Mantle metasomatic enrichment in LILE of basalt magma sources beneath the Northeast Japan arc, as indicated by the LILE/Y-Zr/Y plots

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The Nb/Y-Zr/Y plot implies that, before mantle metasomatism, magma sources for NE Japan arc basalts were more depleted in incompatible elements as a result of previous extraction of magmas than those for the N-type mid-oceanic ridge basalts. The LILE/Y-Zr/Y plots imply that the NE Japan arc basalts are derivatives from mantle regions metasomatically enriched in LILE probably by fluids released from the subduction zone. The degree of alkali (Rb and K) enrichment is lower in the trench side than in the back-arc side, but that of Sr enrichment is higher in the trench side. The alkalic volcanism in the back-arc region, with the exception of that belonging to the Chokai volcanic zone, is affected by mantle metasomatism to limited extents.

Introduction

The high field strength elements (HFSE) such as Zr and Y are similarly resistant to secondary mobility, while large ion lithophile elements (LILE) such as K and Rb are very easily removed by hydrothermal and other alteration processes (e.g. Cann, 1970; Ludden *et al.*, 1982). In addition, recent dehydration experiments on serpentine at 850°C and 12 kbar show that the LILE are much more easily transported by water vapor than Y (Tatsumi *et al.*, 1986). Therefore, the behavior of LILE must be significantly different from that of HFSE during metasomatism under upper mantle conditions (Pearce, 1983; Tatsumi *et al.*, 1986). On the other hand, both the HFSE and LILE behave similarly during mafic magmatism under crustal and upper mantle conditions (e.g. Wood *et al.*, 1979; Pearce, 1983); they are collectively said to be "incompatible".

The purpose of this paper is to evaluate the effects of mantle metasomatism in the

petrogenesis of basalts from the Northeast Japan arc and the adjacent volcanoes using available analyses on the HFSE and LILE. The conclusions reached are partly different from those by previous studies (Nakamura *et al.*, 1985; Sakuyama and Nesbitt, 1986). The elements Nb, Zr and Y are chosen here as representatives of HFSE, and Rb, K and Sr as representatives of LILE, because abundant reliable data sets are available for these elements. The following assumptions are made to simplify the discussions: (1) elemental diffusion does not cause heterogeneity of a magmatic liquid; and (2) no separate fluid phase is present during generation and differentiation of basic magmas. In this paper, "metasomatism" refers restrictedly to alteration of rocks not by magmatic melts but by fluids.

Rb/Y-Zr/Y plot

The significance of the Rb/Y-Zr/Y plot, a representative of the LILE/Y-Zr/Y plots, is first studied.

Theory

Crystal-liquid reactions: The tops of arrows in Fig. 1a indicate representative compositions calculated for liquid and solid phases yielded by melting of a primitive mantle (PM, $Rb/Y=0.17$ and $Zr/Y=2.7$; Taylor and McLennan, 1981). Recently estimated compositions for primitive (or primordial) mantle are fairly similar with respect to the Rb/Y and Zr/Y ratios among several investigators ($Rb/Y=$

Table 1. Crystal-liquid partition coefficients used in model calculations

	Ol	Opx	Cpx	Gar	Amph	Pl
Rb	0.0098	0.022	0.031	0.042	0.29	0.071
K	0.0068	0.014	0.038	0.015	0.96	0.17
Sr	0.014	0.017	0.12	0.012	0.46	1.83
Nb	0.01	0.15	0.1	0.1	0.8	0.01
Zr	0.01	0.03	0.1	0.3	0.5	0.01
Y	0.01	0.2	0.5	2.0	1.0	0.03

Data sources: Arth (1976) and Pearce and Norry (1979). Abbreviations: as in Fig. 1.

Fig. 1. Theoretical compositional vectors for (a) partial melting of a mantle source, (b) crystallization differentiation of a basalt magma, and (c) mantle metasomatism. Used crystal-liquid partition coefficients: Table 1. Abbreviations: Amph, amphibole; Cpx, clinopyroxene; Opx, orthopyroxene; Gar, garnet; Ol, olivine; Pl, plagioclase; PM, primitive mantle ($Rb=0.48$ ppm, $Zr=7.8$ ppm and $Y=2.9$ ppm; Taylor and McLennan, 1981); and PMMZ, primitive mantle magmatism zone (see text).

(a) Compositions calculated using the batch partial melting equation (eq. 4 in Arth, 1976) for liquid and solid phases formed by 20% melting leaving a harzburgite residue ($Ol_{70}Opx_{25}Cpx_5$) are respectively at the tops of solid line arrows, and those by 1% melting leaving a garnet lherzolite residue ($Ol_{85}Opx_{20}Cpx_{10}Gar_5$) are at the tops of broken line arrows. Dotted line: mixing trend between the residue of 20% melting and liquid of 1% melting, with numerals near open circles indicating wt. % of the liquid to mix.

(b) Composition calculated using the Rayleigh fractionation equation (eq. 8 in Arth, 1976) for a liquid by 50% fractional crystallization of each mineral is at the arrow top. Vectors for olivine and plagioclase: covered by solid circles showing the parental basalt composition.

(c) Vector for enrichment of primitive mantle in incompatible elements is indicated by an open arrow on the assumption that Zr and Y are equally least mobile during mantle metasomatism.

0.16–0.18 and $Zr/Y = 2.3\text{--}2.7$ (e.g. Wood *et al.*, 1979; Taylor and McLennan, 1981, 1985)). Since the bulk solid-liquid partition coefficient, D is calculated to be $Rb \leq Zr < Y < 1$ for Fig. 1a, the liquid phase inevitably lies at the upper right side, and the residual solid phase at the lower left side, of the source in the $\log Rb/Y$ – $\log Zr/Y$ plot.

The presence of accessory minerals in the refractory residue may considerably modify the D values for incompatible elements. As for the NE Japan arc magmagenesis, however, this possible effect of accessory minerals is likely to be negligible (Sakuyama and Nesbitt, 1986; Ujike, in preparation).

The dotted curve indicates a compositional trend for materials formed by mixing of two end members; a refractory residual phase produced by 20% melting with a liquid phase produced by 1% melting (Fig. 1a). This curve is applicable to the case in which a mantle source which had undergone previous extraction of a magma is newly enriched in incompatible elements by injection of a magma ascended from a deeper mantle region. Significantly, the compositions so formed are close to those represented by a straight line connecting the two end members.

During differentiation of basic magmas by fractional crystallization of common phenocrystic minerals, the liquid composition varies toward the upper right side by separation of amphibole, clinopyroxene and/or orthopyroxene in the $\log Rb/Y$ – $\log Zr/Y$ plot (Fig. 1b). The liquid composition remains nearly constant in terms of Rb/Y – Zr/Y by separation of olivine and plagioclase. In Fig. 1b, vectors illustrating fractionation effects are drawn for melts from which as much as 50% of each mineral has precipitated. A vector for magmatic differentiation must be shorter than that for fractionation of amphibole and clinopyroxene, because the major phenocrysts are commonly

olivine and/or plagioclase in basalts. It is likely, therefore, that a compositional difference of basalts from their source on the Rb/Y – Zr/Y plot is more dependent on the degree of partial melting than on magmatic differentiation.

It is clear from the above considerations that all derivatives by magmatic crystal-liquid reactions from the primitive mantle, as well as their mixing products, fall within a tight zone with a slope close to 45° in the $\log Rb/Y$ – $\log Zr/Y$ plot. This compositional zone, including the derivatives from a primitive mantle by one or two steps of magmatic reactions, is here called the primitive mantle magmatism zone (PMMZ, Fig. 1c). The slope of PMMZ is estimated to be the same as the broken line arrow pointing the composition of the 1% melt in Fig. 1a.

Mantle metasomatism: As discussed above, the mobility of elements by metasomatic fluids is Rb (and other LILE) $\gg Y \approx Zr$ under upper mantle conditions. Therefore, a mantle material can be enriched or depleted in Rb by metasomatism, while the Zr and Y contents will be only slightly changed. In the $\log Rb/Y$ – $\log Zr/Y$ plot, the vectors for mantle metasomatism are vertical and considerably different from PMMZ (Fig. 1c). The compositions of magmas derived from regions enriched in incompatible elements by mantle metasomatism will plot above PMMZ.

Parenthetically, it should be added here, however, that a sample which plots within PMMZ is not always a product from a primitive mantle. For example, a source region which had previously lost Rb , and then regained Rb by two distinct episodes of mantle metasomatism, may generate a magma with a composition within PMMZ, but this magma is by no means generated from a *primitive* mantle.

Observation

In order to test the validity of vectors for crystal-liquid reactions and PMMZ both theoretically estimated above, data from well-

documented igneous suites are plotted in the Rb/Y-Zr/Y diagram below.

Magmatic differentiation: Two examples of *in situ* differentiation products, the tholeiitic Karroo dolerite sill (Eales and Robey, 1976) and an alkalic sill, Hunter Valley (Gamble, 1984), are cited in Fig. 2. Despite the fact that the

samples represent two distinct magma series, the plots both show rather short elongations subparallel to PMMZ. Thus the vectors in Fig. 1b and their average slopes appear to be applicable to the natural magmatic differentiation.

Magma from a primitive mantle: A

Fig. 2. Rb/Y-Zr/Y relations for some igneous rocks, compared with PMMZ. Intrusive rocks from Karroo, Africa (Eales and Robey, 1976) and Hunter Valley, New South Wales, Australia (Gamble, 1984): *in situ* differentiation products. Selected high-K volcanic rocks with $\text{MgO} > 5.0\%$ from the Roman Region (Appleton, 1972; Cundari, 1979; Rogers *et al.*, 1985): derivatives from mantle sources metasomatically enriched in incompatible elements. Arrows: compositional variations during processes through which the magmas formed (directions are of particular significance, but positions are only of approximation). Mean composition of 7 Archean volcanic rocks (excluding No. 16 affected by strong K-metasomatism) with $\text{SiO}_2 = 54-65\%$ and $[\text{La}/\text{Yb}]_N = 1.0-3.0$ from Noranda (Ujike and Goodwin, 1987): derivative from a primitive mantle source. PM and PMMZ: as in Fig. 1.

detailed geochemical study (Ujike and Goodwin, 1987) on immobile trace elements indicates that Archean subalkalic volcanic rocks with chondrite-normalized La/Yb ratios between 1 and 3 in the Central Noranda area are products by partial melting of a primitive mantle source and subsequent fractional crystallization. Although the individual Rb analyses are unlikely to represent the magmatic composition due to the high secondary mobility of Rb, if severely altered samples are omitted, characteristic values of Rb content are defineable for each volcanic sequence (Ludden *et al.*, 1982). The mean composition of selected intermediate volcanic rocks from Noranda, those least affected by alteration, falls within PMMZ as expected (Fig. 2).

Magmas from highly enriched mantle regions: Recent isotopic and geochemical studies on high-K magmas erupted in the Roman Region, Italy, indicate that these magmas were generated from mantle sources metasomatically enriched in incompatible elements (Holm and Munksgaard, 1982; Rogers *et al.*, 1985). Such magmas should have significantly higher Rb/Y ratios than PMMZ. The Roman high-K volcanic rocks (Appleton, 1972; Cundari, 1979; Rogers *et al.*, 1985) plot upward away from PMMZ as expected (Fig. 2). A possible compositional path through which the volcanic rocks formed is also shown in Fig. 2, although the starting point of such a path is uncertain.

Applications

It has been indicated above that using the Rb/Y-Zr/Y plot, a magma genetically affected by mantle metasomatism is generally distinguishable from that produced by crystal-liquid reactions from a primitive mantle. Thus the Rb/Y-Zr/Y plot is applied to basaltic rocks ($\text{SiO}_2 \leq 53\%$ and $\text{SI} = 100\text{MgO}/(\text{MgO} + \text{FeO} + 0.9\text{Fe}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O}) > 30$) from the NE

Japan arc and the adjacent volcanoes. In addition, other incompatible element ratios (Nb/Y, K/Y and Sr/Y) are also examined to study (1) the ultimate nature of mantle regions which have melted to produce the basaltic magmas and (2) the relative behavior of individual LILE in the magmagenesis. The Sr/Y ratio of basalts will be significantly modified from that of the magma sources if abundant plagioclase is fractionally crystallized. However, the used analyses are those on less differentiated basalts, and therefore they would be affected by fractional crystallization of plagioclase to limited extents.

Quaternary volcanoes on the NE Japan arc have been divided geographically into two zones, the Nasu zone on the Pacific (=trench) side and the Chokai zone on the Japan Sea (=back arc) side (Fig. 3). The analyses cited from the literature (see the caption to Fig. 4 for the sources) are those on tholeiites from the Nasu zone, high-alumina, calc-alkali and alkali basalts from the Chokai zone, and alkali basaltic rocks from volcanic islands in and around the Japan Sea (Fig. 3). Although alkali basalt occurs on the Oshima-oshima Island which does situate in the Japan Sea, the Oshima-oshima analysis is excluded from the category of "alkalic rocks around the Japan Sea" in this paper because it belongs to the Chokai zone.

Nb/Y-Zr/Y plot

The nature of the magma source mantle regions is studied in terms of the Nb/Y-Zr/Y relation (Fig. 4) which is least changed by mantle metasomatism (e.g. Pearce, 1983). Also shown in Fig. 4 is a compositional zone (NMSMZ: N-type MORB source magmatism zone) within which derivatives by crystal-liquid reactions from an average source of N-type mid-oceanic ridge basalts (N-MORB SOURCE, open star: Wood *et al.*, 1979) will fall. The NMSMZ has been estimated with similar calcu-

Fig. 3. Outline map showing location of the Chokai and Nasu volcanic zones and alkalic volcanic islands around the Japan Sea. Alkalic volcanic islands, not all of which were formed in the Quaternary, around the Japan Sea (solid stars): DG, Dog (or Take); JJ, Jeju; ODZ, Oki-Doen; RI, Rishiri; and UL, Ulreung. Depth contour of deep-earthquake foci: Yoshii (1978).

lations to those for PMMZ. Significantly, the majority of the NE Japan basalts, from both of the Nasu and Chokai zones, falls within this zone (NMSMZ). It is indicated, therefore, that the ultimate sources for the NE Japan basalts could be compositionally analogous to the N-type MORB source, in agreement with arguments by earlier workers (e.g. Sakuyama and Nesbitt, 1986).

However, most of the Nasu zone analyses are lower in Zr/Y than the average N-type MORB source. This feature indicates that the mantle sources are more depleted by previous extraction of magmas for the NE Japan arc basalts than for N-type MORB. A mantle wedge beneath the NE Japan arc, which was essentially MORB type, may have repeatedly

served as a magma source.

All the analyses on alkali basaltic rocks around the Japan Sea (ALK-JS in Fig. 4; see Fig. 3 for the localities) but that from the Rishiri Island (RI) have Nb/Y even higher than PMMZ. The Rishiri basalt close to NMSMZ in Fig. 4 is likely to have been derived from an N-MORB type source as the Nasu and Chokai zone basalts. On the other hand, other alkali basaltic rocks around the Japan Sea, with high Nb/Y, are likely to have been generated from a source similar to that of ocean island basalts (OIB: Nakamura *et al.*, 1985).

LILE/Y-Zr/Y plots

In the Rb/Y- and K/Y-Zr/Y plots (Figs. 5a, b), basalts from the Chokai zone plot farther above NMSMZ than those from the Nasu zone.

Fig. 4. Nb/Y-Zr/Y relations for basalts ($\text{SiO}_2 \leq 53\%$ and $\text{SI} > 30$) from the NE Japan arc (Chokai and Nasu zones) and alkalic volcanic rocks around the Japan Sea (ALK-JS). Data sources: Lee *et al.* (1982), Yoshida *et al.* (1982, 1983), Kaneko *et al.* (1983), Hayashi *et al.* (1984), Ishikawa *et al.* (1984), Sakayori *et al.* (1984), Yoshida and Aoki (1984), Nakagawa *et al.* (1985), Oba *et al.* (1985), Sasaki *et al.* (1985, 1986), Kim (1986), Kim *et al.* (1986) and Sakuyama and Nesbitt (1986). Abbreviations: as in Fig. 1; RI, Rishiri; and NMSMZ, magmatism zone within which derivatives from N-MORB SOURCE (average composition of sources for N-type MORB estimated by Wood *et al.*, 1979) will lie. Crystal-liquid partition coefficients used to draw PMMZ and NMSMZ: Table 1.

The simplest interpretation is that relative to the Nasu-zone magmas, the Chokai-zone magmas were produced (1) from mantle sources metasomatically more enriched in Rb and K, as suggested by the higher Rb/Y and K/Y, and (2) by smaller degrees of partial melting, as suggested by the higher Zr/Y. This interpretation

is consistent with petrologic evidence that magmas from the Chokai zone contain more water than those on the Nasu zone (Sakuyama, 1979). The water content in a magma may be increased either by hydration of the magma source due to metasomatism, or by decreased degree of partial melting due to the small D for

Fig. 5. LILE/Y-Zr/Y relations for basalts from the NE Japan arc (Chokai and Nasu zones) and alkalic volcanic rocks around the Japan Sea (ALK-JS). Data sources and Abbreviations: as in Fig. 4.

H₂O or OH. A possible mechanism for mantle metasomatism beneath the Northeast Japan arc is the recycled addition of LILE to the mantle wedge overlying the subduction zone, from crustal materials of the subducting Pacific slab (e.g. Nakamura *et al.*, 1985; Sakuyama and Nesbitt, 1986).

It is difficult to evaluate the effects of contamination of magmas by a continental crust through which the magmas ascended to the surface, since the composition of the contaminant is unknown. However, crustal contamination is unlikely to have caused high Rb/Y and K/Y in the Chokai basalts, since the continental igneous rocks seem to be characterized by Rb/Y close to PMMZ in general (e.g. Hunter Valley and Karroo, Fig. 2). Thus the systematic difference in Rb/Y and K/Y

between the Chokai-zone and Nasu-zone volcanics (Figs. 5a, b) will not be ascribed to the difference in degree of crustal contamination.

In the Sr/Y-Zr/Y plot (Fig. 5c), the analyses of Nasu basalts are farther above NMSMZ than those of the Chokai basalts. This implies that, with respect to Sr, the degree of metasomatic enrichment is greater for the Nasu magma sources than for the Chokai sources, being in agreement with a result of a Sr isotope study (Notsu, 1983). It is highly probable that Sr behaves differently from Rb and K during mantle metasomatism beneath the NE Japan arc.

The above interpretation is totally different from that proposed by Sakuyama and Nesbitt (1986), where the NE Japan magma sources are estimated to have been

homogeneously enriched. Mineralogic control on the metasomatic fluid composition has been proposed as the process by which individual LILE are differentially added to the magma sources during mantle metasomatism (Nakagawa *et al.*, 1986; Ujike, 1986). This matter will be fully discussed in a separate paper (Ujike, in preparation).

The volcanism of the Chokai zone is traditionally considered (e.g. Kuno, 1959) to be intermediate between the Nasu zone and the alkalic volcanism around the Japan Sea, in chemistry. However, all the alkalic rock analyses (ALK-JS) plot closer to NMSMZ than the Chokai zone analyses in the LILE/Y-Zr/Y plots (Fig. 5). It is indicated that, except for the Oshima-oshima Island basalt belonging to the Chokai zone, mantle metasomatism played less important roles in the generation of alkalic magmas erupted around the Japan Sea than in that of the magmas of the NE Japan arc. In other words, with the exception of the Oshima-oshima volcanism, the alkalic volcanism around the Japan Sea is in no way a consequence of extreme mantle metasomatism.

The low LILE/Y ratios of the Rishiri basalt (Fig. 5) suggest that its source was affected by mantle metasomatism to a minimal extent if any. The magmagenesis of Rishiri basalt is probably characterized by a small degree of partial melting, as opposed to metasomatic enrichment, of the source mantle.

It has been stated on the basis of the K/(Ta+La) ratio and Pb isotopic data (Nakamura *et al.*, 1985) that the effect of metasomatism becomes weaker with increasing distance from the trench in the mantle beneath the Japan Sea and Japanese islands region. The interpretation of Fig. 5 is generally in agreement, in that the alkalic magmas seem to be less affected by mantle metasomatism than the Chokai zone magmas. However, the effect of mantle metasomatism does *not* monotonously decrease

towards the Japan Sea, as implied from the fact that the Chokai-zone analyses plot farther above NMSMZ than the Nasu-zone ones in Figs. 5a, b. The conclusion reached by Nakamura *et al.* (1985) may be oversimplified, possibly due to the sparse density of data points on the NE Japan arc.

The depth of the deep-earthquake foci (Yoshii, 1978) is ≥ 300 km under the localities of the alkalic rocks (solid stars in Fig. 3), whereas it is ≤ 200 km under the Chokai zone. Therefore, the above observation suggests that the mantle wedge above the deep-earthquake foci with depths ≥ 300 km are not appreciably enriched by mantle metasomatism. The subducting Pacific slab is likely to be incapable of generating sufficient metasomatic fluids at depths ≥ 300 km. This consideration is in harmony with a result of a Sr isotope study (Notsu and Kobayashi, 1985): they concluded that material derived from the subducted Pacific slab contributed to negligible extents in the magmagenesis of volcanoes in Hokkaido above the deep-earthquake foci with depths > 200 km. Also, results of melting experiments support this view in that no major hydrous minerals including amphibole, mica, serpentine, talc, etc. are known to be stable at pressures ≥ 100 kb (e.g. Allen *et al.*, 1972; Tatsumi, 1986).

Summary

Fig. 6 summarizes the results of consideration using the Nb/Y- and LILE/Y-Zr/Y plots. Basaltic magmas of the Nasu and Chokai zones were generated from mantle regions enriched in LILE probably transported by metasomatic fluids from the subducted Pacific slab. These NE Japan arc magma sources were, before mantle metasomatism, broadly similar in chemistry to an average N-type MORB source mantle, but the NE Japan arc magma sources were more depleted in incompatible elements by previous magmatism than the N-type

		NASU	CHOKAI	RISHIRI	WJS*
Depth of deep-earthquake foci		100-200km		300km	>400km
Basalt series	{ Subalkali Alkali				
Mantle metasomatic enrichment**	{ Rb K Sr				?
					?
					?
Type of source mantle before metasomatism	{ N-MORB OIB Previous depletion***	Yes	Yes	Yes	?

Fig. 6. Summary of examination using Nb/Y- and LILE/Y-Zr/Y plots. *, alkalic volcanic islands in and around the western part of the Japan Sea (DG, JJ, ODZ and UL in Fig. 3). **, relative degree of enrichment shown by the height of box. ***, due to magmatic extraction and relative to the average N-type MORB source (Wood *et al.*, 1979).

MORB source. Mantle regions below the Nasu zone were metasomatically added more Sr but less Rb and K, relative to those below the Chokai zone.

Alkalic rocks studied are likely to have been derived from three distinct magma sources. Alkali basalts from the Chokai zone (Oshima-oshima) and the Rishiri Island were both generated from mantle regions basically analogous to the N-type MORB source: the Oshima-oshima source with significant mantle metasomatism, whereas the Rishiri one with slight mantle metasomatism. Alkalic magmas erupted on islands in the western part of the Japan Sea, > 400 km above the subduction zone, may be derivatives from mantle regions similar to the ocean island basalt (OIB) source.

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東北日本弧玄武岩マグマの根源物質への LILE の添加—— LILE/Y-Zr/Y 図によるマントル交代作用の検討

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液相濃集元素のうち LILE (Rb, Sr, K) はマントル交代作用で移動しやすく、Nb, Zr および Y は移動しにくい。故に玄武岩の組成を Nb/Y-Zr/Y 図と LILE/Y-Zr/Y 図上で検討すれば、マントルの特徴を知り得る。東北日本弧および利尻火山下のマントルは本来、N 型中央海嶺玄武岩の根源物質に類似する。ただし東北日本弧と利尻火山下のマントルは、かつての火成作用のために後者よりも液相濃集元素に乏しくなっている。東北日本弧の玄武岩マグマは、マントルが交代作用を受けて LILE に富化した後に生じる。交代作用の結果として海溝側（那須火山帯）の玄武岩は相対的に Sr に富み、背弧側（鳥海火山帯）のそれは Rb と K に富む。利尻島下のマントルは交代作用を殆んど受けていない。

島名の英和対照 Dog, 独島（又は竹島）; Jeju, 濟州島; Oki-Dozen, 隠岐島前; Oshima-oshima, 渡島大島; Rishiri, 利尻島; Ulreung, 鬱陵島。