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TRACE ELEMENT ABUNDANCES OF THE LATE TERTIARY VOLCANIC ROCKS IN SOUTHWEST HOKKAIDO

by

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(with 11 text-figures and 1 table)

Abstract

Abundances of trace elements including REE were determined for 22 Late Tertiary (Late Miocene to Pliocene) volcanic rocks from southwest Hokkaido. Geochemical features of the Late Tertiary volcanic rocks show that LIL elements, K_2O and Th, increase at a given SiO_2 content across the arc from the Pacific to the Japan Sea side, being similarly to those of the Quaternary volcanic rocks in the same region. However, the Late Tertiary volcanic rocks are higher in Hf/Yb ratios than the Quaternary rocks. This geochemical feature suggests that the former rocks may have derived from a more enriched source mantle than the source material of the latter ones.

Introduction

It is well established that the consistent increase in incompatible element concentrations of volcanic rocks away from the trench occur for many arc systems (Kuno, 1959, 1966; Katsui, 1961; Dickinson and Hatherton, 1967; Hatherton and Dickinson, 1969; Katsui et al., 1978). It has also been claimed that, during the development of an arc system, there is a consistent change with time from primitive island arc tholeiites to alkalic or shoshonitic rocks through calc-alkali rocks (Jakeš and Gill, 1970; Jakeš and White, 1972; Ringwood, 1974). Arculus and Johnson (1978), however, pointed out several exceptions to the above tendency of change in arc magmas with time, i.e. incompatible element contents at a given SiO_2 content either increase often with time in Fiji (Gill, 1970) and Mariana-Bonin arc (Shiraki et al., 1978), or decrease with time in the Cascades (White and McBirney, 1979) and Antilles (Gunn et al., 1974).

Abundant Late Cenozoic volcanic rocks occur in southwest Hokkaido in the northern part of the north Honshu arc. The Early to Middle Miocene volcanic rocks have the nature of non-orogenic volcanic rocks as evidenced by their incompatible element abundances, i.e. high P_2O_5, Hf and TiO_2 contents (Okamura et al., 1984). The Miocene volcanic rocks exhibit a striking contrast in geochemical aspects to the Quaternary volcanic rocks of typical island arc in the same region (Katsui et al., 1978). Thus, an important problem of when the island arc system commenced in southwest Hokkaido arises in studying magmatism as well as tectonics evolution.

Recently the author determined trace element abundances of the Late Miocene to Pliocene volcanic rocks from southwest Hokkaido. In the present paper, their com-

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positional variations in space and time are discussed in comparison with both the Early Miocene volcanic rocks and the Quaternary rocks from the same region.

Geological setting and samples

Southwest Hokkaido is located in the northern part of the north Honshu arc, and characterized by the extensive distribution of the Late Miocene to Pliocene volcanic rocks as well as the Quaternary ones (Text-fig. 1). These Late Tertiary volcanic rocks are arranged in a few zones of NW-SE trend. Matsui (1955) pointed out that the geologic structure with NW-SE trend has been formed during Late Miocene and Pliocene time (the Kuromatsunai stage). These volcanic rocks are composed mainly of andesitic hyaloclastites which intercalate basaltic and rhyolitic hyaloclastites. The volcanic activity which yielded such a large amount of volcanic materials occurred generally under shallow submarine environment, in terms of both central (Okamura, 1984) and fissure type of eruption (Yamagishi, 1982).

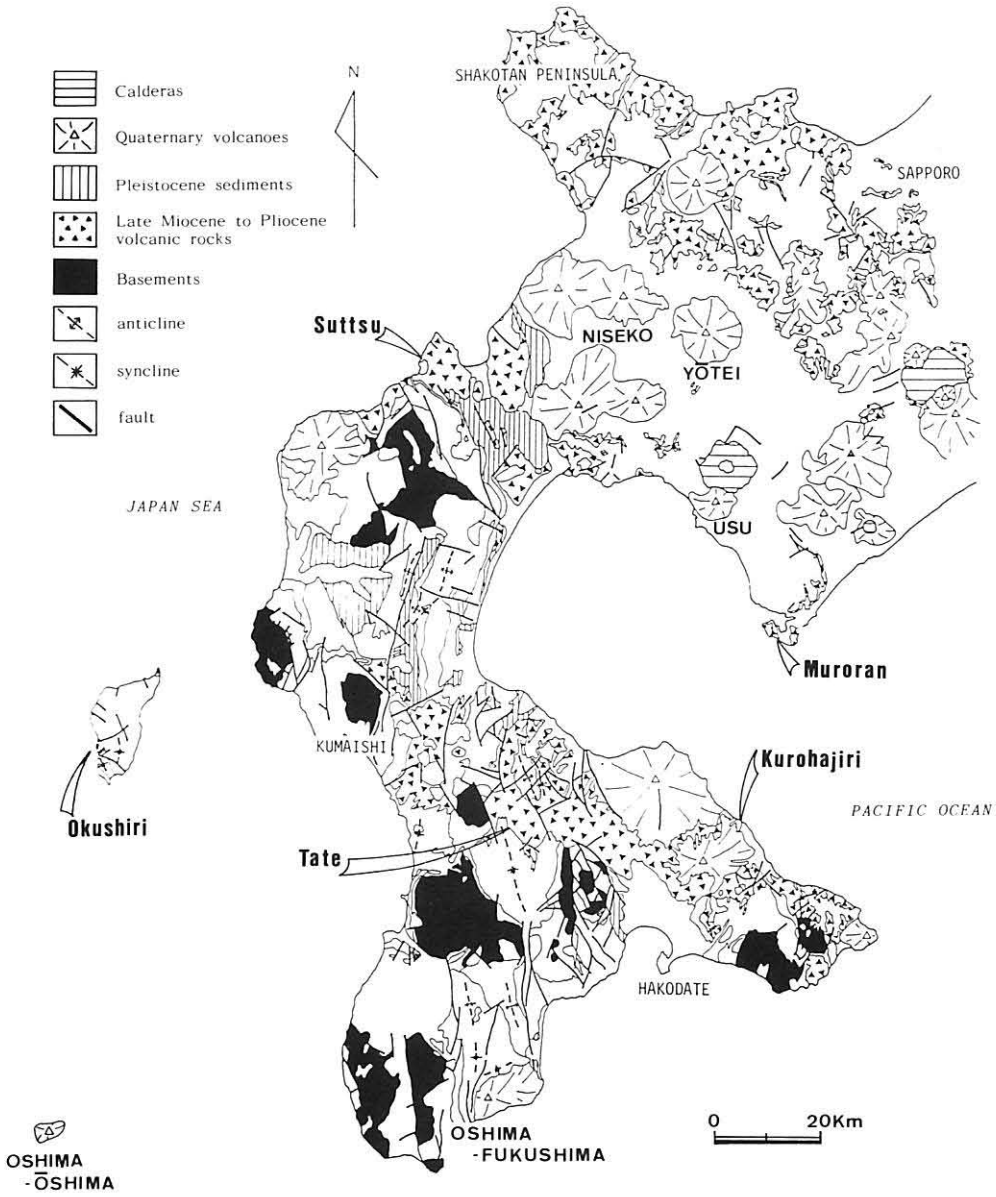
The samples of the Late Miocene to Pliocene volcanic rocks were collected for chemical analysis from Suttsu, Okushiri and Tate on the Japan Sea side, and Muroran and Kurohajiri on the Pacific side (Text-fig. 1). Major and trace elements were analyzed for 22 volcanic rocks. Most of the rocks, except the Kurohajiri rocks, belong to the hypersthenic rock series (Kuno, 1954) which is regarded as the calc-alkali rock series. The Kurohajiri rocks, however, are comprised to the pigeonitic rock series (ditto) which is regarded as the tholeiitic rock series. The rocks of the Japan Sea side carry commonly hornblende and biotite phenocrysts, on the other hand, those of the Pacific side do not. The petrographic character of these Late Miocene to Pliocene volcanic rocks is similar to that of the Quaternary volcanic rocks (Katsui et al., 1978).

Major elements were analyzed by X-ray flourescence. The abundances of trace elements including REE were determined by instrumental neutron activation analysis at the Atomic Energy Research Laboratory of the Musashi Institute of Technology following the method of Masuda et al. (1975).

Results and discussion

The analytical data are listed in Table 1. The Japan Sea side volcanic rocks show a strong increase in K_2O and Th abundances with increasing SiO_2, but the Pacific side volcanic rocks bear only a slight increase (Text-figs. 2 and 3). As shown in Text-figs. 2 and 3, at any given SiO_2 content, there is a progressive increase in both K_2O and Th contents of volcanic rocks from the Pacific side to the Japan Sea side. This increasing tendency of K_2O and Th contents toward the Japan Sea side is found also in the Quaternary volcanic rocks in the same region (Katsui et al., 1978).

The chondrite-normalized patterns of rare earth elements (REE patterns) for the Suttsu, Tate and Kurohajiri volcanic rocks are shown in Text-figs. 4, 5 and 6, respectively. The Suttsu and Tate volcanic rocks on the Japan Sea side are characteristically enriched in light REE, whereas the Kurohajiri volcanic rocks on the Pacific side show



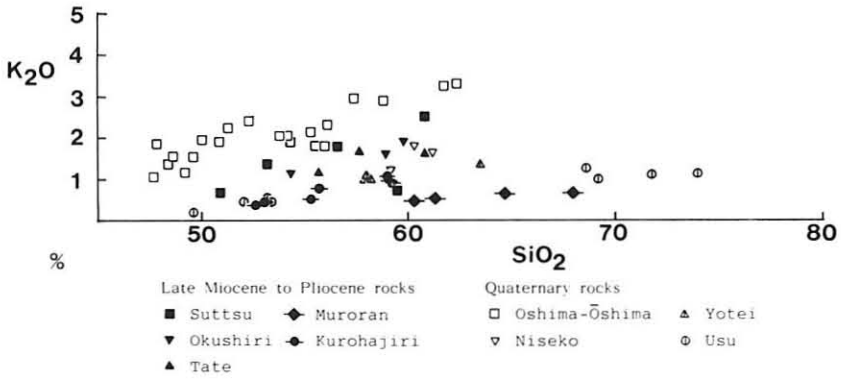
Text-fig. 1 Simplified geologic map of southwest Hokkaido (modified from Takahashi ed., 1980).

flat patterns subparallel to the chondritic REE patterns of the island arc tholeiites as defined by Jakeš and Gill (1970). An increase in light REE relative to heavy REE of the rocks from the Pacific to the Japan Sea side is also found in the Quaternary volcanic rocks in southwest Hokkaido (Katsui et al., 1978). However, in respect to other chemical features, the Early to Middle Miocene volcanic rocks in southwest Hokkaido

Table 1 Trace element abundances in the Late Tertiary volcanic rocks from southwest Hokkaido

Sample	S-21	S-16	S-18	S-25	S-23	T-30	T-28	T-25	Ok-07	Ok-10	Ok-11
wt%											
SiO_2	51.02	53.26	60.91	56.61	59.52	55.70	57.78	60.87	54.30	59.79	58.91
K_2O	0.65	1.34	2.52	1.77	0.62	1.12	1.62	1.61	1.10	1.90	1.56
ppm											
Hf	2.49	3.28	4.06	3.91	3.21	2.85	2.32	2.92	2.98	2.07	2.59
Th	2.82	3.98	7.02	4.46	5.12	3.99	4.64	4.96	3.70	6.88	7.14
La	12.84	9.92	17.25	12.66	11.94	10.93	16.81	13.45	13.37	19.62	15.43
Ce	20.49	26.27	37.88	40.11	23.69	36.90	38.56	40.64	35.53	44.58	35.55
Sm	3.22	2.52	4.36	3.46	3.11	2.41	4.07	2.94	4.38	4.11	3.18
Eu	1.07	0.87	1.18	1.28	0.86	0.73	0.91	0.79	1.98	1.45	1.47
Yb	2.03	1.19	2.90	2.41	2.02	1.82	2.77	2.12	1.58	2.66	2.36
Lu	0.49	0.40	0.61	0.56	0.46	0.31	0.35	0.43	0.34	0.39	0.33

S-21 Augite-olivine basalt, Suttsu. (Tsukikoshi Pyroclastics Formation; Okamura, 1984)
S-16 Augite-olivine basalt, Suttsu. (ditto)
S-18 Augite-hypersthene andesite, Suttsu. (ditto)
S-25 Hypersthene-augite-andesite, Suttsu. (ditto)
S-23 Augite-hornblende andesite, Suttsu. (ditto)
T-30 Augite-hypersthene andesite, Tate. (Annoro Pyroclastic Rocks; Ishida et al., 1975)
T-28 Hypersthene-augite andesite, Tate. (ditto)
T-25 Hypersthene-augite andesite, Tate. (ditto)
Ok-07 Hypersthene-augite andesite, Okushiri. (Yoneoka Formation; Hata et al., 1982)
Ok-10 Hypersthene-augite andesite, Okushiri. (ditto)
Ok-11 Hypersthene-augite-hornblende andesite, Okushiri. (ditto)



Text-fig. 2 K_2O versus SiO_2 diagram for the Late Tertiary and Quaternary volcanic rocks in southwest Hokkaido. The Quaternary volcanic rocks are from Katsui et al. (1978) and Yamamoto (1984).

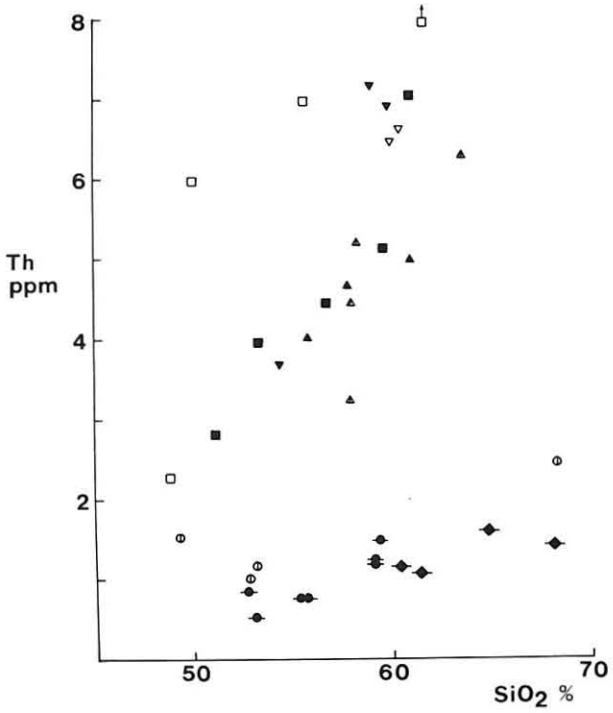
have considerably different nature from the island arc volcanic rocks. Okamura et al. (1984) showed that the Oshima-Fukushima district at the southwestern end of southwest Hokkaido, are distinguished from island arc basalts by the high TiO_2, Hf contents and low Th/Hf ratios, but similar to some non-orogenic alkali basalts in continental region. The subsequent Middle Miocene volcanic rocks in the same regin, however, consist of low-K andesites which are characteristically high in Na_2O and TiO_2 contents and enriched in LREE, which distinguishes them from island arc tholeiites.

Table 1 (continued)

K-45	K-46	K-47	K-48	K-51	K-52	K-53	M-76	M-77	M-78	M-79	Sample
											wt%
52.71	53.10	55.39	55.78	59.08	59.03	59.36	68.01	64.80	61.41	60.36	SiO_2
0.36	0.43	0.51	0.76	0.95	1.05	0.82	0.69	0.69	0.58	0.42	K_2O
											ppm
2.33	2.28	2.71	2.34	3.16	5.89	4.98	8.02	3.30	2.43	2.74	Hf
0.86	0.51	0.78	0.73	1.21	1.19	1.48	1.41	1.59	1.05	1.15	Th
4.97	3.83	3.27	6.23	8.86	19.07	16.14	32.66	11.54	5.07	5.89	La
10.70	n.d.	10.43	17.54	20.94	53.71	38.95	33.92	20.54	15.36	13.16	Ce
2.75	2.18	1.90	2.59	3.89	9.21	7.39	9.75	4.40	2.62	2.59	Sm
0.88	0.81	0.59	n.d.	1.15	2.26	n.d.	2.39	0.76	0.67	0.70	Eu
2.54	1.18	1.53	1.87	2.63	8.26	4.64	4.90	3.09	2.09	2.11	Yb
0.27	0.27	0.51	0.36	0.46	1.42	0.81	0.63	0.61	0.41	0.27	Lu

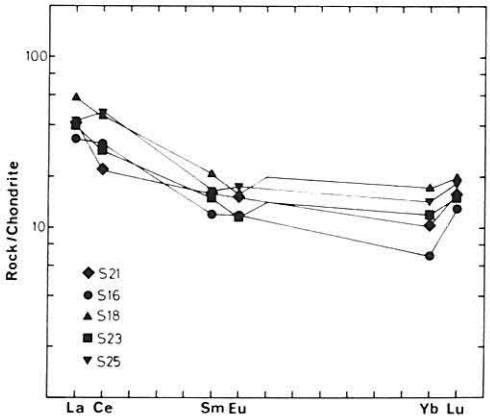
n.d. = not determined.

K-45 Augite-hypersthene basalt, Kurohajiri. (Kurohajiri Agglomerates; Konoya et al., 1967)
K-46 Augite-hypersthene basalt, Kurohajiri. (ditto)
K-47 Plagioclase-phyric andesite, Kurohajiri. (ditto)
K-48 Plagioclase-phyric andesite, Kurohajiri. (ditto)
K-51 Plagioclase-phyric andestie, Kurohajiri. (ditto)
K-52 Hypersthene-augite andesite, Kurohajiri. (ditto)
K-53 Augite-hypersthene andesite, Kurohajiri. (ditto)
M-76 Hypersthene-augite dacite, Muroran. (Muroran Formation; Osanai and Sako, 1953)
M-77 Augite andesite, Muroran. (ditto)
M-78 Hypersthene-augite andesite, Muroran. (ditto)
M-79 Hypersthene-augite andesite, Muroran. (ditto)

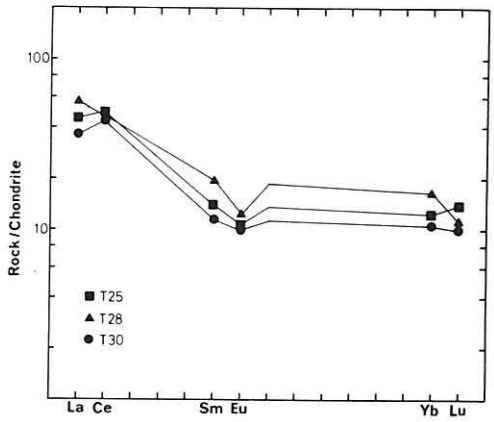


Text-fig. 3 Th versus SiO_2 diagram. Symbols same as in Text-fig. 2

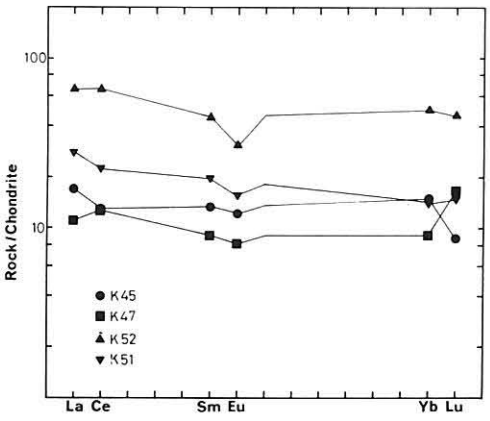
These geochemical variations of volcanic rocks with geologic time suggest that arc magmas have commenced to erupt since Late Miocene in southwest Hokkaido,



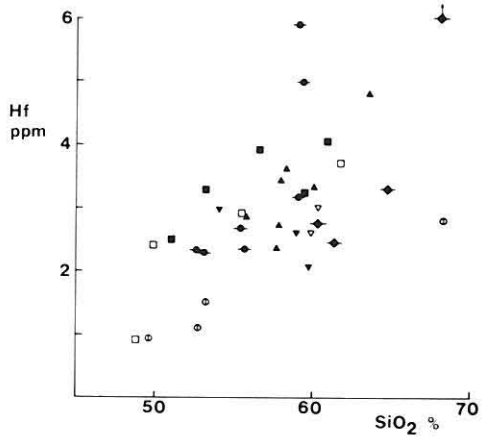
Text-fig. 4 Chondrite-normalized REE patterns for the Late Tertiary volcanic rocks from Suttsu.



Text-fig. 5 Chondrite-normalized REE patterns for the Late Tertiary volcanic rocks from Tate.



Text-fig. 6 Chondrite-normalized REE patterns for the Late Tertiary volcanic rocks from Kurohajiri.

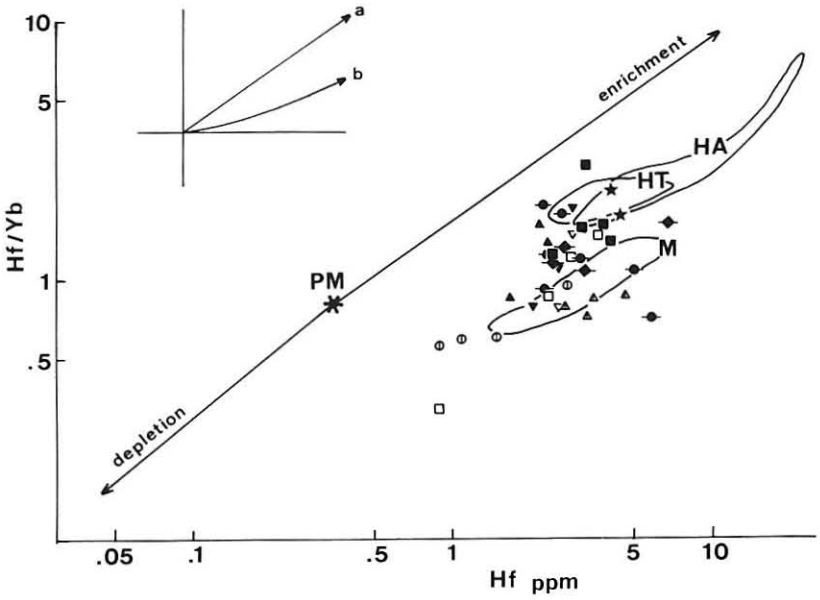


Text-fig. 7 Hf versus SiO_2 diagram. Symbols same as in Text-fig. 2.

although the tectonic movement to build the island arc system may have preceded to this age.

The Hf-SiO_2 relation is shown in Text-fig. 7. Hf has a low radius/charge ratio and is comprised in high field strength (HFS) elements. Consequently it behaves as an incompatible element in magmatic systems (Saunders et al., 1980). Both the Late Tertiary and Quaternary volcanic rocks exhibit no apparent spatial variations in Hf abundances, but the former rocks are generally higher in Hf content at the SiO_2 poor range than the latter.

The Hf/Yb-Hf relation is shown in Text-fig. 8. This kind of diagram was originally used by Pearce and Norry (1979) as Zr/Y versus Zr diagram. Hf and Zr are grouped

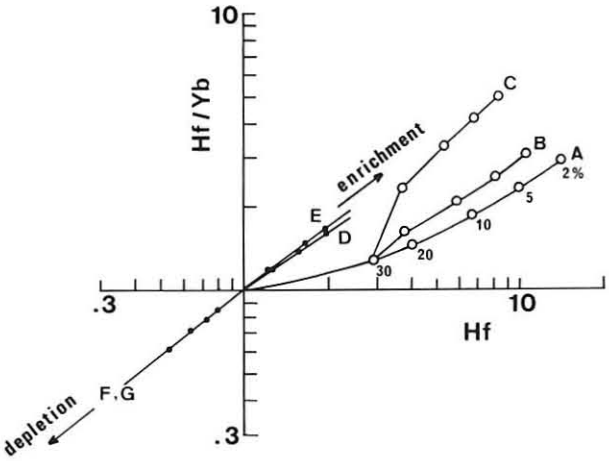


Text-fig. 8 Hf/Yb versus Hf diagram. Star = Early Miocene basalts from Oshima-Fukushima district (Okamura et al., 1984); HA = Hawaii alkali series basalts; HT = Hawaii tholeiites (Basaltic Volcanism Study Project, 1980); M = Reykjanes Ridge basalts (Wood et al., 1979); PM = Primordial Mantle (Wood et al., 1979 and Ganapathy et al., 1974). a = source enrichment (no garnet); b = partial melting trend (no garnet). Other symbols same as in Text-fig. 2.

with the HFS elements, and Yb and Y appear to occupy an intermediate position between large ion lithophile (LIL) and HFS elements (Saunders et al., 1980). Text-fig. 9 shows petrogenetic modeling observed when plotted Hf/Yb against Hf following the method of Pearce and Norry. A, B and C indicate equilibrium partial melting trends, and give composition of melts derived from a mantle source at various degree of partial melting, assuming the melt remains in equilibrium with the residue prior to segregation. The trend A represents the melt composition from a fertile mantle (starting composition) with an assemblage $Ol_{0.6}Opx_{0.2}Cpx_{0.1}Pl_{0.1}$, and the trend B does that with an assemblage $Ol_{0.6}Opx_{0.2}Cpx_{0.2}$. Similarly, the trend C does that with an assemblage $Ol_{0.6}Opx_{0.2}Cpx_{0.1}Gt_{0.1}$. The implication of the model is that, unless garnet is a residual phase ($D^{Yb}_{Gt/\ell} > 4$ for basic melt composition; Cox et al., 1979), the degree of partial melting will affect significantly the Hf content of the melt but only slightly the Hf/Yb ratio.

D, E, F and G indicate source heterogeneity trends. The enrichment trends D and E show changing source composition as successive melt increments are added, while the depletion trends F and G show changing source composition as successive melt increments are removed. The trends D and F represent the melt composition from a mantle with an assemblage $Ol_{0.6}Opx_{0.2}Cpx_{0.1}Pl_{0.1}$, and the trends E and G do that with an assemblage $Ol_{0.6}Opx_{0.2}Cpx_{0.2}$.

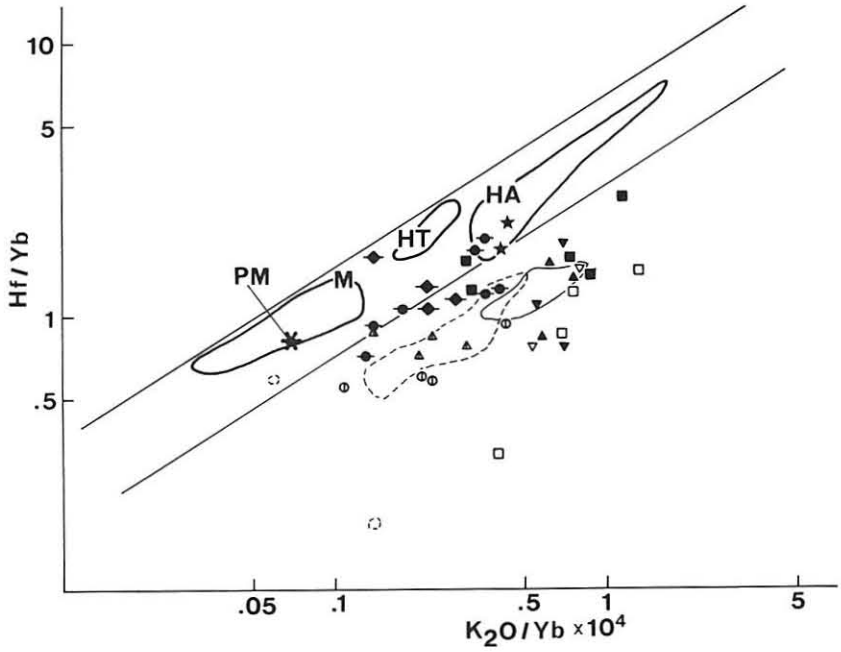
As shown in Text-fig. 8, the Late Tertiary and Quaternary volcanic rocks in



Text-fig. 9 Petrogenetic model showing the effects of some key variables on Hf/Yb ratios and Hf concentration. A, B and C = equilibrium partial melting trends. Calculation based on equation $C^1/C^\circ = 1/(F+D(1-F))$, C^1 and C° are element concentrations in melt and source, respectively. F is the weight proportion of melt, and D is the bulk distribution coefficient. D, E, F and G = source heterogeneity trends. Enrichment events D and E are based on equation $C^\circ(\text{enr.}) = (C^\circ + FC^1) / (1+F)$. Depletion events F and G are based on equation $C^\circ(\text{depl.}) = (C^\circ - FC^1) / (1-F)$.

southwest Hokkaido increase in Hf/Yb ratios with increasing Hf. The Late Tertiary basalts and basaltic andesites are clearly higher in Hf/Yb ratios than the Quaternary rocks (Usu and Oshima-Oshima). This trend can be explained from the modeling in two ways; a systematic heterogeneity of source materials, and variation in degree of partial melting of a garnet lherzolite source material (Text-fig. 9). If the source material is spinel lherzolite, the Late Tertiary volcanic rocks may have originated from more enriched mantle than the source material of Quaternary rocks. If the source material is garnet lherzolite, heavy REE abundances should decrease rapidly from high to low degree of partial melting. However, both the Late Tertiary and Quaternary basalts exhibit practically no apparent differences in heavy REE abundances (Table 1; Katsui et al., 1978, Table 6). Accordingly, the partial melting solution of garnet lherzolite source seems implausible. It is worthy to note that the Early Miocene basalts from Oshima-Fukushima district are higher in Hf/Yb ratios than the Late Tertiary and Quaternary basalts (Text-fig. 8). This suggests that the source material of the volcanic rocks in southwest Hokkaido has become more depleted with geologic time since Early Miocene.

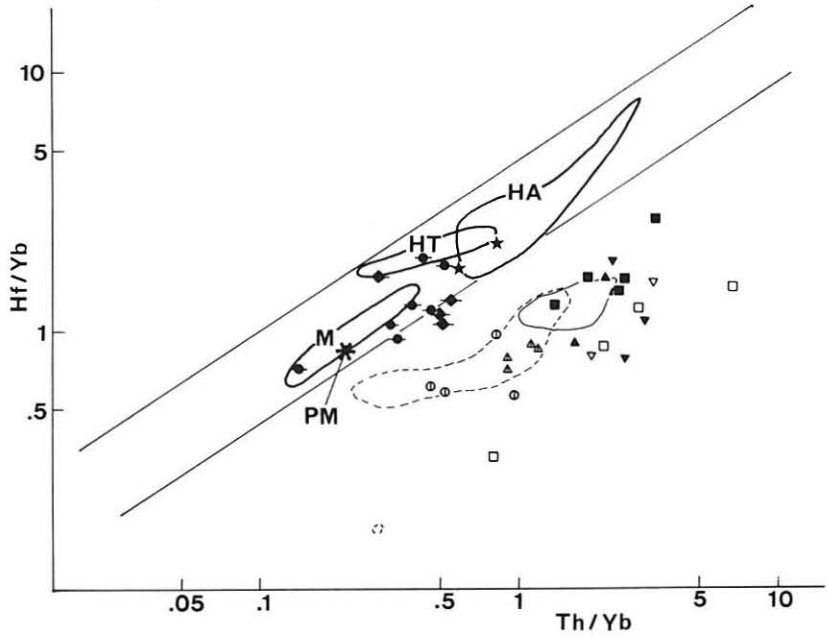
Text-figs. 10 and 11 show Hf/Yb-K_2O/Yb and Th/Yb relations, respectively. The normalized factor used (Yb) is, as in the previous diagrams, effective in largely eliminating variations due to partial melting and fractional crystallization (Pearce, 1982). K_2O and Th are characterized by their low ionic potential (charge/radius) and hence their greater tendency to be mobilized by aqueous fluids (Pearce, 1982, 1983). Pearce pointed out that since within plate enrichment events enrich Hf, K and Th equally, its vector on these diagrams has a slope of unity, whereas the subduction events which cause transport of LIL element via aqueous fluids affects K and Th but not Hf. The Late Tertiary volcanic rocks exhibit a shift to higher K_2O/Yb and Th/Yb ratios relative to the restricted band of non-orogenic basalts (mid-ocean ridges and ocean islands) and the primordial mantle composition estimated by Wood (1979) (Text-fig. 10). This band is termed "array of basalts from non-subduction settings" by



Text-fig. 10 Hf/Yb versus K_2O/Yb diagram. Compositional fields of the Quaternary volcanic rocks from the Chokai Zone (dashed line) and from the Nasu Zone (solid line) of northeast Japan (Masuda, 1979) are shown. The band shown by two parallel lines is the array of basalts from non-subduction settings of Pearce (1983). Other symbols same as in Text-figs. 2 and 8

Pearce (1983), and accounts for mantle variations. The Quaternary basalts exhibit a shift to higher K_2O/Yb and Th/Yb ratios than the Late Tertiary basalts from the array. This evidence can be interpreted presumably in terms of the introduction of more aqueous fluids into the mantle source region in Quaternary than in Late Tertiary. The Kurohajiri and Muroran volcanic rocks on the Pacific side are plotted close to the array, and hence seem to have been only slightly affected by the subduction events of Pearce (1983). The Early Miocene basalts from Oshima-Fukushima district plot in the array (Text-figs. 10 and 11), suggesting that the Early Miocene volcanism have not yet been affected by the subduction events.

Accordingly, during the development of an arc system in southwest Hokkaido, the effects of aqueous fluids into the mantle source region have increased from Late Miocene to Quaternary. The Early Miocene basalts in southwest Hokkaido are distinguished from island arc basalts by high TiO_2, Hf and low Th/Hf, but similar to some non-orogenic alkali basalts in continental region (Okamura et al., 1984). Hence, it is considered that the magmatism from Early Miocene to Quaternary has occurred possibly in relation to the geotectonic movement which converted from a continental environment to an island arc one.



Text-fig. 11 Hf/Yb versus Th/Yb diagram. Symbols same as in Text-figs. 2, 8 and 10.

Conclusion

The Late Miocene to Pliocene volcanic rocks in southwest Hokkaido increase in K/Si, Th/Si and LREE/HREE ratios away from the present trench. A similar pattern of variation in LIL element abundances is also found in the Quaternary volcanic rocks in the same region.

The basalts of Late Miocene to Pliocene have high Hf/Yb ratios relative to the Quaternary basalts in southwest Hokkaido. The difference in Hf/Yb ratios between both of these basalts is probably related to source heterogeneities.

The Quaternary volcanic rocks exhibit a shift to higher K_2O/Yb and Th/Yb ratios than the Late Tertiary volcanic rocks from the array of basalts from non-subductin settings of Pearce (1983). This tendency suggests increasing of mobile elements into the mantle source region with time.

The Early to Middle Miocene volcanic rocks in southwest Hokkaido have different nature from the island arc volcanic rocks, especially the Early Miocene basalts are similar to non-orogenic alkali basalts in continental region (Okamura et al., 1984). This suggests that arc magmas have commenced to erupt since Late Miocene.

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