Significance of Cs/Rb ratios in volcanic rocks as exemplified by the Nohi Rhyolite complex, Central Japan

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Abstract–Cs/Rb ratio was examined on the Nohi rhyolite complex in Central Japan, which is composed mainly of rhyolitic ignimbrite. The variation in the Cs/Rb ratio of the magma through crystallization differentiation was estimated using a Rayleigh fractionation model and available distribution coefficients between phenocryst minerals and co-existing groundmass. It was found that the K/Rb ratio of this complex decreased from 273 to 125, but the Cs/Rb ratio remained constant through the differentiation process. The initial Cs/Rb ratio of this magma is 0.030 ± 0.005 . The value is very close to the average Cs/Rb ratio of the ignimbrite in the Taupo region, Northern New Zealand, but is distinctly heigher than that of ocean floor basalts. To estimate the effect of contamination the Rb-Sr whole rock isochron method was used.

INTRODUCTION

In studies on differentiated volcanic rocks, many workers have been engaged in finding some chemical parameters which have remained constant during crystallization differentiation, because such parameters are expected to provide us with a significant clue to the source material or the magma generation mechanism. Strontium and lead isotopic ratios have been frequently used for such an index. AHRENS *et al.* (1952) proposed K/Rb ratio as such a parameter. SHAW (1968) critically examined the K/Rb fractionation trends in many rock suites by covariance analysis. He found in his "main trend" that the K/Rb ratio decreases, and potassium and rubidium have a linear relation on a loglog diagram. He attributed the decrease in the K/Rb ratio to crystallization of mica and/ or hornblende.

On the other hand, a number of data have been published recently on the distribution coefficients of trace elements between phenocrysts and co-existing groundmass in volcanic rocks (ONUMA *et al.*, 1968 and others). PHILPOTTS and SCHNETZLER (1970) have suggested that the decrease of the K/Rb ratio during differentiation is due to the difference in distribution coefficients of potassium and rubidium between feldspars and magma. Therefore, the mode of decrease of the K/Rb ratio should depend on the relative quantities of crystallizing minerals. Thus the K/Rb ratio is not a suitable parameter for studying the source material or the magma generation mechanism on a considerably differentiated rocks. In the present paper we discuss whether the Cs/Rb ratio could be used as such a parameter in the crystallization differentiation, using the available distribution coefficient data between phenocrysts and groundmass, and new analyses of the Nohi rhyolite complex

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erupted about 73 million years ago in Central Japan.

THEORETICAL CONSIDERATION

The relationship between concentrations of a trace element A in the parental and differentiated melts, ${}^{0}C_{melt}^{A}$ and C_{melt}^{A} , can be expressed by a Rayleigh equation (equation (1)). (NEWMAN *et al.*, 1954).

$$C_{\text{melt}}^{\text{A}} = {}^{0}C_{\text{melt}}^{\text{A}} \left(1 - x \right)^{\left(D \right)^{\text{A}}} - 1 \right)$$
(1)

where

$$x = \frac{\text{quantity of crystallized solid}}{\text{quantity of parental melt}}$$

The following conditions have been required in obtaining the relation expressed by equation (1):

(1) The diffusion in the melt is complete, but that in the solid is virtually prohibited, (2) Surface equilibrium between the liquid and solid is maintained with a constant distribution coefficient, D^{A} .

If more than two solid phases are crystallizing simultaneously under the above conditions, D^A in equation (1) has to be substituted by an overall distribution coefficient defined as:

$$D^{\mathrm{A}} = \sum_{i}^{\mathrm{n}} W_{i} D_{i}^{\mathrm{A}} \tag{2}$$

where

$$W_i = \frac{\text{quantity of crystallizing solid i}}{\text{total quantity of crystallizing solids}}; \quad \sum_{i=1}^{n} W_i = 1$$

When two species of trace elements, A and B, satisfy the above conditions, we have the following equation:

$$\frac{C_{\text{melt}}^{\text{A}}}{C_{\text{melt}}^{\text{B}}} = \left(\frac{{}^{0}C_{\text{melt}}^{\text{A}}}{{}^{0}C_{\text{melt}}^{\text{B}}}\right) (1 - x)^{(D^{\text{A}} - D^{\text{B}})}$$
(3)

or

$$\left(\frac{C_{\text{melt}}^{A}}{C_{\text{melt}}^{B}}\right) / \left(\frac{{}^{0}C_{\text{melt}}^{A}}{{}^{0}C_{\text{melt}}^{B}}\right) = (1 - x)^{(D^{A} - D^{B})}$$
(4)

where D^{A} and D^{B} stand for the overall distribution coefficients defined by equation (2). This equation shows that the concentration ratio A/B in the melt is a simple function of the degree of crystallization. The left-hand side of equation (4) will be denoted hereafter by $R_{A/B}$. The variation of $R_{A/B}$ is shown in Fig. 1 as a function of the degree of crystallization (x) for some given values of $(D^{A} - D^{B})$. The figure shows that the change in the $R_{A/B}$ value does not exceed 5 percent up to 98 per cent solidification of the melt, when $|D^{A} - D^{B}|$ is smaller than 0.02.

EWART and TAYLOR (1969), PHILPOTTS and SCHNETZLER (1971), HIGUCHI (1973) and HART and BROOKS (1974) determined the distribution coefficients, D_i^{Cs} , D_i^{Rb} and D_i^K for a variety of phenocryst groundmass pairs of volcanic rocks. Their data are compiled in Table 1. In the table, D_i^{Rb} values of okivine, pyroxenes and plagioclase are smaller than 0.01 or in the same order of magnitude. The D_i^{Cs} for olivine, pyroxenes and plagioclase

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Fig. 1. Variation of $R_{A/B}$ at a degree of solidification of a magma. The $R_{A/B}$ shows a variation degree of concentration ratio of A and B trace elements in a differentiated magma from that of the parental magma. It is expressed as $R_{A/B} = (1 - x)^{(D^A - D^B)}$. Some given values of $(D^A - D^B)$ are shown on each curve.

are also almost equal to or less than D_i^{Rb} . Although the data on these minerals in Table 1 are insufficient to estimate D_i^{Cs} directly for a wide range of chemical composition of magma, it has been thought that D_i^{Cs} of these minerals are smaller than or about of the same order of magnitude as D_i^{Rb} from the view point of ionic radii of these elements (JENSEN, 1973). Then the order of magnitude of $|D_i^{Cs} - D_i^{Rb}|$ is estimated to be smaller than 0.01 for these minerals in the composition range from basalt to dacite. Consequently $R_{Cs/Rb}$ does not change substantially in the course of crystallization differentiation when olivine, pyroxenes and plagioclase are the principal minerals crystallizing from the magma. Since D_i^{Rb} , $D_i^{Cs} - D_i^{Rb}|$ of hornblende, biotite and K-feldspar are con-

siderably larger than those of olivine, pyroxenes and plagioclase, $R_{Cs/Rb}$ may change significantly when these minerals crystallize. In a particular case, when W_i in equation (2) for hornblende, biotite and K-feldspar are less than 0.01, the overall distribution coefficient defined by equation (2) is not expected to vary significantly by the presence of such minerals.

SAMPLES AND ANALYTICAL PROCEDURES

Rock samples investigated in this work were collected from the Nohi Rhyolite complex which consists mainly of salic ignimbrite erupted in Late-Cretaceous period in Central Japan. The complex is composed of many stratigraphic units. The samples were taken from the successive three units from the bottom upwards, named Higashimata, Akaishi and Yumoriyama welded tuff sheets, all of which belong to "Stage II" of the Nohi Rhyolite volcanism (YAMADA *et al.*, 1971). No field evidences indicating large time gaps between these three units have been found, so they are assumed to be products of successive eruptions.

The welded tuffs have fragmental phenocrysts of $40 \sim 50$ volume per cent and cryptocrystalline groundmass. Phenocrysts are mainly plagioclase (An₅₀ ~ An₂₀), quartz and K-feldspar (perthite) with small amounts (below 2 volume per cent) of altered mafic minerals (biotite, hornblende, augite and hypersthene) excpet for a specimen NY-10. Each phenocryst is considerably altered to clay minerals, epidotes, calcite, etc. Alteration of mafic minerals is so high that only their pseudomorphs are usually observed. Hydrous mafic minerals are considered from their pseudomorphs to amount to about 0.5 per cent. Modes of phenocrysts are presented in Table 2, in which alteration products are expressed, as far as possible, as their primary minerals.

Major elements were analyzed by the rapid methods of MATSUI (1963) and MgO was determined by atomic absorption spectrophotometry, when its concentration was low. Rb

				_	Plagioclase					
-	2	9	7	8	6	10	11	12	13	14
	0.0109									0.0248
0.016	0.0173	0.041	0.016	0.011	0.02	0.03	0.08	0.007	0.008	0.0262
.302	0.110	0.10	0.076	0.16	0.10	0.05	0.14	0.09	0.10	0.180
	-0.0064									-0.0014
	Hypersthene				Augite			Pigeon.	Olivine	K-Felds.
2	6	2	2	14-1	14-2	15	16	17	14	18
0.0052			0.0258	0.0039	0.0015	0.0179			0.0004	
0.0062	0.0027	0.00048	0.0245	0.0033	0.0014	0.0094	0.0326	0.00031	0.0002	0.659
0.0057	0.0023	0.00055	0.0275	0.0028	0.0017	0.0052	0.0448	0.00344	0.0002	1.49
0.0010			0.0013	0.0006	0.0001	0.0085			0.0002	
	Horn	blende		Biotite						
e	4	6	7	5						
0.046	0.198			2.35						
0.31	0.404	0.014	0.0077	3.53						
0.60	0.86	0.081	0.065	2.50						
0.264	-0.206			-1.18						

Symbols are as follows; Pigeon. = pigeonite, K-Felds. The same numbered phenocrysts are separated from the same rock samples. potassium feldspar. Data sources are as follows;

1) Alkali olivine basalt, Takashima, Kyushu, Japan, HIGUCHI (1973).

Onioshidashi andesite, Asama Volcano, Japan, ibid.

Alkali basalt, Iki-Island, Kyushu, Japan, ibid.

Sanogawa andesite, Shizuoka, Japan, ibid.

Dacite, Sunshine Canyon, Colorado, U.S.A., ibid.

Torihama dacite, Southern Kyushu, Japan, NAGASAWA and SCHNETZLER (1971).

Kakuto dacite, Southern Kyushu, Japan, ibid.

Rhyolitic biotite hornblende hypersthene ignimbrite, Central North Island, New Zealand, EwaRT and TAYLOR (1969).

1954 Nagauruhoe lava, augite hypersthene basaltic andesite, Taupo, New Zealand, ibid.

Augite hypersthene acidic andesite, Taupo, New Zealand, ibid. 10)

Hornblende hypersthene andesite, Taupo, New Zealand, ibid. 11)

Hornblende hypersthene rhyolite, Taupo, New Zealand, ibid. Biotite hornblende rhyolite, Taupo, New Zealand, ibid. 12)

(continued)

Ankeramite, northwest flank of Pico Volcano, Pico Island, Azores, HART and BROOKS (1974). (14-1); Green clinopyroxene, (14-2) Brown clinopyroxene (4)

- Andesitic basalt, Kozori, Sano, Japan, ibid.
 - Oceanite-ankeramite, Tahiti, ibid.
- Ferropigeonite-ferrohortonolite andesite, Kuzo-zawa and Asozawa, Asio, Japan, ibid. 15) 16) 17) 18)
 - Porphiritic rhyodacite, Monte di Proceda, Phlegrean, Fields, Naples, Italy, ibid.

۵	0.39	0.01	0.06	0.04	0.04	0.006	0.04	0.04	0.04	0.04	0.004								
×	72.24	0.25	14.21	0.36	1.64	0.06	0.73	2.21	3.38	3.95	0.10								
This work	72.02	0.30	14.23	0.38	1.54	0.07	0.62	2.17	3.40	4.11	0.07								
NA-2 K-Felds.		trace	18.22			trace	trace		3.60	11.67	trace				9.69	649	8.76	0.013	149
NY-10	57.46	0.38	14.13	1.90	7.71	0.19	3.40	5.80	1.91	0.92	0.14	3.66	0.80	99.44	0.81	35.23			230
NY1	70.29	0.34	13.95	0.78	1.58	0.05	0.46	2.36	3.47	3.79	0.04	1.47	1.56	100.14	3.24	149.2			217
NA-4	74.30	0.17	12.93	1.06	0.64	0.03	0.14	1.50	2.97	4.61	0.02	0.91	1.10	100.38	3.89	212.8	5.97	0.028	183
NA-3	75.33	0.11	12.59	0.58	0.96	0.03	0.11	0.90	2.66	4.87	0.02	1.12	0.26	99.55	4.12	270.9	9.57	0.035	152
NA-2	75.17	0.10	12.21	0.66	0.81	0.03	0.08	1.03	3.08	5.10	0.03	0.83	0.40	99.53	4.31	344.3	9.02	0.026	125
NA-1	75.62	0.16	12.48	0.62	1.24	0.05	0.09	1.76	3.14	4.66	0.02	0.73	0.74	101.32	3.88	254.7			152
NH-5	70.03	0.34	14.47	0.69	2.08	0.05	0.60	2.53	3.50	3.83	0.04	1.00	1.49	100.65	3.24	123.7	3.84	0.031	262
NH-1	65.88	0.57	14.74	0.36	3.54	0.09	1.01	3.36	3.51	3.14	0.11	1.96	2.18	100.45	2.73	100.0	2.95	0.030	273
	SiO ₂	TiO ₂	Al ₂ O ₃	Fe_2O_3	FeO	OuM	MgO	CaO	Na ₂ O	K20	P205	H_2O	c02	Total	K 10 ⁴ ppm	Rb ppm	Cs ppm	Cs/Rb	K/Rb
	$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	NH-1 NH-5 NA-1 NA-2 NA-3 NA-4 NY-10 NA-2 This x σ SiO2 65.88 70.03 75.62 75.17 75.33 74.30 70.29 57.46 72.02 72.24 0.39	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{l l l l l l l l l l l l l l l l l l l $								

Chemical and model compositions of the Nohi Rhyolite complex Table 2.

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			\overline{x} ; Mean value of JG-1 by ANDO	et al. (1971)	σ; Standard deviation in this work			
	NY-10	7.6	46.1	I	17.4	2.2	0.7	25.9
AODES	NY-1	20.0	20.5	9.9	2.0	9.0	0.2	46.7
	NA-4	22.8	11.4	13.2	1.2	0.3	0.2	51.2
	NA-3	15.2	9.7	14.3	0.7	0.2	0.1	59.9
	NA-2	19.3	8.4	11.9	0.2	0.1	trace	59.9
V	NA-1	16.3	9.0	17.3	0.7	0.2	trace	56.1
	NH-5	9.4	25.6	5.7	2.8	0.2	0.1	55.6
	NH-1	1.8	34.9	I	2.8	0.5	trace	58.4
		Quartz.	Plagio.	K-Felds.	Altered mafic.	Opaques	Accessory minerals	Groundmass

Cs contents are below 2 and 5 per cent, respectively. Symbols of mineral names are as follows; Plagio. = element analyses are given in the JG-I analysis and in the last column, respectively. The latter is the mean standard deviation calculated from twice or three-time determinations on each sample. Accuracies of Rb and plagioclase, K-Felds. = potassium feldspar (perthite), Altered mafic. = alteration products from mafic minerals, such as biotite, hornblende, hypersthene and augite, plus fresh mafic minerals mentioned above, Opaques. = magnetites plus ilumenites. Accessory minerals are such as apatite, zircon and allanite. Symbols of sample names are as follows; "NH-" = the Higashimata, "NA-" = the Akaishi and "NY-" = the Yumoriyama welded A K-feldspar, separated from specimen NA-2, was partially analyzed. Accuracies and precisions of major tuff sheets. and Sr were determined by isotope dilution procedures described by ISHIZAKA (1971), and Cs by the instrumental neutron activation analysis, using "JG-1" with 10.1 ppm Cs content (ANDO *et al.*, 1971) as a reference standard. Analytical errors for Rb, Sr, Cs contents and Sr isotopic ratio are less than 2, 2, 5 and 0.1 per cent, respectively, and those of major elements are listed in Table 2. One K-feldspar (perthite) was separated by a Frantz Isodynamic separator and heavy liquids.

RESULTS AND DISCUSSION

Analytical data on major elements are given in Table 2. Each element of them falls on a smooth curve in the silica-variation diagram, and hence it indicates that the effect of alteration on the chemical compositions of the samples as whole rocks was small enough to be neglected. Comparison of these data with those of YAMADA *et al.* (1971) reveals that sample NH-1 corresponds to the most basic welded tuff and NA-2 to the most acidic one. Thus the samples investigated in this work cover the whole range of chemical composition of the complex. Sample NY-10, which has distinctly basic composition, is excluded from genetical discussions, because it has an extreme amount of phenocrysts (more than 74 per cent).

The Rb and Sr isotopic compositions were determined on six whole rock samples, NA-1, NA-2, NA-3, NA-4, NY-1 and NY-10, and the results are given in Table 3. The data are plotted on the whole rock isochron diagram in Fig. 2, in which they define a single isochron with 72.9 ± 2.4 million years in age and with the initial 87 Sr/ 86 Sr ratio of 0.7088 ± 0.0007. The age is in harmony with the available geologic and K-Ar ages of $97 \sim 67$ million years (KAWADA, 1971). Thus, these rocks are supposed to have been not appreciably contaminated by foreign materials. These data also suggest that the whole rock chemical compositions of the samples were not substantially affected by the alteration of their phenocryst minerals.

The Cs, Rb and K contents are given in Table 2, and K-Rb and Cs-Rb relations are shown in Fig. 3. Rubidium and potassium contents were determined for 11 more samples, and are also plotted in the same diagram^{*}. The K/Rb ratio of this complex decreases from 273 to 125 with increasing K and Rb contents, and $d(\log K)/d(\log Rb)$ is smaller than that of the "main trend" by SHAW. On the other hand, the Cs/Rb ratio is nearly constant within a value of 0.030 ± 0.005 .

In our samples, K-feldspar is a major phenocryst, so its influence on the $R_{Cs/Rb}$ must be taken into account. Since it was impossible to obtain fresh K-feldspars from our samples, one of the least altered K-feldspar was separated from specimen NA-2, and chemically analyzed, the results being given in Table 2. The values $D_{K-feldspar}^{Cs}$, $D_{K-feldspar}^{Rb}$

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Sample			⁸⁷ Rb/ ⁸⁶ Sr	⁸⁷ Sr/ ⁸⁶ Sı
NA-1	72.13	10.92	6.530	0.7173
NA- 2	97.50	3.855	25.00	0.7355
NA- 3	76.68	7.192	10.54	0.7241
NA- 4	60.26	10.20	5.843	0.7141
NY- 1	40.48	20.29	1.972	0.7107
NY-10	9.976	31.63	0.3118	0.7097

Table 3.	Rb and Sr isotopic compositions and their isotopic	2
	ratios of the Nohi Rhyolite complex	

Analytical errors of these analyses, Rb, Sr and Sr isotopic ratios, are below 2, 2, and 0.1 per cent, respectively. Symbols of sample names are the same as in Table 2.

* These data are available upon request to the first-named author (K.O.).



Rb-Sr whole rock isochron of the "Stage Fig. 2. II" Nohi Rhyolite complex. Defined age is 72.9 ± 2.4 million years and the initial Sr isotopic ratio is 0.7088 ± 0.0007.

Fig. 3. K-Rb and Cs-Rb relations of the Nohi Rhyolite complex plotted on a log-log diagram. The upper portion shows the K-Rb relations and the lower shows Cs-Rb relations. Lines labelled 0.026 and 0.035 refer to constant Cs/Rb ratios. Symbols showing K-Rb relation are as follows; solid circles = welded tuffs belonging to "Stage II", squares = dyke rocks intruding the "Stage II" welded tuffs.



and $D_{K-feldspar}^{K}$ are calculated to be 0.62, 0.97 and 1.23, respectively, using the estimated composition of groundmass (NA-2). The composition of the groundmass was calculated from the whole rock analysis and phenocryst modes on the assumption that these elements are absent in the other phenocryst minerals. Thus the distribution coefficients obtained may be smaller than the actual ones, particularly for potassium, because sodic plagioclase should contain considerable amount of potassium. The calculated $|D_{K-feldspar}^{Cs} - D_{K-feldspar}^{Rb}|$ equals to 0.35, and $|D^{Cs} - D^{Rb}|$ in equation (4), overall difference of the distribution coefficient, is about 0.1, provided that W_i in equation (2) could be approximated by the relative abundances of phenocrysts in the rock.

The degree of solidification of magma, x in equation (4), is expressed approximately as $x = (1 - C_{melt}^{Rb} / C_{melt}^{Rb})$, as far as D^{Rb} is very close to zero. Now we can estimate the degree of solidification of magma, and calculated result is that the rock NA-2 corresponds to the 71 per cent solidification of magma starting from the magma of the composition of the rock NH-1. This is a minimum estimate of the degree of solidification because D^{Rb} is actually not equal to zero. Therefore an increase of more than 10 per cent in Cs/Rb ratio of these rocks is expected during the crystallization differentiation from NH-1 to NA-2 as judged from theoretical values shown in Fig. 1. The contribution of biotite and hornblende phenocrysts to the overall change of Cs/Rb ratio can be neglected because the amount of biotite and hornblende phenocrysts in samples is less than 1 per cent in mode, and W_{biotite} and $W_{\text{hornblende}}$ in equation (2) are only 0.01 or less. Nevertheless the theoretically expected increase of 10 per cent is not observed in the Cs/Rb ratio as shown in Fig. 3. The discrepancy cannot be ascribed to analytical errors. Apparent constant value of Cs/Rb ratio for this complex may be attributed to the overestimation of $|D^{Cs} D^{Rb}$ for the magmatic condition because of the alteration of K-feldspars. Alternatively the fractionation of magma by crystallization of K-feldspar might be not so effective to the increase in Cs/Rb ratio as that by other minerals because of, for example, incomplete

removal of K-feldspars from the melt by the floating of K-feldspars.

The Cs/Rb ratio is near 0.030 in the Rb-poor side rocks in Fig. 3 and Table 2, in which the amount of crystallizing K-feldspar is small (as is seen in Table 2), and thus, both of $|D^{Cs} - D^{Rb}|$ and x in equation (4) will become smaller and so the effect of them on the change in $R_{Cs/Rb}$ will become smaller during crystallization. Mafic minerals of specimen NY-10, which is considered to represent a crystal accumulated portion of this magma at an early stage of crystallization, are augite and hypersthene. And it suggests that this magma did not crystallize significant amounts of hornblende and/or biotite. Therefore, even if the most basic welded tuff (NH-1) itself was the product of differentiation from more basic magma, Cs/Rb ratio should change little from that of the parental magma with initial Cs/Rb ratio of 0.030 ± 0.005.

CONCLUDING REMARKS

Potassium and rubidium contents of the Nohi Rhyolite complex do not have a linear relation on a log-log diagram, and thus it is not a suitable parameter to specify the parental magma of this complex. On the other hand, Cs/Rb ratio is kept nearly constant during fractional crystallization differentiation, suggesting that the parental magma of this complex has a Cs/Rb ratio of 0.030 ± 0.005 and an initial Sr isotopic ratio of 0.7088 ± 0.0007 . Few systematic analyses of Cs and Rb of other salic volcanic rocks or ignimbrites have been published. Only one example is the ignimbrite in the Taupo region of Northern New Zealand, in which the average Cs/Rb ratio (0.03, EWART *et al.*, 1968) is very close to that of the Nohi Rhyolite complex. On the other hand, the average Cs/Rb ratio of ocean floor basalts (HART *et al.*, 1972) is 0.0145 and is distinctly lower than those of ignimbrites. Thus the Cs/Rb ratio is supposed to be useful to distinguish and specify different volcanic rock suites or rock series.

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