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Sulfur and carbon contents and $\delta^{34}S$ ratio of Miocene ilmenite-series granitoids: Osumi and Shibi-san plutons, Kyushu, SW Japan

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Abstract: Total sulfur and carbon were analyzed by infrared absorption spectrometry for 117 specimens from two Miocene ilmenite-series granodiorite-monzogranitic plutons, Osumi (426 km^2) and Shibi-san (18 km^2) of SW Japan, and whole rock $\delta^{34}S$ ratio was also determined for 9 selected specimens. The results are displayed in the rock-facies maps to identify their areal distribution and local heterogeneity. About the total sulfur, the Osumi pluton gives a range of 10-3,550 ppm with a high average value of 493 ppm (104 samples). The content is high in the Koyama type which contains restitic garnet and the Heda-Okawa type which contains abundant sedimentary enclaves. The total carbon content varies 30-670 ppm and the average is 191 ppm (n=103). The carbon content is high in the Koyama type and some parts of the Oura SW body and Hanaze types; thus S/C ratio is the highest in the Heda-Okawa type.

In the Shibi-san pluton (13 samples), total sulfur content is generally low as 20-70 ppm with two high values (>1,500 ppm). The content is generally high in the Hirabae type of the northern half. Total carbon content is high in the southwestern part of the Kusubae type; thus S/C ratio is generally high in the north.

$\delta^{34}S$ data of the Osumi granitoids with various S contents, reveal constant values of −8.3 to −7.0 permil, while those of the Shibi-san granitoids variy from −6.2 to −5.7 permil. Thus, sulfur is considered isotopically homogenized at depth. Presence of many sedimentary enclaves in the granitoids and the negative $\delta^{34}S$ values strongly indicate contribution of sedimentary sulfur. In referring to $\delta^{34}S$ values of the Shimanto Supergroup of western Shikoku (−9.4‰, 2,400 ppm S), the sulfur of the granitoids is considered mostly originated in metamorphic equivalent of the Shimanto Supergroup; 10-20 percent mixing of the sedimentary rocks can produce the observed $\delta^{34}S$ values of the Osumi granitoids, and 5-10 percent mixing is necessary to produce the Shibi-san granitoids.

1. Introduction

In the Cretaceous-Paleogene accretional sedimentary terrane (Shimanto Supergroup) of the Outer Zone of SW Japan, there occur two types of ilmenite-series granitoids of Miocene age: one characterized with a narrow range of SiO_2 contents, such as Osumi and Shibi-san plutons, while the other having a wide variation in SiO_2 contents, as exemplified by Okue-yama and Takakuma-yama bodies (S. Ishihara, unpublished data). The latter granitoids have high silica rocks at the top and center of the plutons, indicating that the wide range of silica contents formed by in-situ magmatic fractionation. The former granitoids are considered un- or less-fractionated.

There are many tin-arsenic-base metal deposits around the northwestern margin of the Okue-yama body. The Takakuma-yama body is associated with similar mineralization (Hamachi and Ishihara, 1958; Ishihara and Kawachi, 1961). Little or no such association, on the other hand, has been known with the Osumi and Shibi-san plutons, which may be typical examples of S- and I-type, respectively, ilmenite-series granitoids (Takahashi *et al*., 1980).

The previous reconnaissance study (Terashima and Ishihara, 1986) indicated that the Osumi granitoids contain the highest average content of total sulfur (average 709 ppm, n=8) among Japanese granitoids. It was considered that sulfur in the granitic magma was originally much in amount and was fixed within the granitoid itself for the very low redox state of the

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original magmas (Ishihara *et al.*, 1988). Yet, the number of sulfur analyses has not been large enough, so we present here additional data of total sulfur and carbon analyses on systematically collected 104 samples for the Osumi pluton and 13 samples for the Shibi-san pluton, in order to confirm the previous conclusions. The sample localities are given in Figures 1 and 4. Rock sulfur isotopic analyses (Sasaki and Ishihara, 1979) are also done on selected samples.

The analyzed granitoids are freshest rocks available on surface, and are those used previously for major and minor chemical analyses (Tateishi *et al.*, 1986; Nishimura and Yamamoto, 1994; Yamamoto *et al.*, 1988). The analytical method is infrared absorption spectrometry of Terashima (1979). Powdered samples of 0.1 and 0.5 g were weighed out into ceramic crucible. The surface was covered by iron powder and tungsten chips added. The crucible was placed on the pedestal of the furnace and introduced to the combustion tube and combusted for 35-60 sec. at ca. 1,800°C. The detection limits are 20 ppm for total carbon and 5 ppm for total sulfur.

Microscopic studies indicate abundant pyrrhotite and some chalcopyrite occurring in the analyzed granitoids. No sulfate sultur has been detected among the selected S-rich samples (Terashima and Ishihara, 1986). Thus, the total sulfur implies the sulfides sulfur. Although carbonate carbon was not analyzed, high total carbon rocks were tested qualitatively by HCl, and only no. 58 sample was found to contain certain amount of carbonate carbon. Thus, the total carbon is considered to indicate graphite carbon in general. Graphite-bearing enclaves have been known in the Osumi pluton (Ishihara, 1982).

2. The Osumi Pluton

The Osumi pluton is located at the southernmost part of the Osumi Peninsula, Kagoshima Prefecture. It is exposed in an area of 426km^2 elongating east-northeast (Fig. 1), which is a common lineament in southwestern Japan, and intrudes Paleogene units of the Shimanto Supergroup. The granitoids have K-Ar age of 14-15 Ma (Sato *et al.*, 1992), and are covered by Plio-Pleistocene ash flow deposits and Alluvium. The pluton varies in composition from granodiorite, mon-

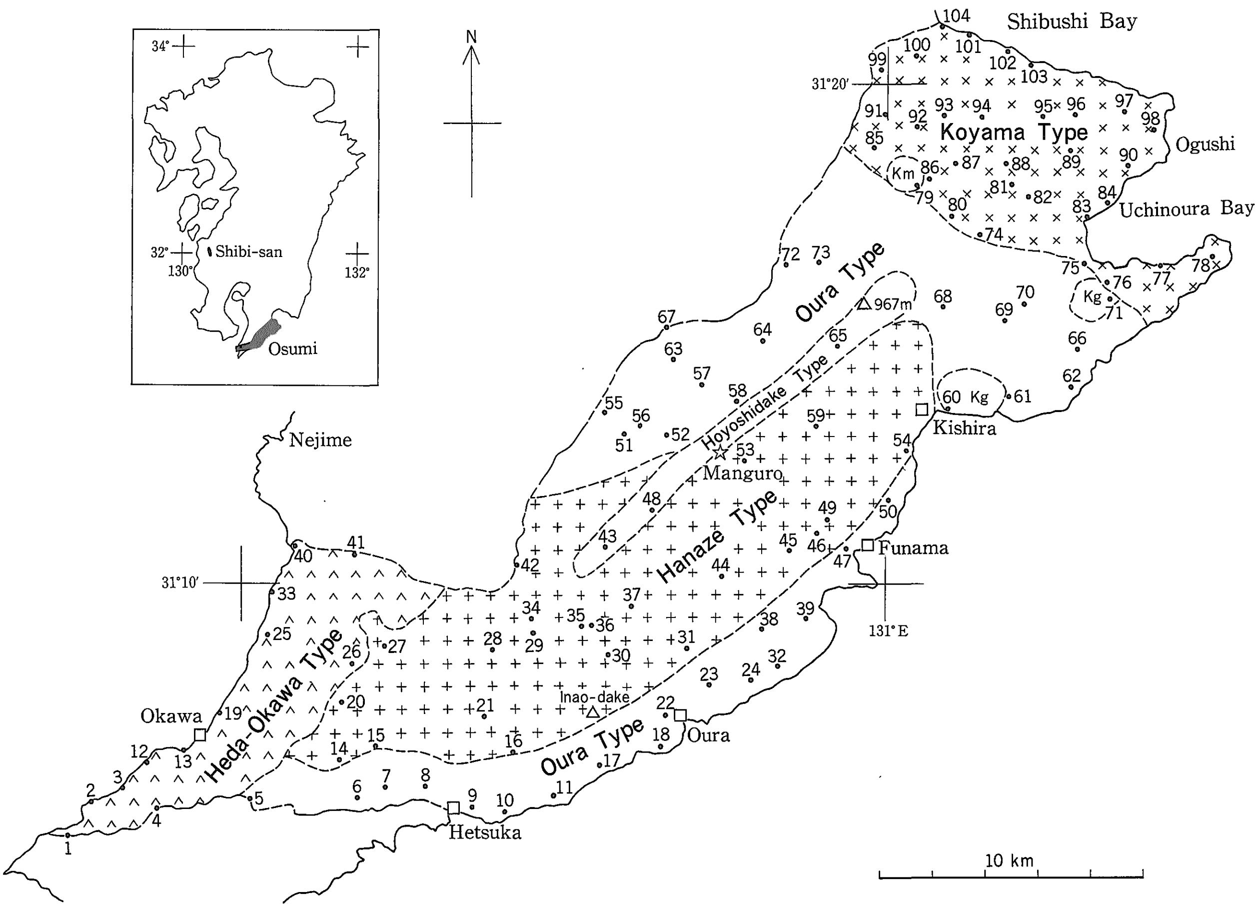


Fig. 1 Sample locality and major rock types of Miocene granitoids in the Osumi pluton. Boundaries of the granitoid types are taken from Yamamoto *et al.* (1983). Kg: Kawaguchi type; Km, Kunimi type. ☆: Locality of graphite-bearing enclave.

zogranite, then to aplitic monzogranite, but the major part is granodiorite, having a narrow range, 66–69%, of SiO_2 contents (Tateishi *et al.*, 1986).

The Osumi granitoids are characterized by their heterogeneous appearance due to small mafic clots (Plate 1), which may be remnants of well-digested restite of sedimentary and mafic igneous protolith, and by the presence of accessory pyrrhotite, which is shown by limonite staining on fresh outcrop (Plate 2A). No Al-silicates but garnet is a common rock-forming mineral in some phases of the granitoids (Plate 2B).

Mafic igneous enclaves, as identified by an igneous texture (Plate 2C) and also the presence of common hornblende and microscopic ophitic texture (Ishihara, 1982), are rare but present. Sedimentary-metamorphic enclaves as identified by their texture and presence of Al-silicates (Plates 2D) and Ca-poor amphiboles, are common throughout the pluton. In some occasions, pyrrhotite and graphite are very dominant in these enclaves (Plate 2E and 2F). Those of the Manguro old adit were said entrained with N–S strike and 50°E dip; sulfides were particularly rich in the well digested enclaves, thus mined during 1943–44 (Ishihara, 1982).

Biotite is a universal mafic silicate. Hornblende is present, and orthopyroxene, cummingtonite, garnet and pyrrhotite, but no magnetite are present in minor amounts. Accessory minerals are allanite, apatite, zircon, cordierite, corundum, titanite, muscovite, tourmaline, axinite, and chalcopyrite (Yamamoto and Oba, 1983). The copper contents give an average of 12.9 ppm for the Osumi granitoids (n=8, Terashima and Ishihara, 1986).

The granitoids are divided into three major and three minor types; all of which change gradually each other. The major types are named, from west to east, Heda-Okawa type, Hanaze and Oura types, and Koyama type (Fig. 1). The Hanaze type is distributed in the center among these types (Yamamoto *et al.*, 1983).

The Heda-Okawa type is hornblende-bearing biotite granodiorite and partly monzogranite, which looks heterogeneous for mafic clots, having biotite±plagioclase and biotite+hornblende+cummingtonite assemblages. Sedimentary-metamorphic enclaves are common in this type (Plate 2B). Xenocrysts of cordierite and corundum have been observed (Yamamoto *et al.*, 1983).

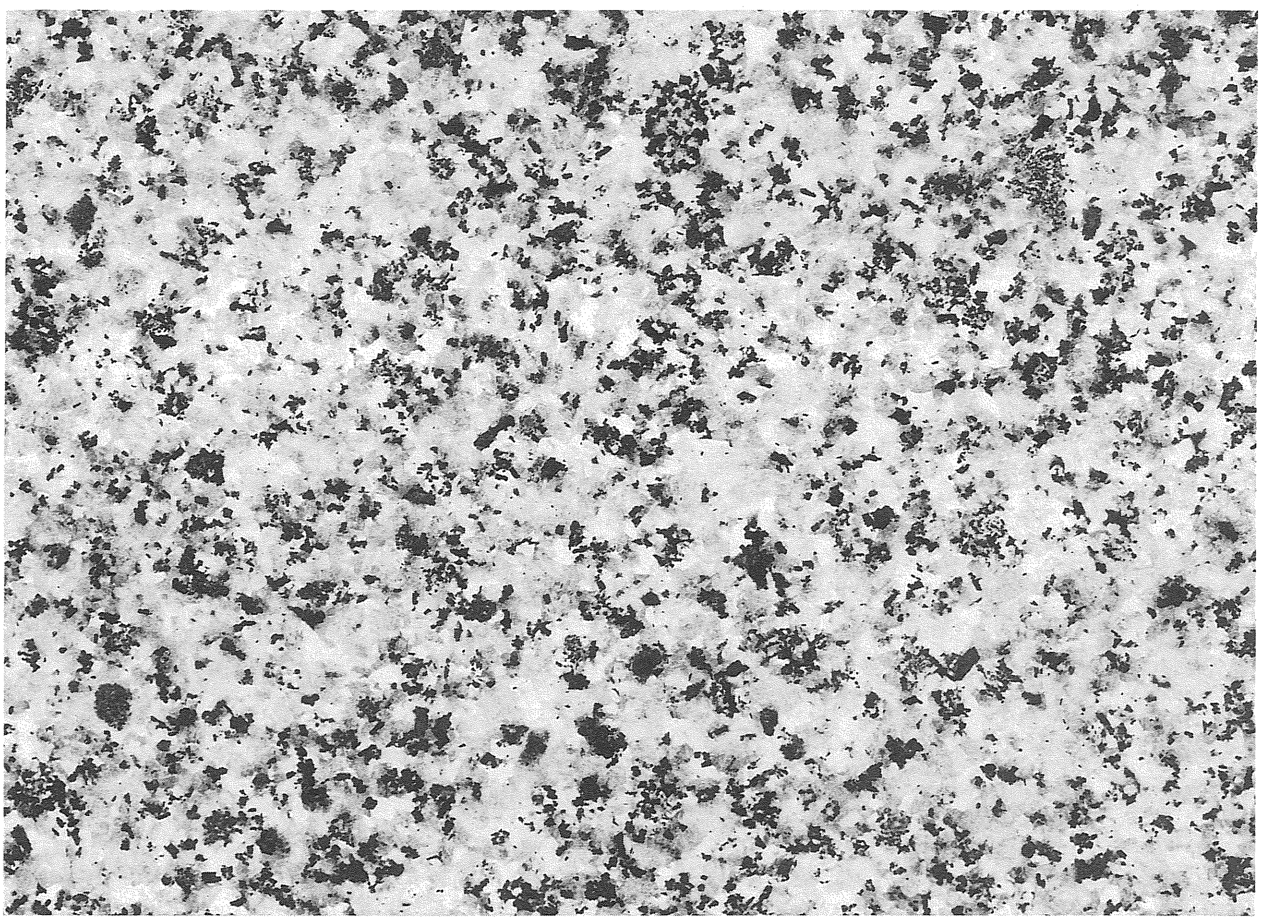


Plate 1 The Oura-type granodiorite from the Kai Industrials quarry at Hetsuka, only stone quarry opened in the Osumi pluton. Note abundant small clots composed of aggregates of mafic silicates which could be well-digested remnant of sedimentary and mafic igneous enclaves (x 1).

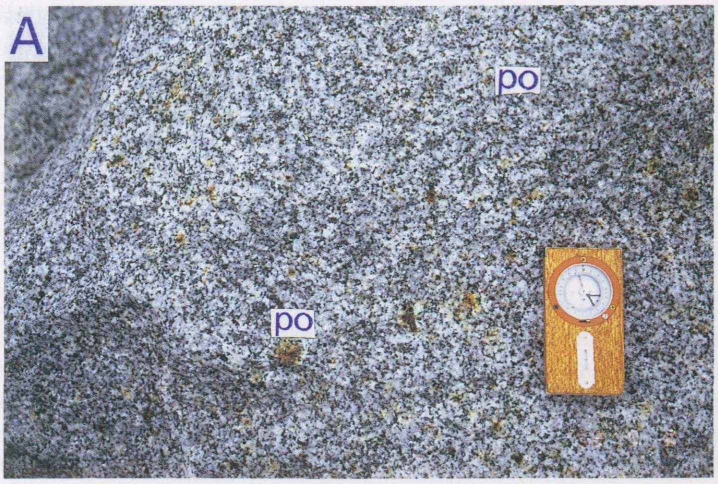


Plate 2A Common appearance of the Osumi granodiorite in terms of limonite spot after decomposition of pyrrhotite (po). The Oura type in Funama. Length of the clinometer is 11.5 cm.

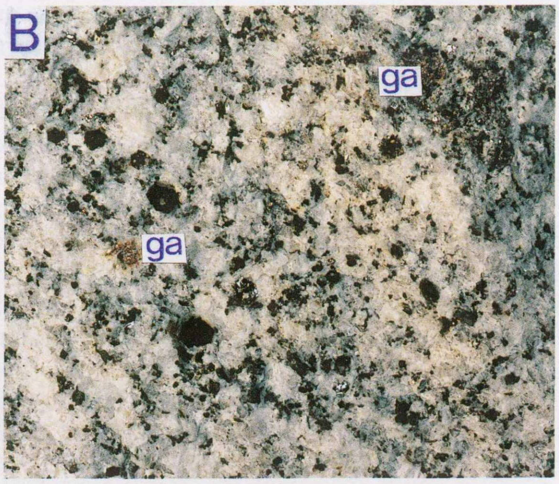


Plate 2B Reddish brown garnet (ga) occurring in the Koyama-type granodiorite. The width is 7.3 cm. Todoroki Fall.

Plate 2C Mafic igneous enclave in the Hanaze type. The igneous origin is shown by the texture and presence of not orthpyroxene and cummingtonite but augite and common hornblende.

Plate 2D Sillimanite needle (sm)-bearing gneiss enclave in the Koyama type granodiorite. Ogushi coast.

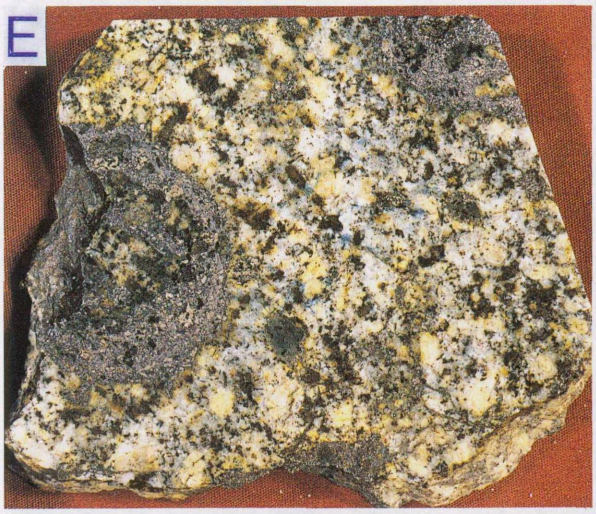


Plate 2E A ball-like pyrrhotite-graphite-bearing enclaves (58MG41) in the Oura type granodiorite, Manguro, Uchinoura township. Chalcopyrite-rich part was mined in 1943. The width is 7.8 cm.

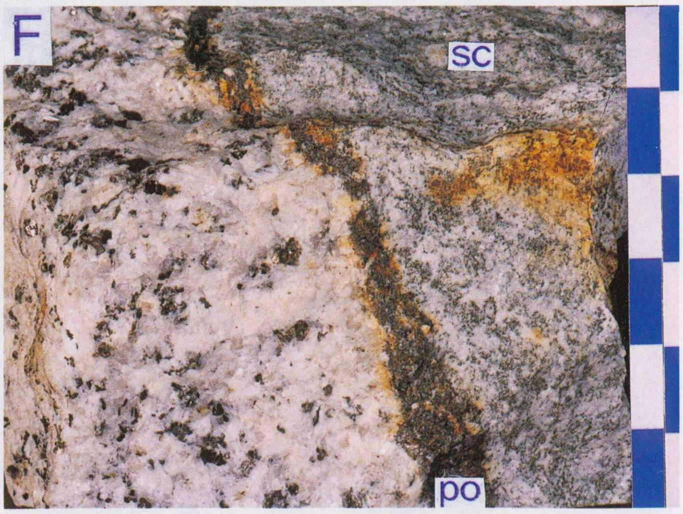


Plate 2F Sheelite-bearing hydrothermally altered enclave (sc) with pyrrhotite ring (po) in the Heda-Okawa type monzogranite (58K17B), Yamamoto, Nejime township. The scale bar is in cm.

The Hanaze and Oura types are orthopyroxene-cummingtonite-bearing biotite granodiorite and monzogranite, which may lack orthopyroxene in some places. These types are also heterogeneous, because of mafic clots containing biotite±plagioclase, biotite+cummingtonite±orthopyroxene, and biotite+garnet, and also sedimentary enclaves. Trace amounts of garnet and tourmaline have been observed (Yamamoto *et al.*, 1983).

The Koyama type is garnet-bearing biotite granodiorite-monzogranite (Plate 2B), and contains small amounts of spinel and tourmaline. This type is also heterogeneous for mafic clots having mineral assemblages of biotite+plagioclase, and biotite+

Table 1 Sulfur and carbon contents of the Osumi granitoids.

Filing no.	Sample no.	Rock type	S (ppm)	C (ppm)	S/C
1	OS-04-40	Heda-Okawa	420	200	2
2	OS-05-41A	do.	390	200	2
3	OS-05-42A	do.	760	110	7
4	OS-05-43	do.	1,340	120	11
5	OS-05-45	Oura-SW	770	70	11
6	OS-05-47A	do.	650	90	7
7	OS-05-48A	do.	310	160	2
8	OS-05-49	do.	220	110	2
9	OS-05-50	do.	590	120	5
10	OS-05-51A	do.	620	100	6
11	OS-05-52	do.	500	80	6
12	OS-06-42	Heda-Okawa	340	80	4
13	OS-06-43	do.	550	80	7
14	OS-06-47	Hanaze	<10	70	<0.1
15	OS-06-48	do.	560	130	4
16	OS-06-51A	Oura-SW	210	70	3
17	OS-06-53A	do.	370	290	1
18	OS-06-54	do.	630	540*	1
19	OS-07-44	Heda-Okawa	590	100	6
20	OS-07-47	Hanaze	800	180	4
21	OS-07-50	do.	110	100	1
22	OS-07-54	Oura-SW	690	300	2
23	OS-07-55B	do.	640	370*	2
24	OS-07-56	do.	20	90	0
25	OS-08-45B	Heda-Okawa 1	1,320	50	26
26	OS-08-47	do.	630	70	9
27	OS-08-48	Hanaze	870	60	15
28	OS-08-50	do.	330	40	8
29	OS-08-51	do.	470	260	2
30	OS-08-53A	do.	560	360	2
31	OS-08-55	do.	30	80	0
32	OS-08-57	Oura-SW	110	350	0
33	OS-09-45A	Heda-Okawa	560	100	6
34	OS-09-51	Hanaze	440	310	1
35	OS-09-52	do.	400	450*	1
36	OS-09-53	do.	1,360	70	19
37	OS-09-54A	do.	980	670*	2
38	OS-09-57	Oura-SW	710	410*	2
39	OS-09-58	do.	880	50	18
40	OS-10-46	Heda-Okawa	450	100	5
41	OS-10-47	do.	770	110	8
42	OS-10-51	Hanaze	440	40	11
43	OS-10-53	Hoyoshidake	100	70	1
44	OS-10-56	Hanaze	190	70	3
45	OS-10-57	do.	90	30	3
46	OS-10-58	do.	280	110	3
47	OS-10-59	Oura-SW	50	130	0
48	OS-11-54	Hoyoshidake	20	170	0
49	OS-11-58	Hanaze	10	80	0
50	OS-11-00	Oura-SW	440	50	9
51	OS-12-53	Oura-NE	180	50	4
52	OS-12-54	do.	60	60	1
53	OS-12-56	Hanaze	30	260	0
54	OS-12-00	do	10	30	0
55	OS-13-53B	Oura-NE	450	80	6
56	OS-13-54	do.	300	50	6
57	OS-13-55	do.	130	50	3
58	OS-13-56	do.	10	660**	n.c.
59	OS-13-58A	Hanaze	610	60	10
60	OS-13-01	Kawaguchi	310	60	5
61	OS-13-02	Oura-NE	170	70	2
62	OS-13-04	do.	210	200	1
63	OS-14-55	do.	920	90	10
64	OS-14-57	do.	220	110	2
65	OS-14-58	Hoyoshidake	70	80	1
66	OS-14-04	Oura-NE	660	130	5
67	OS-15-54	do.	470	100	5
68	OS-15-01	do.	70	70	1
69	OS-15-02	do.	420	80	5
70	OS-15-03	do.	240	80	3
71	OS-15-05B	Kawaguchi	550	330	2
72	OS-16-57	Oura-NE	110	120	1
73	OS-16-58	do.	360	110	3
74	OS-16-02	Koyama	940	350	3
75	OS-16-04	do.	30	60	1
76	OS-16-05	do.	440	230	2
77	OS-16-06A	do.	750	280	3
78	OS-16-07A	do.	1,040	200	5
79	OS-17-00	do.	30	210	0
80	OS-17-01	do.	280	410*	1
81	OS-17-02	do.	930	370	3
82	OS-17-03A	do.	640	300	2
83	OS-17-04	do.	570	60	10
84	OS-17-05	do.	1,000	260	4
85	OS-18-59A	do.	570	260	2
86	OS-18-00	do.	510	320	2
87	OS-18-01	do.	610	300	2
88	OS-18-02A	do.	710	330	2
89	OS-18-04A	do.	850	220	4
90	OS-18-05B	do.	450	370	1
91	OS-19-59	do.	630	180	4
92	OS-19-00C	do.	890	330	3
93	OS-19-01A	do.	900	380	2
94	OS-19-02B	do.	710	340	2
95	OS-19-03A	do.	590	330	2
96	OS-19-04B	do.	640	390	2
97	OS-19-05A	do.	770	240	3
98	OS-19-06	do.	600	340	2
99	OS-20-59C	do.	210	280	1
100	OS-20-00	do.	870	290	3
101	OS-20-01	do.	890	290	3
102	OS-20-02	do.	700	350	2
103	OS-20-03	do.	860	300	3
104	OS-21-01	do.	80	580	0

* some CO_2, ** much CO_2 examined by HCl; n.c., not calculated.

spinel+plagioclase. This type is most aluminous and least calcic in ACF diagram (Tateishi *et al.*, 1986).

Among minor types, the Kawaguchi type (Kg in Fig. 1) occurs only in the Uchinoura area and is orthopyroxene-cummingtonite-bearing biotite granodiorite, i.e., slightly more mafic than the Hanaze and Oura types.

Hoyoshidake type occurring at topographically high levels, on the other hand, is more leucocratic than the Hanaze and Oura types, being generally porphyritic biotite monzogranite containing trace amounts of muscovite and garnet. This type is considered one of the latest phases of the Osumi granitoids. Kunimi type (Km in Fig. 1) occurs as small stocks and is fine-grained garnet-bearing muscovite-biotite aplitic monzogranite, which is well fractionated and the latest phase among various types of the Osumi granitoids.

3. Sulfur and Carbon Contents of the Osumi Pluton

Total sulfur and carbon contents and S/C ratio of the Osumi granitoids are listed in Table 1. They are geographically plotted in Figure 2.

The total sulfur content is high in the northeasternmost part and westernmost portions. These regional distribution trends are also reflected on the individual types.

Sulfur content is generally high in the Heda-Okawa type (average 677 ppm, n=12) and Koyama type (677 ppm, n=29). The former contains a strange enclave which is composed of scheelite-disseminated altered core with pyrrhotite crust in fresh biotite granite (Plate 2F), and the latter is rich in restitic garnet (Plate 2B).

Oura type is geographically divided into the southwestern body and the northeastern body. The former is higher in sulfur content (482 ppm, n=17) than the latter (278 ppm, n=18). The Hanaze type is averaged as 422 ppm (n=24). Among minor types, Hoyoshidake type is very low in the total sulfur content (average <50 ppm, n=4).

The total carbon content, on the other hand, is generally high in the northeastern-most part where the Koyama type is distributed and the central coast area of the Oura type.

Total sulfur and carbon contents are poorly correlated (Fig. 3); the correlation coefficient is 0.205. The carbon content is the highest in the Koyama type (average 306 ppm, n=29). Thus, the Koyama type gives

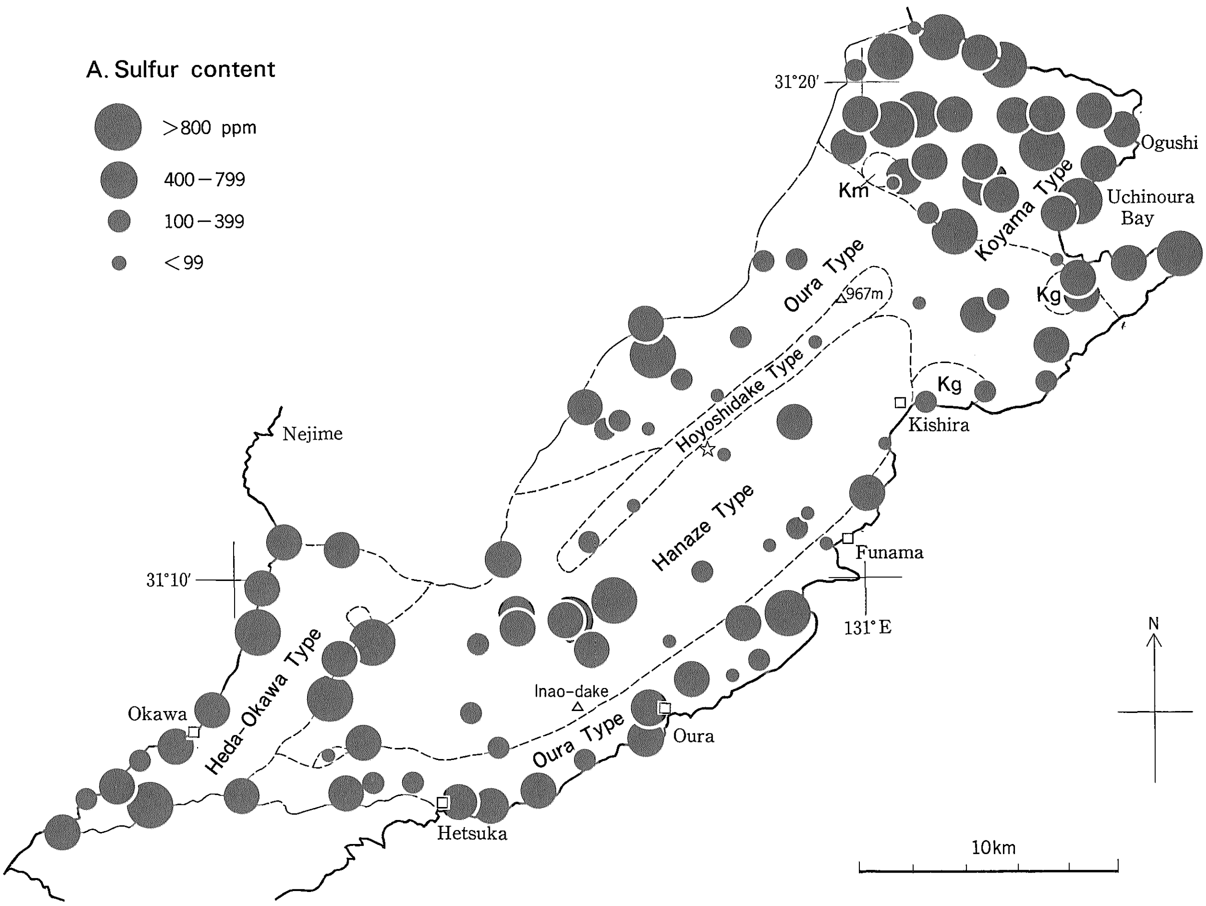
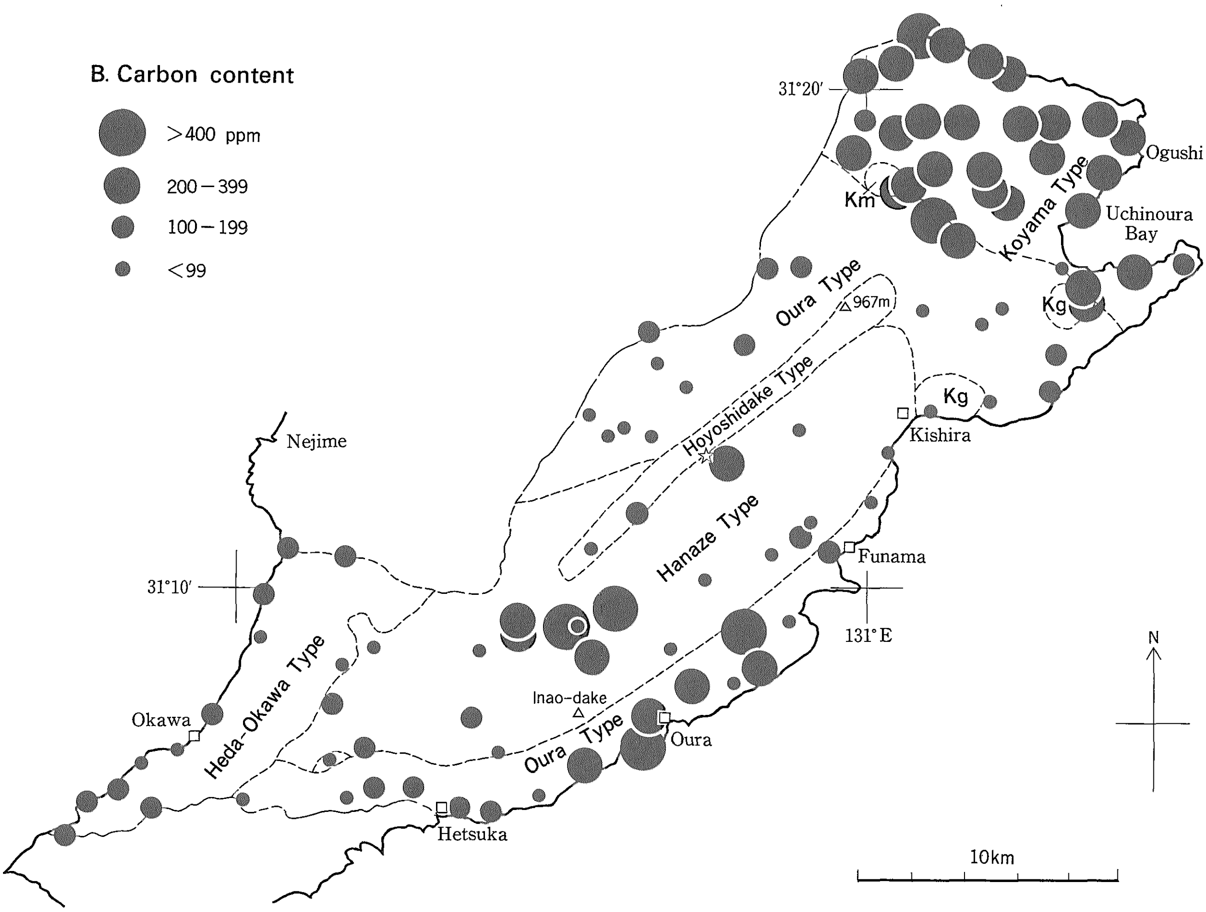
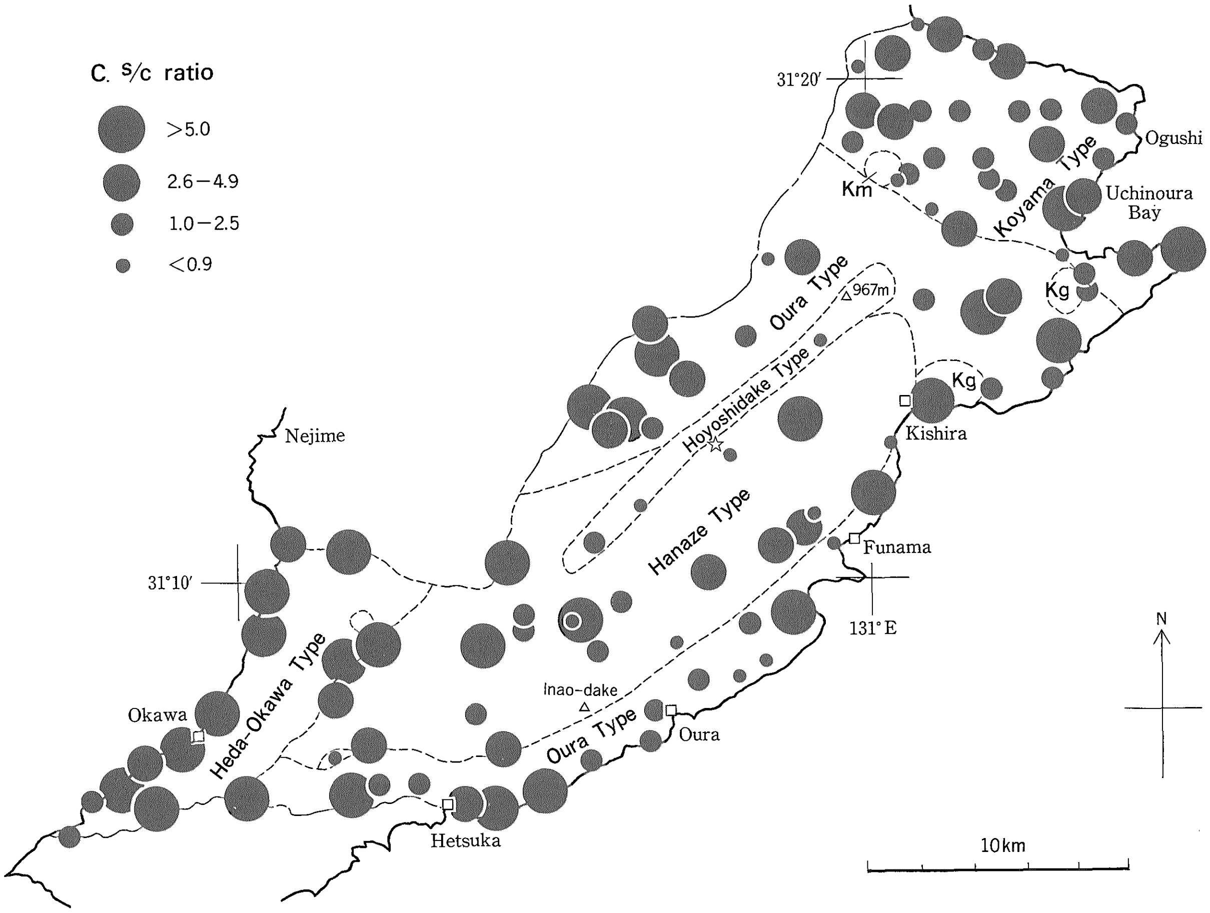


Fig. 2 Distribution of total sulfur (A) and carbon (B), and S/C ratio (C) in the Osumi pluton.





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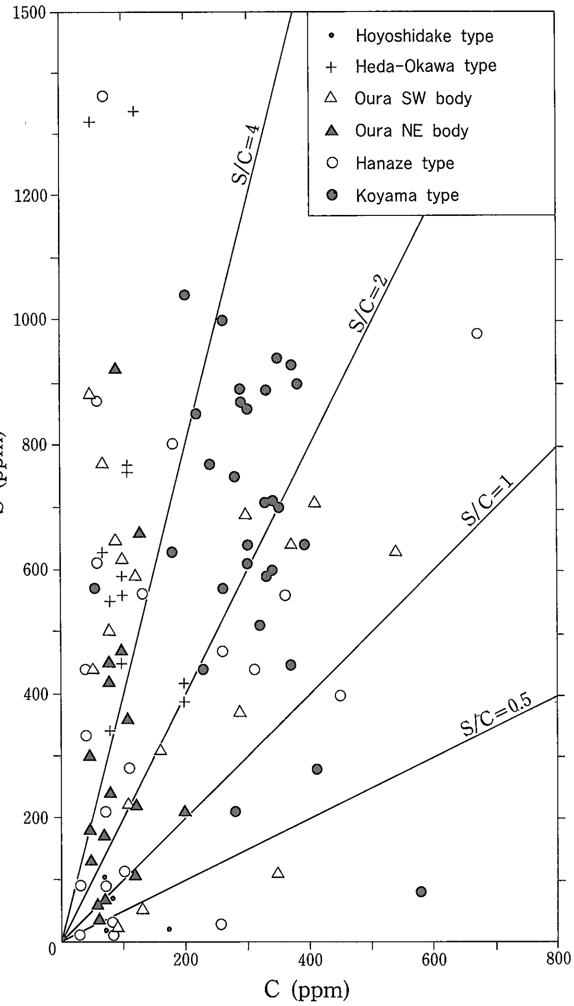

Fig. 3 Total sulfur vs. total carbon plots of the Osumi granitoids.

a low S/C ratio (average 2.2, n=29). Carbon content is high in the Oura SW body (average 195 ppm, n=17) and low in the NE body (average 89 ppm, n=17; No. 58 was excluded from the calculation), but S/C ratios are similar (Fig. 3): average 2.5 (n=17) for the SW body and average 3.1 (n=17) for the NE body.

The carbon contents are moderate in the Hanaze type (average 165 ppm, n=21; average S/C=2.4, n=24) and are low in the Heda-Okawa type (110 ppm, n=12). For the high sulfur and low carbon contents, the Heda-Okawa type is plotted in unique high S/C field (Fig. 3), the average S/C ratio is 6.2 (n=12). On the contrary, the Hoyoshidake type gives the lowest S/C ratio of <0.5 (n=4).

4. Shibi-san Pluton and its Sulfur and Carbon Contents

This is a small granitic body (18 km^2) intruding along an anticlinal axis of Cretaceous units of the Shimanto Supergroup in the northern Kagoshima Prefecture (see index map of Fig. 1). It is a granodiorite body divisible into two types. Kusubae type occurring in the southern half is hornblende-biotite granodiorite-monzogranite (SiO_2 66.4-67.8%), while Hirabae type occupying the northern half of the granitic body (Fig. 4), is hornblende-bearing biotite granodiorite (SiO_2 64.0-66.5%) (Yamamoto *et al.*, 1988). Aplite dike and vein cutting these two types are not uncommon, and a few granodiorite porphyry dikes which predate the granitic intrusion are seen around the western margin of the granitic body.

Kusubae type is a homogenous medium-grained rock containing both biotite and hornblende, but the Hirabae type is a heterogeneous rock containing biotite and little hornblende as major mafic silicate phase. Both types are partly porphyritic due to megaphenocrysts of K-feldspar, especially along the transitional zone of the two types. Accessory minerals are clinopyroxene, garnet, cummingtonite, actinolite, apatite, zircon and pyrrhotite. Garnet is trace in amount but the content increases toward the boundary between the two types.

Two kinds of enclaves have been reported. One has a porphyritic igneous texture or microgranular texture called autolith, and the other is sedimentary rocks from the host Shimanto Supergroup, which is called xenolith (Yamamoto *et al.*, 1988). The former is more common than the latter.

The analytical results of 13 samples are listed in Table 2. Sulfur content is generally high in the northern half, and total carbon is rich in the southern half, so that S/C ratio tends to be high in the north.

Total sulfur contents of the Kusubae type in the south is 20-50 ppm, whereas the total carbon contents range from 130 to 510 ppm, averaging 37.5 ppm and 285 ppm, respectively (S/C ratio=0.13).

The Hirabae type in the north, on the contrary, gives low sulfur values (30-70 ppm) similar to those of

Table 2 Sulfur and carbon contents of the Shibi-san grnitoids.

Filing no.	Sample no.	Rock type	S (ppm)	C (ppm)	S/C
1	SB-55-22B	Kusubae	50	300	0.2
2	SB-56-20	do.	20	510	0.04
3	SB-56-21B	do.	60	430	0.1
4	SB-56-22E	do.	20	180	0.1
5	SB-57-20A	do.	30	300	0.1
6	SB-57-21F	do.	20	130	0.2
7	SB-57-22E	do.	50	260	0.2
8	SB-58-20F	do.	50	170	0.3
9	SB-58-21G	Hirabae	70	250	0.3
10	SB-59-20C	do.	1,540	330	4.7
11	SB-59-21B	do.	50	130	0.4
12	SB-00-21B	do.	30	140	0.2
13	SB-01-21C	do.	3,550	260	13.7

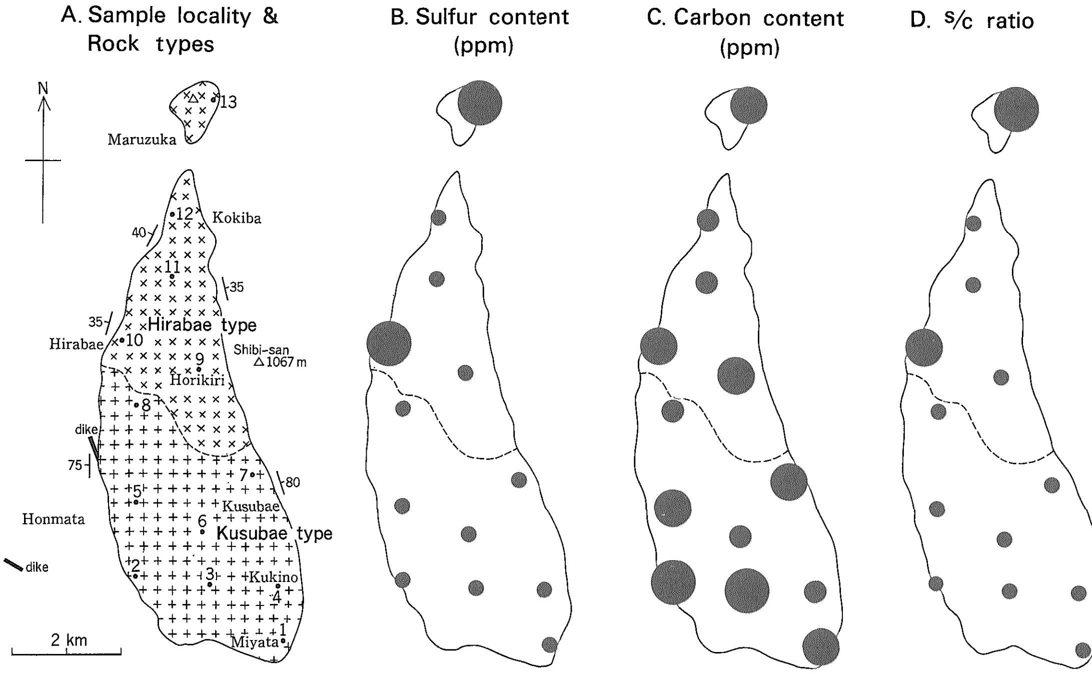

Fig. 4 Sample locality, major rock types, total sulfur and carbon content, and S/C ratio in the Shibi-san pluton. Size expressions by solid circles are the same es those of Fig. 3. Boundaries of the granitoid types are taken from Yamamoto et al. (1988).

the Kusubae type, and two high values (1,540 and 3,550 ppm S). The 3,550 ppm rock is taken from just below the roof sedimentary wall in the northern apophysis. Average values excluding the 3,550 ppm rock are 423 ppm S and 213 ppm C (S/C ratio=2.0).

5. Sulfur Isotopic Ratio

Rock sulfur isotope ratio was determined by the method reported in Sasaki *et al.* (1979). The results are listed in Table 3. Among the Osumi granitoids studied, the samples with varying amounts of sulfur were selected. The δ^{34}S values fall in a narrow range as -8.3 to -7.0 permil, and are consistent with the previously reported value of the Heda-Okawa type of -7.5 permil, which contains 705 ppm S (Ishihara *et al.*, 1988). Thus, no systematic difference can be seen in the δ^{34}S values of different S contents.

In the Shibi-san body, the δ^{34}S values are -5.7 and -6.2 permil, which are similar to the previously reported value of -5.0 permil (1,100 ppm S, Sasaki and Ishihara, 1979). Thus, the Shibi-san granitoids have their own values (average -5.6 ‰), which are about 2 permil higher than those of the Osumi granitoids (average -7.7 ‰).

Table 3 Rock sulfur isotopic ratio (δ^{34}S) of the Osumi and Shibi-san granitoids.

Sample no.	Rock type	δ^{34}S(‰)	S (ppm)
Osumi granitoids			
OS-09-53	Hanaze	-7.7	1,360
OS-08-45B	Heda-Okawa	-7.0	1,320
OS-05-45	Oura	-7.5	770
OS-17-04	Koyama	-8.3	570
OS-15-02	Oura	-7.9	420
OS-06-51A	do.	-7.9	210
OS-14-58	Hoyoshi-dake	-7.4	70
Shibi-san granitoids			
SB-01-21C	Hirabae	-5.7	3,550
SB-59-20C	do.	-6.2	1,540

6. Discussion

Although sedimentary enclaves are generally common in the Osumi granitoids, the presence of hornblende-bearing clots and mafic enclaves with an ophitic texture (c.f., Fig. 8, Ishihara, 1982) may indicate involvement of mafic igneous rocks. For the average $+11.5$ permil δ^{18}O values of the Osumi granitoids, Ishihara and Matsuhisa (1999) suggested a mixed source of 45 % mafic magmas from the depth and 55 % felsic magma generated within the Shimanto Supergroup for the Osumi pluton.

Nakamura *et al.* (1986) studied garnet of the Osumi and Shibi-san granitoids, and found a compositional zoning of low Mg core and high Mg rim, which is an evidence of progressive metamorphism. The analyzed garnets fall also in the metamorphic field in TiO_2-CaO diagram. Thus, they concluded that the garnets are metamorphic in origin and are xenocryst from the protolith.

Garnet is most abundant in the Koyama type up to 0.5 vol. %. Nishimura and Yamamoto (1994) found in and around the Koyama type that garnet occurs at the coastal level, while orthopyroxene-cummingtonite in the middle topographic level, and cummingtonite at the highest level. They refer that the minerals are restites and the mineral zoning represents metamorphic grades of the lower continental crust during an ultra-metamorphim (White and Chappell, 1977), whereby the granitic magmas were generated.

The present study indicates that the Koyama type is consistently high in both the total sulfur and carbon contents. This fact would be explained by predominance of sedimentary components involved in this type, because sulfur and carbon contents are much higher in sedimentary, especially of pelitic, rocks than mafic igneous materials. The Shimanto Supergroup contains roughly equal amounts of sandstone and shale, and the following numbers are calculated as the averages on the rocks of western Shikoku: -9.4 permil $\delta^{34}S$, 0.24 % S and 0.44 % C (Sasaki and Ishihara, 1979; Ishihara *et al.*, 1985). Metamorphic equivalents of the Shimanto Supergroup are most possible candidates for the source of sulfur.

Quaternary basalts and andesites of the Japanese Island Arc contain total sulfur lower than 40 ppm with average $\delta^{34}S$ of $+4.4 \pm 2.1$ permil (Ueda and Sakai, 1984). Simple mixing calculation between $+4.4$ permil $\delta^{34}S$ (40 ppm) magma and -9.4 permil $\delta^{34}S$ (2,400 ppm) metamorphic rocks indicates 10 % sediments contribution to produce -7.7 permil $\delta^{34}S$ of the Osumi granitoids, and only 5 % of sediments contribution can produce -5.5 permil $\delta^{34}S$ of the Shibi-san granitoids.

Mafic volcanic rocks of the Mariana Island Arc have the mean $\delta^{34}S$ value of $+3.8$ permil and mean S content of 100 ppm (Alt *et al.*, 1993). In this case, 20 % of sediments contribution is necessary to produce -7.7 permil $\delta^{34}S$ of the Osumi granitoids, and 10 % of sediments contribution for -5.5 permil $\delta^{34}S$ of the Shibi-san granitoids. Thus, sulfur is considered mostly derived from the accretionary sediments of the Shimanto Supergroup.

Homogeneous trend in the $\delta^{34}S$ values and no positive correlation between S contents and $\delta^{34}S$ data observed in the Osumi granitoids indicate that sulfur was homogenized in the magma chamber at depth. The large variation in the abundance is considered limited mobility of sulfur species in reduced granitic magmas.

7. Conclusions

Sulfur contents of the Osumi granitoids are found to be consistently high: average of the major types varies from 677 ppm (Heda-Okawa and Koyama types) to 278 ppm (Oura NE body), and the whole average is 428 ppm (N=99), except local leucocratic phase of the Hoyoshidake type (average <50 ppm, n=4). Both total sulfur and carbon are the highest in the Koyama type, which contains restitic garnet (average 677 ppm S and 306 ppm C, n=29). The magma of the Osumi granitoids has likely been incorporated with high-S and C sedimentary rocks at depth, and homogenized in terms of $\delta^{34}S$ values at depth, and has kept most of the sulfur within the granitoid for its strongly reducing atmosphere.

On the other hand, the Shibi-san granitoids are generally low in sulfur as 30–70 ppm, except sporadic high values near the wall rocks, because of less sedimentary components involved in the original magma. Sulfur in these reduced ilmenite-series granitoids is considered to reflect the original contents; the two granitoids are different in the source-rock ratio of igneous and sedimentary rocks.

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九州の大隅・紫尾山岩体における中新世チタン鉄鉱系花崗岩類の炭素・硫黄含有量と硫黄同位体比

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要　旨

大隅岩体(426 km^2)，紫尾山岩体(18 km^2)を構成する中新世チタン鉄鉱系花崗岩類117個について，全硫黄，全炭素含有量を赤外吸収分光法で分析し，硫黄含有量が異なる岩石9個についてδ^{34}S比を質量分析器で測定した．硫黄・炭素の結果は岩相図に表示し，地域的変化を明らかにした．

硫黄含有量は大隅花崗岩類では<10-3,550 ppm，平均493 ppm（n=104)であり，岩相タイプ別ではレスタイト柘榴石を持つ高山型や堆積岩源エンクレイブが多い辺田-大川型で高い．炭素含有量は30-670 ppm，平均191 ppm（n=103)であり，高山型と大浦型南西岩体および花瀬型の一部で高い．従って，S/C比は辺田-大川型で最も大きい．

紫尾山岩体(13試料)では，硫黄は一般に低く20-70 ppmであるが，北部の平八重型では局部的に高いものがある．炭素含有量は岩体南西部の楠八重型で高く，従ってS/C比は一般に北部で高い．

硫黄含有量が異なる試料の硫黄同位体比(δ^{34}S)は大きな変化を示さず，大隅岩体では-8.3～-7.0‰

の範囲にある．紫尾山岩体でも一定した負の値(-6.2～-5.7‰)を持つ．従って硫黄同位体比はマグマ生成時に均質化され，岩体ごとにに固有の値を持つに至った．堆積岩源のエンクレイブの多いこと，負の硫黄同位体比などは，花崗岩類の硫黄が四万十層群の変成岩相硫黄の影響を強く受けていることを示す．四国西部の堆積岩類の加重平均値(-9.4‰ $\delta^{34}S$, 2,400 ppm S)とマリアナ弧の硫黄データに基づく単純な混合モデルによれば，大隅花崗岩類では10-20%，紫尾山花崗岩類では5-10%の堆積岩硫黄の混入で実測値を説明できる．