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MINOR ELEMENT GEOCHEMISTRY OF THE ROCKS FROM MASHU VOLCANO, EASTERN HOKKAIDO

by

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(with 5 Figures and 3 Tables)

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Abstract

Ten minor elements, such as Ga, Cr, Li, Ni, Co, V, Zn, Y, Cu and Sr of 29 samples of the Mashu rocks were analysed by means of emission spectroscopy and atomic absorption spectroscopy. The Mashu rocks are generally depleted in K, Li and Sr. This chemical feature and other evidences suggest that they have been derived from a low-K tholeiitic magma through fractional crystallization. The Mashu rocks vary in rock series from the tholeiitic (the older somma lavas) to the calc-alkaline (the caldera and central cone products) through a transitional one (the younger somma lavas). V together with Fe is markedly enriched in the first series, but depleted in the later ones, suggesting that increase (or constancy) of oxygen partial pressure in the magma occurred in the middle to later stage of fractional crystallization. This may be responsible to the change in the rock series towards the later volcanic activity.

Introduction

The behavior of minor elements during magmatic differentiation has been well established in some stratified, differentiated plutonic rocks, such as the Skaergaard intrusion (Wager and Mitchell, 1951) and the South California batholith (Sen, Nockolds and Allen, 1959). In volcanic rocks, however, such investigation may encounter with difficulties, because these rocks do not always erupt in the order of differentiation of magma.

Mashu volcano, located at eastern Hokkaido, is one of the few volcanoes whose history, together with chronological variation in the major element chemistry of its products, has been investigated in detail (Katsui, Ando and Inaba, in press).

In this paper, the author reports the result of chemical analyses of minor elements, such as Ga, Cr, Li, Ni, Co, V, Zn, Y, Cu and Sr in the Mashu rocks and discusses their behavior from the chronological point of view.

Summary of Geology and Petrography

Mashu volcano was constructed on the eastern wall of the Kutcharo caldera, during the later Pleistocene to Holocene. A full account of the history of Mashu volcano is given by Katsui, Ando and Inaba (in press). The history of Mashu volcano can be divided into the following three stages, on the basis of the sequence of the somma lavas and the stratigraphy of the pyroclastic deposits:

- 1) Stratovolcano building stage: ca. $17 - 12 \times 10^3$ y. ago
- 2) Caldera building stage: $11 - 7 \times 10^3$ y. ago
- 3) Central cone stage: ca. $4 - 1 \times 10^3$ y. ago.

The somma lavas are subdivided by a quiescent epoch into the older and the younger somma lavas. The older somma lavas are composed of pyroxene andesite and basalt (types Ic and Vc*). The earlier lavas (Older somma lava 1) are hypersthene andesite of nearly aphyric variety (type Ic). Then, the lavas became porphyritic and more mafic in composition (Older somma lava 2), such as augite hypersthene andesite (type Vc) and hypersthene basalt (type Ic). Their groundmass, however, is persistently the same and consists of plagioclase, clinopyroxene, iron ore and cristobalite, commonly showing an intersertal texture.

The younger somma lavas are mostly composed of hypersthene andesite (type Id→c) and augite hypersthene andesite (type Vd→c), most of which are intermediate in composition except the earlier felsic pumice and dykes. Their groundmass is made up of plagioclase, clinopyroxene, orthopyroxene with a reaction rim of clinopyroxene, iron ore and cristobalite showing an intersertal texture.

After the formation of the stratovolcano, felsic andesite pumice (Ma-1) and ash (Ma-k) were erupted about 11,000 years ago. Then, about 7,000 years ago, the culminating activity that led to the formation of the Mashu caldera occurred with the ejection of ash fall (Ma-j), followed by successive eruptions of pumice falls (Ma-i, Ma-h and Ma-g) and a pyroclastic flow (Ma-f). All of these pumices are augite hypersthene felsic andesite (type V) except that of Ma-f which is more mafic in composition.

After a long quiescence, about 4,000 years ago, the Kamuinupuri volcano was formed in the southeast side of the caldera. About one thousand years ago, an explosive activity occurred, ejecting successive pumices (Ma-b5 to Ma-b1), and formed a large crater or small caldera. During the formation of the Kamuinupuri, a dacite lava dome, the Kamuishu, extruded at the center of the

* Symbols of Kuno's classification based on the ferromagnesian silicate mineral assemblage (Kuno, 1950).

caldera. The first lava of the Kamuinupuri is augite hypersthene andesite (type Vd), and the second lava is augite hypersthene felsic andesite (type Vd). The Kamuishu dome lava is augite hypersthene dacite (type Vd). The groundmass of the above lavas is composed of plagioclase, cristobalite and a small amount of alkali-feldspar, iron ore, orthopyroxene, clinopyroxene and apatite, showing a felsitic texture.

In reviewing the petrography of the Mashu rocks, it is noticed that they vary in rock series from the tholeiitic (the older somma lavas) to the calc-alkaline (the caldera and central cone products), through a transitional one (the younger somma lavas) (Katsui, Ando and Inaba, in press).

Table 1 Minor element abundances of the older somma lavas.

No.	1	2	3	4	5	6	7	8	9	10
	M-38	M-39	M-40	M-41	M-42	M-43	M-2	M-44	M-45	M-46
SiO ₂	56.24	57.43	54.79	58.16	56.08	54.56	52.51	52.80	53.53	52.39
K ₂ O	.28	.32	.23	.32	.22	.19	.22	.19	.20	.16
S. I.	17.9	16.3	18.1	19.1	16.4	23.3	25.7	25.2	24.9	25.5
D. I.	38.0	40.7	36.5	43.1	39.4	31.6	26.9	28.7	30.8	28.6
C. I.	33.3	30.1	31.7	30.2	26.3	31.3	31.4	32.1	35.2	34.2
Ga	12	14	12	12	13	13	13	13	15	13
Cr	13.0	11.0	20.0	18.5	14.0	14.0	49.9	25.0	48.0	48.0
Li	4.0	4.5	3.0	3.5	6.0	3.0	4.3	5.0	5.0	9.0
Ni	35.0	31.0	35.0	28.5	35.0	34.0	35.0	43.0	46.0	41.0
Co	44.4	42.5	43.5	41.0	44.5	42.5	43.5	51.4	57.6	48.0
V	250	265	295	275	175	320	295	300	300	300
Zn	111	117	102	111	98	96	93	93	95	102
Y	23	21	19	19	17	22	21	20	17	18
Cu	55.3	50.5	66.5	39.5	32.0	104.0	64.4	56.9	61.6	57.5
Sr	100	112	113	106	119	94	119	112	125	106

- 1 - 4 Hypersthene andesite (nearly aphyric), lava, foot of the northwestern part of the caldera.
- 5 - 6 Hypersthene andesite (porphyritic), lava, loc. ditto.
- 7 Hypersthene andesite (porphyritic), dyke, loc. ditto.
- 8 Hypersthene andesite (porphyritic), lava, loc. ditto.
- 9 Augite-bearing hypersthene basalt (porphyritic), lava, loc. ditto.
- 10 Augite hypersthene mafic andesite (porphyritic), lava, loc. ditto.

Table 2 Minor element abundances of the younger somma lavas.

No.	11	12	13	14	15	16	17	18	19	20
	pumice	dyke	M-48	M-49	M-50	M-51	M-52	M-53	M-54	M-55
SiO ₂	66.64	65.82	55.89	60.81	59.73	60.18	59.89	59.43	61.20	59.41
K ₂ O	.45	.46	.20	.36	.26	.27	.25	.26	.24	.23
S. I.	12.8	13.8	19.7	16.1	17.5	18.4	16.2	16.3	16.5	13.1
D. I.	62.1	61.6	38.4	50.1	46.4	48.1	47.9	48.4	49.2	48.2
C. I.	12.5	17.2	23.8	25.8	19.7	19.8	19.4	18.7	18.2	21.2
Ga	13	12	15	13	13	13	14	12	14	13
Cr	9.0	6.5	12.0	11.0	11.0	11.0	9.5	9.0	11.0	6.5
Li	9.0	7.0	5.0	5.5	4.2	8.0	4.5	5.0	5.5	5.0
Ni	26.0	23.5	34.0	30.5	31.0	33.5	29.5	28.9	27.5	28.5
Co	25.0	17.0	34.0	30.5	22.0	37.0	25.0	28.9	25.0	35.0
V	43	42	175	109	72	113	110	103	135	125
Zn	114	110	104	122	119	107	141	110	110	107
Y	14	18	19	18	13	18	17	19	23	18
Cu	5.0	17.0	31.5	28.5	25.0	25.0	30.5	38.9	32.0	24.5
Sr	125	100	113	125	125	144	125	125	125	150

11 Hypersthene andesite (nearly aphyric), welded pumice fall deposit, north-western part of the caldera.

12 Aphyric andesite, dyke, loc. ditto.

13 Hypersthene andesite (porphyritic), lava, loc. ditto.

14 Aphyric andesite, lava, loc. ditto.

15 - 20 Augite-bearing hypersthene andesite (porphyritic), lava, loc. ditto.

Analytical Methods

Ga, V and Y were determined by emission spectroscopy using a Shimadzu grating spectrograph. Equal portions of sample, NaCl and graphite powders containing indium and palladium black as internal standards are thoroughly mixed and then excited with a 10A d.c. arc. The standard samples G-2, GSP-1, AGV-1, BCR-1, DTS-1 and PCC-1 were used for preparation of working curves (Flanagan, 1969). Lines were read on a Shimadzu microphotometer. The following lines were read: Ga 2943.64, V 3185.40, Y 3242.28, In 3039.36, Pd 3242.71 (A). Precision expressed as relative deviations is as follows: Ga $\pm 11\%$, V $\pm 19\%$, Y $\pm 13\%$. All samples were analysed in quadruple.

Cr, Li, Ni, Co, Zn, Cu and Sr were determined by atomic absorption spectroscopy after the method developed by Terashima (1971), using a Hitachi 207 type spectrograph.

Table 3 Minor element abundances of the volcanic products of the caldera and central cone stages.

No.	Caldera building stage				Central cone stage				
	21 Ma-1 pumice	22 Ma-i pumice	23 Ma-h pumice	24 Ma-f pumice	25 Ma-d pumice	26 Kamui- nupuri	27 Kamui- nupuri	28 Ma-b ₅ pumice	29 Ma-b ₃ pumice
SiO ₂	65.79	67.57	66.65	65.73	63.55	69.73	72.96	65.53	66.37
K ₂ O	.46	.91	.78	.75	.51	.60	1.07	.85	.67
S. I.	12.9	13.8	14.5	16.3	13.9	11.0	10.1	12.9	13.6
D. I.	64.7	69.4	66.9	64.3	58.8	72.3	76.4	64.0	64.3
C. I.	11.5	10.4	11.9	13.6	14.4	11.1	8.5	11.9	13.2
Ga	13	12	15	14	12	11	12	15	12
Cr	9.0	9.0	2.0	3.0	9.0	8.0	33.5	9.0	8.0
Li	8.0	9.5	9.5	9.5	8.5	6.5	8.0	9.0	8.5
Ni	22.0	20.0	20.0	16.0	20.5	22.0	36.5	22.5	22.5
Co	18.5	14.5	18.0	12.0	18.0	19.5	12.0	21.0	20.0
V	18	7	6	27	34	tr	8	29	33
Zn	98	95	96	99	92	88	82	93	93
Y	20	13	10	13	25	tr	11	7	12
Cu	2.5	2.0	2.0	3.0	9.0	1.2	1.5	3.0	3.0
Sr	94	113	119	106	100	106	94	106	113

- 21 Augite hypersthene felsic andesite, pumice from the Ma-1 pumice fall deposit. Kennebetsu, east of the Mashū caldera (on the basis of H₂O free).
- 22 Augite hypersthene felsic andesite, pumice from the Ma-i pumice fall deposit, Nijibetsu, east of the Mashū caldera.
- 23 Augite hypersthene felsic andesite, pumice from the Ma-h pumice fall deposit, loc. ditto.
- 24 Augite hypersthene andesite, pumice from the Ma-f pumice flow deposit, loc. ditto.
- 25 Augite hypersthene andesite, pumice from the Ma-d pumice fall deposit, top of the northwestern part of the Mashū caldera. (on the basis of H₂O free).
- 26 Augite hypersthene felsic andesite (nearly aphyric), the 2nd lava, summit of Kamuinupuri.
- 27 Augite hypersthene dacite (nearly aphyric), dome lava, Kamuish island.
- 28 Augite hypersthene felsic andesite, pumice from the Ma-b₅ pumice fall deposit, top of the Mashū caldera.
- 29 Augite hypersthene felsic andesite, pumice from the Ma-b₃ pumice fall deposit, top of the Mashū caldera.

Analytical Results

The analytical results of the Mashu rocks are shown in Tables 1 to 3 with their SiO_2 content and solidification, differentiation and color indices. Their variation diagrams of minor element abundances versus silica content are given in Figs. 1 and 2.

Lithium: Li content is generally low, and increases with SiO_2 . Such behavior of lithium is similar to that of barium (Ando, 1972). Mason (1966) showed that the Li/Mg ratio increases with differentiation of magma and that this ratio is a good indicator of magmatic differentiation. In this respect, the Li/Mg ratio of the Mashu rocks shows a steady increase towards the later felsic rocks.

Chromium, Nickel, Cobalt and Copper: The abundances of these elements decrease steadily with increasing silica. The Kamuishu dome lava, however, is exceptionally high in Cr and Ni, though it is the most felsic lava (SiO_2 72.96%).

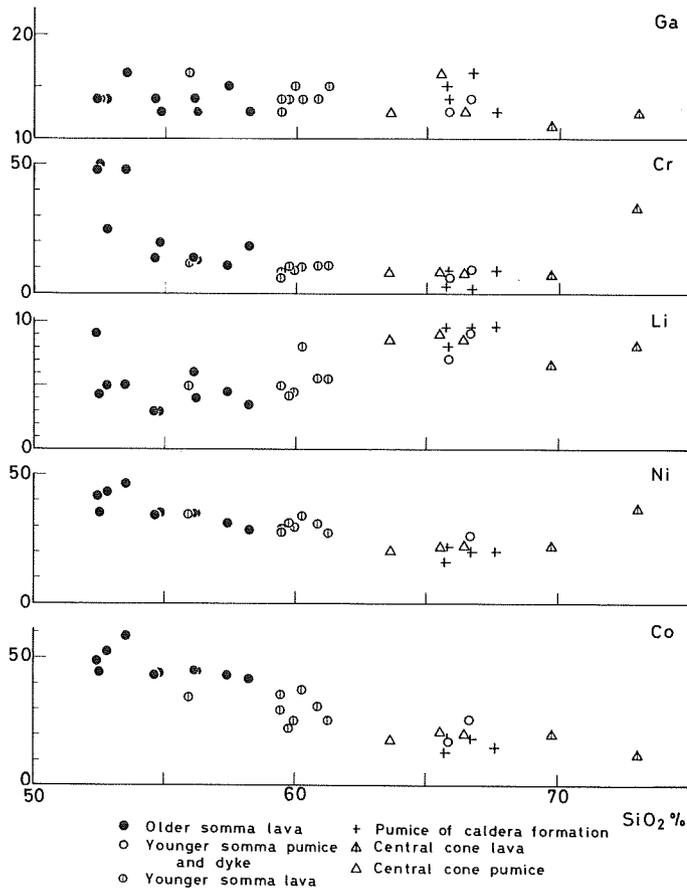


Fig. 1 Variation in Ga, Cr, Li, Ni and Co values in ppm against SiO_2 .

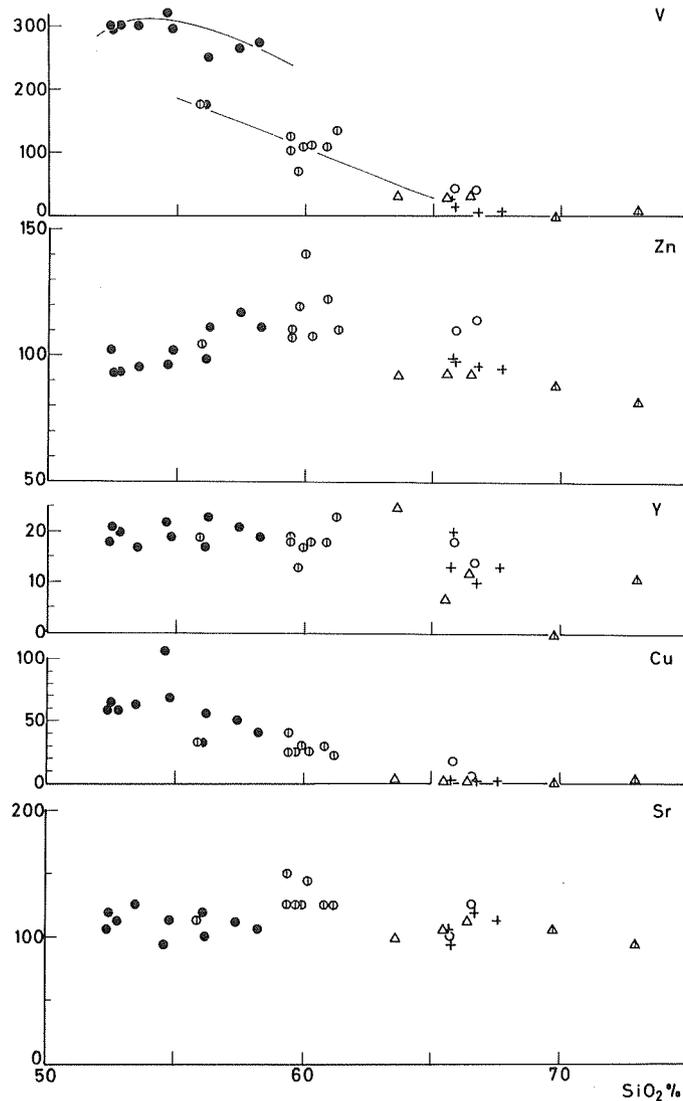


Fig. 2 Variation in V, Zn, Y, Cu and Sr values in ppm against SiO₂. Symbols as same as Fig. 1.

Vanadium: V content decreases with increasing silica, but two types of variation can be recognized. The older somma lavas have a maximum in vanadium content at the middle stage of differentiation. On the contrary, the vanadium content in the younger somma lavas and the later pumices and lavas, persistently decreases with increasing silica. Such behavior of vanadium is similar to that of iron. The former type is a characteristic feature of the tholeiitic series, while the latter type is of the cala-alkaline series (Ando, 1971,

1972). The same behavior is also noticed in scandium (Masuda, et al., in press). Gallium: Ga abundance is nearly constant ranging from 11 to 15 ppm over the whole series of products. This behavior of Ga is similar to that of Al, as expected from their resemblance in ionic radii.

Yttrium: Y displays an irregular variation but generally decreases from the mafic to felsic ends.

Zinc: Zn content slightly increases from the basic to the intermediate rocks, but decreases gradually towards the felsic rocks.

Strontium: Sr content is nearly constant throughout the studied rocks, ranging from 97 to 150 ppm. Sr/Ca ratios in igneous rocks increase during fractional crystallization of magma (Taylor, 1966). In the Mashu rocks, this ratio increases gradually from 1.3 in mafic rocks to 4.0 in felsic rocks.

Chronological Variation of Minor Elements

As shown in Fig. 3, a general trend from mafic to felsic composition can be traced throughout the whole products (long term variation), while a compositional variation in the reverse order, from felsic to mafic, is also noticed within a short period of activity (short term variation) (Katsui, Ando and Inaba, in press).

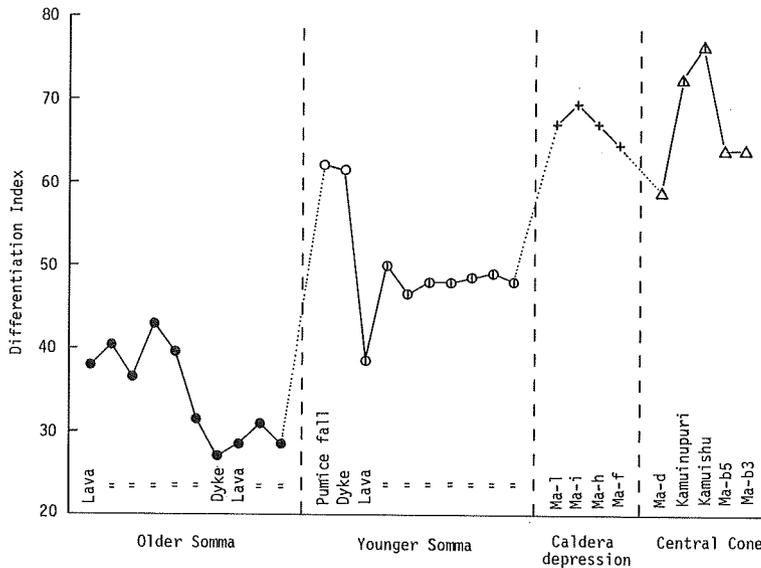


Fig. 3 Relation between the differentiation indices of the rocks and the order of eruption. Symbols as same as Fig. 1 (after Katsui, Ando and Inaba, in press).

A similar variation is also noticed in minor element abundances as shown in Figs. 4 and 5. Li content increases gradually from the older somma lavas to the later products. On the other hand, Ni, Co, V and Cu contents steadily decrease. However, no remarkable variation in respect to the long term variation is found in other elements, such as Ga, Cr, Zn, Y and Sr. With respect to the short term variation, a similar compositional variation as found in the major elements can be recognized in many elements especially in Cr, Li, V and Sr, and Li/Mg and Sr/Ca ratios.

The long term variation has been interpreted by persistent differentiation of a basaltic magma taking place without any supply of new magma, and the short term variation in terms of a zoned magma chamber. The chronological variation of minor elements, as above mentioned, may support the result obtained by major element chemistry.

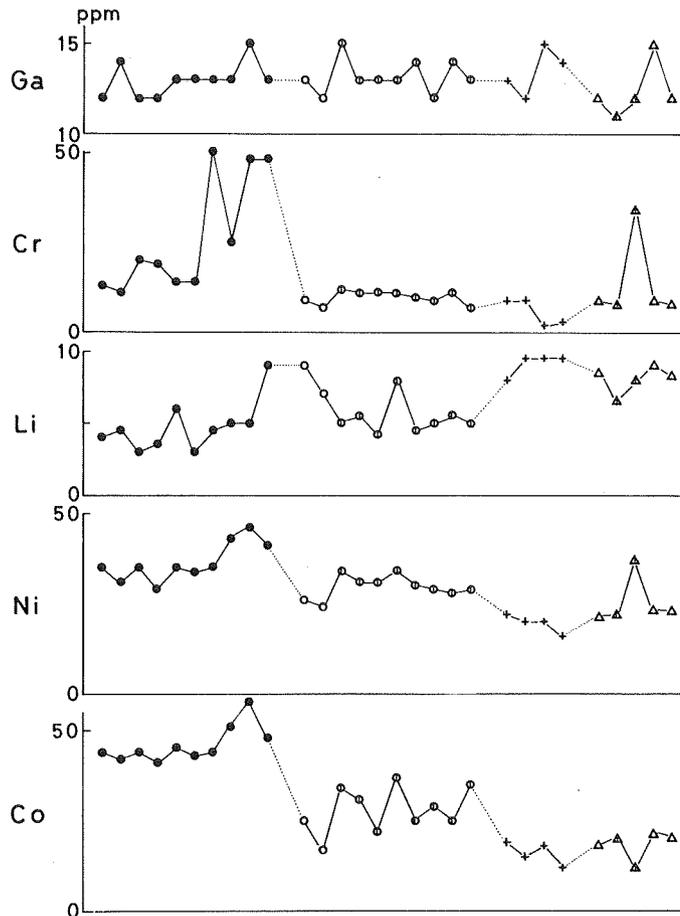


Fig. 4 Plots of Ga, Cr, Li, Ni and Co vs. the order of eruption. Symbols as same as Fig. 1.

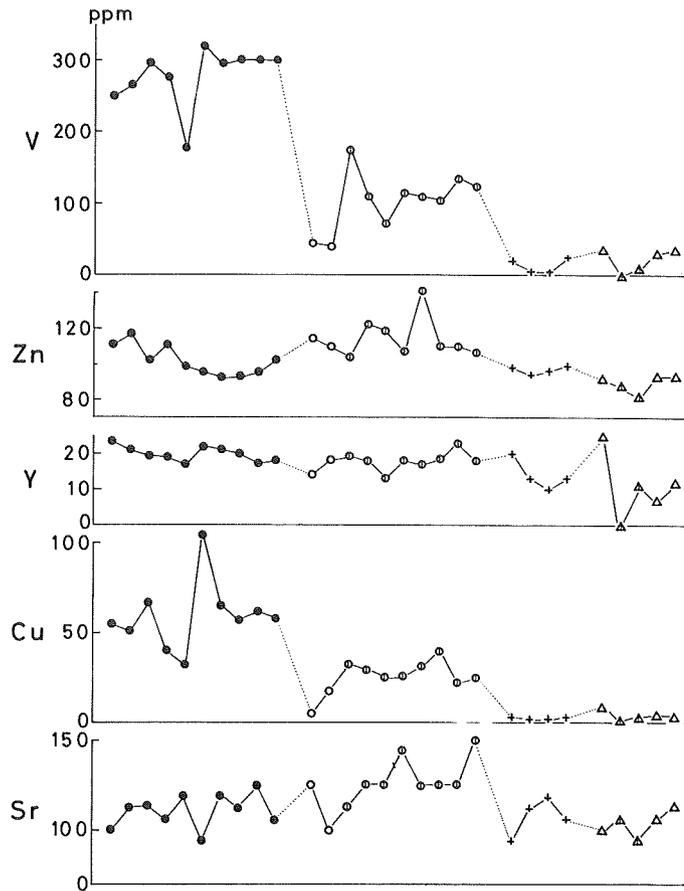


Fig 5 Plots of V, Zn, Y, Cu and Sr vs. the order of eruption. Symbols as same as Fig. 1.

The older somma lavas display the tholeiitic trend characterized by the enrichment of vanadium together with iron, whereas the products of the caldera and central cone stages exhibit the calc-alkaline trend represented by increasing SiO_2 without enrichment of vanadium and iron. The younger somma lavas, in turn, may be a transitional type between the above two trends. Such behavior of vanadium may be interpreted in terms of crystallization of magnetite in magma, since vanadium enters magnetite preferentially than any other minerals as shown in the Skaergaard intrusion (Wager and Mitchell, 1951) and other volcanic rocks (Duncan and Taylor, 1968; Taylor, et al., 1969; Matsumoto, 1969). In the calc-alkaline series, vanadium enters early crystallized magnetite, so its content in rocks decreases gradually towards the later stage. On the other hand, in the tholeiitic series, vanadium remains in the magma until magnetite begins to crystallize. This consideration may be

supported by the occurrence of magnetite, i.e. phenocrystic magnetite does not appear until the younger somma stage. Crystallization of magnetite might be controlled mainly by the oxygen partial pressure in the magma as suggested by Osborn (1959). Increase (or constancy) of oxygen partial pressure in the magma may be suggested in the middle to later stage of fractional crystallization. This may be responsible to the change in the rock series towards the later stage.

Conclusions

The minor element abundances in the Mashu rocks were examined. The Mashu rocks are characterized by low K, Li and Sr contents and by high total iron and V, having a similar feature to those of the low-K tholeiitic or the island arc tholeiitic series of Jakeš and Gill (1970) and Jakeš and White (1972). The measured concentrations of Ni and Cr are also overlapped with those of the island arc tholeiitic series.

The variation of minor element abundances versus silica, displays in most cases, a nearly smooth curve for each element without significant scattering.

Increase of Li content and decrease of Ni, Co, V and Cu contents are found from the older somma lavas towards the later products, in harmony with the long term variation of major elements. However, within a short period of activity, minor elements, such as, Cr, Li, V and Sr, and Li/Mg and Sr/Ca ratios, significantly change in accordance with the major element variation in the reverse order, felsic to mafic. Vanadium accompanied with iron shows a marked enrichment in the older somma lavas of the tholeiitic series, but not in the later volcanic products of the calc-alkaline and transitional series.

The facts mentioned above, may suggest that the Mashu rocks have been derived through fractional crystallization from a magma of the low-K tholeiitic or island arc tholeiitic type. The conversion of rock series from the tholeiitic to the calc-alkaline series may be ascribed to the increase (or constancy) of oxygen partial pressure in the magma, but not to new supply of calc-alkaline magma or to crustal contamination. This consideration may be supported by the chondrite normalized REE patterns (Masuda, et al., in press) and Sr⁸⁷/Sr⁸⁶ ratios (Katsui, et al., in press) of four samples from the Mashu rocks.

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