\text{Nb}/\text{La} = 1.33 \pm 0.06$, 1.27 ± 0.03 , 1.33 ± 0.03 ; and $\text{Zr}/\text{Hf} = 38.68 \pm 5.37$, 40.08 ± 2 , 36.97 ± 3.15 , for historical, Langevin and Remparts series, respectively [Albarède *et al.*, 1997]). This strongly suggests that the sources of alkaline and tholeiitic series are similar in terms of trace element composition, but are not identical in Nd isotopic composition. This scheme contrasts with eruption sequences at Mauna Kea [Yang *et al.*, 1996; Lassiter *et al.*, 1996] and Haleakala [Chen *et al.*, 1991] in Hawaii where intercalated alkaline and tholeiitic lavas have similar Nd isotopic ratios, indicative of various degrees of melting of an homogeneous source. It is also distinct from the Maui volcanic sequences in Hawaii [Chen and Frey, 1985], where early eruptive stages have tholeiitic affinities and lower Nd isotopic ratios than the late alkaline stages, reflecting a multicomponent mixing model involving a temporal decrease of the plume component in favor of the MORB component.

[25] The negative correlation between $^{143}\text{Nd}/^{144}\text{Nd}$ and Th/La (Figure 6b), and in general between $^{143}\text{Nd}/^{144}\text{Nd}$ and others ratios involving trace elements enriched in the crust (e.g., Ba/La) may indicate that a low $^{143}\text{Nd}/^{144}\text{Nd}$ and high Th/La , Ba/La component (an enriched component) is necessary to explain the $^{143}\text{Nd}/^{144}\text{Nd}$ and trace element ratios of the Remparts samples.

5.4. Nature of the Mantle Source Under Réunion Volcano

[26] The temporal Nd isotopic evolution of the Réunion volcanic products demonstrates their

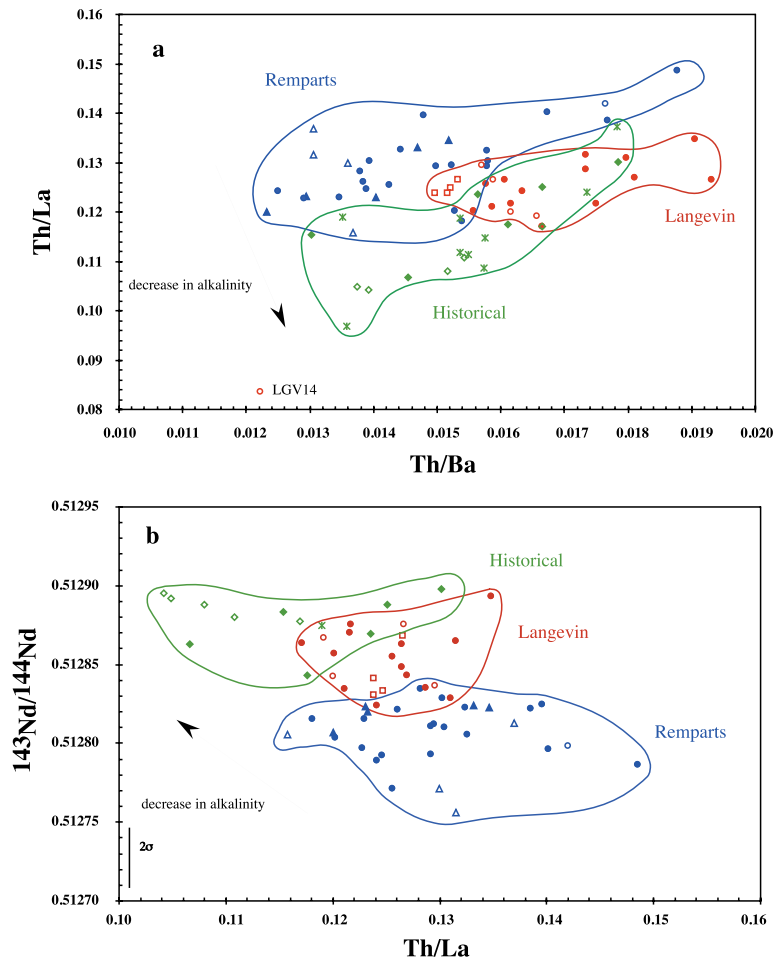


Figure 6. (a) Th/La versus Th/Ba and (b) $^{143}\text{Nd}/^{144}\text{Nd}$ versus Th/La variations for the Piton de la Fournaise samples (symbols as in Figure 2). Decrease with time in alkalinity from the Remparts to the historical series is associated with a decrease in Th/La ratios and an increase in $^{143}\text{Nd}/^{144}\text{Nd}$ ratios, indicative of a change in source composition. Note the low Th/La ratio for sample LGV14, because of low Th contents; trace element data from *Albarède et al.* [1997].

derivation from an heterogeneous source, and raises the question of the composition of the plume component and the nature of the mantle source.

[27] At the scale of Indian Ocean OIB and MORB isotopic compositions (Figure 4a), the $^{143}\text{Nd}/^{144}\text{Nd}$ - $^{87}\text{Sr}/^{86}\text{Sr}$ evolution from Remparts to historical SS basalts does not follow the main MORB-OIB negative trend, in that the increase in $^{143}\text{Nd}/^{144}\text{Nd}$ is not accompanied by a decrease in $^{87}\text{Sr}/^{86}\text{Sr}$. Therefore the Piton de la Fournaise series define a vertical isotopic evolution. It is unlikely that an initial Nd-Sr isotopic correlation following the main MORB-OIB array would have been largely destroyed by secondary processes: e.g., hydrothermal alteration. In Figure 7, a distinct negative

correlation between Cs/Th ratios, also an indicator of hydrothermal alteration, and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios can be seen only for Langevin SSB, where the least altered samples have $^{87}\text{Sr}/^{86}\text{Sr}$ values of about 0.70404. However, under this hypothesis, alteration would have slightly increased the $^{87}\text{Sr}/^{86}\text{Sr}$ of the Langevin samples, whereas much lower $^{87}\text{Sr}/^{86}\text{Sr}$ values (from 0.7037 to 0.7039) are necessary to fit the MORB-OIB trend. Considering the small age difference between these three series, decoupling of Sr and Nd isotopic ratios cannot reflect an “evolving plume source” [Class *et al.*, 1993].

[28] The lack of correlation between Nd and Sr isotopes induced by the increase in $^{143}\text{Nd}/^{144}\text{Nd}$ ratios would reflect change in the mantle source

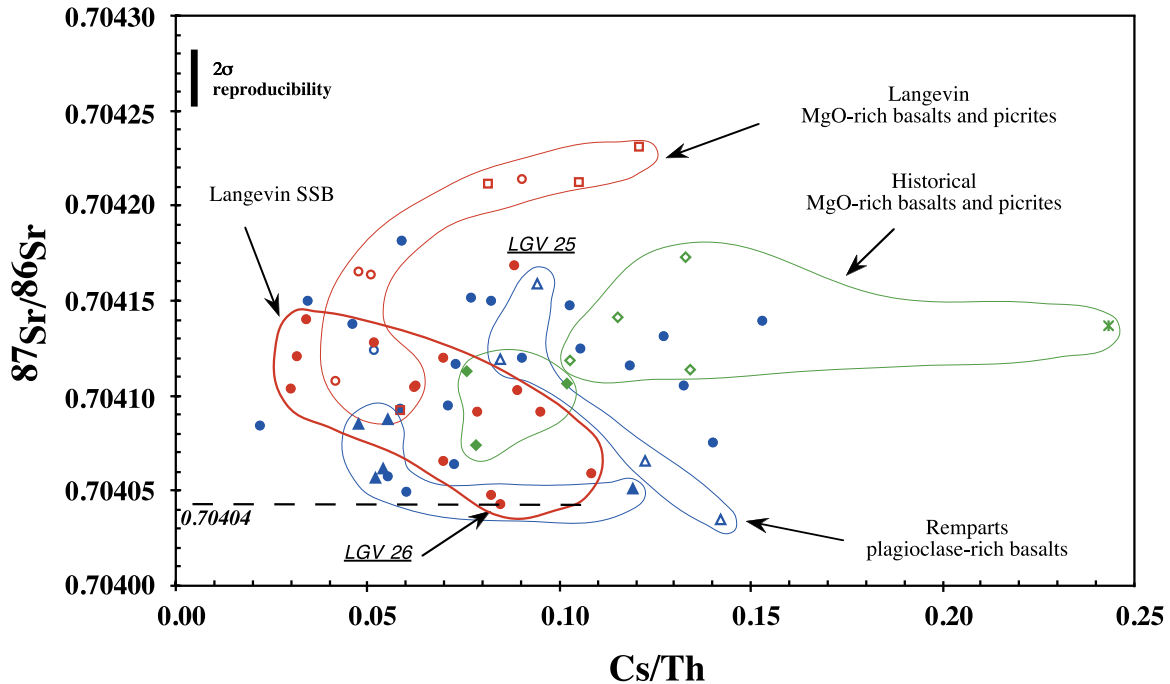


Figure 7. $^{87}\text{Sr}/^{86}\text{Sr}$ versus Sc/Th diagram for the Piton de la Fournaise samples. Symbols as in Figure 2. The sample LGV26 with the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.70404 could represent the least hydrothermally altered sample. Whereas the negative correlation between $^{87}\text{Sr}/^{86}\text{Sr}$ and Sc/Th for the Langevin SSB results from hydrothermal alteration, the negative correlation for the plagioclase-rich basalts would reflect the increase in $^{87}\text{Sr}/^{86}\text{Sr}$ ratio and the enrichment in Th concentrations because of contamination s.l. No well-defined field can be drawn for the Remparts SSB. Trace element data from *Albarède et al.* [1997].

with time, possibly resulting from mixing processes in the mantle between a plume component and an enriched component. The temporal constancy in Sr isotopes indicates that Réunion volcanism does not result from MORB-OIB interaction, in that no MORB component would be involved in the source of these lavas, in agreement with high He values [*Hanyu et al.*, 2001]. This model is consistent with *White et al.* [1990] in the temporal decrease in the asthenospheric contribution to the plume, with decreasing age of the Réunion OIB hot spot products. The Réunion mantle source would contain virtually no asthenosphere component, and the narrow range in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.70397–0.704298) and the $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of the historical samples would then characterize the isotopic composition of the Réunion plume.

[29] However, if the two components of the mixing have similar $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and parent-daughter element ratios, but variable $^{143}\text{Nd}/^{144}\text{Nd}$, simple

mixing could indeed occur without leading to a correlation. Considering that one component is the Réunion plume, the other component must have an enriched Nd isotopic signature in order to account for the low $^{143}\text{Nd}/^{144}\text{Nd}$ signature of the Remparts samples. It can be then ascribed to a mafic, and probably old material. It would be too much speculation to precise its nature. However, the elevated $^3\text{He}/^4\text{He}$ ratios of all Réunion samples [*Hanyu et al.*, 2001] preclude an HIMU component with low and uniform $^3\text{He}/^4\text{He}$ ratios [*Hanyu and Kaneoka*, 1997].

[30] In this model, the decrease in $^{143}\text{Nd}/^{144}\text{Nd}$ ratios with time in Piton de la Fournaise series results from a temporal decrease of the proportion of the enriched component in the mantle source. It is mostly involved in the generation of the plagioclase-rich basalts. However, this evolution is restricted to Piton de la Fournaise and does not hold for the old Piton de Neiges with $^{143}\text{Nd}/^{144}\text{Nd}$ signature [*Fisk et al.*, 1988; *Deniel*,



1990] similar to Langevin and historical series. This would imply that the enriched component in the mantle source beneath Réunion hot spot is not uniformly distributed and survive as isolated blobs or fragments because of inefficient mixing.

6. Conclusions

[31] The detailed isotopic stratigraphy of Piton de la Fournaise volcanics from the oldest exposed lavas of Remparts (527–290 ka) and Langevin cliffs (70–40 ka) to historical lavas from Grand Brûlé has demonstrated a significant variability in Nd isotopic compositions which extend toward low values the range given by *Newsom et al.* [1986] and *Fisk et al.* [1988]. The overall increase with time in $^{143}\text{Nd}/^{144}\text{Nd}$ from 0.512751 to 0.512902 ($\varepsilon_{\text{Nd}} = +2.21$ to $+5.16$) is inversely correlated with the alkalinity index which decreases from Remparts (average %alk. = 0.936) to Langevin (%alk. = 0.0886) and historical samples (%alk. = -0.0728).

[32] No correlation between Nd and Sr isotopes is observed. The vertical Nd-Sr isotopic evolution does not follow the Indian MORB-OIB array. The increase in $^{143}\text{Nd}/^{144}\text{Nd}$ from Remparts to Langevin to historical samples can be explained by a temporal decrease in the contribution of an enriched low $^{143}\text{Nd}/^{144}\text{Nd}$ old mafic component in the mantle source of Piton de la Fournaise lavas.

Acknowledgments

[33] I wish to thank F. Albarède for having initiated this work, and for his comments on the first draft of the manuscript. I am grateful to M. Semet and L. Reisberg for constructive comments and English corrections. Many thanks to C. Zimmermann (CRPG, Nancy) for running Sr isotopic analyses of some historical samples. The Editors B. White and C. Chauvel, and D. Paul and an anonymous reviewer are thanked for comments and careful revision of the manuscript. This work was partly supported by EEC Grant EVSV-CT-92-0189 to F.A. This is CRPG contribution n° 1645.

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