Distinctive REE patterns for tholeiitic and calc-alkaline magma series co-occurring at Adatara volcano, Northeast Japan

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(Received July 12, 1991; Accepted February 26, 1992)

Rare earth element (REE) concentrations were precisely determined by inductively coupled argon plasma/atomic emission spectrometry (ICP) for 9 tholeiitic and 13 associated calc-alkaline rocks from Adatara volcano, located at the volcanic front of Northeast Japan arc. The tholeiitic samples commonly display nearly flat REE patterns, whereas the calc-alkaline samples characteristically show concave light REE-enriched patterns. In both suites, the REEs, with the exception of Eu, are positively correlated to varying extents with K_2O and other incompatible elements. Eu concentrations in the calc-alkaline suite do not vary significantly, resulting in significant negative Eu anomalies. In the tholeiitic suite, chondrite-normalized $(Ce/Yb)_{CN}$ ratios are low and show little variation (1.54–1.80) over a significant range in SiO_2 content (52–62 wt%). In contrast, $(Ce/Yb)_{CN}$ in the calc-alkaline suite show much greater variation (2.17 to 2.94) over a smaller range in SiO_2 content (56 to 62 wt%) and increase systematically with SiO_2 .

REE data for the tholeiitic suite are consistent with a crystal fractionation model that successfully reproduces the observed major- and trace-element variations. A similar fractionation model is compatible with major-element and some trace-element variations for the calc-alkaline suite, but results in excessively high heavy REE contents, and fails to reproduce the elevated $(Ce/Yb)_{CN}$ ratios for the most acidic samples. The preferential enrichment of light REEs may reflect additional processes such as: (1) assimilation, (2) incorporation of a differentiated liquid (or evolved magma) that has undergone fractionation of heavy REE-rich minerals, and (3) mixing of primary magma derived by a small degree of partial melting from a common (amphibolitic or hornblende gabbroic) source.

Although the tholeiitic and calc-alkaline suites possess distinct $^{87}Sr/^{86}Sr$, Rb/Ba and Zr/Nb ratios, different magma sources are not required to produce the distinct REE patterns of the coexisting suites. An equilibrium batch partial melting model can reproduce both the flat (tholeiitic) and concave (calc-alkaline) REE patterns from a common peridotitic source by larger (10–15%) and smaller (5%) degrees of partial melting, respectively. Therefore, both tholeiitic and calc-alkaline suites could have been derived from a common lherzolitic magma source, provided that the calc-alkaline suite underwent incorporation (assimilation) of isotopically distinct (lower crust-derived) materials prior to evolution.

INTRODUCTION

Island-arc tholeiitic suites (Jakes and Gill, 1970) are closely associated with calc-alkaline suites in the outer volcanic zone of the Northeast Japan arc (Nasu volcanic zone; NVZ) (Kawano and Aoki, 1960, Kawano *et al.*, 1961, Yoshida and Aoki, 1984). As reviewed by Fujinawa (1988), several petrologists have tried to determine the evolutionary processes for each magma suite and/or a possible genetic relationship between the suites. On the basis of trace-element

compositions of tholeiitic and calc-alkaline lavas from the NVZ, Masuda and Aoki (1979) argued that these two magma suites could not have been derived from a common parental magma. They instead proposed that both tholeiitic and calc-alkaline parental magmas were generated from the same peridotitic mantle through higher (about 20%) and lower (about 3%) degrees of partial melting. In contrast, based on petrography and mineralogy, Sakuyama (1981, 1983) and Wada (1981, 1985) cited magma mixing as an important process in the evolution of

calc-alkaline magma suites.

Fujinawa (1988) investigated the major-element, trace-element and Sr-isotope geochemistry of lavas at Adatara volcano located in the NVZ, and concluded that: (1) members of the tholeiitic suite are related principally through fractional crystallization of the observed phenocryst phases; (2) a similar fractionation mechanism is dominant in the evolution of the calc-alkaline suite, with subordinate magma mixing as an additional mechanism; and (3) tholeiitic and calc-alkaline lavas were probably derived from different parental magmas, with postulated calc-alkaline parental magma having higher Rb/Ba, Zr/Nb and lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratios than tholeiitic parental magma. Mineralogical data for these samples (Fujinawa, 1990a) are compatible with these conclusions. It may be noteworthy that the fractionation-magma mixing model of Fujinawa (1988) is in accord with recent general discussions on the origin of calc-alkaline suites in island-arc regions (Lühr and Carmichael, 1980, 1985; Kosco, 1981; Grove *et al.*, 1982, 1983; Kay *et al.*, 1982; Grove and Baker, 1984). Recently, Fujinawa (1990b, 1991) argued that high Rb/Ba, Zr/Nb and, possibly, low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the calc-alkaline parental magma could well have been caused by anatexis of lower crust and subsequent incorporation (assimilation) of the anatectic melt into mantle derived basaltic magma, presuming that lower crust beneath the Northeast Japan arc has amphibolitic or hydrous gabbroic composition with $^{87}\text{Sr}/^{86}\text{Sr}$ as low as 0.7030.

Previous work concerning the REE compositions of lavas from the NVZ is apparently not in agreement with the conclusions of Fujinawa (1988, 1990a). By averaging the available REE data of NVZ lavas, Masuda and Aoki (1979) demonstrated that: (1) flat REE patterns are common among the tholeiitic basalts; (2) calc-alkaline andesites are characterized by light REE [LREE]-enriched concave patterns; and (3) the concave upward REE patterns for tholeiitic andesites are virtually indistinguishable from the patterns for calc-alkaline andesites. Unfortunately, Masuda and Aoki did not interpret these

varieties (or variations) in detail. Fujimaki (1980) emphasized the similarity in REE patterns among two tholeiitic samples and their intimately associated one calc-alkaline sample from Iwate volcano located in the northern region of the NVZ, and concluded that temporally and spatially associating tholeiitic and calc-alkaline magmas could have been derived from a common parental magma.

The aim of this paper is to reevaluate, using previously published and new REE data, the magmatic evolution within both tholeiitic and calc-alkaline magma suites and the relationship between these suites. To achieve these goals, it is important to determine: (1) differences (or similarities) in REE patterns between the two suites; and (2) parallelism of REE patterns [i.e. constant $(\text{Ce}/\text{Yb})_{\text{CN}}$ ratios] among the members of each suite. Based on REE data, fractionation models were constructed to examine the role of crystal fractionation in the magmatic evolution of each rock suite. Also, equilibrium partial melting models were examined to test consonance between the two suites.

SAMPLE DESCRIPTIONS

The analyzed samples include 9 tholeiitic and 13 calc-alkaline lavas from Adatara volcano. Major- and trace-element compositions of these samples were presented in previous papers (Fujinawa, 1980, 1984, 1988) along with their petrography and modal compositions. A detailed description of the mineralogy of these samples is available in Fujinawa (1990a). Sample numbers are consistent with those used previously (Fujinawa, 1988, 1990a). Tholeiitic samples range from basalt (SiO_2 ; 52 wt%) through andesite (SiO_2 ; 53%-61.7%), and calc-alkaline samples from basic andesite (SiO_2 ; 55.9%) to acidic andesite (SiO_2 ; 61.7%). Tholeiitic samples commonly contain plagioclase as the most abundant phenocryst phase, followed by hypersthene, augite and titanomagnetite, in order of abundance. Olivine phenocrysts are found in the basic tholeiites, i.e. those containing < 55 wt% SiO_2 . Calc-alkaline samples com-

monly contain plagioclase > hypersthene > augite > titanomagnetite as the dominant phenocryst phases. Rare phenocrysts of olivine, ilmenite and quartz occur in some of the calc-alkaline samples. Quartz phenocrysts are partially resorbed.

ANALYTICAL PROCEDURE AND RESULTS

REE contents were determined by inductively coupled argon plasma/optical emission spectroscopy (ICAP/OES) following the method of Tagiri and Fujinawa (1988) which is based on the technique of Crock and Lichte (1982). The ICAP emission spectrometer employed is a Jarrell-Ash Model 575 set up at Ibaraki University. Two or three replicate analyses were made of each sample (see Table 1 for results of replicate analyses).

Results of REE analyses together with SiO₂ and K₂O contents are listed in Table 2, and chondrite-normalized REE patterns for representative samples are presented in Figs. 1 and 2. Tholeiitic and calc-alkaline suite samples can be easily distinguished from each other on the basis of REE compositions. Tholeiitic samples commonly display nearly flat REE patterns, whereas calc-alkaline samples are characterized by LREE-enriched concave patterns.

In the tholeiitic suite, both LREE and heavy

REEs (HREE) vary sympathetically with K₂O and other incompatible elements. As SiO₂ increases from 52 to 62 wt%, Ce and Yb abundances increase from 14 to 23 ppm and from 2.15 to 3.3 ppm, respectively. However, relative to the increase in REE abundances, chondrite-normalized Ce/Yb ratios [(Ce/Yb)_{CN}] increase only slightly, varying within a narrow range (1.54 and 1.80).

In the calc-alkaline suite, on the other hand, LREEs and, to a lesser extent, HREEs increase with increasing SiO₂, whereas Eu remains almost constant, resulting in significant negative Eu anomalies in the most silicic lavas. As SiO₂ increases from 56 to 62 wt%, (Ce/Yb)_{CN} ratios gradually increase from 2.17 to 2.94.

DISCUSSION

Fractional crystallization models

(1) Tholeiitic suite

A general consensus has emerged among petrologists favoring the generation of island arc tholeiitic suite lavas by fractional crystallization of basaltic parental magmas (Ewart *et al.* 1973; Brown *et al.* 1977; Togashi 1977; Depaolo and Johnson 1979; Masuda and Aoki 1979; Sakuyama 1981, 1983; Gill 1981; Sakuyama and Nesbitt 1986). Major- and trace-element variations for the tholeiitic suite at Adatara volcano

Table 1. Results of replicate ICP analyses for particular samples

Spl. No.	3-4					3-8				
	1	2	3	Ave.	STD	1	2	3	Ave.	STD
La	8.58	8.63	8.62	8.61	0.0216	9.46	10.00	9.58	9.68	0.2315
Ce	22.40	21.90	22.64	22.31	0.3082	23.60	23.70	22.20	23.17	0.6847
Nd	13.40	13.00	12.48	12.96	0.3766	13.20	12.90	12.12	12.74	0.4551
Sm	3.97	3.85	3.91	3.91	0.0490	3.76	3.79	3.56	3.70	0.1020
Eu	1.04	1.03	0.94	1.00	0.0450	0.95	0.99	0.89	0.94	0.0410
Gd	4.07	4.00	4.13	4.07	0.0531	3.78	3.64	3.71	3.71	0.0571
Dy	4.25	4.15	4.17	4.19	0.0432	4.21	4.21	3.89	4.10	0.1508
Er	—	2.81	2.76	2.79	0.025	—	3.27	2.69	2.98	0.29
Yb	2.50	2.53	2.61	2.55	0.0464	2.61	2.63	2.56	2.60	0.0294
Lu	—	0.42	0.38	0.40	0.02	—	0.42	0.39	0.41	0.015
(Ce/Yb) _{CN}	2.29	2.21	2.21	2.24	0.0377	2.31	2.30	2.21	2.27	0.0449

Spl. No.; sample number. (Ce/Yb)_{CN}; chondrite-normalized Ce/Yb ratios. Chondritic values from Masuda *et al.* (1973). STD; standard deviation (σ).

Table 2. Wet chemically determined SiO_2 and K_2O contents (wt%) and REEs analyses (ppm) of whole rock samples by ICAP/OES

Spl. No.	2-1 TH	2-2 TH	2-3 TH	2-4 TH	2-5 TH	2-6 TH	2-7 TH	2-9 TH	2-10 TH	3-1 CA	3-2 CA
SiO_2	51.99	53.14	53.94	54.23	54.52	54.62	56.60	56.85	61.74	55.92	57.12
K_2O	0.28	0.40	0.34	0.29	0.40	0.37	0.42	0.59	0.79	1.17	1.05
La	5.52	7.31	5.59	5.72	5.10	6.09	6.90	7.46	9.28	7.66	8.73
Ce	13.89	16.14	14.53	15.10	14.05	14.66	17.13	18.61	23.19	21.40	23.82
Nd	10.32	12.79	10.53	10.95	9.89	11.99	12.03	13.35	15.32	12.04	13.68
Sm	3.20	3.73	3.28	3.51	3.30	3.57	3.96	3.99	4.82	3.76	4.24
Eu	1.02	1.16	1.00	1.12	0.99	1.15	1.13	1.12	1.35	0.93	1.12
Gd	3.51	3.91	3.56	3.58	3.63	3.97	4.23	4.38	5.26	3.80	4.28
Dy	3.73	4.30	3.94	3.92	3.89	4.30	4.66	4.63	5.79	4.19	4.48
Er	2.37	2.75	2.47	2.67	2.49	2.82	3.19	3.01	3.63	2.75	3.02
Yb	2.16	2.32	2.26	2.19	2.19	2.43	2.69	2.72	3.30	2.51	2.69
Lu	0.28	0.29	0.28	0.29	0.29	0.29	0.36	0.34	0.42	0.35	0.41
$(\text{Ce}/\text{Yb})_{\text{CN}}$	1.64	1.78	1.64	1.76	1.64	1.54	1.62	1.74	1.80	2.17	2.26

Abbreviations are same as those in Table 1. TH; tholeiitic series. CA; calc-alkaline series. Source of SiO_2 and K_2O data is Fujinawa (1988).

Fig. 1. Chondrite-normalized REE plot referenced to the Leedey chondrite (Masuda et al., 1973), showing the patterns for tholeiitic suite samples from Adatara volcano, Japan.

are also consistent with simple crystal fractionation of observed phenocryst assemblages, with 68–60 wt% plagioclase, 21–25% hypersthene, 5–8% augite and 5–8% titanomagnetite $\pm$ olivine as fractionated phases (Fujinawa, 1988). Such a model is supported by Sr-isotopic data, in that $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the tholeiitic lavas lie within a limited range 0.7057 ± 0.0001 (Fujinawa, 1988).

Also, there are mineralogical observations concurring with the crystal fractionation hypothesis. These include: (1) complete absence of olivine in the tholeiitic lavas containing $>55\%$ SiO_2 ; (2) positive correlations of Fe/Mg ratios of mafic silicates and Ca/Na of plagioclase with those of host lavas; and (3) increase in Usp mol% of magnetite phenocryst cores with increasing

Table 2. (continued)

Spl. No.	3-3 CA	3-4 CA	3-5 CA	3-6 CA	3-8 CA	3-9 CA	3-10 CA	3-11 CA	3-12 CA	3-13 CA	3-14 CA
SiO ₂	52.28	57.72	59.02	59.13	59.48	60.11	60.17	60.75	61.00	61.56	61.69
K ₂ O	1.24	1.14	1.16	1.63	1.53	1.58	1.47	1.71	1.37	1.68	1.98
La	8.47	8.61	10.66	10.03	9.68	11.25	11.76	12.40	12.13	10.75	13.18
Ce	22.47	22.31	25.80	25.61	23.17	27.85	28.50	30.07	30.34	26.48	32.28
Nd	12.70	12.96	13.93	13.66	12.74	16.20	15.84	16.58	17.58	14.97	16.93
Sm	3.77	3.91	4.11	3.93	3.70	4.79	4.53	4.35	4.79	4.15	4.74
Eu	0.96	1.00	0.94	0.88	0.94	1.16	0.99	1.01	1.14	0.87	0.93
Gd	3.89	4.07	4.11	3.82	3.71	4.70	4.37	4.28	4.81	4.07	4.38
Dy	4.27	4.19	4.44	4.05	4.10	5.05	4.64	4.84	5.23	4.38	4.58
Er	2.77	2.79	2.84	2.65	2.98	3.22	3.88	3.09	3.31	2.81	2.92
Yb	2.65	2.55	2.82	2.43	2.60	3.09	2.78	2.95	3.14	2.72	2.80
Lu	0.38	0.40	0.41	0.40	0.41	0.45	0.35	0.45	0.46	0.39	0.42
(Ce/Yb) _{CN}	2.16	2.24	2.33	2.68	2.27	2.30	2.61	2.60	2.47	2.48	2.94

Fig. 2. Chondrite-normalized REE plot showing the patterns for selected calc-alkaline suite samples from Adatara volcano.

whole rock SiO₂ (Fujinawa, 1990a). The nearly parallel REE patterns of the tholeiitic samples provide further evidence that these lavas are related through crystal fractionation, because the content of augite, the only phase which would fractionate LREE and HREE in the fractionated phases, was estimated to be as low as 5–8 wt% (Table 3).

In order to test the fractionation hypothesis

of Fujinawa (1988), REE variations in the tholeiitic suite were modelled assuming Rayleigh fractionation of observed phenocryst phases. The weight proportion of phases that must be removed from chosen parent magmas to derive daughter magmas is estimated by averaging successful results of least-squares calculations after the method of Wright and Doherty (1970) (Table 3; Ave. B and Ave. A). Mineral compositions

Table 3. Results of least-squares calculation (after Wright and Doherty, 1970) and weight% of crystals in the fractionated phases

	Parent	Dauthter	MC	Sum(R ²)	Ol	Opx	Cpx	Pl	Liquid	
Tholeiitic series										
1	2-1	2-5	B	0.0520	1.43 (5.8)	1.83 7.4	--- ---	20.51 83.0	0.93 3.8)	74.34
2	2-1	2-6	B	0.0134	--- (---)	5.27 15.6	0.72 2.1	26.33 78.0	1.42 4.2)	66.05
3	2-1	2-7	B	0.0084	--- (---)	6.08 16.0	1.35 3.6	28.10 74.1	2.38 6.3)	60.91
4	2-2	2-6	B	0.0275	--- (---)	6.24 30.9	0.96 4.8	12.34 61.1	0.65 3.2)	79.22
5	2-2	2-7	B	0.0262	--- (---)	7.21 28.6	1.72 6.8	14.47 57.4	1.80 7.1)	73.05
6	2-3	2-7	B	0.0217	--- (---)	7.46 29.0	0.16 0.6	17.00 66.2	1.07 4.2)	73.55
7	2-4	2-7	B	0.0071	--- (---)	4.26 22.4	3.26 17.2	10.52 55.4	0.96 5.1)	79.84
Ave. B					(0.8)	21.4	5.0	67.9	4.8)	
8	2-4	2-7	A	0.0071	--- (---)	5.45 24.7	3.08 14.0	12.73 57.7	0.79 3.6)	76.57
9	2-4	2-9	A	0.0735	--- (---)	7.42 31.8	2.11 9.0	12.48 53.4	1.36 5.8)	75.47
10	2-5	2-7	A	0.0657	--- (---)	2.85 17.0	1.93 11.5	10.12 60.4	1.86 11.1)	83.00
11	2-5	2-9	A	0.0187	--- (---)	4.90 27.2	0.90 5.0	9.71 54.0	2.48 13.8)	82.00
12	2-8	2-10	A	0.1498	--- (---)	8.92 23.8	--- ---	26.99 72.1	1.52 4.1)	62.49
Ave. A					(---)	24.9	7.9	59.5	7.7)	
Calc-alkaline series										
1	3-1	3-6	B	0.0735	--- (---)	3.85 20.5	0.09 0.5	11.75 62.7	3.06 16.3)	81.52
2	3-1	3-7	B	0.0126	1.53 (7.6)	4.50 22.3	1.40 6.9	9.80 48.5	2.98 14.7)	80.36
3	3-5	3-9	B	0.0358	--- (---)	3.95 28.6	1.71 12.4	7.75 56.2	0.39 2.8)	87.10
Ave. B					(2.5)	23.8	6.6	55.8	11.3)	
4	3-5	3-9	A	0.0325	--- (---)	4.26 26.4	1.84 11.4	9.58 59.3	0.47 2.9)	84.81
5	3-5	3-10	A	0.0511	--- (---)	3.86 52.3	0.38 5.1	2.22 30.1	0.92 12.5)	92.56
6	3-5	3-13	A	0.0890	--- (---)	4.31 28.5	0.41 2.7	8.96 59.2	1.45 9.6)	84.45
7	3-5	3-16	A	0.1178	--- (---)	7.24 28.0	1.45 5.6	15.00 58.0	2.16 8.4)	75.60
8	3-10	3-14	A	0.0752	--- (---)	1.77 11.8	0.09 0.6	12.03 80.3	1.09 7.3)	86.11
9	3-10	3-16	A	0.0494	--- (---)	3.64 18.3	1.15 5.8	13.79 69.2	1.34 6.7)	81.71
10	3-11	3-15	A	0.0196	--- (---)	0.14 2.2	2.61 41.2	2.48 39.1	1.11 17.5)	93.91
Ave. A					(0.0)	23.9	10.3	56.5	9.3)	

B and A at the column MC indicates the set of chemical compositions of basic and acidic fractionated phase assemblages, respectively. They are available in Table 6 of Fujinawa (1988). Values in parentheses represent weight% of the crystals in the fractionated phases. Sample Nos. correspond to those in Table 2. Data sources: Fujinawa (1988).

Table 4. Estimates of Ce and Yb concentrations in residual magmas

Tholeiitic series											
SiO ₂ (residual magma)	52	53	54	55	56	57	58	59	60	61	62
SiO ₂ (fractionated phases)			47.0	/				47.2			
DFTb	1.000	0.833	0.714	0.625							
DFTa				1.000	0.886	0.796	0.722	0.661	0.609	0.565	0.527
Ce (MIN) (ppm)	13.1	15.7	18.2	20.7	16.1	18.1	20.1	22.1	24.1	26.1	28.1
Ce (MAX)	13.1	14.8	16.3	17.8	17.4	18.6	19.7	20.9	21.9	23.0	24.0
Yb (MIN)	2.06	2.46	2.85	3.24	2.72	3.02	3.32	3.61	3.91	4.20	4.49
Yb (MAX)	2.06	2.27	2.46	2.64	2.43	2.57	2.70	2.82	2.94	3.05	3.16
(Ce/Yb) _{CN} (MIN)	1.62	1.63	1.63	1.63	1.69	1.70	1.70	1.70	1.70	1.71	1.70
(Ce/Yb) _{CN} (MAX)	1.62	1.66	1.69	1.72	1.69	1.73	1.76	1.78	1.81	1.83	1.86
											1.88
Calc-alkaline series											
SrO ₂ (residual magma)	55.5	56	57	58	59	60	61	62	63		
SiO ₂ (fractionated phases)			43.8		/		46.8				
DFCb	1.000	0.959	0.886	0.824	0.770						
DFCa					1.000	0.924	0.859	0.803	0.753		
Ce (MIN) (ppm)	19.9	20.7	22.4	24.0	25.7						
Ce (MAX)	19.9	20.4	21.4	22.4	23.4	27.6	29.7	31.7	33.7		
Yb (MIN)	2.48	2.58	2.78	2.98	3.18	25.6	26.9	28.1	29.3	30.4	
Yb (MAX)	2.48	2.53	2.62	2.71	2.73	2.94	3.16	3.37	3.58		
(Ce/Yb) _{CN} (MIN)	2.05	2.05	2.06	2.05	2.06	2.82	2.91	3.00	3.08		
(Ce/Yb) _{CN} (MAX)	2.05	2.06	2.08	2.11	2.13	2.39	2.40	2.40	2.40	2.40	
					2.39	2.43	2.46	2.49	2.52		

Degrees of fractionation, DFTb, DFTa, DFCb, and DFCa, are estimated in terms of weight fractions of residual magmas relative to chosen parent magmas, Tb, Ta, Cb, and Ca, respectively. Data source of the degree of fractionation is Fujinawa (1988).

used are the average phenocryst core compositions listed in Table 5 of Fujinawa (1988). Thus, the values of weight proportion are identical to those used for examining trace-element variations by Fujinawa (1988). Degrees of fractionation are estimated in terms of weight fractions of residual magmas relative to chosen parent magmas, which is also the same as that employed to examine the trace-element variations (Table 4). These are calculated by an addition-subtraction method using SiO₂ contents in both whole rock

samples and fractionated phases (cf. Fujinawa, 1988). Tholeiitic basalt magma (LDM of Fujinawa, 1988) containing 52 wt% SiO₂ and andesite magma with 55 wt% SiO₂ are chosen as the parents for basaltic to andesitic residual (daughter) magmas < 55 wt% SiO₂ and andesitic residual magmas > 55 wt% SiO₂, respectively, in that olivine phenocryst perfectly disappears in tholeiitic lavas containing > 55 wt% SiO₂. For the calculations, Ce and Yb were chosen to represent the LREEs and HREEs, respectively (Table

4). Ce and Yb abundances for the 2 chosen parents are approximated on the basis of best fits through all the data plots on silica variation diagrams (Table 4). Crystal-liquid partition coefficients are from the literature (Table 5). Maximum and minimum trends in Fig. 3 are drawn assuming maximum and minimum values of D_{Ce} , D_{Yb} (Table 5; column A), respectively.

Data plots of the tholeiitic lavas come very close to the estimated variation trends, especially, maximum trend lines (Fig. 3), indicating that the REE data also support the crystal fractionation hypothesis. Furthermore, it should be noted that the successful fractionation model requires the highest values for both D_{Ce} and D_{Yb} quoted in Table 5. These partition coefficients (D_s) are considerably higher than the D_s used in fractionation modelling of other incompatible elements in the earlier paper (Fujinawa, 1988).

(2) Calc-alkaline suite

The role of crystal fractionation in the evolution of the calc-alkaline suite was examined with the same method used for modelling the tholeiitic suite. The least differentiated magma (LDM of Fujinawa, 1988) with 55.5 wt% SiO_2

and an arbitrarily chosen andesite magma containing 59 wt% SiO_2 are regarded as the parents for basic to intermediate residual (daughter) magmas < 59 wt% SiO_2 and acidic residual magmas > 59 wt% SiO_2 , respectively. Ce and Yb abundances employed for these parents are also approximated values based on the best fits on the silica variation diagrams. In spite of the general success in reproducing the major- and some trace-element variations (Fujinawa, 1988), fractionation models consistently failed to reproduce the low HREE contents and the relatively high $(Ce/Yb)_{CN}$ ratios observed for acidic members of the suite (Fig. 3). Predicted fractionated mineral assemblages containing < 12 wt% augite (Table 3) are not supported by the REE data, because augite is the only phase which can significantly fractionate Ce/Yb ratios (Schnetzler and Philpotts, 1970; Henderson, 1984; Fujimaki *et al.*, 1984). Thus, compositional variations in the calc-alkaline suite cannot be explained solely by crystal fractionation of observed phenocrystic phases.

A significant contribution by other process(es) is required to explain the observed enrichment of LREEs relative to HREEs. Several proc-

Table 5. Partition coefficients employed in fractionation model

	Ce		Eu		Yb	
	A	B	A	B	A	B
olivine	0.008–0.03	0 (0–0.005)	0.005–0.05	0 (0–0.005)	0.03–0.07	0 (0–0.004)
ortho-pyroxene	0.03–0.33	0.006 (0.001–0.01)	0.03–0.42	0.02 (0.0036–0.05)	0.1–0.67	0.08 (0.029–0.2)
clino-pyroxene	0.043–0.65	0.05 (0.033–0.17)	0.09–2.0	0.48 (0.14–0.75)	0.09–1.5	0.6 (0.16–1.0)
plagioclase	0.02–0.3	0.12	0.02–2.11 0.02–0.4 (TH) 0.5–1.0 (CA)	0.2	0.006–0.3	0.067
magnetite	0.06–0.82		0.06–0.66		0.09–1.0	
spinel		0.015 (0–0.03)		0.025 (0–0.05)		0.05 (0–0.1)

A and B; partition coefficients for crystal/andesitic liquid and crystal/basaltic liquid, respectively. TH; tholeiitic series, CA; calc-alkaline series. Values in parentheses are ranges of partition coefficients for crystal/basaltic liquid. Data sources: Onuma *et al.* (1968), Schnetzler and Philpotts (1970), Bornhorst (1980), Nicholls and Harris (1980), Gill (1981), Villemant *et al.* (1981), Dostal *et al.* (1983), Fujimaki *et al.* (1984), Henderson (1984), and Luhr and Carmichael (1985) for A; Baxter *et al.* (1985), Feigenson *et al.* (1983), and Irving and Frey (1984) for B.

Fig. 3. $(\text{Ce}/\text{Yb})_{\text{CN}}$ vs. Ce diagram showing plots of whole rock analyses and fractionation paths predicted on the basis of Rayleigh fractionation models. Solid circles=tholeiitic samples, open circles=calc-alkaline samples. Solid and broken lines indicate the trend lines for the tholeiitic and calc-alkaline magma series, respectively. Tb, Ta, Cb, and Ca, are 4 chosen parent magmas: Tb and Cb refer to tholeiitic and calc-alkaline LDMs of Fujinawa (1988), Ta=tholeiitic andesite containing 55 wt% SiO_2 , and Ca=calc-alkaline andesite with 59 wt% SiO_2 . Each fractionation trend is denoted according to SiO_2 contents of the residual magmas.

esses could produce such an enrichment, including: (1) assimilation; (2) incorporation of liquid (or highly evolved magma) that has undergone fractionation of HREE-rich minerals into differentiating magma; and (3) mixing of small partial melts derived from a common source material. In each case, the two end-members, i.e. host magma and assimilant or end-member magmas, need to possess similar $^{87}\text{Sr}/^{86}\text{Sr}$, Rb/Ba and Zr/Nb ratios, since these ratios remain almost constant through the suite.

Model (3) is not plausible without an assumption that lower crust-derived mafic materials such as amphibolite or hornblende gabbro are the principal source of calc-alkaline magmas (Takahashi and Tomiya, 1990), because simultaneous increase of both $(\text{Ce}/\text{Yb})_{\text{CN}}$ and Ce with SiO_2 would be brought about either by some fractionation mechanism or mixing of acidic component. Ultramafic source can not generate acidic (dacitic to rhyolitic) end-member magma by partial melting under probable

melting conditions beneath NVZ.

The negative Eu anomalies observed for calc-alkaline suite lavas likely reflect both the dominance of plagioclase in the fractionated phase assemblages (Table 3) and the high D_{Eu} value for plagioclase. Petrographically observed apparent partition coefficients (separated plagioclase phenocryst/groundmass) for Eu range as high as 1.0 for calc-alkaline samples from the NVZ (Odaka, unpublished data). In contrast, the apparent D_{Eu} values for plagioclase are consistently low ($D_{\text{Eu}}=0.2\text{--}0.4$) for NVZ tholeiites. D_{Eu} is known to be dependent on the redox state of magmas; under extreme reducing conditions, the D_{Eu} approaches a value similar to that of D_{Sr} (Drake and Weill, 1975). However, the apparent D_{Eu} s are quite opposite to what is expected from the inferred $f\text{O}_2$ conditions for the two magma series at Adatara volcano. Fujinawa (1990a) considered, on the bases of mineralogy and phase relations of phenocrystic oxides, that the tholeiitic magma evolved under

lower fO_2 and higher temperature (100 °C difference) conditions than the calc-alkaline magma. The difference of apparent D_{Eu} s might reflect temperature dependence more than fO_2 dependence, because a linear relationship between $\ln D_{Eu}$ and inverse temperature is experimentally observed in oceanic ridge basalt (Sun *et al.*, 1974).

Batch partial melting model

An equilibrium partial melting model was applied to test whether the distinct REE patterns of tholeiitic and calc-alkaline suite lavas could have been generated from a common magma source by different degrees of partial melting. Spinel lherzolite is employed as the source material, because low-alkali tholeiitic magmas in the NVZ are believed to segregate from underlying lherzolitic upper mantle at the pressure around 10 kb, where spinel is thought to be the most probable stable aluminous phase (Tatsumi, 1986; Nakagawa *et al.*, 1988). The modelling is based on the following assumptions: (1) the spinel lherzolite consists of 58% olivine, 25% orthopyroxene, 15% clinopyroxene and 2% spinel by weight, corresponding to the modal composition of KLB-1, the most undepleted spinel lherzolite xenolith from Kilborne Hole, New Mexico (cf. Takahashi, 1986a); (2) the lherzolite has a flat REE pattern, and contains tentatively the same REE abundances as the Leedey chondrite (Masuda *et al.*, 1973); (3) REE partitioning between source material (C_0) and partial melt (C_L) is adequately described by the equation $C_L/C_0 = 1/[D_0 + F(1 - P)]$, where F is the degree

of partial melting, and D_0 and is the bulk partition coefficients related to modal source composition, and P is modal melt composition (i.e. modal composition of minerals entering into melt), respectively (Shaw, 1970); and (4) the melting relations and ratio of the phases incorporated into the melt in each melting interval are those listed in Table 6.

Results indicate that varying degrees of partial melting of a common lherzolitic source can account for the distinct REE patterns displayed by tholeiitic and calc-alkaline suite lavas (Fig. 4). Less than 5% partial melting can generate concave REE patterns similar to those observed for calc-alkaline lavas, whereas greater than 10% partial melting can produce virtually flat REE patterns similar to those observed for tholeiitic lavas. These results coincide with the findings of Masuda and Aoki (1979).

The fact that the postulated parental magmas are lower in REE abundances than the most mafic lavas observed (eg. 2-1, 3-1) can well be explained by presuming a source with higher REE abundances and/or earlier (deep-level) olivine-dominated fractionation of primary magmas. Indeed, several estimates of mantle compositions (Jagoutz *et al.*, 1979; Hart and Zindler, 1986) have 1.5 to 2.5 times chondritic REE abundances. In view of the fact that Mg and Ni contents are consistently low in both rock suites, it is likely that even the most basic lavas underwent such fractionation before their eruption. Several petrologists have favored such a mechanism to explain the low Mg and Ni contents in basalts and andesites from the NVZ (Tatsumi *et al.*,

Table 6. Assumed melting relations among the phases in spinel lherzolite and the ratio of phases incorporated into the melt in the melting intervals from 0–5%, 5–25%, and >25%

Degree of partial melting	Phase			
	olivine	orthopyroxene	clinopyroxene	spinel
0–5%	1	1	4	4
5–25%	1	spinel out	4	
>25%	1	clinopyroxene out		
		1

Fig. 4. Chondrite-normalized Ce, Eu and Yb plot showing predicted REE patterns based on an equilibrium partial melting model. Chondritic values from Masuda *et al.* (1973).

1983; Yoshida and Aoki, 1988; Sakuyama and Nesbitt, 1986; Nakagawa *et al.*, 1988; Fujinawa, 1988).

Implications for magmagenesis

Despite the REE evidence presented, distinct $^{87}\text{Sr}/^{86}\text{Sr}$, Rb/Ba and Nb/Zr ratios between tholeiitic and calc-alkaline lavas (Fujinawa, 1988) are apparently inconsistent with the view that the two parental magmas beneath Adatara volcano were directly produced from a common lherzolitic mantle source.

Fujinawa (1990b, 1991) proposed anatexis of lower crust and subsequent incorporation of the anatectic melt into an initial magma segregated from a mantle diapir as the most likely process to cause such high Rb/Ba, Zr/Nb and, possibly, low $^{87}\text{Sr}/^{86}\text{Sr}$ values in postulated calc-alkaline parental magma. The proposal is based on an anatexis model assuming: (1) lower crust beneath Northeast Japan possesses amphibolitic or hydrous gabbroic composition (Kuno and Aoki, 1970; Aoki, 1971, 1973; Takahashi, 1978; Arai and Saeki, 1980; Tanaka and Aoki, 1980), with its $^{87}\text{Sr}/^{86}\text{Sr}$ as low as 0.7030 (Zashu *et al.*, 1980); and (2) hydrous mafic rocks in the lowermost crust are fairly close to their melting tem-

peratures (Takahashi, 1986b).

Provided that the REE concentrations in the magmas are buffered by dominant component, whereas $^{87}\text{Sr}/^{86}\text{Sr}$, Rb/Ba and Zr/Nb are sensitive to minor component, the tholeiitic and calc-alkaline suites could very well have been derived from a common source, with the calc-alkaline suite undergoing assimilation of isotopically distinct materials during ascent to the magma chamber in which the magma evolved. The same magma-generation model would be plausible in case the isotopically distinct magma sources contain similar REE concentrations.

A similar contrastive nature of the tholeiitic and calc-alkaline magmas is commonly recognized among the volcanoes of NVZ, and this model implies that chemical characteristics of both primary basalt and anatectic melt should be approximately constant in the generation of calc-alkaline parental magmas beneath the NVZ. Moreover, the roughly bimodal nature of these magma series in NVZ suggests that mixing proportion of the primary basalt and the anatectic melt should be approximately constant.

A possible explanation of such constancy is that amphibolitic or hornblende-gabbroic lower

crust, as well as its underlain lherzolitic upper mantle, has analogous melting behavior to some multidimensional synthetic batch melting system. Namely, on a temperature vs. melting degree diagram its melting curve displays a segmented nature, with eutectic-like partial melt during an initial stage of melting, and phases that disappear sequentially at greater degrees of partial melting, causing inflections in the melting curve. Under a similar tectonic, geochemical and geothermal environment such as the NVZ, the individual uprise of mantle diapir may cause a similar manner of partial melting and anatexis underneath the volcanic centers, resulting in the crudely constant proportion of mantle-derived melt and lower crust-derived anatectic melt mentioned above.

An alternative model concordant with these geochemical data is that the tholeiites were derived from relatively shallow lithospheric mantle, whereas the calc-alkaline parental magmas were derived from the mantle wedge beneath the lithosphere, with the addition of a subducting "crustal" component contributed by melts from underlying oceanic slab. However, this model seems inconsistent with the Sr isotopic data of Adatara volcano, in that such a subducting component would be highly enriched in radiogenic Sr.

A possible scenario that comprehensively explains the distinct geochemical characteristics of the tholeiitic and calc-alkaline parental magmas is as follows:

(1) Lherzolitic diapir, a possible common magma source, may have suffered enrichment of radiogenic Sr ($^{87}\text{Sr}/^{86}\text{Sr}$ around 0.7056) near the bottom of wedge mantle beneath Adatara volcano.

(2) The diapir rises within the upper mantle and begins to melt.

(3) Tholeiitic parental magma is produced by relatively high degree partial melting, segregating from the diapir under 10 kb pressure.

(4) When the partially molten lherzolitic diapir rises without segregating partial melt because of smaller degree of partial fusion, the

diapir comes up to crust-mantle boundary, and stops rising by a balance of buoyancy. Emplacement of the hot diapir causes anatexis of lower crust. Eventually, the melt is squeezed out of the diapir, penetrates into the lower crust and incorporates liquid of anatexis origin, which gives rise to calc-alkaline parental magma.

SUMMARY

Flat REE patterns are common among the tholeiitic lavas of Adatara volcano, whereas associated calc-alkaline lavas are characterized by LREE-enriched, concave REE patterns. $(\text{Ce}/\text{Yb})_{\text{CN}}$ ratios remain almost constant through the tholeiitic suite, whereas in the calc-alkaline suite, $(\text{Ce}/\text{Yb})_{\text{CN}}$ ratios increase systematically with increasing SiO_2 .

REE, major- and trace-element variations for the tholeiitic suite support the argument that these lavas are related principally by fractional crystallization of the observed phenocryst phases. Fractionation models are successful in reproducing the major- and some trace-element variations for the calc-alkaline suite, but fail to adequately predict HREE contents or $(\text{Ce}/\text{Yb})_{\text{CN}}$ ratios for the most acidic lavas. The observed LREE enrichment may be the result of crystal fractionation combined with (1) assimilation, (2) incorporation of liquid (or residual magma) that has undergone fractionation of HREE-enriched minerals into differentiation magma, or (3) mixing of small partial melts originated from a common (amphibolitic or hb-gabbroic) source.

In view of the distinct $^{87}\text{Sr}/^{86}\text{Sr}$, Rb/Ba and Nb/Zr ratios between the two suites, tholeiitic and calc-alkaline magmas must be derived from different magma sources. However, the relevant magma sources do not necessarily possess distinct REE compositions. Equilibrium batch partial melting can successfully produce both the lavas with flat and lavas with concave REE patterns from a common lherzolitic source by different degrees of partial melting. A possible model of magma generation is that both tholeiitic and calc-alkaline magma suites are originated from a common lherzolitic diapir,

with calc-alkaline suite undergoing incorporation of partial melt produced by anatexis of lower crust prior to evolution.

Acknowledgments—The late Prof. Naoki Onuma of Ibaraki Univ. kindly introduced the ICP facilities into our laboratory. Prof. M. Tagiri of Ibaraki Univ. continuously supported this study. Miss A. Odaka of Ibaraki Univ. kindly offered her unpublished data. Dr. H. B. West of University of Hawaii at Manoa critically read the manuscript and helpfully advised. The author is greatly indebted to them for their kind cooperation, constructive criticism and improvement of the manuscript. This work was supported by a Grant-in-Aid for Scientific Research (Nos. 63540643, 02640620) from the Ministry of Education, Science and Culture of Japan.

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