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Copper, lead, zinc, arsenic and sulfur of the Japanese granitoids (3) : Green Tuff Belt of Northeast Japan and Outer Zone of Southwest Japan

Shigeru TERASHIMA* and Shunso ISHIHARA**

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Abstract : Late Cenozoic granitoids occurring in the Green Tuff Belt of Northeast Japan and in the Outer Zone of Southwest Japan were analyzed for sulfur, copper, lead, zinc and arsenic, and the results were discussed, together with our previous data, in terms of the granitoid series classification, the relationship to the mode of occurrence and mineralization, and also of the time sequence.

Sulfur contents are higher in ilmenite series granitoids than magnetite-series granitoids, reflecting the contribution of sedimentary sulfur from the basement to the granitoids, and also high in small stocks related to sulfide mineralizations. Within single body or terrane, sulfur is contained more in mafic rocks than in felsic rocks. The late Cenozoic granitoids studied are significantly higher in sulfur content than the Cretaceous-Paleogene granitoids previously studied.

Copper is generally correlated positively with sulfur. It is also enriched in small mineralized stocks. The copper content of the late Cenozoic Green Tuff granitoids and Cretaceous Kitakami granitoids are higher than those of the other granitoids, whereas zinc contents have little regional variation. Both elements decrease with increasing of the differentiation index. Lead is more abundant in ilmenite-series granitoids than in magnetite-series granitoids in general. There is a clear positive correlation between the lead content and the differentiation index (hence K_2O content) in the ilmenite series but no correlation in the magnetite-series granitoids.

Arsenic has a great regional variation and is particularly high in the ilmenite-series Outer Zone granitoids. The content is very low in Cretaceous-Paleogene ilmenite-series granitoids of regional metamorphic terranes such as the Ryoke Belt and Abukuma Highland. Small granitic stocks associated with arsenic and tin mineralizations have the arsenic content higher than those unrelated.

Average values of the studied elements of the Japanese granitoids are more or less similar to those reported from other regions of the world. However, granitoids series and age and mode of occurrence of granitoids must be taken into consideration in the estimation of abundance of sulfur and arsenic.

* Geochemistry and Technical Service Department
** Mineral Deposit Department

Introduction

In order to understand metallogenesis of magmatic hydrothermal ore deposits of granitic terranes, we need chemical data of the ore components in their host rocks (TERASHIMA and ISHIHARA, 1974) and related granitoids

(TERASHIMA and ISHIHARA, 1976). About the ore elements concerned here, we have presented analytical results on granitoids from the Kitakami Mountains and Abukuma Highland of northern Honshu (TERASHIMA and ISHIHARA, 1983) and also those of the Inner Zone of Southwest Japan (TERASHIMA and ISHIHARA, 1984). This paper is the third one which describes the results of late Cenozoic granitoids occurring in the Green Tuff Belt of Northeast Japan and the Outer Zone of Southwest Japan, and compares the data with those of the previous works.

The samples analyzed were collected throughout the island arcs, as shown in Figure 1. Least altered rocks available on surface and underground were selected for the analyses. The analytical methods are atomic absorption for copper, lead and zinc, and modified atomic absorption for arsenic; and combution infrared absorption for sulfur, which are the same methods as those described in our previous papers (e.g., TERASHIMA, 1978, 1979).

Geologic Background

Granitoids of the Green Tuff Belt of Northeast Japan are distributed along the back-arc side of the islands, more or less parallel to the Quaternary volcanic chains. The granitoids have K-Ar mineral ages between 15 and 5 Ma.

They occur as small stocks in the most part but are seen as larger bodies, up to 500 km^2, in the Fossa Magna region of central Honshu where the late Cenozoic igneous activity cut across the older basement (Fig. 1). The granitoids intrude into late Cenozoic volcanic rocks in general, and locally into sedimentary rocks of the basement.

The late Cenozoic granitoids have generally magnetite-series characteristics and their composition varies from gabbro to monzogranite but is mostly tonalite-granodiorite. These rocks are hydrothermally altered in many cases, especially in small stocks. Most of the stocks are associated with base metal and manganese vein deposits. The mineralization is minor in and near the large bodies in the Fossa Magna region.

Granitoids of the Outer Zone of Southwest Japan, on the contrary, are distributed in the fore-arc side of the Japanese islands. The exposed areas are similar to those of the Green Tuff Belt, varying from more than 400 km^2 of batholithic size to less than 1 km^2 of stock size. Almost all the granitoids intrude into sedimentary basement.

The granitoids have narrow range of K-Ar mineral age around 12 Ma (SHIBATA, 1979). The composition is generally granodiorite-monzogranite having the most mafic facies of quartz diorite. The granitoids belong solely to ilmenite series, except for one alkaline-series pluton at Ashizurimisaki. Those distributed in the southern half of the Outer Zone have cordierite-bearing S type, but those in the northern half have cordierite-free I type characteristics (TAKAHASHI *et al.*, 1980).

The granitoids are associated with intense tin mineralization of both vein and skarn types in Kyushu. Base metal mineralizations are known to occur sporadically (ISHIHARA, 1978). The granitoids are relatively unaltered throughout these bodies.

Studied Plutons

Location of the studied plutons are illustrated in Figure 1. In the Green Tuff region of Hokkaido, four small stocks were selected from Neogene volcanic areas. Jozankei granodiorite porphyry is located in the center of base metal mineralization in the Toyoha mine area and is considered to be genetically related to the mineralization (YAJIMA, 1977). Small stocks at Oe and Jokoku mines are associated with rhodochrosite veins which may be accompanied by some amount of Ag-

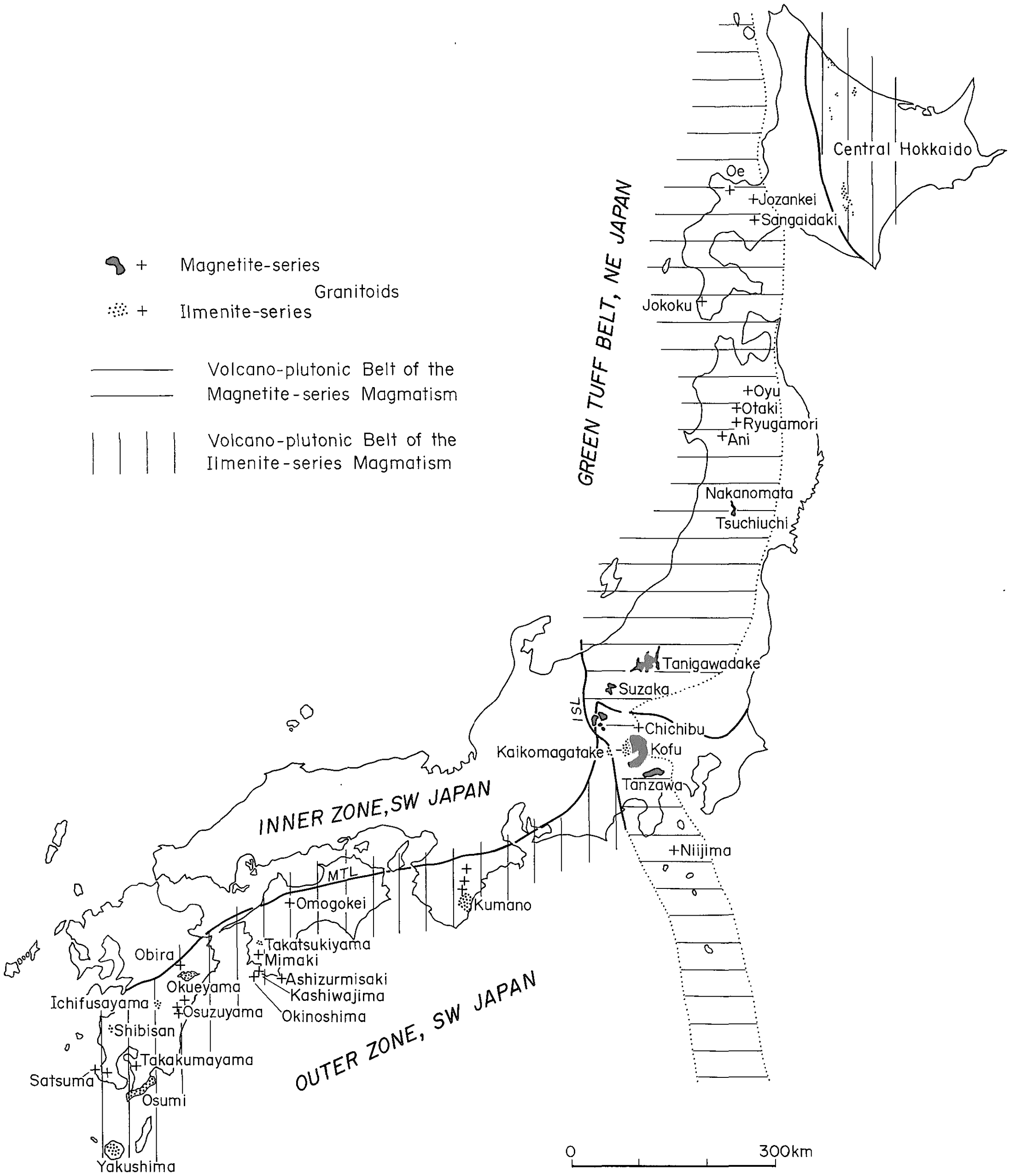

Fig. 1 Distribution of the studied granitic bodies in the Outer Zone of Southwest Japan and Green Tuff Belt of Northeast Japan. Among the Outer Zone granitoids, cordierite-free I-type bodies are Kaikomagatake, Omogokei, Okueyama, Ichifusayama, Shibisan and Satsuma peninsula ; cordierite-bearing S-type granitoids are Kumano, Takatsukiyama, Kashiwajima, Okinoshima, Osuzuyama, Takakumayama, Osumi, and Yakushima.

Table 1 Analytical results for magnetite-series plutonic rocks of the Green Tuff region (1): Hokkaido-Tohoku District

Sample No.	Rock description	DI (%)	S (ppm)	Cu (ppm)	Pb (ppm)	Zn (ppm)	As (ppm)
Sangaidaki							
MY Qd-91	Bt-Hb Qd*(chl>epd=cc)	52.6	110	45	11	74	n.d.
MY Adm 1 A	Px-Hb Tn*(epd>chl>sph)	58.1	70	36	12	62	n.d.
MY Gd 2	Bt-Hb Tn*(ser≫chl)	65.3	20	14	10	50	n.d.
MY Gd 1	Bt-Hb-Px Gd*(cc>ser)	72.3	40	23	15	48	n.d.
MY Adm 2 B	(Hb-) Bt MzG	83.0	20	6	20	53	n.d.
Jozankei							
74 HK-1	Hb Gd porphyry** (cc>ser)	66.3	80	16	11	61	n.d.
Oe mine							
74 HK 109	Hb Diot*	31.2	2,050	9	12	48	n.d.
74 HK 110	Bt-Hb Gd**(cc>chl=ser)	75.4	40	1	20	89	n.d.
Jokoku mine							
KK 76701401	Hb-Bt Qd	43.5	23	9	10	73	n.d.
KK 76702102	Hb-Bt Gd	69.4	30	21	11	39	n.d.
Oyu							
70 Ⅲ-1	Px Gd porphyry*(cc=chl)	61.1	50	24	12	67	0.8
70 Ⅲ-2	do.	62.8	50	69	13	71	1.2
Otaki							
77102706	Bt Gd, granophyre	72.6	20	2	4	65	n.d.
Ryugamori							
YF 81	Hb Diot**(ser>chl>epd)	30.2	1,480	78	22	111	2.0
YF 92-2	do.	30.3	1,520	40	17	84	3.1
YF 188	do.	37.8	30	34	20	106	1.2
YF 68	(Px-)Bt-Hb Diot	41.6	140	33	14	88	0.9
YF 69081905	Hb-Bt MzG*(chl>ser>cc)	82.0	20	16	18	22	0.4
YF 191	(Hb-) Bt MzG	84.1	20	31	4	34	0.4
Ani mine							
70 AN 20	Bt-Hb Diot*(chl=epd>cc)	48.5	910	96	35	28	n.d.
70 AN 2	(Hb-)Bt Gd*(chl>ser>epd)	75.0	32	8	8	60	0.3
70 AN 3	Bt MzG	78.2	82	99	20	66	0.4
70 AN 7	Bt MzG, porphyritic	79.2	470	96	5	83	0.3
Nakanomata mine							
70 AN 29	Hb Qd, granophyre	52.5	28	60	15	94	7.3
70 AN 28	do.**(cc>chl)	70.8	20	54	15	78	7.2
Tsuchiuchi							
70 AN 35	Hb Diot*	34.6	1,800	38	18	113	4.0
70 AN 32	Hb Qd*(ser)	50.2	20	25	14	104	6.2
70 AN 34	do. (ser>chl)	52.8	31	43	12	73	3.0
70 AN 31	Hb Tn (dike)*(cc>ser>chl)	56.0	10	70	13	68	1.4

Abbreviations for Tables 1-4 : Diot, diorite ; Qd, quartz diorite ; Tn, tonalite ; Gd, granodiorite ; MzG, monzogranite ; SyG, syenogranite ; Px, pyroxene ; Hb, amphibole ; Bt, biotite ; Epd, epidote ; Sph, sphene ; Cd, cordierite ; Gt garnet ; Tm, tourmaline ; Qz, quartz ; Qmd, quartz monzodiorite ; Mz, monzonite ; Ms, muscovite ; ser, sericite ; chl, chlorite ; epd, epidote ; cc, calcite ; sph, sphene ; (p), porphyritic ; [ilm], local ilmenite-series rock in the Green Tuff region. **strongly, *moderately altered rocks with the alteration mineral assemblage in parenthesis.

Table 2 Analytical results for magnetite-series plutonic rocks of the Green Tuff region (2): Fossa Magna area

Sample No.	Rock description		DI(%)	S(ppm)	Cu(ppm)	Pb(ppm)	Zn(ppm)	As(ppm)
Tanigawadake								
76 TN 147	Hb Diot-Qd		33.4	60	17	17	78	n.d.
76 TN 165	Px-Hb Tn		58.2	40	69	12	51	n.d.
76 TN 154	Bt-Hb Gd		61.8	85	38	12	65	n.d.
76 TN 152	Px-Bt-Hb Gd		63.3	70	38	15	50	n.d.
76 TN 158	Hb-Bt-MzG, granophyric		81.8	30	5	9	22	n.d.
76 TN 153	Sph-epd-chl MzG		87.8	20	4	10	22	n.d.
76 TN 166	Bt MzG porphyry		82.8	10	3	7	29	n.d.
Suzaka								
75 SZ 88	Hb Qd		53.2	10	2	4	21	1.1
76 SZ 131	do.*(epd>chl)		55.0	15	3	10	20	n.d.
76 SZ 132	Hb Tn, porphyritic		60.9	210	59	15	104	n.d.
76 SZ 127	Hb Qd, albitized		73.5	20	13	9	26	n.d.
Wadatoge								
75 WD 3	Px-Hb Diot**(epd>ser)		45.1	15	12	10	67	1.9
75 WD 9	Hb-Bt Gd		66.6	50	33	10	32	0.4
75 WD 11	Bt-Hb Gd		68.8	820	3	7	28	n.d.
76 WD 113	Hb-Bt Gd		69.4	20	5	8	36	n.d.
76 WD 100	Bt-Hb Gd		70.0	5	4	8	38	n.d.
76 WD 103	Hb-Bt Gd		81.7	35	3	9	40	n.d.
76 WD 101	Bt MzG(dike)		93.7	920	3	9	22	n.d.
Chichibu mine								
75 CB 93	Px-Bt Qd		39.6	50	22	17	82	n.d.
75 CB 90	(Px-)Bt-Hb Qd		47.7	220	46	28	95	n.d.
75 CB 83	(Tm-Bt)Hb Tn		56.1	3,250	30	9	30	11.7
75 CB 84	Hb Tn		56.2	960	38	6	31	11.3
75 CB 88	Bt-Hb Tn	[ilm]	56.4	160	16	16	71	n.d.
75 CB 73	Hb-Bt Tn		57.3	30	28	9	63	0.9
75 CB 74	do.	[ilm]	57.4	15	15	16	92	0.7
75 CB 82	(Tm-)Bt Gd		78.0	20	4	4	15	10.7
Kofu, Tokuwa-Kogarasu type								
75 KO 48	Hb Tn		39.7	50	57	64	150	n.d.
75 KO 32	Bt-Hb Tn		42.7	30	5	9	107	n.d.
75 KO 86	Hb-Bt Tn	[ilm]	51.2	830	14	18	69	n.d.
75 KO 84	Hb-Bt Tn		53.9	640	36	17	64	n.d.
80 KO 93	Hb-Bt Tn	[ilm]	56.0	1,320	18	15	72	n.d.
80 KO 99	do.	[ilm]	58.4	3,860	47	12	70	n.d.
75 KO 19	Px-Hb Tn		59.6	40	88	20	65	n.d.
75 KO 89	Bt-Hb Tn Gd		61.1	50	21	18	56	n.d.
80 KO 98	Hb-Bt Gd	[ilm]	61.6	80	9	14	52	n.d.
80 KO 84	do.*(cc>chl>epd)	[ilm]	64.1	70	8	11	43	n.d.
80 KO 105	Bt-Hb Gd		65.8	50	10	12	48	n.d.
Kofu, Mitake type								
74 KO 8	Hb-Bt MzG		85.7	30	2	20	47	0.4

(Table 2 continued)

Sample No.	Rock description	DI(%)	S(ppm)	Cu(ppm)	Pb(ppm)	Zn(ppm)	As(ppm)
74 KO 1	Bt MzG	89.1	20	1	19	13	0.2
74 KO 6	do.(granophyric)	92.6	20	1	19	22	0.7
74 KO 31	do.	93.7	40	2	32	25	0.4
Kofu, Ashigawa type							
75 KO 36	Hb Tn	61.6	10	3	3	20	n.d.
75 KO 38	(Bt−)Hb Tn	69.3	10	8	6	62	1.5
75 KO 51	Hb Tn	79.6	20	21	5	17	0.5
80 KO 102	Green Bt Gd, granophyric	83.2	30	1	2	2	n.d.
80 KO 103	Aplite dike(4 m wide)	91.8	10	2	2	1	n.d.
Tanzawa							
75 TA 7	Hb gabbro	12.5	680	31	13	79	n.d.
75 TA 16	(Epd−)Hb Diot	14.0	420	41	10	69	n.d.
75 TA 6	(Bt−)Hb Qd	36.6	10	25	8	108	n.d.
75 TA 17	do.	44.0	40	29	8	75	n.d.
75 TA 9	(Bt−)Hb Tn	51.3	80	25	5	58	0.4
75 TA 3	Hb−Bt Tn	69.8	30	3	4	30	0.2
75 TA 33	Epd−Bt Tn	75.3	20	2	6	34	0.9
75 TA 25	do.	78.4	120	19	6	4	0.1
Niijima							
NI 71061107	Bt Tn	81.3	60	3	5	31	n.d.
AM 2−1	do.	88.8	30	2	5	17	n.d.

Pb-Zn sulfides.

In the Tohoku District of northern Honshu, six stocks were selected. Ani and Nakanomata stocks are host rocks for copper vein-type deposits. Small copper vein is also related to Otaki granodiorite stock and Oyu granodiorite porphyry plug. Ryugamori stock is different from the others in that they belong to ilmenite-series which contain small amount of magnetite locally and intrude into the basement of sedimentary and older granitic rocks.

There are two distinctive features in the granitoids of the Fossa Magna region. Kofu batholith and Chichibu mine stock occur in sedimentary basement ; each body has a large proportion of local ilmenite-series granitoids, especially in area close to pelitic basement (ISHIHARA *et al.*, 1976). Presence of low-K_2O tonalites is another characteristic in the region, which is found in the southernmost part, such as in the Tanzawa body and in the southern part of the Kofu body. Intensive base metal mineralization is known only around calcic-series granitoids of the Chichibu mine stock (ISHIHARA *et al*, 1987).

Ilmenite-series granitoids of the Outer Zone of Southwest Japan are distributed in the area westward from Kaikomagatake of central Honshu. All but one occur in sedimentary basement. The Kaikomagatake body contains some amount of weakly magnetic, intermediate-series granitoids (ISHIHARA *et al.*, 1984). At Kumano, the monzogranite porphyry contains abundant alumino-silicates as xenocrysts. In Shikoku, many small stocks occur in the western part. The Omogokei stock is only body with igneous basement of the Sanbagawa metamorphic rocks, while the others occur in sandstone and shale of the Shimanto Supergroup.

Ashizurimisaki stock is composed of monzonite-syenite and their quartz-bearing variety (HAYASHI *et al.*, 1969), both of which may or may not contain some amount of magnetite, thus belonging to the intermediate series. The granitoids have very low initial ^{87}Sr / ^{86}Sr ratio (SHIBATA and ISHIHARA, 1979), and then may be considered as hot-spot type intrusion independent from the other calc-alkaline granitoids of the Outer Zone.

In Kyushu, the late Cenozoic granitoids occur widely, being S type ilmenite-series granitoids of Osumi and Yakushima as the

Table 3 Analytical results for the Outer Zone granitoids (1): Central Honshu–Shikoku

Sample No.	Rock description	DI (%)	S (ppm)	Cu (ppm)	Pb (ppm)	Zn (ppm)	As (ppm)
Kaikomagatake							
74 KO 61	Hb–Bt MzG	80.6	10	2	9	31	0.9
74 KO 68	Bt MzG	83.3	20	1	12	32	1.1
74 KO 70	do.	86.8	20	1	10	23	0.7
Kumano							
KUMA–3	Bt MzG (p)	79.2	1,550	17	21	45	8.6
KUMA–4	do.	80.6	950	13	19	34	9.0
KUMA–5	do.	89.1	30	3	21	26	1.4
Omogokei							
70 S 303	Hb–Bt Gd	69.4	60	9	16	61	0.4
70 S 304	Bt MzG	89.4	20	2	30	63	0.9
Takatsukiyama							
75 TS 44 W	Hb–Bt Gd	69.9	680	14	13	65	5.2
75 TS 50	do.	71.4	1,680	14	11	35	4.2
75 TS 26	do.	71.7	620	23	23	57	2.2
75 TS 38	Bt Gd	71.9	800	17	75	130	2.0
75 TS 33	do.	72.9	290	24	16	59	1.2
58 A 336	Bt MzG	80.8	40	2	13	22	0.5
75 TS 44 B–G	6 Xenoliths, Composite	73.0	3,180	73	11	65	16.0
Mimaki							
70 S–97	Bt MzG	87.5	30	1	22	25	0.5
Kashiwajima							
58 A 331	Bt MzG	79.9	270	4	50	71	3.5
58 A 328	do.	86.7	380	4	20	69	6.2
Okinoshima							
58 A 303	Cd–Hy–Bt Gd	69.3	50	8	17	87	n.d.
KS 59061501	do.	73.0	20	21	17	81	6.3
58 A 302	do.	75.6	10	17	23	76	n.d.
58 A 309	Bt MzG	92.3	510	2	27	46	n.d.
KS 59060809	Gt–Tm–Bt MzG	92.2	1,270	15	26	345	140
Ashizurimisaki							
58 A 158	Hb–Bt–Mz Diot	40.1	690	33	21	104	3.2
NM S–2	Syenite	86.2	56	10	18	121	0.9
NM S–5	do.	88.7	72	8	10	86	1.4
58 A 128	Bt–Hb Qz Syenite	90.4	56	3	10	86	2.7
NM I–3	Bt MzG	91.3	64	8	9	46	1.3
NM I–4	do.	91.5	40	7	9	30	1.6
58 A 130	do.	93.1	120	3	10	35	1.4
58 A 152	do.	95.2	30	2	11	25	0.6

Table 4 Analytical results for the Outer Zone granitoids (2) : Southern Kyushu

Sample No.	Rock description	DI(%)	S(ppm)	Cu(ppm)	Pb(ppm)	Zn(ppm)	As(ppm)
Obira Body							
75 OB 60	Bt–Hb Qmd	49.0	140	17	20	76	3.3
75 OB 65	Hb–Bt Gd	76.1	10	8	13	73	6.3
75 OB 59	Bt MzG	92.5	<10	2	27	19	0.5
Okueyama Body							
75 OK 89	Hb–Bt Gd	69.9	200	6	18	55	6.8
TN 1807	Bt MzG(dike)	93.0	5	2	33	12	0.6
TN 1806	Hb–Bt Gd	79.5	25	4	15	35	1.0
MT 10–450	do.	77.2	42	5	30	65	2.0
MT 9–360	do.	75.8	120	4	13	50	4.3
MT 8–560	do.	75.2	42	6	31	58	2.0
MT 7–650	do.	78.6	32	4	15	41	0.4
TN 1804	do.	71.3	50	6	15	60	1.4
TN 1803	do.	76.4	43	3	12	61	0.7
TN 1801	do.	70.6	10	7	24	63	1.4
MT 6–770	do.	75.9	38	2	13	51	0.4
MT 5–870	do.	77.2	78	4	12	44	1.3
MT 4–920	do.	77.0	40	5	16	45	4.7
75 OK–95	Bt MzG	86.4	1,180	26	16	16	3.4
TN 1611	do.	80.6	30	3	14	38	0.9
MT 3–1050	do.	82.5	20	2	12	30	1.0
MT 25–2	do. aplitic	91***	90	4	22	10	0.9
75 OK 100	do.	92.5	190	5	25	11	1.6
MT 24–5	do.	85***	24	2	19	28	0.2
75 OK 98	do.	96.6	2,260	29	39	2	2.5
MT 25–7	do.	84***	250	5	22	10	4.7
MT 2–1130	do.	84.1	40	6	16	25	0.4
MT 1–1250	do.	90.4	10	6	20	10	0.8
MT 26–2	do.	95***	22	5	25	8	0.2
MT 1015–4	Hb Mz	55.4	30	2	13	140	0.6
Osuzuyama							
75 KY 107	Bt Gd	75.0	640	4	22	31	31.0
75 KY 108	do.	75***	810	n.d.	n.d.	n.d.	18.0
75 KY 109	do.	74.1	1,100	12	25	54	31.0
75 KY 110	Mz granophyre	93.3	220	1	24	19	6.6
75 KY 111	do.	93***	2,480	n.d.	n.d.	n.d.	68.7
75 KY 112	do.	82.4	1,090	11	21	50	9.0
Ichifusayama							
ICD –8 B(10 c)	Bt Gd	67.8	800	19	20	80	2.2
75 KY 115	do.	67.9	1,570	21	21	65	2.0
75 KY 118	do.	66.5	1,020	22	22	80	1.6
75 KY 117	do.	68***	1,400	n.d.	n.d.	n.d.	1.0
75 KY 116	Bt MzG	87.2	720	4	16	28	0.4

(Table 4 continued)

Sample No.	Rock description	DI (%)	S (ppm)	Cu (ppm)	Pb (ppm)	Zn (ppm)	As (ppm)
Shibisan							
SB 1705	Bt Gd	75.6	40	3	18	53	4.7
SB 1703	do.	76.0	720	15	26	65	7.7
SB 1701	do.	76.8	30	2	17	49	1.3
SB 1602	do.	76.8	40	16	24	60	2.0
SB 1601	do.	76.8	1,180	15	20	42	12.5
Satsuma Peninsula							
Kinpo 3−1	Hb−Bt Gd	74.2	310	16	20	64	9.5
Mukaeyama 1	Bt Gd	76.1	1,620	66	31	72	24.0
Kaseda 13	Bt MzG	89.0	190	6	29	38	12.5
Takakumayama							
AT 604	Bt MzG	85.3	70	2	34	25	86.0
59080201	do.	87***	<10	4	52	21	2.0
57 Z 27 (2c)	do.	88.9	40	5	45	30	2.8
3372002	do.	89.1	20	18	38	35	7.4
AT 703	do. aplitic	89.8	10	3	40	26	0.8
AT 706	Aplitic MzG	91.3	20	3	44	27	4.0
3372109	do.	91.9	20	52	60	21	3.5
57 Y 07	do.	93.1	240	7	49	11	87.0
3352808	do.	93.9	10	10	48	21	2.4
Osumi							
64030405	Bt Gd	66.6	210	11	28	70	0.6
62030706	do.	71.3	1,010	16	26	90	1.6
620307011 A	do.	71.4	950	12	26	86	3.4
62022403	do.	72.1	1,010	16	25	80	2.4
62030801	do.	72.9	820	11	23	80	2.5
62022001	do.	74.0	310	8	24	55	4.8
K 34	do.	74.6	800	18	26	81	2.2
AT 407	do.	74.9	560	11	29	77	2.8
Yakushima							
YK 0110	Bt MzG(p)	76.5	470	7	21	60	n.d.
YK 1011 A	do.	78.2	940	11	20	61	48.0
YK 1003	do.	78.4	570	12	21	60	4.1
YK 0415	do.	79.3	385	7	21	55	n.d.

*** Values estimated from the K_2O contents (TERASHIMA and ISHIHARA, 1983).

largest in exposure. Intense tin and some base metal mineralizations are known to occur around satellitic stocks at northwestern side of the Okueyama body of I type ilmenite-series granitoids. Granitoids of the other bodies are accompanied by some tin and arsenic mineralizations.

Description of the studied samples and their analytical results are listed in Tables 1–4.

Sulfur content

Sulfur contents are plotted against differentiation index of THORNTON and TUTTLE (1960) in Figures 2 and 3. General distribution trend of Cretaceous magnetite-series granitoids of the Kitakami Mountains is shown in the figures for comparison, because these granitoids appear to represent a typical root zone of island-arc andesite.

In Figure 2, the Green Tuff granitoids are shown with two designations : One group of granitic bodies is that intruded into late Cenozoic volcanic rocks, while the other is that intruded into sedimentary basement and locally into older granitoids. The sediments-hosted granitic bodies have often local ilmenite-series granitoids which contain sulfur derived from the host rocks. This sulfur is often dispersed into magnetite-series granitoids to some distance (ISHIHARA *et al.*, 1985 ; 1987).

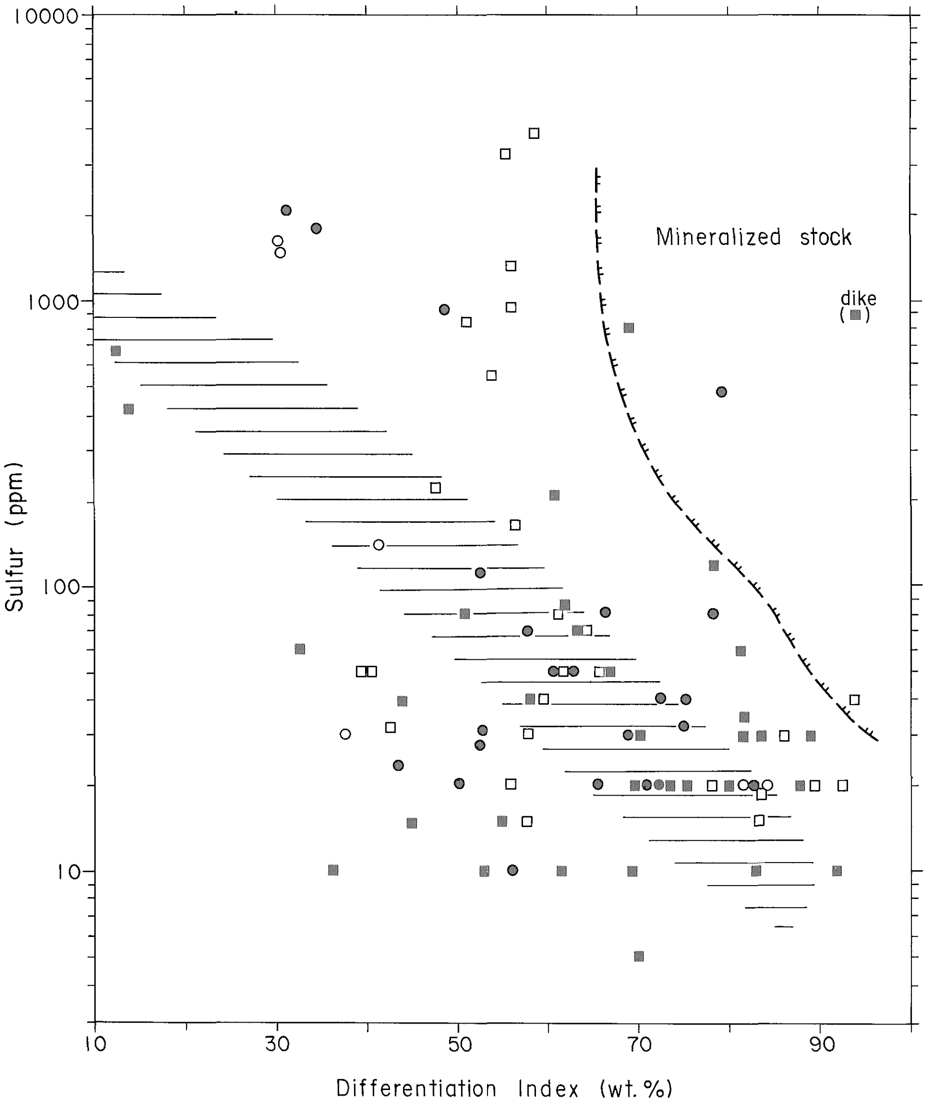

Fig. 2 Sulfur contents vs. differentiation index of the Green Tuff granitoids. (●) Volcanic hosted, (○) Sedimentary hosted bodies in the Tohoku-Hokkaido region. (■) Volcanic hosted, (□) Sedimentary hosted bodies in the Fossa Magna region. Shaded is general area for the magnetite-series granitoids of the Kitakami Mountains (after ISHIHARA *et al.*, 1983 ; TERASHIMA and ISHIHARA, 1983).

Plots of the Green Tuff granitoids are widely scattered as compared with those of Cretaceous-Paleogene granitoids previously reported (TERASHIMA and ISHIHARA, 1983, 1984). One of the reasons may be pervasive alteration on the studied granitoids, which are described in Tables 1 and 2. Granitoids of the Ani stock are biased to sulfur rich area, while those of the Nakanomata and Tsuchiuchi bodies are characterized by low sulfur contents.

Among unaltered granitoids of the Fossa Magna region, low K_2O tonalite of the Tanzawa type is consistently low in the sulfur content, except for the well differentiated rocks. Local ilmenite-series granitoids of the

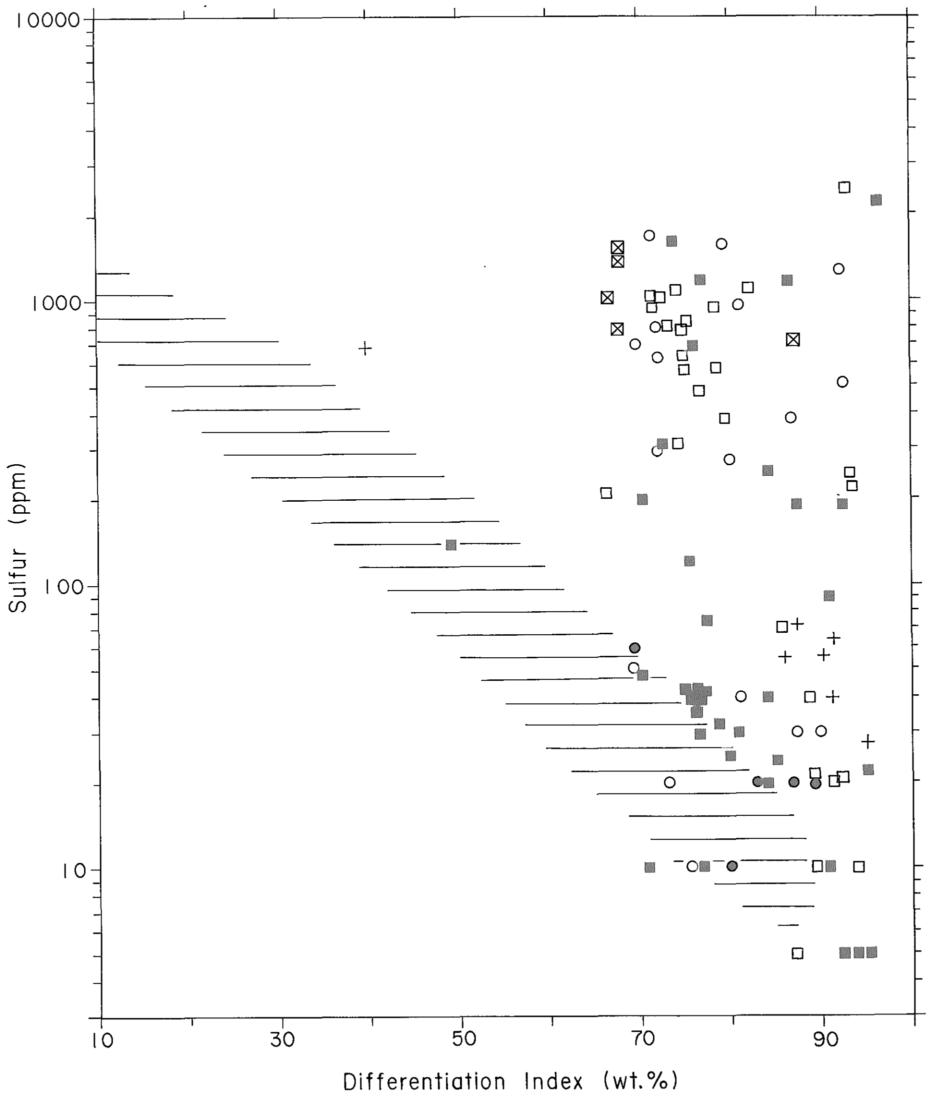

Fig.3 Sulfur contents vs. differentiation index of the Outer Zone granitoids. (●) I type, (○) S type in the central Japan-Shikoku ; (+) Intermediate series of Ashizurimisaki ; (■) I type, (□) S type in Kyushu. (⊠) Ichifusayama body. The content less than 10 ppm is arbitrary plotted as 5 ppm S.

sediments-hosted granitic bodies have sporadic high values.

Plots of the Outer Zone granitoids are completely different from those of the Green Tuff granitoids (Fig. 3). The granitoids have narrow range of differentiation index but a great variety in the sulfur content. Most of the granitoids have much higher values than the Green Tuff granitoids; the values are particularly high in S type granitoids. Sedimentary xenoliths are commonly seen in most of the high sulfur granitoids, such as in Takatsukiyama, Osuzuyama, Ichifusayama, Shibisan bodies. It is noteworthy to point out that homogeneous granitoids of Osumi and Yakushima batholiths have constantly high contents of sulfur.

Base Metal Content

Base metal contents of the Green Tuff granitoids are shown in Figure 4. Zinc contents have generally good positive correlation to FeO contents, thus negatively to the differentiation index (TERASHIMA and ISHIHARA, 1983, 1984). However, the Green Tuff granitoids have widely scattered plots in the zinc-differentiation index diagram, due possibly to decomposition of the original mafic silicates. Zinc contents of the Fossa Magna granitoids are lower than those of the Tohoku-Hokkaido granitoids.

Copper contents are generally high in mafic rocks and are seen under the ore-microscope as proportional to the chalcopyrite contents. The Green Tuff granitoids have again wide scattering (Fig. 4), because of hydrothermal alteration pervasive in them. Copper contents of the Tohoku-Hokkaido granitoids are generally higher than those of the Fossa Magna granitoids, and anomalously high contents are found in the Ani stock.

Lead contents are generally low in all the range of differentiation index, which is one of the characteristics of magnetite-series granitoids (ISHIHARA and TERASHIMA, 1977).

Zinc contents of the Outer Zone granitoids are shown in Figure 5. Among the granitoids of Shikoku and central Honshu, the contents are low at Kaikomagatake but are high at Okinoshima and Kashiwajima, and are extremely high in monzonite-syenite of Ashizurimisaki.

At Okueyama of Kyushu, zinc contents decrease greatly with increasing of differentiation index. The granodiorite has the contents higher than that of the average Japanese granitoids, but the monzogranite is very depleted in the element. Among the other granitoids, the S type has generally higher content than the I type. Local monzonite of the Okueyama body has very high zinc content (Fig. 5).

Copper contents of the Outer Zone granitoids are generally lower than those of the Japanese average (Fig. 5). They are especially low in the I type granitoids of Kaikomagatake, Omogokei and Okueyama bodies. There are a few extremely high values in both the I and S types of well differentiated facies in the Okueyama and Takakumayama bodies, which are due to chalcopyrite contained in them.

Lead contents are again higher in the S types than in the I types and the contents increase greatly with increasing of differentiation index, especially within monzogranite composition of the Okueyama and Takakumayama bodies. Alkaline rocks of Ashizurimisaki body have low contents of lead indicating K-feldspar of this body is depleted in lead.

Arsenic Content

Regional variation of arsenic in Japanese granitoids was discussed previously (TERASHIMA and ISHIHARA, 1976). Among late

Cenozoic granitoids studied, those of the Green Tuff Belt are lower in arsenic content than those of the Outer Zone. Among the Outer Zone granitoids, intermediate-series granitoids of the Ashizurimisaki and I type ilmenite-series granitoids of the Kaikomagatake and Omogokei are the lowest, ranging from 0.4 to 3.2 ppm As.

I type ilmenite-series granitoids of the Okueyama and Ichifusayama (0.2–6.8 ppm As)

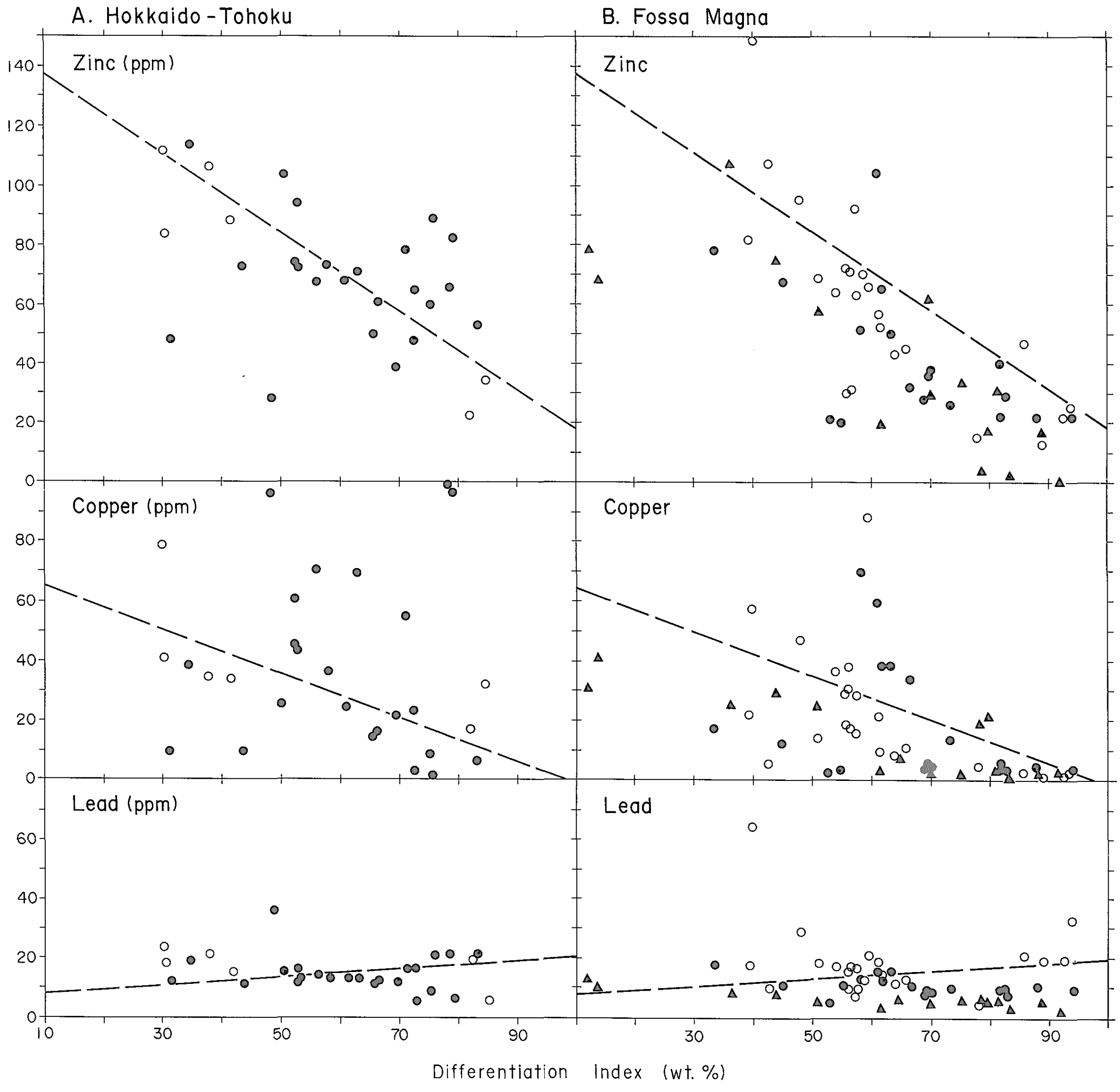

Fig.4 Base metal contents vs. differentiation index of the Green Tuff granitoids. A : Tohoku-Hokkaido region, (●) Volcanics-hosted (most of the bodies other than Ryugamori) ; (○) Sediments-hosted (Ryugamori). B : Fossa Magna region, (●) Volcanics-hosted (Tanigawadake, Suzaka and Wadatoge), (▲) Tanzawa type (Tanzawa, Niijima and Ashigawa of Kofu body), (○) Sediments-hosted (Chichibu mine and Mitake and Tokuwa types of Kofu body). Broken line is average composition of Japanese granitoids (ISHIHARA *et al.*, unpublished).

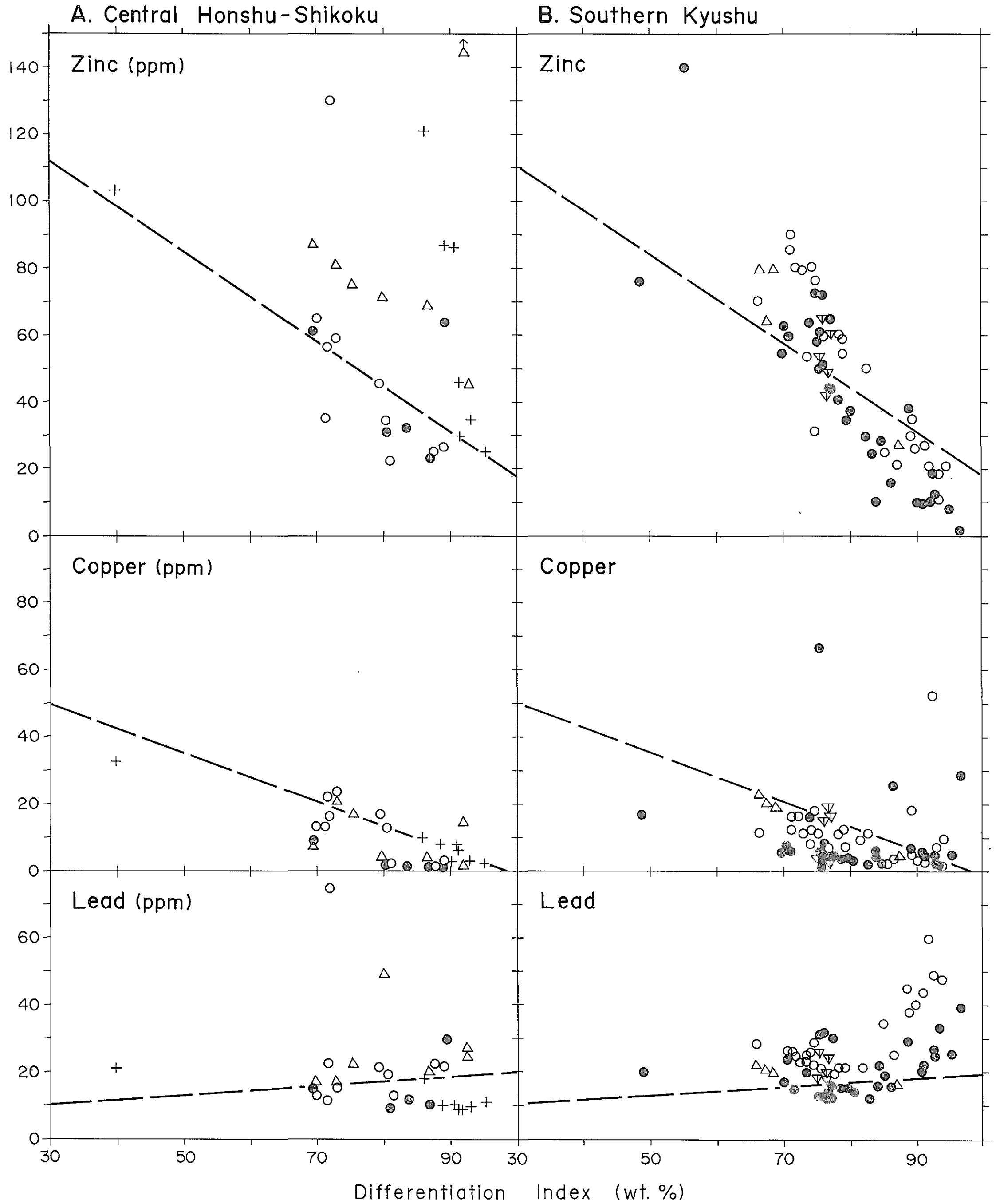

Fig. 5 Base metal contents vs. differentiation index of the Outer Zone granitoids. A : Central Honshu-Shikoku region, (●) I type (Kaikomagatake and Omogokei), (○) S type (Takatsukiyama-Mimaki and Kumano) ; (△) Kashiwajima and Okinoshima ; (+) Intermediate series of Ashizuri-misaki. B : Kyushu, (●) I type (Okueyama, Obira and Satsuma Peninsula) ; (▽) Shibisan ; (△) Ichifusayama ; (○) S type (Osuzuyama, Takakumayama, Osumi and Yakushima).

may be ranked as the next, and the highest values are observed in S type ilmenite-series granidtoids of small stocks such as of the Osuzuyama and Takakumayama.

Average Contents and Comparison with the Previous Works

Areal variation in the average contents of sulfur, copper, lead, zinc and arsenic in the studied granitoids are listed in Table 5.

From the Green Tuff Belt, only the granitic bodies of the Fossa Magna region were selected, because the other bodies in the Hokkaido-Tohoku district are small in size, often altered and related to mineralization. Within

Table 5 Areal variation of average contents for sulfur, copper, lead, zinc and arsenic in the granitoids.

Area	(n)	D.I. (%)	S (ppm)	Cu (ppm)	Pb (ppm)	Zn (ppm)	As (ppm)
Green Tuff Belt, Fossa Magna							
Tanigawadake	(7)	67.0	45	25	12	45	n.d.
Suzaka	(3)	62.5	80	25	9	50	n.d.
Wadatoge	(5)	71.3	186	10	8	35	n.d.
Kofu, Tokuwa–Kogarasu type							
Magnetite–series	(6)	53.8	143	36	23	82	n.d.
Ilmenite–series	(4)	56.8	1,523	22	15	66	n.d.
Kofu, Mitake type (Ilm.–series)	(4)	90.3	28	2	23	27	0.4 (4)
Kofu, Ashigawa type	(5)	77.1	16	7	4	20	1.0 (2)
Tanzawa	(8)	47.7	175	22	8	57	0.4 (4)
Niijima	(2)	85.1	45	3	5	24	n.d.
Average (magnetite–series)	(36)	63.2	109	20	11	48	0.6 (6)
All analyses	(44)	65.0	230	19	12	48	0.5 (10)
Outer Zone, Central Honshu–Shikoku							
Kaikomagatake	(3)	83.6	17	1	10	29	0.6 (3)
Kumano	(3)	83.0	843	11	20	35	6.3 (3)
Omogokei	(2)	79.4	40	6	23	62	0.7 (2)
Takatsukiyama–Mimaki	(7)	75.2	591	14	25	56	2.3 (7)
Kashiwajima	(2)	83.3	325	4	35	70	4.9 (2)
Okinoshima	(5)	80.5	372	13	22	127	73.2 (2)
All analyses	(22)	79.7	423	10	22	67	10.2 (19)
Outer Zone, Southern Kyushu							
Obira	(3)	72.5	52	9	20	56	3.4 (3)
Okueyama	(23)	81.4	210	7	19	36	1.9 (23)
Osuzuyama	(6)	82.1	1,057	7 (4)	23 (4)	39 (4)	27.4 (6)
Ichifusayama	(5)	71.5	1,102	17 (4)	20 (4)	63 (4)	1.4 (5)
Shibisan	(5)	76.4	402	10	21	54	5.6 (5)
Satsuma Peninsula	(3)	79.8	707	29	27	58	15.3 (3)
Takakumayama	(9)	90.0	48	12	46	24	21.8 (9)
Osumi	(8)	72.2	709	13	26	77	2.5 (8)
Yakushima	(4)	78.1	591	9	21	59	26.1 (2)
All analyses	(66)	79.7	446	10 (63)	25 (63)	46 (63)	9.9 (64)

Number of samples in parenthesis. n.d., not determined.

single body, altered rock and local uncommon rocks were excluded.

Average content of sulfur in the magnetite-series Green Tuff granitoids of the Fossa Magna region is 109 ppm (n=36). This value is clearly lower than that of the ilmenite-series Outer Zone granitoids of the Central Shikoku-Honshu (423ppm, n=22) and the southern Kyushu (446 ppm, n=66) regions. The same tendency is seen in the Tokuwa-Kogarasu type granodiorite of the Kofu batholith of the Fossa Magna region, where the magnetite-series granodiorite is averaged as 143 ppm (n=6), while the ilmenite-series one yields an average of 1523 ppm (n=4).

Among all the studied granitoids, anomalously high zinc content of 345 ppm was found in the Okinoshima monzogranite, which may be due to sulfide concentration during the deuteric stage for its high sulfur and arsenic contents (see Table 3). For the other samples, zinc contents were found to have little regional variation. The average contents of arsenic vary widely from 0.4 to 73.2 ppm, and the values of Central Honshu-Shikoku (10.2 ppm, n=19) and southern Kyushu (9.9 ppm, n=64) regions are distinctly higher than those of the Fossa Magna region (0.5 ppm, n=10). There is no clear positive or negative correlation between arsenic and other element contents.

Average values of sulfur, copper, lead, zinc and arsenic of major granitoids belts of Japan, which were observed by the present and previous works (TERASHIMA and ISHIHARA, 1983, 1984 ISHIHARA and TERASHIMA,1985) are given in Table 6, together with some reference data of TUREKIAN and WEDEPOHL (1961) and MASON (1958). These granitoids are Cretaceous to Paleogene in the Kitakami Mountains, Abukuma Highland, Ryoke Belt, Sanyo Belt and Sanin Belt but are Eocene to Miocene in the Hidaka Belt of central Hokkaido, Green Tuff Belt and Outer Zone of Southwest

Table 6 Comparison of the results in average of this study with references data.

	(n)	D.I. (%)	S (ppm)	Cu (ppm)	Pb (ppm)	Zn (ppm)	As (ppm)
Cretaceous–Paleogene granitoids							
Kitakami Mountains[1)]	(80)	64.3	89 (56)	27	11	59	2.1 (37)
Abukuma Highland[1)]	(25)	76.3	75 (24)	6	16	59	0.6
Ryoke Belt[2)]	(52)	77.9	84	7	20	53	0.7 (45)
Sanyo Belt[2)]	(53)	81.4	32	6	22	47	2.8 (48)
Sanin Belt[2)]	(44)	79.4	64 (43)	7	12	43	1.3
Eocene–Miocene granitoids							
Central Hokkaido[3)]	(22)	71.3	402	12	16	65	2.5 (18)
Green Tuff Belt, Fossa Magna[4)]	(44)	65.0	230	19	12	48	0.5 (10)
Outer Zone, Central Honshu–Shikoku[4)]	(22)	79.7	423	10	22	67	10.2 (19)
Outer Zone, Southern Kyushu[4)]	(66)	79.7	446	10 (63)	25 (63)	46 (63)	9.9 (64)
Cretaceous–Paleogene granitoids[4)]	(254)	74.4	68 (228)	13	16	52	1.6 (199)
Eocene–Miocene granitoids[4)]	(154)	74.3	375	13 (151)	19 (151)	52 (151)	7.9 (111)
Japanese granitoids[1)–4)]	(408)	74.4	192 (382)	13 (405)	17 (405)	52 (405)	3.8 (310)
High–Ca granitoids[5)]	n.g.	n.g.	300	30	15	60	1.9
Low–Ca granitoids[5)]	n.g.	n.g.	300	10	19	39	1.5
The earth crust[6)]	n.g.	n.g.	520	45	15	65	2

Number of samples in parenthesis. n.g., not given. Data source 1) TERASHIMA & ISHIHARA (1983), 2) TERASHIMA & ISHIHARA (1984), 3) ISHIHARA & TERASHIMA (1985), 4) This study, 5) TUREKIAN & WEDEPOHL (1961), 6) MASON (1958)

Table 7 Sulfur contents of Paleozoic to Cenozoic sedimentary rocks and Recent sediments.

Age	Area	(n)	S (%)
Paleozoic	Chugoku district	(67)	0.105
	Shikoku district	(10)	0.186
Mesozoic	Chugoku district	(69)	0.231
	Shikoku district	(22)	0.226
Cenozoic	Shikoku district	(12)	0.320
Recent	Marine & Non-marine	(228)	0.502*

Data taken from TERASHIMA *et al.* (1981 and 1983).
* Excluding for brackish-water sediments (average 1.40%, n = 39).

Japan. They belong generally to magnetite-series in the Kitakami Mountains, Sanin Belt and Green Tuff Belt, but mainly to ilmenite-series in the other terranes.

Sulfur contents of the Cretaceous-Paleogene granitoids vary from 32 ppm (Sanyo Belt) to 89 ppm (Kitakami Mountains), and their average is 68 ppm (n=228). The contents of the Eocene-Miocene granitoids range from 230 ppm (Green Tuff Belt) to 446 ppm (southern Kyushu), and their average of 375 ppm (n=154) is about five times higher than that of the Cretaceous-Paleogene granitoids. Sulfur is generally abound in mafic rocks than felsic ones, but in the regional comparison, spatial hence temporal variation is much more distinct, being sulfur predominant in the younger granitoids.

Sulfur contents of sedimentary rocks from the Chugoku and Shikoku districts of Southwest Japan (TERASHIMA *et al.*, 1981) and those of Recent sediments from several lakes and sea areas around the Japanese Islands (TERASHIMA *et al.*, 1983) have been previously reported. The results are summarized in Table 7. There is a clear tendency that the sulfur contents increase as their ages decrease ; the Recent sediments being most dominant in sulfur. The coincidence is considered not accidental but genetical, because a majority of the studied granitoids are ilmenite-series which has been reduced by sedimentary materials.

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地　名

Sangaidaki	三　階　滝
Oe (mine)	大江（鉱山）
Jokoku (mine)	上国（鉱山）

Oyu	大　湯	Mimaki	御　槇
Otaki	大　滝	Kashiwajima	柏　島
Ryugamori	竜ヶ森	Okinoshima	沖ノ島
Ani (mine)	阿仁（鉱山）	Obira	尾　平
Nakanomata (mine)	中股（鉱山）	Okueyama	大崩山
Tsuchiuchi	土　内	Osuzuyama	尾鈴山
Suzaka	須　坂	Ichifusayama	市房山
Ashigawa	芦　川	Shibisan	紫尾山
Tokuwa—Kogarasu	徳和—小烏	Kinpo	金　峰
Mitake	御　岳	Mukaeyama	向江山
Wadatoge	和田峠	Kaseda	加世田
Kaikomagatake	甲斐駒ヶ岳	Takakumayama	高隅山
Omogokei	面河渓	Osumi	大　隅
Takatsukiyama	高月山		

日本の花崗岩類中の銅，鉛，亜鉛，ヒ素と硫黄（3）
グリーンタフ帯と西南日本外帯

寺島　滋・石原舜三

要　旨

東北日本のグリーンタフ帯と西南日本外帯から得られた新世代後期の花崗岩類184試料について硫黄，銅，鉛，亜鉛，ヒ素を分析し，これら元素の広域的な分布や岩石の産状，鉱化作用との関連等についてすでに報告した他地域の結果と合せて検討し，次の結果を得た．

硫黄は，磁鉄鉱系花崗岩類よりもチタン鉄鉱系に多く含有される傾向があり，これは主として硫黄に富む堆積岩類の影響と考えられた．鉱化に関連するストック状の花崗岩体も硫黄に富む場合が多く，また白亜紀～古第三紀の花崗岩類（北上山地，阿武隈高地，西南日本内帯）に比べて新生代の花崗岩類には高い硫黄含有量を示す試料が多かった．

銅も鉱化に関係する岩体では高い特徴があり，多くの場合硫黄含有量とは正の相関を示した．銅含有量の平均値は，グリーンタフ帯と北上山地が他地域よりも高い傾向を示すが，亜鉛含有量には明瞭な地域差はなく，またこの両元素については硫黄で見られるような時代的な相違は認められなかった．銅，亜鉛の含有量はいずれも分化指数の増加に伴って減少する．鉛は，磁鉄鉱系花崗岩類よりもチタン鉄鉱系に，Ｉタイプ花崗岩質岩よりもＳタイプにより多く含有される傾向があった．分化指数との関係では，チタン鉄鉱系花崗岩類では正の相関が認められたが，磁鉄鉱系では認められなかった．

ヒ素含有量は，岩体や地域差が極めて大きい特徴があり，広域変成帯である領家帯や阿武隈高地では低く，チタン鉄鉱系花崗岩類の西南日本外帯で最も高い値が得られた．ヒ素やスズ鉱床に関係する花崗岩体も高いヒ素含有量を示した．

日本の花崗岩類中の銅，鉛，亜鉛含有量の平均値は，世界の花崗岩質岩石の平均値とほぼ良好な一致を示した．そしてヒ素は世界の平均よりもやや高く，硫黄は若干低い傾向が認められた．しかし，これら両元素の含有量は地域や産状，年代等によって大きく異なるため，これらを充分考慮した存在量評価を行う必要があろう．

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