

Rare Earth Contribution to the Origin of Hawaiian Lavas

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Received April 14, 1969

Abstract. Rare earth abundances were determined by neutron activation in twenty Hawaiian lavas and one diabase of known chemical and mineralogical compositions. These results demonstrate a systematic relationship between the absolute or relative rare earth abundances and the petrochemistry of these rocks. Three distinct lava groups are recognized. These correspond to: (1) tholeiites, (2) alkali series, (3) nepheline-melilite basalts.

Based on rare earths: a) The hawaiites and mugearite of the alkali series represent residual melts derived from alkali olivine basalts, most likely by fractional crystallization; the trachyte, however, seems to have a more complicated history. b) Fractional crystallization models linking nephelinites or alkali olivine basalts to tholeiites are possible. However, production of these three lava groups, independently, by various degrees of partial melting of the mantle is equally likely and cannot be distinguished from these fractional crystallization models. c) Daly limestone syntexis hypothesis to produce the nephelinites is unlikely.

Introduction

Smooth rare earth relative abundance patterns ($_{57}\text{La}$ to $_{71}\text{Lu}$ including $_{39}\text{Y}$) have consistently been observed in crustal rocks. These regularities have been tentatively interpreted as to reflect past chemical history of the rock studied without, however, too much direct evidence in support. It thus remained to show that these regularities were not the product of randomness.

In a preliminary report, concentrating on volcanism, a close relationship between the rare earth fractionation patterns and lava-types was qualitatively demonstrated for a suite of twenty genetically related Hawaiian volcanic rocks, well petrochemically documented (SCHILLING and WINCHESTER, 1966). Meanwhile, similar correlations were obtained by BALASHOV and NESTERENKO (1966); PAVLENKO *et al.* (1966); MASUDA (1966, 1968), and more recently by HERRMANN (1968), for volcanic rocks from the Siberian Platform, Central Armenia, the Japanese Islands and Central Germany respectively. In this report our final results and quantitative correlation between rare earth contents and petrochemistry of these rocks are presented and some genetic relationships briefly discussed.

Samples and Analytical Procedure

The samples analyzed are listed in Table 1 and are described in detail elsewhere (SCHILLING, 1966). The petrology, petrography and genesis of these very same samples have been discussed in great detail in the literature (CROSS, 1915; MACDONALD and KATSURA, 1964; MACDONALD and POWERS, 1946; MUIR and

Table 1. *Rare earth concentration in parts per million*

Type	Sample	Locality	Source of data ^a	⁵⁷ La	⁵⁸ Ce	⁵⁹ Pr
Tholeiitic series	9948	Koolau s.	11	5.3	—	2.6
	10398(diabase)	Koolau s.	11	6.2	—	2.9
	JP12(2201)	Mauna Loa	4	7.2	25.8	3.5
	10396	Koolau s.	11	8.4	—	3.9
	10403	Koolau s.	11	8.8	—	4.3
	Pele's Hair	Halemaumau	2,9+	11.4	33.7	4.9
Alkalic series	1801 flow	Hualalai	10+	13.3	—	4.9
	1101	Haleakala c.	5	13.9	—	5.7
	JP14	Keauhou b.	6+	17.8	—	4.6
	JP11(C-32)	Honolulu s.	4	14.5	36.6	4.9
	9982	Honolulu s.	12	18.1	51	6.3
	9962	Honolulu s.	12	21.3	62	7.9
	1106	Haleakala c.	5	24.6	—	9.9
	JP10(C-72)	Mauna Kea	4	31.6	100	13.0
	1102	Haleakala c.	5	36.1	—	13.1
	1104	Haleakala c.	5	48	112	14.2
	JP15	Puu Anahulu	10+	44	117	13.7
Nepheline-melilite basalts	GF30	Honolulu s.	7	41	—	14.5
	JP13(9961)	Honolulu s.	12	52	129	15.2
	JP16	Honolulu s.	1+	63	147	17.5
	9960	Honolulu s.	12	78	188	21.5
	G-1	Westerly, R. I.	3	89	208	20.0
	W-1	Centerville, Va.	3	8.5	27	2.9
	Av. chondrites		8	0.30	0.84	0.12
% Std. dev.		—	4	10	4	
Conc. ratio ^b		—	15	7	8	

^a Major element analyses and sample descriptions are from: 1. (CROSS, 1915); 2. (DALY, 1944); 3. (FAIRBAIRN *et al.*, 1951); 4. (MACDONALD and KATSURA, 1964); 5. (MACDONALD and POWERS, 1946); 6. (MUIR and TILLEY, 1961); 7. (POWELL *et al.*, 1965); 8. (SCHMITT *et al.*, 1963, 1964); 9. (TILLEY, 1960); 10. (WASHINGTON, 1923); 11. (WENTWORTH and

TILLEY, 1961; WASHINGTON, 1923; WENTWORTH and WINCHELL, 1947; WINCHELL, 1947). The strontium isotopic composition for the eight samples numbered JP and GF are known (POWELL, FAURE, and HURLEY, 1965), as well as for samples 9960, 9961, 9982, 9962, 10396, 9948, 10403 (POWELL and DELONG, 1966). The U, Th, Pb content and Pb isotopic composition of samples JP16, JP15, JP10, JP14 are given by TATSUMOTO (1963) under sample number HMc2, HMc3, HMc4, HMc5 respectively. Finally, all these samples have been analyzed for rubidium and strontium as well as the strontium isotopic composition (BENCE, 1966), except samples GF-30, the Pele's Hair, and Hualalai alkali olivine basalt. The potassium and silica content of these samples is shown in a K₂O-SiO₂ variation diagram in Fig. 1.

The neutron activation procedure is basically that used in previous work (TOWELL, 1963; SPIRN, 1965) but with some modifications (TOWELL *et al.*, 1969). The accuracy of the procedure as applied to the analyses of rock standards G-1 and W-1 is discussed by SCHILLING (1966). Reproducibility is in the range of

^{60}Nd	^{62}Sm	^{63}Eu	^{64}Gd	^{65}Tb	^{66}Dy	^{67}Ho	^{69}Tm	^{70}Yb	^{71}Lu	^{39}Y
12.8	3.7	1.28	—	0.66	3.7	0.57	0.20	1.26	0.19	16.4
14.6	4.1	1.31	—	0.73	4.2	0.67	0.22	1.45	0.20	18.8
20.1	4.9	1.77	—	0.85	3.1	0.85	0.32	1.85	0.30	15.2
19.3	5.3	2.03	—	1.00	4.8	0.75	0.27	1.81	0.25	22.4
20.9	5.1	1.77	—	0.87	5.0	0.79	0.27	2.06	0.29	22.1
20.1	5.4	1.81	—	0.90	5.2	0.82	—	1.67	0.24	28.6
21.5	4.8	1.55	—	0.69	3.8	0.68	—	1.70	0.28	19.3
25.8	5.3	1.80	—	0.79	4.6	0.64	0.20	1.40	0.20	18.5
20.7	4.4	1.48	—	0.66	4.0	0.68	0.27	1.63	0.24	18.8
22.1	4.9	1.73	—	0.74	4.3	0.66	0.19	1.14	0.18	17.7
27.0	6.0	2.07	—	0.89	4.8	0.76	0.21	1.23	0.25	19.5
34.7	7.7	2.60	—	1.07	5.8	0.96	0.29	1.82	0.26	24.5
44	9.0	2.83	—	1.36	7.5	1.23	0.35	2.40	0.39	31.8
61	13.2	4.3	—	1.63	10.8	1.52	0.42	2.74	0.43	40.4
62	11.3	3.4	—	1.42	8.8	1.26	0.38	2.44	0.35	33.5
55	11.2	3.6	—	1.25	7.6	1.33	0.42	3.08	0.41	35.7
45	8.7	2.4	—	1.37	8.0	1.43	0.68	4.6	0.68	42.5
58	13.5	4.0	—	1.10	6.4	0.93	0.14	1.88	0.25	26.1
53	11.1	3.7	12	1.53	7.9	1.03	0.41	2.01	0.26	28.0
74	16.0	5.0	15	1.67	5.9	0.87	0.21	0.90	0.13	19.8
86	15.6	4.9	—	1.88	9.3	1.20	0.30	1.96	0.26	—
58	6.8	1.20	3.6	0.57	2.9	0.49	0.153	1.04	0.165	13.1
12.1	3.2	1.03	—	0.60	3.8	0.69	0.26	2.18	0.32	22.4
0.58	0.21	0.074	0.32	0.049	0.31	0.057 ^c	0.025 ^c	0.17	0.031	1.8
6	5	6	15	9	6	7	8	9	9	6
7	4	4	—	3	3	3	5	5	5	3

WINCHELL, 1947); 12. (WINCHELL, 1947); rare earth analyses were carried out on the same hand specimen as that used for published major element analyses except where indicated by +.
^b Max/min concentration ratios obtained within the 21 rocks.
^c Values adopted for best smooth curve normalized abundance patterns.

±4—10% depending on the element. Given in Table 1 are the estimated precisions for individual rare earths on the basis of 9 samples run with two reference solutions, combinations of 5 sets of duplicates, 3 sets of triplicates and 1 quadruplicate of the same or different sample solutions. The steps in the procedure consisted of dissolution of a 0.5 g sample, group isolation of the rare earths, neutron activation, reversed-phase partition chromatography, and radioactivity counting. Reagent blank corrections were negligible.

Results

The rare earth content in ppm of twenty lavas and one diabase are listed in Table 1. These are listed approximately in the order of increasingly greater content of rare earths, particularly the light rare earths. The ratios of maximum to minimum concentrations for each rare earth in the 20 basalts and the diabase are found to vary from 15 to 3, generally being greater for the lower Z elements.

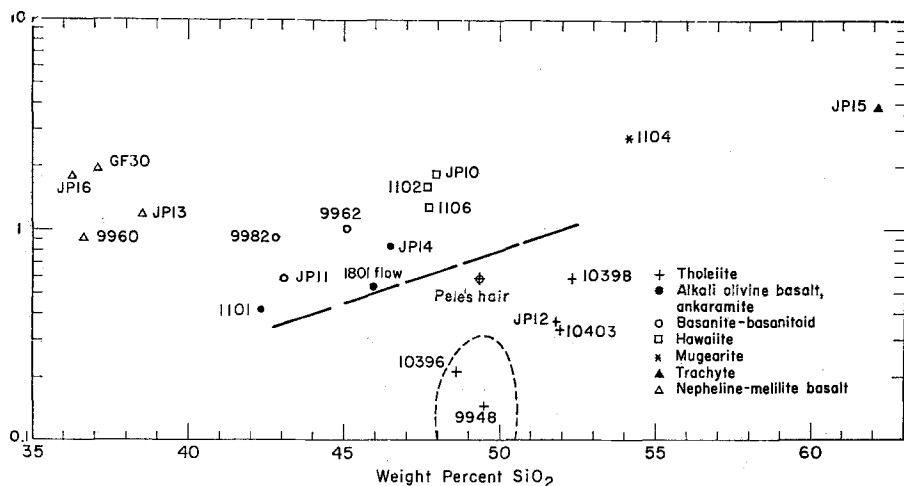


Fig. 1. K₂O content (weight percent) of 20 Hawaiian lavas and one diabase plotted on a logarithmic scale as a function of SiO₂ content (weight percent). Field above the dashed line represents the alkalic series and field below represents the tholeiitic series. The small field delineated by the dotted curve represents the oceanic basalt (low K₂O) field suggested by ENGEL *et al.* (1965). Source of data is given in Table 1

All concentrations in these lavas are considerably greater than the average rare earth abundances in 20 chondritic meteorites (SCHMITT *et al.*, 1963, 1964; HASKIN *et al.*, 1967). Finally, the rare earth concentrations of G-1 and W-1 determined during this investigation, are shown in Table 1 for inter-laboratory comparison.

Discussion

On a Masuda-Coryell plot (MASUDA, 1962; CORYELL *et al.*, 1963) the rare earth pattern of each of the 21 lavas is smoothly and progressively enriched relative to the chondritic meteorites. The lighter rare earths are preferred while in the heavy region the concentrations stay relatively more constant. Lanthanum shows the maximum range of variation and will be used to illustrate quantitatively these variations with respect to the petrochemistry of these rocks. Fig. 2 shows the La abundance as a function of the commonly used differentiation index $\text{SiO}_2/3 + \text{K}_2\text{O} - \text{MgO} - \text{FeO} - \text{CaO}$. Three main groupings are apparent: 1. In the low La concentration region, without apparent relationship to the index of differentiation, are the tholeiites. 2. At intermediate La content, and systematically and logarithmically related to the index of differentiation, are the lavas of the alkali series. 3. At high La content and low values of index of differentiation, are the strongly silica undersaturated nepheline-melilite basalts.

These three groupings, corresponding to three different lava series, perhaps suggest an independent origin for each group rather than being interrelated by a line of descent. Within the alkali series, however, a genetic relationship between the lavas that compose this series is implied. Variation diagrams showing smooth trends, as it is evident for the alkali series in Fig. 2, have been interpreted to

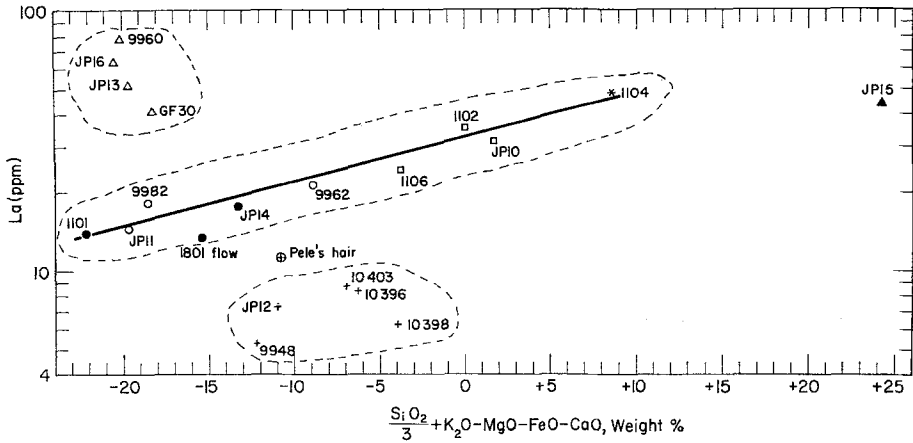


Fig. 2. Variation diagram of La content (in ppm) as a function of the index of fractionation $\text{SiO}_2/3 + \text{K}_2\text{O} - \text{MgO} - \text{FeO} - \text{CaO}$ (weight percent). Cross = tholeiites; black dot = alkali olivine basalts and ankaramite; open circle = basanite-basanitoids; open triangle = nepheline-melilite basalts; open square = hawaiites; star = mugearite; filled triangle = trachyte

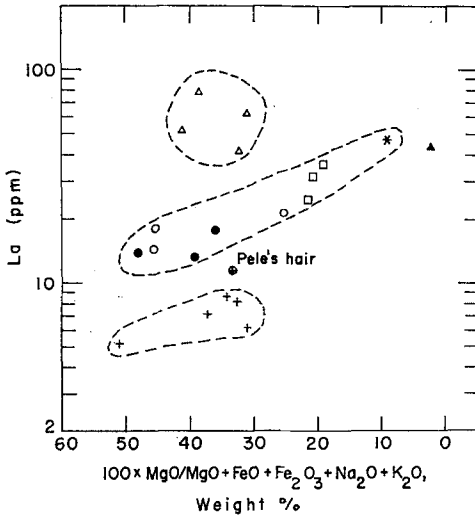


Fig. 3. Variation diagram of La content (in ppm) as a function of Kuno's index of fractionation $100 \text{MgO}/\text{MgO} + \text{FeO} + \text{Fe}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O}$ (weight percent). The same symbols as given in Fig. 2 are used

indicate that the individual rock types have been linked by a process of fractional crystallization. The trachyte, although part of the alkali series but standing by itself in this diagram, suggests further complications and will be discussed later.

The same relation as that in Fig. 2 can be obtained with other commonly used indices of fractionation (KUNO *et al.*, 1957; POLDERVAART and PARKER, 1964). As an additional example, Fig. 3 shows La variation as a function of the Kuno index. Similar plots with the same groupings are obtained for other rare earths, although less and less evident as the abundance variation decreases progressively for rare earths of decreasing trivalent ionic size.

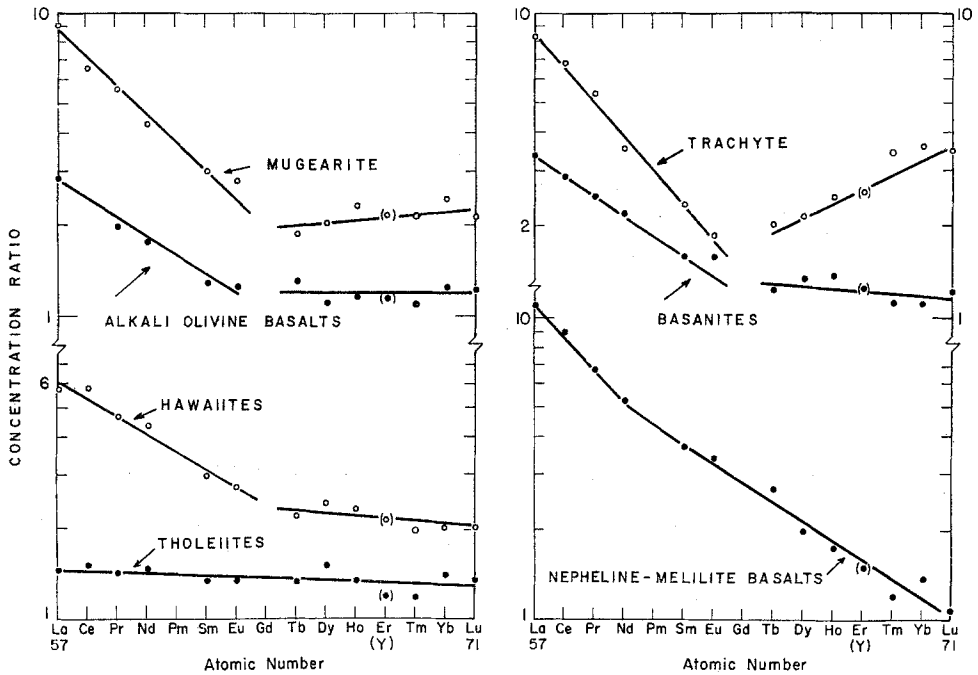


Fig. 4. Rare earth abundance averages of mineralogically recognized rock groups relative to olivine tholeiite 9948

In our earlier report (SCHILLING and WINCHESTER, 1966), the same groupings corresponding to mineralogically well identified rock types were obtained from the rare earth patterns as a whole, as shown in Fig. 4. These groups are: The tholeiites with the lowest rare earth content, followed by the rocks of the alkali series showing a break in the middle of the patterns and the nepheline-melilite basalts with the light rare earths strongly enriched and fractionated relative to tholeiites.

In summary, these three figures show a systematic relationship between the absolute or relative rare earth abundances and the petrochemistry of these rocks; confirming earlier suggestions that the rare earths fractionation may reflect the past chemical history of volcanic rocks.

Genetic Relationships

The three main rock types, representing three possible magma types, can now be considered. These are (1) tholeiites, (2) the alkali olivine basalts including the slightly silica undersaturated basanite-basanitoids, and (3) the nepheline-melilite basalts. Their average rare earth contents are tabulated in Table 2 and the fractionation patterns are shown in Fig. 5. In these averages, the hawaiiites, mugearite and trachyte are not included, as there is very little doubt that these lavas represent derivative melts from the alkali olivine basalts (see variation diagram, Figs. 2 and 3).

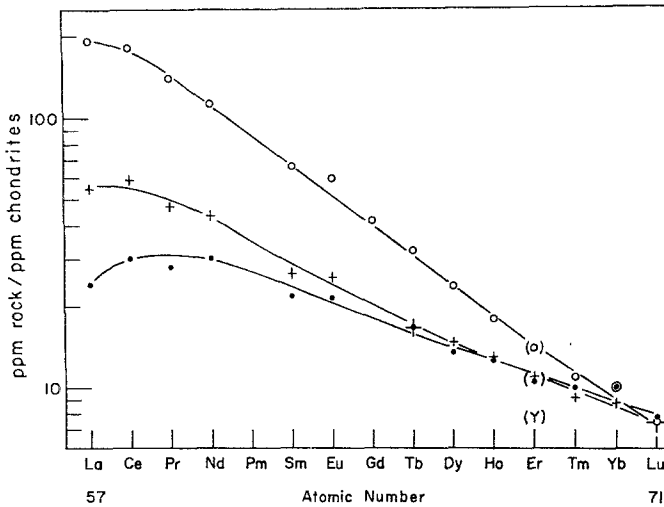


Fig. 5. Rare earth abundance averages relative to 20 chondrites (SCHMITT *et al.*, 1963 and 1964) in: tholeiites, 48–53% SiO₂ (black dots); alkali basalts, 42–47% SiO₂ (crosses); and nepheline-melilite basalts, 36–39% SiO₂ (open circles), see Table 1 and 2 for data.

Table 2. Rare earth concentration averages in parts per million^a

	Tholeiites	Alkali basalts	Nephelinites
SiO ₂ %	48–53	42–47	36–39
Avg. of	5	6	4
⁵⁷ La	7.2 ± 1.1	16.5 ± 2.6	58.4 ± 11.9
⁵⁸ Ce	25.8	49.8 ± 8.8	154 ± 22
⁵⁹ Pr	3.4 ± 0.6	5.7 ± 0.9	17.2 ± 2.3
⁶⁰ Nd	17.5 ± 3.1	25.3 ± 3.8	67.8 ± 12.5
⁶² Sm	4.6 ± 0.6	5.5 ± 0.9	14.0 ± 1.7
⁶³ Eu	1.6 ± 0.3	1.9 ± 0.3	4.4 ± 0.5
⁶⁴ Gd	—	—	13.4 ± 1.4
⁶⁵ Tb	0.82 ± 0.10	0.81 ± 0.12	1.58 ± 0.03
⁶⁶ Dy	4.2 ± 0.6	4.5 ± 0.5	7.3 ± 1.2
⁶⁷ Ho	0.72 ± 0.08	0.73 ± 0.08	1.0 ± 0.1
⁶⁹ Tm	0.25 ± 0.04	0.23 ± 0.04	0.27 ± 0.09
⁷⁰ Yb	1.7 ± 0.3	1.5 ± 0.2	1.7 ± 0.4
⁷¹ Lu	0.24 ± 0.04	0.23 ± 0.03	0.23 ± 0.05
³⁹ Y	18.9 ± 2.6	19.7 ± 1.6	24.6 ± 6.1

^a (±) values are mean deviations.

Models for linking the tholeiites and the alkali olivine basalts, or the nepheline-melilite basalt by fractional crystallization were suggested on the basis of meager rare earth data of common rock-forming silicate mineral available at the time, or crystal-chemistry reasoning (SCHILLING and WINCHESTER, 1966). Since then, rare earth partitioning factors between “phenocryst-volcanic matrix” applicable to this study, have been determined by SCHNETZLER *et al.* (1967). Their results on augite and feldspar from the Mauna Loa ankaramite are particularly pertinent to this study and support the model we previously suggested for deriving alkali

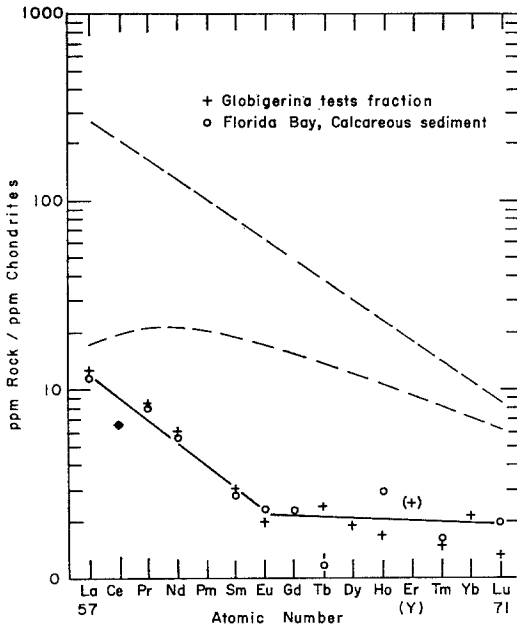


Fig. 6. Abundances of the rare-earth elements relative to 20 chondrites (SCHMITT *et al.*, 1963 and 1964) in two sedimentary materials plotted on logarithmic scale as a function of the atomic number. The two dashed curves represent the olivine tholeiite (lower curve) and a nepheline-melilite basalt (upper curve) from the Hawaiian Islands. Data for the globigerina tests are from SPIRN (1965) and for the Florida Bay sample from HASKIN and GEHL (1962)

basalts from tholeiitic melts by extraction of augite, olivine and a limited amount of plagioclase feldspar. No adequate rare earth data on orthopyroxenes are yet available for checking the olivine-orthopyroxene extraction model we suggested for producing a derivative nepheline-melilite rich melt from a tholeiitic melt. Other fractional crystallization models for production of such strongly silica undersaturated melts are also plausible on the basis of rare earth patterns. For example, extraction from a tholeiitic melt at depth, of garnet whose lattice strongly discriminates against the light rare earths (GOLDSCHMIDT and PETERS, 1931; SAHAMA, 1936; GAVRILOVA and TURANSKAYA, 1958; and HASKIN *et al.*, 1966), could produce a residual melt with the nepheline-melilite rare earth pattern. Although plausible, these two models cannot yet be distinguished with our present knowledge. Rare earth data on additional phenocrysts and ultramafic inclusions found in some of these lavas may help the choice of mechanisms.

As we have previously demonstrated theoretically (SCHILLING, 1966; SCHILLING and WINCHESTER, 1967), mechanisms other than fractional crystallization for deriving independently the three magma types are also plausible. The characteristic rare earth patterns shown in Fig. 5 can apparently also be obtained by different *degrees of melting* of mantle material. The tholeiite would be derived by a relatively slightly larger degree of melting than the alkali olivine basalt and the nepheline-melilite basalt by a very small degree of melting. However the two magmatic processes (i.e. *fractional crystallization* and *partial melting* models) are on the rare earth basis alone, at present, undistinguishable and equally likely.

DALY's (1944) suggestion that nepheline bearing melts (e.g. nepheline-melilite basalts) could be produced simply by assimilation of calcareous sediments can easily be tested by rare earth scrutiny. The rare earths in carbonate-bearing sediments and limestones are shown in Fig. 6 with the olivine tholeiite, and one

nepheline-melilite basalt as well. The limestones are consistently very low in rare earth abundances, and assimilation of such sediments by basalts in any proportions can hardly produce the high light rare earth enrichment and extreme fractionation observed in the nephelinites. The resulting rare earth pattern for mixing alone would have to be intermediate between the original melt (e.g. tholeiite) and assimilated material (e.g. limestone).

Finally, the noticeable increase of the heavy rare earths with decreasing ionic size found for the trachyte merit brief comments. The same increase, although less marked, is observable for the late stage differentiates such as the mugearite, syenitic and granitic rocks. Volatiles may be important in determining the distribution of rare earths inasmuch as the smaller rare earth ions form stronger complexes in solution. Pegmatites which involve volatile transport show also a strong enrichment of the heavy rare earths (SAHAMA and VÄHÄTALO, 1939). Hence the rare earth data would appear to be in no contradiction with SHAND (1933) and TURNER and VERHOOGEN (1960) suggesting that volatile transport may have played an important role in the formation of trachyte.

In conclusion, it should be realized that the rare earth patterns observed in these lavas can significantly be influenced by (1) rare earth composition of source rocks in the mantle (i.e. "source effect"), (2) minerals involved during processes of partial melting and ascent of lavas to the surface (i.e. "mineral effect"), (3) mechanism of magma formation, fractionation and evolution including mixing processes (i.e. "dynamical or kinetic effects"), and possibly but to a lesser extent by (4) selective formation and transport of volatile rare earth complexes (i.e. "complexing effect").

This study establishes that the rare earth abundances in these lavas are closely and systematically related to their petrochemistry. Once factors influencing "whole rock" rare earth patterns in lava are better understood, important inferences on mantle composition and origin of volcanic rocks may be gained.

Acknowledgements. We are indebted to G. FAURE, J. L. POWELL, and J. B. THOMPSON, for furnishing samples analyzed. The technical assistance of GEORGE CIHOCKI, Miss MARY UNTZ, Miss CHARLOTTE EMERY, and PETER FOLEY is greatly appreciated. The experimental work was supported in part by the U.S. Office of Naval Research, contracts Nonr 1841 (74), and Nonr 396 (08), and by the National Aeronautics and Space Administration, grant NsG-496 administered through the Center for Space Research at M.I.T., Cambridge, Massachusetts.

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