

## Paired Sulfur Isotopic Belts: Late Cretaceous-Paleogene Ore Deposits of Southwest Japan

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**Abstract:** Sulfides from late Cretaceous-Paleogene ore deposits of the Inner Zone of Southwest Japan were analyzed for  $^{34}\text{S}/^{32}\text{S}$  ratios ( $\delta^{34}\text{S}_{\text{CDT}}$  values) on 5 samples from the Ryoke Belt, 18 from the Sanyo-Naegi Belt, 15 from the Uetsu-Kanto District and 47 samples from the Sanin-Shirakawa Belt. Together with the published data, the average  $\delta^{34}\text{S}$  values are calculated for individual deposits of the whole region; 109 localities for the ilmenite-series granitic belt and 56 localities for the magnetite-series granitic belt.

The sulfur isotopic ratios vary regionally rather than with sulfide species or types and commodities of the ore deposits. The  $\delta^{34}\text{S}$  values are generally negative in the fore-arc, ilmenite-series belt, and positive in the back-arc, magnetite-series belt. This regional variation is considered to reflect the ilmenite-/magnetite-series pairing of the genetically related granitic or volcanic activities, in which the positive  $\delta^{34}\text{S}$  ore sulfur was derived from a deep igneous source, but the negative  $\delta^{34}\text{S}$  sulfur was originated in biogenic sulfur of the accreted sedimentary complex. Within each terrane,  $\delta^{34}\text{S}$  values and mineral commodities vary locally, which is called domains in this paper. A few negative  $\delta^{34}\text{S}$  minima are observed in the Kinki, Hiroshima and Mino domains. These anomalies may have been brought by the most sediments-dominant granitic magmas generated within the accretionary sedimentary complex.

The paired  $\delta^{34}\text{S}$  belts, called the Japanese-type pattern, may be unique to island arcs where their fore-arc sediments have been accreted in the arc-front associated with the back-arc rifting.

**Keywords:** Southwest Japan, Inner Zone, Late Cretaceous-Paleogene, ilmenite series, magnetite series, granitoids, paired belt, ore deposits, sulfur isotopes.

### 1. Introduction

Late Cretaceous-Paleogene granitic rocks occur widely in the Inner Zone of Southwest Japan, which can be called the Inner Zone Batholith (Ishihara, 1990). Magmatic-hydrothermal deposits of vein and skarn types related to the granitic activities are also widespread in the region (Fig. 1). These ore deposits show a paired asymmetrical zoning in sulfur isotopes, low  $^{34}\text{S}/^{32}\text{S}$  ratios in the fore-arc zone, but high  $^{34}\text{S}/^{32}\text{S}$  ratios in the back-arc zone (Ishihara and Sasaki, 1991), similarly to that of the Late Cenozoic ore deposits (Ishihara *et al.*, 2000b).

In this paper, we report new analytical data of sulfur isotopes for ore deposits on 61 localities in the Inner Zone of Southwest Japan, and compile all the available data published to date. The regional variation is examined in some detail, and the whole region is subdivided into several domains, based upon the mineral commodities and sulfur isotopic ratios.

The analyzed samples are single minerals hand-picked or composite samples which may represent an

average value of any given ore deposit. The results of the two sampling methods agree well on individual ore deposits (e.g., Sasaki and Ishihara, 1980). Analytical methods are the conventional  $\text{SO}_2$  method described elsewhere (Sasaki *et al.*, 1979; Sasaki and Ishihara, 1979). Analytical data are shown in  $\delta^{34}\text{S}_{\text{CDT}}$  notation ( $^{34}\text{S}/^{32}\text{S}$  ratios relative to that of the CDT standard). Analytical error is estimated as  $\pm 0.2$  permil ( $2\sigma$ ). The analytical results are listed in Table 1.

In the following chapters, the analytical results are classified into the Uetsu-Kanto District, Ryoke Belt, Sanyo-Naegi Belt, and Sanin-Shirakawa Belt (Fig. 1). The Ryoke Belt is equivalent to the Ryoke metamorphic belt, which is solely composed of ilmenite-series granitoids and associated with no ore deposits. A few sulfide occurrences in granitoid quarries have been discovered and analyzed in this study. Granitoids of the Uetsu-Kanto District and the Sanyo-Naegi Belts are dominantly of ilmenite series, but the Sanin-Shirakawa Belt is composed mostly of magnetite-series granitoids (Ishihara, 1977).

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## 2 Uetsu- Kanto District

Granitic rocks of the Uetsu-Kanto District are consistent in being generally of ilmenite series. The district can be separated into two domains, the Uetsu and the Kanto (Fig. 1), according to their mode of W/Mo mineralizations and the  $\delta^{34}\text{S}$  trends. The ore deposits of the northern, Uetsu domain, have approximately equal amounts of W and Mo production and high Mn/Fe ratios of wolframite (Ishihara *et al.*, 1983), which are unique to this region. On the contrary, the ore deposits from the southern, Kanto domain are tungsten dominant and cassiterite bearing, which are characteristics of tungsten deposits in the Sanyo-Naegi Belt of the southwestern Inner Zone, but sulfur isotopic ratios of the ore sulfur are somewhat different from those of the Sanyo-Naegi Belt, indicating an independent Kanto domain.

Muscovite is ubiquitous alteration mineral, but no Li-mica and topaz are present in W and Mo deposits of the Uetsu domain. Trace amounts of sulfides contained in these ore deposits have  $\delta^{34}\text{S}$  values between -1.7 permil (no.2, Table 1) and +2.0 permil (no. 3, Table 2). Molybdenites from Tagokura, Fukushima Prefec-

ture (Ishihara, 1972), and Yamamoto-Fudou, Ibaraki Prefecture, have similar  $\delta^{34}\text{S}$  values of -1.5 permil and -0.6 permil, respectively (nos. 4, 5, Table 1).

Among local magnetite-bearing granitoids of the Uetsu domain, Wasada granodiorite hosts a unique ore deposit of disseminated type along cooling joints and fractures at Obari mine, Yamagata Prefecture. Ore minerals such as chalcopyrite, bithmuthinite, arsenopyrite, pyrite, tetrahedrite, sphalerite and molybdenite are disseminated in the strongly muscovitized granodiorite. The ore grade is said to be locally very high as Cu 10 %, Bi 7 %, Au 200 g/t, Ag 270 g/t (Shimazu and Kawachi, 1961). The chalcopyrite was determined to have  $\delta^{34}\text{S}$  value of +0.9 permil.

In conclusion, all the ore deposits in the Uetsu domain have  $\delta^{34}\text{S}$  values around 0 permil. Magnetite-series ore, though found at only one locality, does not show any particularly high values.

All the late Cretaceous granitoids of the Kanto domain belong to ilmenite series. Related to the most fractionated phase of the granitoids, tin-bearing wolframite quartz vein deposits occur with "greisenization", which is shown by Li-bearing muscovite, topaz and quartz at the Takatori mine, or mus-

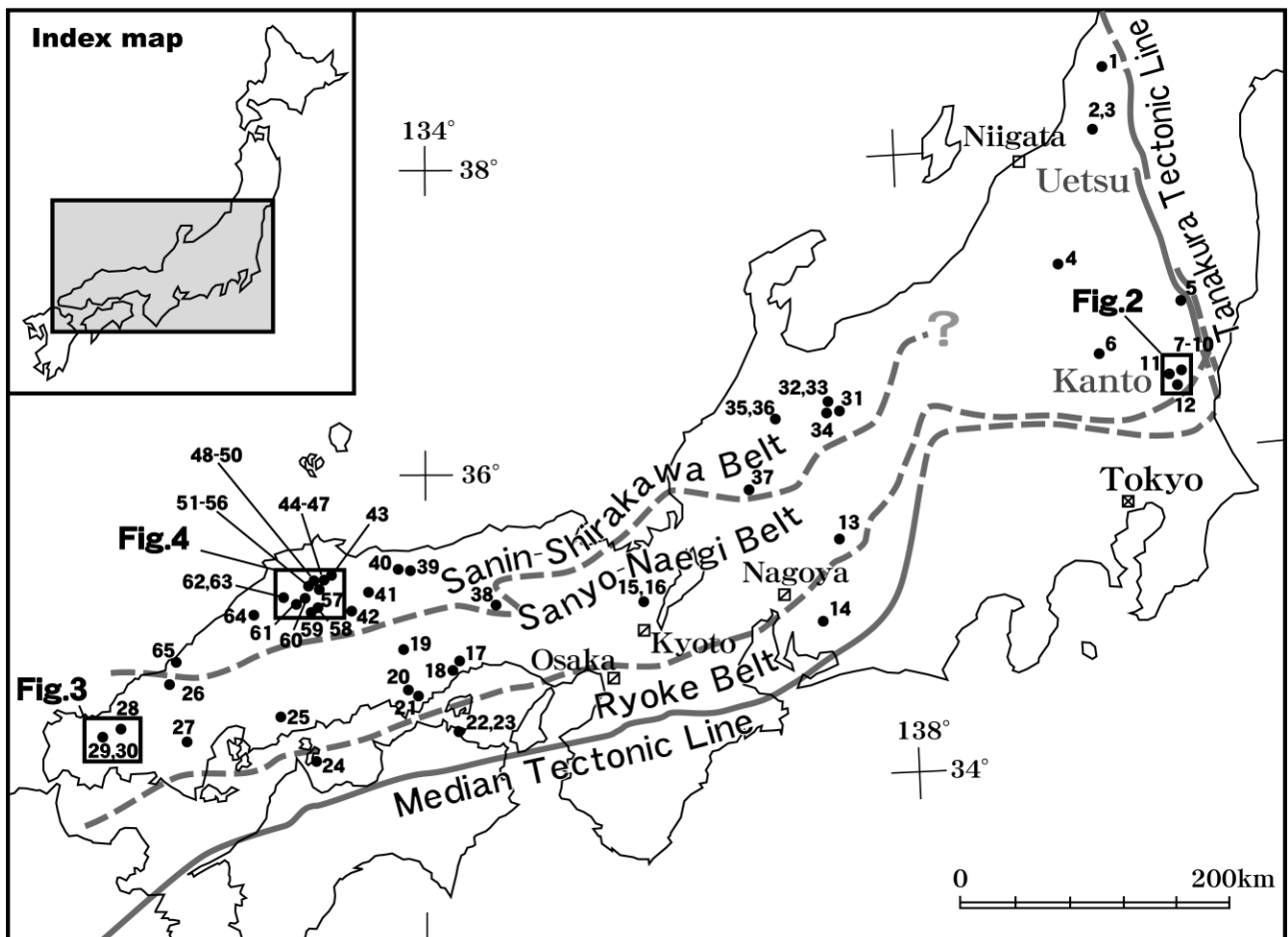


Fig. 1 Geotectonic units and localities of the studied samples. Numerals correspond to the sample nos. in Table 1.

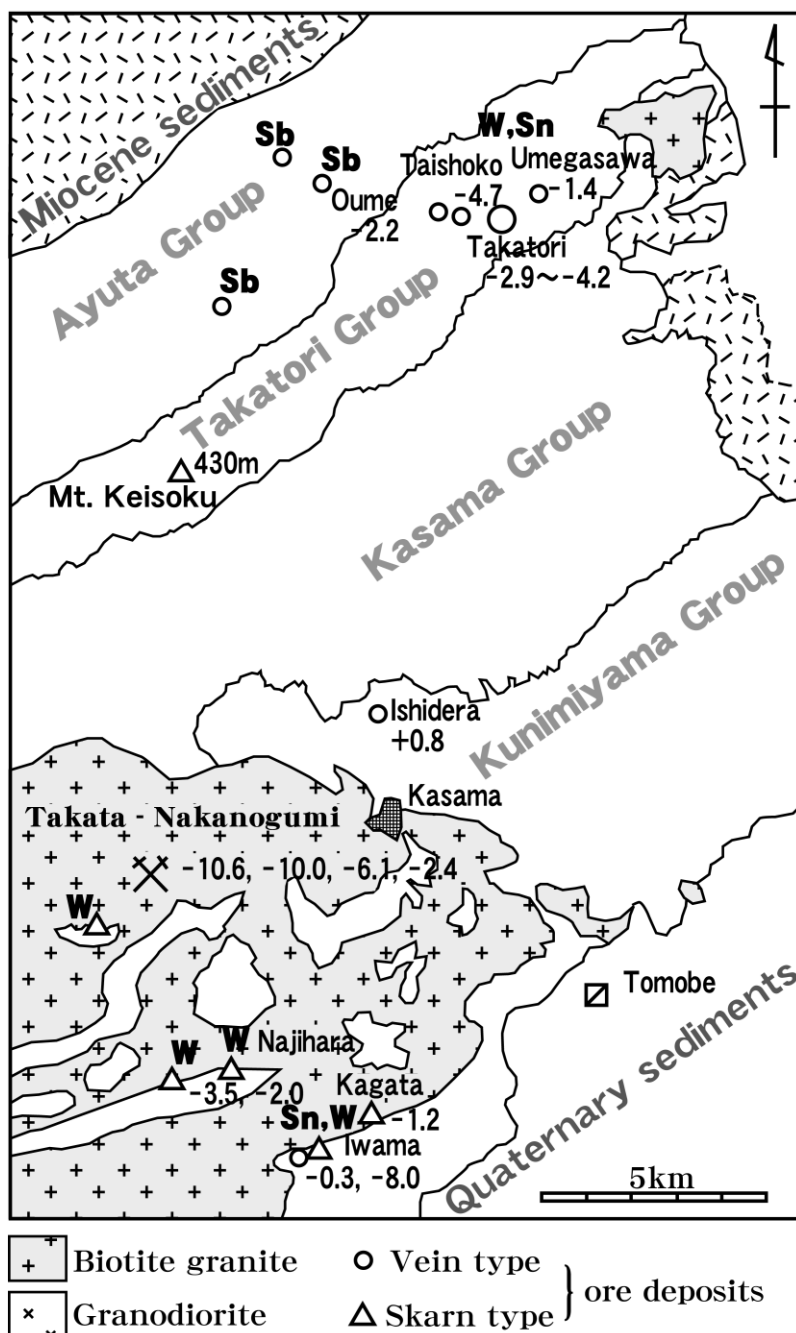


Fig. 2 Geological outline and  $\delta^{34}\text{S}$  ‰ values of the Takatori-Kagata mining area, Ibaraki Prefecture, Kanto District. Ayuta through Kunimiyama Groups are Jurassic-Cretaceous accretionary complex which were intruded by late Cretaceous granitoids. Chemical symbols imply main ore commodities of individual mines and prospects. Numerals indicate  $\delta^{34}\text{S}$  ‰ values. Data source, this study and Ishihara and Kono (2000).

covite, andalusite, corundum and quartz at Itaga mine (Ishihara and Sasaki, 1995).

The most representative example of the Takatori deposits consists of N55°W-75°S vein system occurring in Jurassic-Cretaceous accretionary complex of the Takatori Group of the Yamizo Supergroup (MITI, 1987). The ore veins are composed of quartz, muscovite, topaz, fluorite, calcite, chlorite and less abundantly Li-mica, monazite and rhodochrosite. The ore minerals are wolframite, cassiterite, chalcocopyrite, arsenopyrite, pyrite, and small amounts of stannite, galena, sphalerite and bismuthinite (JMIA, 1968). Similar ore deposits occur in the surrounding area and small

stibnite veins are known to the west of the Sn-W ore deposits (Fig. 2).

Pyrite concentrates separated by Wilfrey tables give  $\delta^{34}\text{S}$  values of -4.2 permil, while arsenopyrite concentrates are determined as -2.9 permil (no. 8, Table 1). Pyrite from the Umegasawa prospect, 1 km ENE of the Takatori mine, gives -1.4 permil (no.7, Table 1), while pyrite of the Taishoko prospect, ca. 1 km west of the Takatori mine, is -4.7 permil (no.9, Table 1).

In Kasama area to the south (Fig. 2), sulfides occurring in granite give  $\delta^{34}\text{S}$  values of -6.1 and -2.4 permil, but pyrrhotites of some sedimentary enclaves are as low as -10.6 permil (Ishihara and Kono, 2000). Sul-

Table 1  $\delta^{34}\text{S}$  values of Cretaceous-Paleogene ore deposits studied from the Inner Zone of Southwest Japan

Locality no.	Sample no. and locality	Main commodity	Ore deposit	Analyzed sample	$\delta^{34}\text{S}$ (‰)	Analysts
<i>Uetsu and Kanto Districts</i>						
1	58S01 Obari, Wasada, Yamagata	Cu, Au	Cp>>apy disseminated type in altered granodiorite	Chalcopyrite	+0.9	Mitsubishi CL
2	59102410Daikoku, Niigata	W, Sn	Wf-cassiterite-quartz vein in biotite granite	ditto	-1.7	A. Sasaki
3	59103110Kanamaru, Niigata	W, Mo	Wf-mly-py dissemination in Kf pegmatite	Pyrite	-0.5	ditto
4	70K167 Tagokura, Fukushima	Mo	Molybdenite in cracks of altered granite	Molybdenite	-1.5	ditto
5	73A03 Yamamoto-Fudou, Ibaraki	Mo	Molybdenite disseminated in biotite granite	Pyrite	-0.6	Mitsubishi CL
6-1	Itaga-NK, Kanuma, Tochigi	W, As	Wf-quartz vein and wf-dissemination in granite and hornfels	Lollingite	-5.1	A. Sasaki
6-2	Itaga-ASditto	ditto	ditto	ditto	-4.5	ditto
7	KB070 Umesawa, Ibaraki	W	Wf-quartz vein in Yamizo Supergroup	Pyrite	-1.4	Mitsubishi CL
8-1	TA108 Takatori, Ibaraki	W, Sn, Cu	Sn-wf-quartz vein with greisen, Apy concentrates	Apy>py	-2.9	ditto
8-2	TA109 ditto	ditto	ditto, chalcopyrite-bearing pyrite concentrates	Pyrite	-4.2	ditto
9	KB015 Taisho-ko, Ibaraki	W	Sn-wf-quartz vein deposit in Yamizo Supergroup	ditto	-4.7	ditto
10	KA012 Oume, Ibaraki	Sb	Stibnite-quartz vein in Yamizo Supergroup	Stibnite	-2.2	ditto
11	000617 Inada, Nakanogumi, Ibaraki	Mo	Molybdenite in biotite granite	Molybdenite	-6.1	A. Sasaki
12-1	KD004 Najihara, Ibaraki	W	Scheelite skarn at contact of Yamizo Supergroup and granite	Pyrite, po	-3.5	ditto
12-2	KD005 ditto	W	ditto	ditto	-2.0	ditto
<i>Sanyo-Naegi and Ryoke Belts</i>						
13-1	69-221 Ebisu, Naegi, Gifu	W, Sn	Mly-quartz vein in sericitized ignimbrites	Molybdenite	-0.3	ditto
13-2	69-222 ditto	ditto	Lollingite-wf quartz vein	Lollingite	-1.5	Mitsubishi CL
14-1	90031804 Okazaki, Aichi	Mo	Kf-quartz pegmatite dike in two-mica granite quarry (Nakane)	Molybdenite	-5.7	A. Sasaki
14-2	RY83 ditto	ditto	ditto	ditto	-6.0	ditto
15	92KURA-Y Yamafuji Qry, Kyoto	Rock	Pyrrhotite-bearing granodiorite ("Kurama stone")	Pyrrhotite	-3.5	ditto
16	92KURA-M Masuda Qry, Kyoto	Rock	Pyrrhotite-bearing quartz diorite ("Kurama stone")	ditto	-4.6	ditto
17	99032903 Mitsuishi-Ohira, Okayama	Pyrophyllite	Fine pyrite nodule in black siltstone in rhyolitic tuffs	Pyrite	-3.9	ditto
18	99032901 Yotsutsujiyama, Okayama	Rock?	Pyrrhotite-bearing biotite granite	Pyrrhotite	-0.3	ditto
19-1	Kamo, Okayama	Mo	Molybdenite-quartz vein in granite	Molybdenite	-1.8	ditto
19-2	ditto	ditto	ditto	Composite (cp, py, sp)	-3.5	Mitsubishi CL
20	Miyoshi, Okayama	W	Wolframite-quartz vein with greisen; pyrite disseminated in altered granite	Pyrite	-5.0	ditto
21-1	GSI-4483 Obie, Okayama	Cu	Chalcopyrite quartz vein veins in Jurassic sediments just above granite	Chalcopyrite	-6.1	A. Sasaki
21-2	ditto (Kanahori)	ditto	pyrite-chloritized rock	Pyrite	-4.8	Mitsubishi CL
22	71TO329B Aji, Kagawa	Cu	Cp>py-quartz veinlet in biotite granite quarry (Terashima)	Chalcopyrite>pyrite	-7.2	A. Sasaki
23	71TO330B Mure, Kagawa	FeS	Molybdenite film in biotite granite quarry (Urushibara)	Molybdenite	-3.7	ditto
24	91040401 IyoOshima, Ehime	Mo	Sulfides-bearing pegmatitic quartz vein (E-W), Oda quarry	Pyrrhotite>cp	-3.8	ditto
25	Shiratoriyaama, Hiroshima	Mo	Molybdenite disseminated in chloritized granite	Molybdenite	-5.8	ditto
26	Hidarigayama, Shimane	As, Cu	Cu-As-Mo-quartz vein in the Masago granite	ditto	-2.6	Mitsubishi CL
27-1	Kawayama, Yamaguchi	Cu, FeS, Zn	Sulfides-filling sheared zone & skarn (50 mL), zinc ore (Zn 8%, S32%)	Marmatite>po	-5.8	ditto
27-2	ditto	ditto	ditto (110 mL), copper-pyrrhotite ore (Cu 1.5%, S 35%)	Pyrrhotite>cp	-5.8	ditto
27-3	ditto	ditto	ditto (140mL) Rievrite-garnet-pyrrhotite ore	Pyrrhotite	-6.0	ditto
28-1	ICN-1, Ichinosaka, Yamaguchi	Ag	Ag-galena ores at Jin-nai-mabu	Galena	-5.0	A. Sasaki
28-2	ICN-2, ditto	ditto	ditto	ditto	-5.5	ditto
29	Naganobori, Yamaguchi	Cu, Co	Copper skarn orebodies around fine biotite granite	Chalcopyrite	-4.2	ditto
29-1	2003, Imori orebody	ditto	ditto, chalcopyrite-carbonate breccia	Galena	-0.6	ditto
29-2	42123, North orebody	ditto	ditto, galena-garnet skarn	Chalcopyrite	-4.2	ditto
30	42001b Ota, Yamaguchi	Cu	ditto, chalcopyrite skarn	Chalcopyrite	-4.2	ditto
<i>Sannin-Shirakawa Belt</i>						
31	19236 Kasagatake, Gifu	Pb, Zn	Pb-Zn veins in late Cretaceous rhyolites, galena in druse	Galena	+2.5	ditto
32	Yoshiki, Gifu	Pb, Zn, Ag	Pb-Zn veins in quartz porphyry dike	ditto	+2.6	Mitsubishi CL
33-1	Kamioka-Tochibora, Gifu	Zn, Pb, Ag	Pb-Zn skarns in Hida meta-rocks. Zn concentrates-1986	Sphalerite	+2.1	ditto
33-2	ditto	ditto	ditto, lead flotation concentrates-1986	Galena	+1.2	ditto
33-3	86111802 ditto	ditto	Pyrite-quartz vein in aplitic granite	Pyrite	+3.7	ditto
33-4	86111810 ditto	ditto	Sphalerite disseminated aplitic granite	Sphalerite	+1.9	ditto

Table 1 Continued

Locality no.	Sample no. and locality	Main commodity	Ore deposit	Analyzed sample	$\delta^{34}\text{S}$ (‰)	Analysts
34-1	Kamioka-Mozumi, Gifu	Zn, Pb, Cu	Pb-Zn skarns in Hida meta-rocks. Zn concentrates-1986	ditto	+2.4	A. Sasaki
34-2	ditto	ditto	ditto, Pb concentrates-1986	Galena	+1.0	ditto
34-3	ditto	ditto	ditto, Cu concentrates-1986	Chalcopyrite	+2.6	ditto
35	090504 Ashikura, Shirakawa, Gifu	Mo	Molybdenite-quartz vein (3 cm wide) in Hatogaya granitoids	Molybdenite	+5.6	R.M. Bai
36	090606 Tsubakihara tunnel, do.	ditto	Molybdenite on crack of Tsubakihara granitoids	ditto	+6.6	ditto
37	20163 Omodani, Fukui	Cu, Pb, Zn	Cp-sp-gn veins in quartz porphyry	ep>>bornite	-0.2	A. Sasaki
38 7	1AR62 Ariga, Hyogo	Cu	Pyrite disseminated in altered quartz diorite	Pyrite	+1.3	Mitsubishi CL
39-1	590798 Endani, Tottori	Mo, U	Mly-bearing coffinite-quartz vein in the Ogamo granite	Molybdenite	+4.0	ditto
39-2	610 ditto	ditto	Pyritized altered granite of the wall rock	Pyrite	+4.9	ditto
40-1	590896 Takashiro, Tottori	Mo	Molybdenite-quartz vein	Molybdenite	+4.5	ditto
40-2	590893 ditto	Mo	Pyritized altered biotite granite (OgamoGr)	Pyrite	+3.0	ditto
41	Ushirodani, Tottori	Zn, Pb	Base metal veins and skarn in slate and limestone/chert	Massive sphalerite	+3.2	ditto
42-1	86S117 Okurayama, Tottori	Zn, Pb	Base metal veins in granodiorite	Sphalerite	+3.8	ditto
42-2	86S119 ditto	ditto	Pyritized altered granodiorite of the wall rock	Pyrite	+4.2	ditto
43	59072101 Iyadani, Shimane	Mo	Fine molybdenite-quartz vein	Molybdenite	+6.1	A. Sasaki
44	59072103 Yamasa, Shimane	Mo	Fine molybdenite-quartz vein in leucogranite (Yamasa)	Mly conc.1959	+4.6	Mitsubishi CL
45	59072418 Takagi, East Adit, Shimane	Mo	Mly-qz vein in leucogranite (Yamasa)	Molybdenite	+6.1	A. Sasaki
46	59072410 Iwakuradani, Shimane	Mo	Very fine mly-qz vein in leucogranite (Yamasa)	ditto	+1.1	ditto
47	59072534 Kami-Yamasa, Shimane	Mo	Mly-qz vein in leucogranite (Yamasa)	ditto	+7.7	ditto
48-1	86S104a Sasagatani prospect, Shimane	Mo	Py-mly disseminated in altered ignimbrites	ditto	+6.5	Mitsubishi CL
48-2	86S104b ditto	Mo	Pyrite occurring in druse of the ignimbrites	Pyrite	+4.9	ditto
49-1	HY795 Higashiyama, Hidari 1, Shimane	Mo	Py-mly-qz vein in leucogranite, late-hi	Molybdenite	+4.5	ditto
49-2	HY776 ditto, Migi 2	ditto	Pyrite pocket in quartz vein	Pyrite	+3.9	ditto
50	66SK13 Seikyu, Chukan-hi 2L, Shimane	Mo	Disseminated mly in albitized granite	Molybdenite	+5.3	A. Sasaki
51	650612 Kanenari, Daito, Shimane	Rock	Schistose hornfels (Kanenari)	Molybdenite	+6.2	Mitsubishi CL
52	60F12 Togiishiyama, Daito, Shimane	Rock	Biotite hybrid (Togiishiyama)	ditto	+4.2	ditto
53	86S101 Mouth of Kanasakani, Daito, Shimane	Rock	Biotite granodiorite (Rengeji)	ditto	+4.8	ditto
54	86S103 Rengeji new road, Daito, Shimane	Rock	Biotite granite (Rengeji)	ditto	+6.0	ditto
55-1	590899 Daito mine, Aug. 1959 concentrates	Mo	Floatation concentrates from the mine	Molybdenite	+4.7	A. Sasaki
55-2	69DT513 ditto Honko vein, Shimane	ditto	Massive pyrite in ore vein	Pyrite	+3.4	Mitsubishi CL
55-3	5905-296 Daito mine, Hinotani veins	ditto	Mly-chlorite vein in aplitic granite	Molybdenite	+5.1	A. Sasaki
56	69OR25 Orisakani 1st vein, Daito mine	Mo	Mly disseminated in pink aplite pool in qz diorite	ditto	+6.9	ditto
57	5907-227 Seikyu-Minami, Shimane	Mo	Molybdenite-quartz vein in Shimokuno aplite	Pyrite	+8.1	ditto
58	59073099a Omaki, Yubune, Shimane	Mo	Molybdenite disseminated in altered granite	Molybdenite	+6.4	ditto
59-1	56IM08 Ichiman OB, Komaki, Shimane	Mo, W	Wf-mly quartz pipe with "greisen"	ditto	+3.9	ditto
59-2	86KM11 ditto	ditto	Wf-mly quartz pipe, py replacing wolframite	Pyrite	+2.7	Mitsubishi CL
59-3	D32-006 Honko OB, Komaki mine	ditto	Mly in garnet quartz rock	Molybdenite	+4.3	A. Sasaki
59-4	5KM164 ditto, #9 Rise	ditto	Mly disseminated in "greisen"	ditto	+3.9	ditto
60	86S110 Kamokura, Shimane	Mo	Molybdenite-pyrite disseminated in host leucogranite	Pyrite	+3.7	Mitsubishi CL
61	86S109 Sogi, Yoshida, Shimane	FeS	Pyrite disseminated in altered granodiorite (Miocene?)	ditto	+2.1	ditto
62	5907341 Osa, Shimane	Mo	Molybdenite-quartz vein in leucogranite	Molybdenite	+3.5	ditto
63	86S124 Kakeya, Shimane	Mo	Pyrite disseminated in altered ignimbrite	Pyrite	+3.4	ditto
64	Sanbesan, Shimane	Mo	Molybdenite-quartz vein in leucogranite	Molybdenite	+4.7	ditto
65	Okami, Shimane	Rock	Pyrrhotite disseminated in granodiorite	Pyrrhotite	-3.2	A. Sasaki

Locality no. corresponds to those of Table 1. Abbreviations: apy, arsenopyrite; bn, bornite; cp, chalcopyrite; conc., concentrates; gn, galena; Gr Qry, granite quarry for building stone; K.f, K-feldspar; meta-rocks, metamorphic rocks; mly, molybdenite; OB, orebody; po, pyrrhotite; py, pyrite; sp, sphalerite; wf, wolframite.

Table 2 Average  $\delta^{34}\text{S}$  values of Cretaceous-Paleogene ore deposits from the ilmenite-series granitic terranes.

Nos.	Locality	Main commodity	$\delta^{34}\text{S}$ (‰)	References
<b>A Uetsu domain</b>				
1	Obari, Yamagata	Cu 4	+0.9	This study
2	Shionomachi, Niigata	Mo	+1.8	Ishihara et al. (1983)
3	Nabekura, Niigata	W	+2.0	ditto
4	Daikoku, Niigata	W	-1.7	This study
5	Kanamaru, Niigata	Pegmatite	-0.5	This study
6	Tagokura, Fukushima	Mo	-1.5	This study
7	Magome, Fukushima	Pb, Zn	+0.5	Shimazaki (1985)
8	Mizuhiki, Fukushima	Zn, Pb	+1.0	ditto
9	Yamamoto-Fudou, Ibaraki	Mo	-0.6	This study
<i>Average of the Uetsu Domain: +0.2 (n=9)</i>				
<b>B Kanto Domain</b>				
10	Itaga, Tochigi	W 4	-4.8 (n=2)	This study
11	Umegasawa, Ibaraki	W	-1.4	This study
12	Takatori, Ibaraki	W 2	-3.6 (n=2)	This study
13	Taisho-ko, Ibaraki	W	-4.7	This study
14	Oume, Ibaraki	Sb	-2.2	This study
15	Ishidera, Ibaraki	As	+0.8	Ishihara & Kono (2000)
16	Inada, Ibaraki	Mo	-6.1	This study
17	Ditto	Enclaves	-10.3 (n=2)	Ishihara & Kono (2000)
18	Najihara, Ibaraki	W	-2.8 (n=2)	This study
19	Kagata, Ibaraki	W	-1.6 (n=2)	Shimazaki & Yamamoto (1979)
20	Iwama, Ibaraki	Cu, As	-4.2 (n=2)	Ishihara & Kono (2000)
<i>Average of the Kanto Domain: -3.1 (n=10)</i>				
<b>C. Mino Domain</b>				
21	Hiragane, Gifu	Cu	-6.4	Shimazaki (1985)
22	Takane, Gifu	Cu	-7.6	ditto
23	Okura, Gifu	Pb-Zn	-10.3	ditto
24	Mansei, Gifu	Zn, Pb	-1.5	ditto
25	Mase, Gifu	Cu, Zn	-2.7	ditto
26	Narai, Gifu	Cu, Zn	-5.4	ditto
27	Hatasa, Gifu	Cu	-2.7	ditto
28	Hiraiwa, Gifu	F	-5.6	ditto
29	Horado, Gifu	Pb-Zn	-6.8	ditto
30	Kurokawa, Gifu	Cu	-2.6	Shimazaki (1985)
31	Ebisu, Gifu	W 3	-0.9	This study
32	Kasugayama, Nagano	W	-5.3	ditto
33	Okazaki, Aichi	Mo	-5.9 (n=2)	This study
34	Inuyama, Aichi	Cu, Zn	-8.4	Shimazaki (1985)
<b>D. Kinki Domain</b>				
35	Nosaka, Fukui	Cu, Zn	-3.2	ditto
36	Oike, Shiga	Cu	-5.9 (n=3)	Miyoshi et al. (1988)
37	Ayukawa, Shiga	Pb-Zn	-3.2	Shimazaki (1985)
38	Ishibe-Kanayama, Shiga	Zn	-4.4	ditto
39-1	Kaneuchi, Kyoto	W	-5.9	Sasaki & Ishihara (1980)
39-2	ditto	W	-7.3 (n=6)	Miyoshi et al. (1988)
<i>Average of the Kaneuchi</i>				
			-7.1 (n=7)	
40-1	Otani, Kyoto	W	-8.4 (n=9)	Sasaki & Ishihara (1980)
40-2	ditto	W	-9.9 (n=14)	Miyoshi et al. (1988)
<i>Average of the Otani</i>				
			-9.3 (n=23)	
41	Tada, Hyogo	Cu	-2.4 (n=10)	ditto
42	Hatano, Osaka	Zn, Pb	-4.1	Shimazaki (1985)
<i>Average of the Mino-Kinki Domain: -5.1 (n=22)</i>				
<b>E. Ikuno-Akenobe Domain</b>				
43	Kabasaka, Hyogo	Cu, Zn	+0.3	This study
44	Dainichi, Hyogo	Zn, Pb	-2.4	Miyoshi et al. (1988)
45-1	Ikuno, Hyogo	Cu, Zn, Sn, Pb	+1.4	Sasaki & Ishihara (1980)
45-2	kuno-Kinsei vein	ditto	+0.7 (n=7)	Miyoshi et al. (1988)
45-3	Ikuno-Tenju vein	ditto	+0.3 (n=2)	ditto
<i>Average of the Ikuno polymetallic veins</i>				
			+0.7 (n=10)	
46	Nii, Hyogo	Cu	-6.2	ditto
47-1	Akenobe, Hyogo	Sn, W, Cu, Zn, Pb	-2.1	Sasaki & Ishihara (1980)
47-2	Akenobe-Ryusei vein	ditto	-2.2 (n=44)	Yamamoto (1974)
47-3	Akenobe-Fudono vein	ditto	-2.0 (n=2)	Ishihara et al. (1981)
47-4	Akenobe-Chiemon vein	ditto	-2.6 (n=6)	ditto
47-5	ditto	ditto	-1.9 (n=11)	Miyoshi et al. (1988)
47-6	Akenobe-Eiko vein	ditto	-1.6 (n=3)	ditto
47-7	Akenobe-Eisei vein	ditto	-1.6 (n=2)	ditto
47-8	Akenobe-Ginsei vein	ditto	-1.6 (n=2)	ditto
47-9	Akenobe-Shichisei vein	ditto	-1.3 (n=12)	ditto
47-10	Akenobe-Shirogane vein	ditto	-1.8 (n=17)	ditto
<i>Average of the Akenobe polymetallic veins</i>				
			-1.9 (n=102)	

Table 2 Continued

Nos.	Locality	Main commodity	$\delta^{34}\text{S}$ (‰)	References
48	Omidani, Hyogo	Ag, Au	-3.2 (n=7)	ditto
49	Asahide, Hyogo	Zn, Pb	+0.8 (n=2)	ditto
50	Hirafuku, Hyogo	Zn, Pb	-0.8	Shimazaki (1985)
<b>F. Okayama Domain</b>				
51	Aji, Kagawa	Cu	-7.2	This study
52	Mure, Kagawa	Mo	-3.7	This study
53	Katsutoyo, Okayama	Zn, Pb	-3.6	Shimazaki (1985)
54	Seto, Okayama	Cu	-5.2	ditto
55	Isobe-Asahi, Hyogo	Au, Ag	-1.5	ditto
56	Tatsuyama, Okayama	Cu	-3.0	ditto
57	Yamate, Okayama	Cu	-2.7	ditto
58	Sano, Okayama	Cu	-2.8	ditto
59	Ida, Okayama	Zn, Pb	-3.3	ditto
60	Nagusa, Okayama	Zn, Pb	-4.8	ditto
61	Eyomi, Okayama	Cu	-4.4	ditto
62	Tsuda, Okayama	Cu	-6.3	ditto
63	Kamo, Okayama	Mo	-2.7 (n=2)	This study
64	Miyoshi, Okayama	W	-5.0	This study
65	Obie, Okayama	Cu	-5.5 (n=2)	This study
66	Takinomaru, Okayama	Fe	+0.3	Shimazaki (1985)
67-1	Yoshioka-Honzan, Okayama	Cu vein	-2.7	ditto
67-2	Yoshioka-Sasano, ditto	Cu skarn	-1.5	Shimazaki & Yamamoto (1979)
68	Motoyama, Okayama	Cu	-0.8	Shimazaki (1985)
69	Sanpo, Okayama	Fe, Cu	-4.5	ditto
70	Kanehira, Okayama	Fe	-3.6	ditto
71	Mihara, Okayama	Cu	-1.0 (n=2)	Yamamoto (1976), Shimazaki (1985)
<i>Average of the Okayama Domain -3.4 (n=22)</i>				
<b>G. Hiroshima Domain</b>				
72	Fukiyadani, Hiroshima	Cu	-1.3	Shimazaki (1985)
73	Hirako, Hiroshima	Cu	-3.1	ditto
74	Jinmu-Mihara, Hiroshima	F, Cu	-4.6	ditto
75	IyoOshima, Ehime	Cu	-3.8	This study
76	Shiratoriyama, Hiroshima	Mo	-5.8	This study
77	Kinmei, Hiroshima	Cu	-5.3	Shimazaki (1985)
78	Kabe, Hiroshima	Cu, Zn	-2.2	ditto
79	Kawayama, Yamaguchi	Cu, Zn	-6.0 (n=5)	This study, Shimazaki (1985)
80	Kuga, Yamaguchi	W	-10.0	Sasaki & Ishihara (1980)
81	Kiwada, Yamaguchi	W	-7.7	Shimazaki & Yamamoto (1979)
82	Fujigatani, Yamaguchi	W	-9.9 (n=2)	Sasaki & Ishihara (1980), Shimazaki & Yamamoto (1979)
<i>Average of the Hiroshima Domain: -5.4 (n=11)</i>				
<b>H. Yamaguchi Domain</b>				
83	Shimane, Shimane	Cu	-0.4	Shimazaki & Yamamoto (1979)
84	Tsumo, Shimane	Cu, Zn	-0.1 (n=2)	Sasaki & Ishihara (1980)
85	Hidarigayama, Shimane	Mo	-2.6	This study
86	Sasagatani, Shimane	Cu	-1.8	Shimazaki (1985)
87	Kitsunozuka, Yamaguchi	Cu	+2.9	Shimazaki & Sakai (1984)
88	Kawayama, Yamaguchi	Cu	+0.0	ditto
89	Sakurago, Yamaguchi	Cu	-0.8	ditto
90	Kujiradake, Yamaguchi	Zn, Pb	-0.6	ditto
91	Omoto, Yamaguchi	Cu	+0.3	ditto
92	Ichinosaka, Yamaguchi	Ag,Pb,Zn	-5.3 (n=2)	This study
93	Kitabira, Yamaguchi	Fe, Cu	-3.9	Shimazaki & Sakai (1984)
94	Naganobori, Yamaguchi	Cu	-3.0 (n=3)	This study, Shimazaki & Sakai (1984)
95	Ota, Yamaguchi	Cu	-4.2	This study
96	Kanagatao, Yamaguchi	Cu	+0.9	Ishihara et al. (1984)
97	Yakuouji, Yamaguchi	Cu	+0.9	ditto
98	Sasanami, Yamaguchi	Cu	+0.6	ditto
99	Hidaka, Yamaguchi	Cu, W	-3.1	ditto
100	Omi, Yamaguchi	Zn	+0.7	ditto
101	Sanjou, Yamaguchi	Cu, W	-1.2	Shimazaki & Sakai (1984)
102	Ofuku, Yamaguchi	Cu	-5.1	ditto
103	Higashi-Kibe, Yamaguchi	Fe	-3.2	ditto
104	Ono, Yamaguchi	Cu	-2.2	ditto
105	Ida, Yamaguchi	Cu	-2.6	ditto
106	Moji, Fukuoka	Cu	+1.4	ditto
107	Ishida-Yokoshiro, Fukuoka	W	+0.4	Sasaki & Ishihara (1980)
108	Yoshihara, Fukuoka	Cu	-0.3	Shimazaki & Sakai (1984)
109	Sannotake, Fukuoka	Cu	-1.5	ditto
<i>Average of the Yamaguchi Domain: -1.3 (n=27)</i>				

fides occurring in veins and lenses in the sedimentary rocks have a wide range of variation between -8.0 and +0.8 permil (Ishihara and Kono, 2000).

### 3. Ryoke Belt

The Ryoke Belt of the Inner Zone of Southwest Japan is known to consist of both foliated and massive granitoids of ilmenite series having no sulfide-ore deposits, probably because of a deep level of the granitoids exposed and their strongly reduced nature. However, minor sulfide occurrences can be expected in limited areas.

Molybdenite is often found in stone quarries if the granite is fractionated one. Such molybdenites were discovered in garnet-bearing two-mica granite at Nakane Quarry, Taki-machi, Okazaki City, Aichi Prefecture (Okazaki stone, Sato and Nakai, 1991), and in a similar rock type of biotite granite in Aji stone mining area, Kagawa Prefecture (Aji stone, Ishihara, 1991a). These molybdenites and other sulfides give  $\delta^{34}\text{S}$  values between -7.2 and -3.7 permil (average -5.1‰, n=5; nos. 14, 22-24, Table 1). These values are somewhat lighter than the host-granitoid  $\delta^{34}\text{S}$  values between -5.0 and -2.1 permil (average -3.5‰, n=3, Sasaki and Ishihara, 1979).

### 4 Sanyo-Naegi Belt

The granitoids of this belt are composed mostly of ilmenite series, but trace amounts of magnetite tend to appear northward and magnetite-series rocks occur sporadically near the northern margin. Their rock  $\delta^{34}\text{S}$  values are typically -4.1~-4.6 permil (Sasaki and Ishihara, 1979).

A small stock or plug, less than 1 km in diameter occurring to the north of Kyoto city contains locally abundant pyrrhotite, which rusts quickly giving iron-oxides color on the weathered surface. This nature has been loved to use the stone for Japanese gardens by tea-ceremony people in Kyoto since the 11th century, giving a special name of Kurama stone after their locality. Sulfur of the pyrrhotite is considered to have been derived from the wall pelitic rocks of the Tanba Supergroup of Jurassic age, and reduced the granitic magma (Ishihara, 1991b). The pyrrhotites give  $\delta^{34}\text{S}$  values of -3.5 and -4.6 permil (nos. 15, 16, Table 1). These values are similar to the rock sulfur value of -4.6 permil of the nearby Otani mine stock (Sasaki and Ishihara, 1979). Pyrrhotite from Yotsutsujiyama, southern Okayama Prefecture (-0.3‰  $\delta^{34}\text{S}$ ), may represent rock sulfur of the host ilmenite-series granite (no. 18, Table 1).

The Sanyo-Naegi belt is a major Sn-bearing tungsten metallogenic province in Japan. Hundreds of middle to small sized sulfide-forming ore deposits are

also known in this belt. Among base metal deposits, copper is dominant relative to lead and zinc (Shimazaki, 1975). Iron sulfides are generally pyrrhotite and the coexisting sphalerites have high iron contents (Tsukimura *et al.*, 1987). The ore deposits are of reduced type and show generally negative  $\delta^{34}\text{S}$  values around -5 permil (Sasaki and Ishihara, 1980; Shimazaki, 1985; Miyoshi *et al.*, 1988), which is also confirmed in this study (Table 1).

On the basis of ore sulfur  $\delta^{34}\text{S}$  data, Shimazaki (1985) argued the genesis of "Kawayama-type copper-pyrrhotite deposits" in the Chugoku district to be epigenetic related to granitoids, instead of volcanogenic sedimentary origin as had been believed sometime. He obtained negative  $\delta^{34}\text{S}$  values for the sulfides from the Kawayama (-6.3‰, n=2), Tsuda (-6.3‰) and Eyomi (-4.4‰) mines, which were similar to those of vein and skarn types in the surrounding areas. Our results of the Kawayama specimens with negative  $\delta^{34}\text{S}$  values of -5.8 to -6.0 permil (nos. 27-1 to 27-3, Table 1) support his interpretation.

Framboidal aggregates of pyrite occurring in local mudstone within the host rhyolitic tuffs of pyrophyllite deposits at Mitsuishi mining area, which may have been formed along a caldera wall (Ishihara and Imaoka, 1999), give a  $\delta^{34}\text{S}$  value of -3.9 permil (no. 17, Table 1). The historically famed silver deposits mined during the early Edo period at Ichinosaka, Yamaguchi Prefecture, are thought to be related to felsic dikes intruded along a cauldron wall of the Sasanami cauldron (Ikawa and Imaoka, 2001). Galenas from sphalerite-galena-rhodochrosite veins yield -5.0 and -5.5 permil  $\delta^{34}\text{S}$  (no. 28, Table 1). Another small vein deposit occurring in the northern margin of the cauldron (Fig. 3) gives +0.3 permil (no. 91, Table 2).

To the south of the Sasanami cauldron, there occurs the Hobenzan stock (13 km<sup>2</sup>) intruding into the Sangun meta- sedimentary rocks and sedimentary rocks of the Oda Supergroup (Fig. 3). The stock belongs to an intermediate series composed of weakly magnetic granodiorite and magnetite-free biotite granite. The granitoids are rich in K<sub>2</sub>O, Cl, S, Cu and B (Ishihara *et al.*, 1984), thus associated with many polymetallic vein- and disseminated-type deposits containing Cu, As, Co, Bi and W in and around the stock. Major ones are granitoid-hosted Yakuoji deposits and sediments-hosted Kanagatao deposits.

Rock sulfur of the granitoids extracted from the samples with a minor sulfides dissemination (pyrrhotite>chalcopyrite>pyrite) ranges from +0.3 to +2.2 permil (average +1.3‰, Sasaki and Ishihara, 1979; Ishihara *et al.*, 1984). Ore sulfur of composite samples varies  $\delta^{34}\text{S}$  values from +0.6 to +0.9 permil in the copper deposits, but -3.1 permil in the Hidaka scheelite veins (Ishihara *et al.*, 1984).

To the west of the Hobenzan stock, fine-grained



granite intrudes into the Akiyoshi limestone plateau (Fig. 3). The granite is exposed in an ovoid shape with E-W 1,360 m by N-S 840 m. Five copper skarn orebodies have been found around the granite cusp including the Naganobori mine at the eastern margin and the Ota mine at the western margin (Ogura, 1927). The Naganobori mine is the oldest copper mine in this country operated since the 8<sup>th</sup> century, and the ore was used in casting the bronze statue of “Great Buddha” in the Nara dynasty (A.D. 757). Chalcopyrites from the Imori and Ota orebodies give  $\delta^{34}\text{S}$  value of -4.2 permil, while galena from the North orebody yields that of -0.6 permil (nos.29 & 30, Table 1).

### 5. Sanin-Shirakawa Belt

This belt is composed mostly of magnetite-series granitoids, which show positive rock  $\delta^{34}\text{S}$  values of +3.9~+8.3 permil (Sasaki and Ishihara, 1979).

In the Daito area of the central Sanin District, two small hybrid masses are known at the northwestern margin of the Rengeji granitoid (Fig. 4), whose fractionated phases are genetically related to molybdenum mineralizations of the major three mines, Daito, Seikyū and Higashiyama. Pyrites separated from the Kanenari schistose hornfels and the Togiishiyama biotite hybrid (Ishihara, 1971a) have  $\delta^{34}\text{S}$  values of +6.2 and +4.2 permil, respectively (nos. 51 & 52, Table 1). The Rengeji granitoid close to these hybrid rocks contain also pyrite, which gives +4.8 and +6.0 permil (nos. 53 & 54, Table 1). These pyrites are considered magmatic

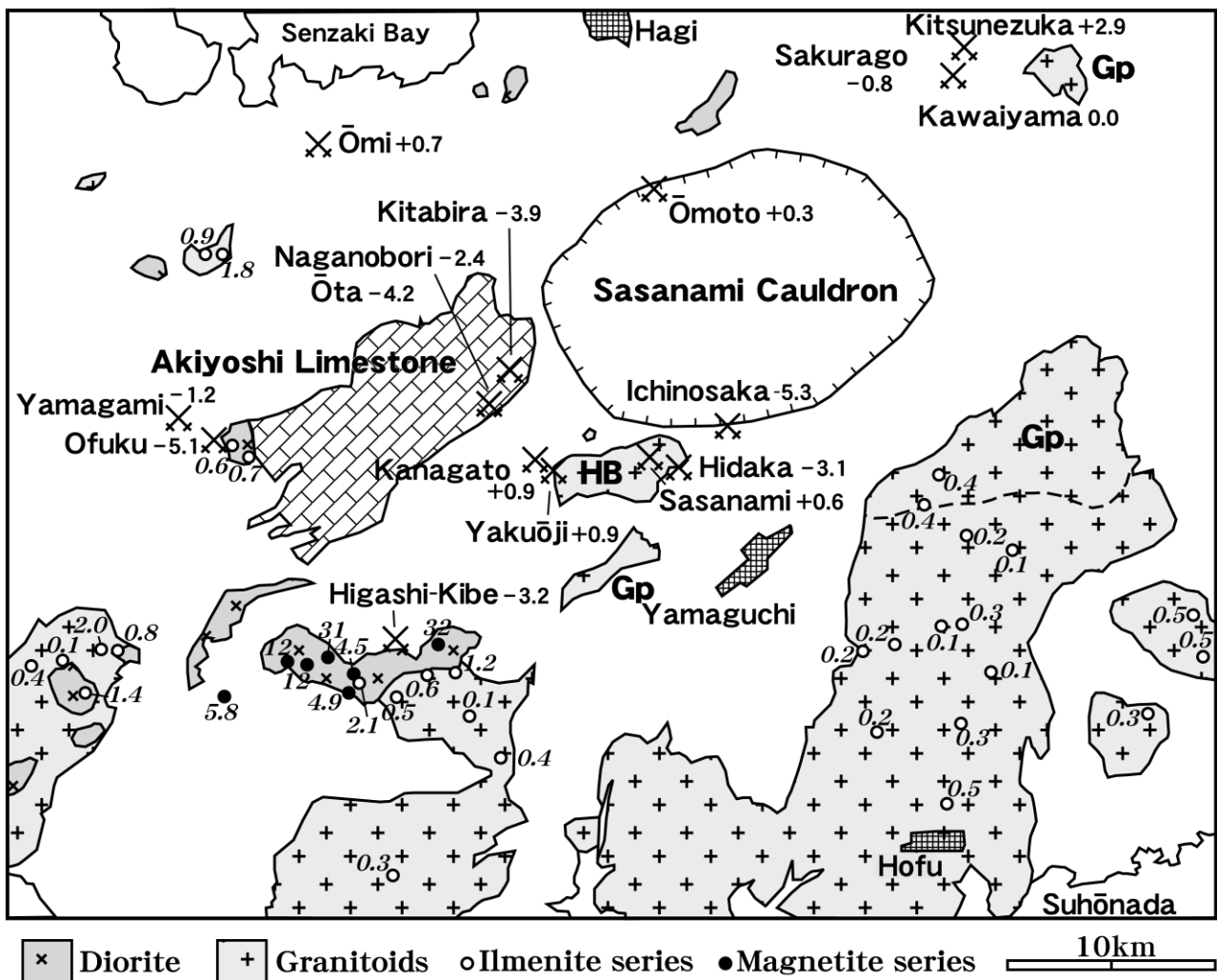


Fig. 3 Magnetic susceptibility of the late Cretaceous granitoids and average  $\delta^{34}\text{S}$  ‰ values of related ore deposits in the central part of Yamaguchi Prefecture, western Sanyo-Naegi Belt (see Fig. 1). Geology simplified from Nishimura *et al.* ed. (1995). Numerals by open and solid circles are magnetic susceptibility measured by Kappameter KT5 device in  $10^{-3}$  SI unit. Those by closed mine symbol are  $\delta^{34}\text{S}$  ‰ values of ore sulfur. Gp implies porphyritic granite. HB, Hobenzan granodiorite-granite stock. Outline of the Sasanami cauldron taken from Ikawa and Imaoka (2001).

in origin, because the mineral is scattered in granitic rocks without any sign of hydrothermal alteration. The measured  $\delta^{34}\text{S}$  values of +4.2 to +6.2 permil are similar to rock  $\delta^{34}\text{S}$  values of +3.9~+6.3 of magnetite-series granitoids of the Sanin District (Sasaki and Ishihara, 1979).

In the westernmost part of the Sanin-Shirakawa Belt (Fig. 1), there occurs a small zoned pluton of Paleogene age called the Okami body intruding into pelitic facies of the Sangun metamorphic rocks at the north-eastern part and into coeval volcanic rocks on the south-western side (Nureki, 1957). The main phase of granodiorite has magnetic susceptibility of  $25\sim 10 \times 10^{-3}$  SI, as measured by the portable device of KT5, and local quartz diorite has the values up to  $40 \times 10^{-3}$  SI.

Thus, the pluton belongs to magnetite series. The north-eastern margin of this pluton becomes granitic and granophyric, and the magnetic susceptibility varies from 14 to  $3 \times 10^{-3}$  SI. Pyrrhotite with iron oxide staining is seen in some parts of the fresh granite, and the stained part is less magnetic as  $7.3\sim 2.0 \times 10^{-3}$  SI. The  $\delta^{34}\text{S}$  value of the pyrrhotite is -3.2 permil (no. 66, Table 1). Proximity to the pelitic wall rocks and absence of sedimentary xenolith indicate sulfur would have come from the wall rocks as hydrogen sulfur and methane gasses and have reduced the granitic magma, converting its  $\delta^{34}\text{S}$  to a negative value.

Toward the eastern end of the Sanin-Shirakawa Belt, large lead-zinc skarn deposits occur in the Kamioka-Tochibora and Kamioka-Mozumi and the Nakatatsu

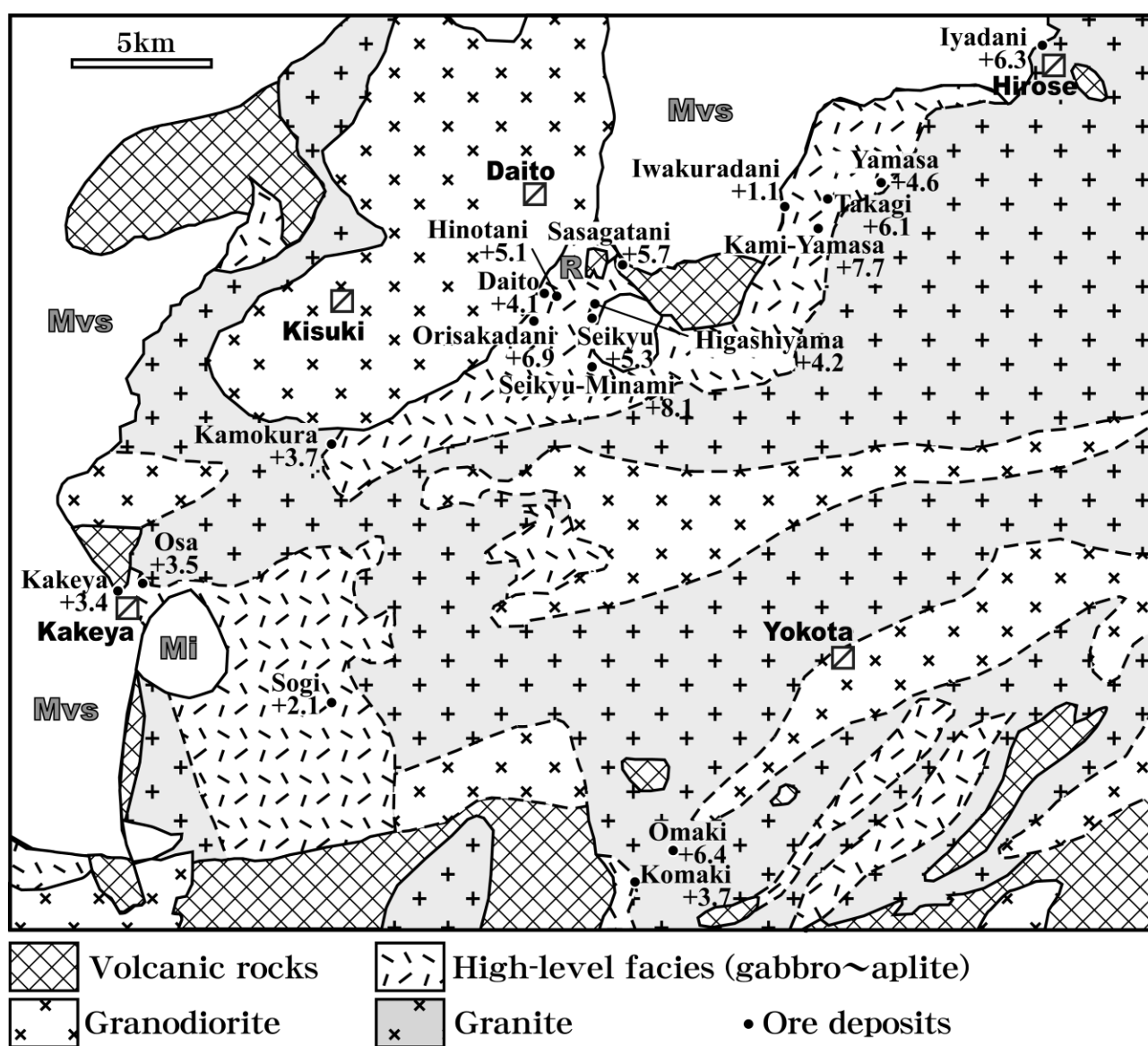


Fig. 4 Geological outline and  $\delta^{34}\text{S}$  ‰ values of the Daito-Komaki area, Shimane Prefecture, western Sanin-Shirakawa Belt (see Fig. 1). All the values are positive. R implies Rengeji Temple around which Kanenari hornfels, Togiishiyama hybrid and Rengeji granite occur. Mvs, Miocene volcanic rocks; Mi, Miocene intrusion. Modified from Ishihara (1971a).

mineralizations.  $\delta^{34}\text{S}$  values for products of the Kamioka dressing plants are +1.0 and +1.2 permil for the lead concentrates, +2.1 and +2.4 permil  $\delta^{34}\text{S}$  for the zinc concentrates and +2.6 permil for copper concentrates from the Mozumi mine (nos. 33 & 34, Table 1).

Molybdenites occur at many places within biotite leucogranites of the Shirakawa granitoids, Gifu Prefecture (+ 5.6 & +6.6  $\delta^{34}\text{S}$  ‰, nos. 35 & 36, Table 1). This mineral is also present with coffinite-quartz vein at Endani, Tottori Prefecture (+4.0 ‰  $\delta^{34}\text{S}$ , no.39, Table 1) and pyrite-quartz vein at Takashiro (+4.5  $\delta^{34}\text{S}$  ‰, no. 40, Table 1). Other productive sulfides in the Sanin district are sphalerites at the Ushirodani mine (+3.2 ‰  $\delta^{34}\text{S}$ , no. 41, Table 1) and Okurayama mine (+3.8 ‰  $\delta^{34}\text{S}$ , no. 42, Table 1)

Many molybdenite-quartz vein deposits are known to occur in the central Sanin District (Fig. 3). They tend to occur in fine-grained leucocratic granites which are marginal facies of coarse-grained biotite granite. Molybdenites from these ore deposits return  $\delta^{34}\text{S}$  values ranging from +1.1 permil at Iwakuradani to +8.1 permil at Seikyū-Minami-ko (nos. 46 & 57, Table 1). These end-member values are obtained from epithermal veins, but the main hypothermal to mesothermal molybdenite veins at Higashiyama, Seikyū and Daito mines have  $\delta^{34}\text{S}$  values of +4.5 to +5.3 permil (nos. 49-50, 55, Table 1).

The Komaki molybdenum mine has some taste of tungsten mineralizations of the Sanyo-Naegi Belt. The host rock is an ilmenite-series two-mica granite, though the surrounding granitoids are of magnetite series. Among several orebodies so far discovered, the Ichimanko orebody contains minable amount of wolframite and scheelite, while the other orebodies solely produced molybdenite. The  $\delta^{34}\text{S}$  values are +3.9 and +2.7 permil for the Ichimanko orebody, and +4.3 and +3.9 permil for the Honko orebody (no. 59, Table 1); these are of typical values for the Sanin-Shirakawa Belt.

## 6. Regional Variation of $\delta^{34}\text{S}$ values of Ore Sulfur and its Genetic Background

Average  $\delta^{34}\text{S}$  values of individual ore deposits in this study and the previous studies are summarized in Tables 2 and 3. These averages are also shown in the histograms of Figs. 5 and 6. Their regional distribution is shown in Fig. 7. The regional variations are combined with regional characteristics of the mineralizations, and several domains are proposed within each belts (Fig. 8).

The  $\delta^{34}\text{S}$  values vary most distinctly across the arc, being low in the ilmenite-series granitic belts to the south (Table 2), but high in the magnetite-series granitic belt to the north (Table 3). Within both the ilmenite-series and magnetite-series belts, moreover, the values are different along the arc (Fig. 5), and together

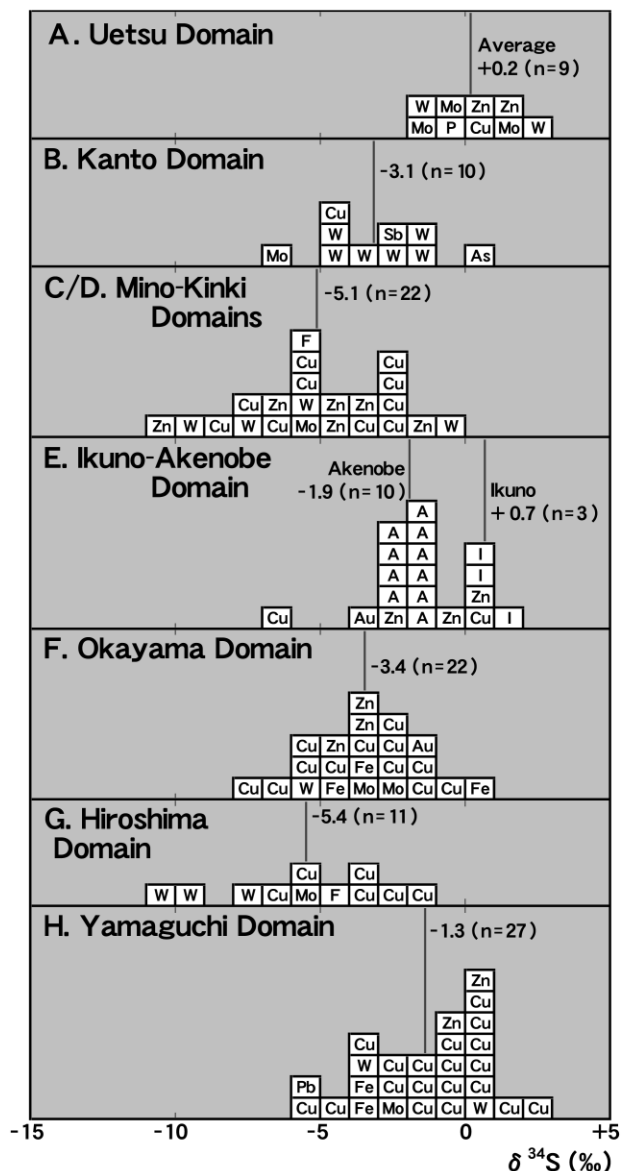


Fig. 5 Histograms of  $\delta^{34}\text{S}$  ‰ values of ore deposits in the ilmenite-series granitic terranes. P, pegmatite; Fe, magnetite(-hematite); Mo, molybdenite; W, wolframite and scheelite, Cu, chalcopyrite mainly; As, arsenopyrite, Zn, spalerite>galena; Pb, galena>sphalerite; Sb, stibnite; Ag, Ag-bearing sulfides, Au, Native gold and electrum, F, fluorite deposits. A and I, Akenobe and Ikuno polymetallic vein deposits, respectively. Average values of each domain are indicated by bar.

with mineral commodities, several domains have been proposed. In the Uetsu domain, for example, the ore deposits are characterized by tungsten and molybdenum and lead-zinc mineralizations, and the  $\delta^{34}\text{S}$  values are around +0.2 permil, but those of the Kanto domain are dominantly tungsten and the  $\delta^{34}\text{S}$  values are averaged as -3.1 permil (Fig. 5).

Table 3 Average  $\delta^{34}\text{S}$  values of Cretaceous-Paleogene ore deposits from the magnetite-series granitic terranes.

Nos.	Locality	Main commodity	$\delta^{34}\text{S}$ (‰)	References
<b>I. Hida Domain</b>				
110	Kokurobe, Toyama	Mo	+4.7	Ishihara et al. (1990)
111	Kaminorouka, Toyama	Mo	+5.7	ditto
112	N. Suishodake, Toyama	Mo	+3.1	ditto
113	Warimozawa, Nagano	Mo	+6.4	ditto
114	Yumata, Nagano	Mo	+6.0	ditto
115	Kamoshikazawa, Nagano	Mo	+3.7	ditto
116	Kurosawa, Nagano	Mo	+3.0	ditto
117	Sarutobi, Gifu	Zn, Pb	+2.1	Shimazaki (1985)
118	Kasagatake, Gifu	Zn, Pb	+2.5	This study
119	Hirayu, Gifu	Zn, Pb	+1.0	Shimazaki (1985)
120	Shimonomoto, Gifu	Ag, Au	+3.0	ditto
121	Yoshiki, Gifu	Pb, Zn	+2.6	This study
122-1	Kamioka-Tochibora, Gifu	Zn, Pb	+1.7 (n=2)	ditto
122-2	ditto	Mo	+2.3 (n=3)	This study, Sasaki & Ishihara (1980)
123-1	Kamioka-Mozumi, Gifu	Zn, Pb	+1.7	This study
123-2	ditto	Cu	+2.6	ditto
124	Kagasawa, Gifu	Fe	+9.5	Shimazaki (1985)
125	Jintsu, Gifu	Fe	+0.4	ditto
126	Amou, Gifu	Au	-0.6	ditto
127	Ashikurasawa	Mo	+5.6	This study
128	Tsubakihara tunnel	Mo	+6.6	ditto
129	Hirase, Gifu	Mo	+5.1	Shimazaki (1985)
130	Ono, Gifu	Fe	+1.7	ditto
131	Mitani, Gifu	Zn, Pb	-3.0	ditto
132	Kutani, Ishikawa	Zn, Pb	+3.6	ditto
133	Bandojima, Fukui	Pb, Zn	+4.1	ditto
134	Omodani, Fukui	Pb, Zn	-0.2	This study
135	Nakatatsu, Fukui	Pb, Zn	+3.5 (n=2)	Sasaki & Ishihara(1980), Shimazaki (1985)
136	Fumuro, Fukui	Pb, Zn	+2.4	Shimazaki (1985)
137	Makitani, Fukui	Zn, Pb	+0.6	ditto
<i>Average of the Hida Domain</i>			<i>+3.1 (n=30)</i>	
<b>J. Tango Domain</b>				
138	Komori, Kyoto	Cu	+1.1 (n=5)	Miyoshi et al. (1988)
139	Umetani, Kyoto	Zn, Pb	+3.5	Shimazaki (1985)
140	Yabu, Hyogo	Ag	+4.5	ditto
141	Asahide, Hyogo	Cu	+0.8 (n=2)	ditto
142	Ariga, Hyogo	Cu	+1.3	This study
<i>Average of the Tango Domain</i>			<i>+2.2 (n=5)</i>	
<b>K. Sanin Domain - Tottori</b>				
143	.Endani, Tottori	Mo, U	+4.5 (n=2)	This study
144	Sekigane, Tottori	W, Mo	+3.9	Shimazaki (1985)
145	Ushirodani, Tottori	Zn, Pb	+2.7(n=2)	This study, Shimazaki (1985)
146	Okurayama, Tottori	Zn, Pb	+4.0 (n=2)	This study
<i>Average of the Tottori Domain</i>			<i>+3.9 (n=4)</i>	
<b>L. Shimane Domain - Shimane</b>				
147	Iyadani, Shimane	Mo	+6.3	This study
148	Yamasa, Shimane	Mo	+4.6	This study
149	Takagi, Shimane	Mo	+6.1	This study
150	Iwakuradani, Shimane	Mo	+1.1	This study
151	Kami-Yamasa, Shimane	Mo	+7.7	This study
152	Sasagatani, Shimane	Mo	+5.7 (n=2)	This study
153	Higashiyama, Shimane	Mo	+4.2 (n=2)	This study
154	Seikyu, Shimane	Mo	+5.3	This study
155	Daito-Honko, Shimane	Mo	+4.1 (n=2)	This study
156	Daito-Hinotani	Mo	+5.1	This study
157	Seikyu-Minami, Shimane	Mo	+8.1	This study
158	Omaki, Shimane	Mo	+6.4	This study
159	Komaki, Shimane	Mo, W	+3.7 (n=4)	This study
160	Kamokura, Shimane	Mo	+3.7	This study
161	Sogi, Shimane	FeS	+2.1	This study
162	Osa, Shimane	Mo	+3.5	This study
163	Kekeya, Shimane	Mo	+3.4	This study
164	Sanbesan, Shimane	Mo	+4.7	This study
<i>Average of the Shimane Domain</i>			<i>+4.8 (n=18)</i>	
<b>M. Kyushu Domain</b>				
165	Fukuoka-Suien, Fukuoka	Mo	+2.2	This study

Abbreviations: apy, arsenopyrite; Gr Qry, granite quarry for building stone; cp, chalcopyrite; Kf, K- feldspar; mly, molybdenite; po, pyrrhotite; py, pyrite; wf, wolframite.

In the Mino-Kinki domain, the  $\delta^{34}\text{S}$  values are similarly negative having an average of -5.1 permil. The ore deposits are characterized by tungsten and copper, and some lead-zinc and fluorite. In the Okayama domain, the ore deposits contain dominantly copper and some magnetite and lead-zinc sulfides. Their  $\delta^{34}\text{S}$  values are averaged as -3.4 permil.

The Hiroshima domain of Hiroshima and eastern Yamaguchi Prefecture is characterized by tungsten and copper, and low  $\delta^{34}\text{S}$  values with an average of -5.4 permil. In the Yamaguchi domain of Yamaguchi Prefecture and northeastern Kyushu, there occur many small copper deposits. They have  $\delta^{34}\text{S}$  values of -1.3 permil on the average.

In the Ikuno-Akenobe area of the central part of Hyogo Prefecture, there occur a number of polymetallic veins known as "subvolcanic type", containing productive amounts of Sn, W, Cu, Zn and Pb, and associated with Au and Ag (Nakamura, 1998). They could belong to an independent unit of the Ikuno-Akenobe domain. The Akenobe vein system has an average  $\delta^{34}\text{S}$  value of -1.9 permil, and the Ikuno veins are averaged as +0.7 permil (Fig. 5). The  $\delta^{34}\text{S}$  values of the subvolcanic type seem to be slightly heavier than those of the plutonic types in the other domains of the Sanyo-Naegi Belt.

In the magnetite-series granitic belt (Fig. 6), the average  $\delta^{34}\text{S}$  value is +3.1 permil in the Hida domain, in which the ore deposits are characterized by molybdenum and lead-zinc mineralizations. On the contrary, the Sanin domain composed of soley molybdenum

deposits has higher  $\delta^{34}\text{S}$  values around +4.8 permil.

Fig. 8 shows regional variation of the averaged  $\delta^{34}\text{S}$  values of the ore deposits in the Inner Zone of Southwest Japan. Boundary between the positive and negative  $\delta^{34}\text{S}$  values is similar to that of the magnetite-series and ilmenite-series granitoids. Within the magnetite-series granitic belt, distribution of ore sulfur  $\delta^{34}\text{S}$  is rather monotonous; the most productive Daito Mo-mineralized area and the second largest Mo-mineralized Shirakawa area in the Hida Belt give similar values. On the contrary, several distinctly negative  $\delta^{34}\text{S}$  centers can be recognized in the ilmenite-series granitic belts. These anomalies are always observed in sedimentary terranes of usually accretionary complex.

The most remarkable minima are seen around two Sn-bearing W-mineralized areas, the Otani - Kaneuchi mine area in Kyoto Prefecture and eastern Yamaguchi Prefecture areas. In the Otani Sn-scheelite quartz vein deposits occurring in a granitic stock, the ore sulfur is averaged as -9.3 permil (n=23) and rock sulfur of the host granodiorite stock is -4.9 permil (Sasaki and Ishihara, 1979). The stock contains often sedimentary enclaves and shows very low bulk  $\text{Fe}_2\text{O}_3/\text{FeO}$  ratios between 0.07 and 0.09 (Ishihara, 1971b). This stock, though granodiorite in composition, was formed by a granitic magma generated in sediments-dominant protolith where organic carbon and biogenic sulfur may have been abundant. The Kaneuchi wolframite-quartz veins (-7.1‰  $\delta^{34}\text{S}$ , n=7) at northwest of the Otani mine are situated in biotite hornfels zone of the host sediments of the Tanba terrane, and may have been formed by ore fluids derived from hidden ilmenite-series granitic magmas.

No granitic rocks are exposed in the Kuga (-10.0‰  $\delta^{34}\text{S}$ ), Kiwada (-7.7‰  $\delta^{34}\text{S}$ ) and Fujigatani (-9.9‰  $\delta^{34}\text{S}$ ) Sn-bearing scheelite skarn deposits area of eastern Yamaguchi Prefecture, but a few small leucocratic granitic bodies have been discovered by drilling and a hidden cupola is assumed (MITI, 1981). A similar sediment-intrusive relationship encountered in the Otani-Kneuchi mine area can be expected here, and the negative  $\delta^{34}\text{S}$  values are considered due to the concealed ilmenite-series granitic bodies.

Among several negative ore sulfur  $\delta^{34}\text{S}$  values reported by Shimazaki (1985) in the Mino sedimentary terrane, the Okura Pb-Zn-Ag deposits (-10.3‰  $\delta^{34}\text{S}$ ) are of quartz porphyry related skarn type occurring in limestone and chert beds. The Takane Cu-Pb-Zn-Ag deposits (-7.6‰  $\delta^{34}\text{S}$ ) are stockworked veins and skarns in slate, chert and limestone intruded by granite porphyry, and the Hiragane Cu-pyrrhotite skarn deposits (-6.4‰  $\delta^{34}\text{S}$ ) occur in slate and chert intruded by felsic dikes. The Horado Pb-Zn-Mo skarn deposits (-6.8‰  $\delta^{34}\text{S}$ ) occur in limestone, slate and sandstone intruded by quartz porphyry (Hida *et al.*, 1956).

Compared to the plutonic type Sn-bearing tungsten

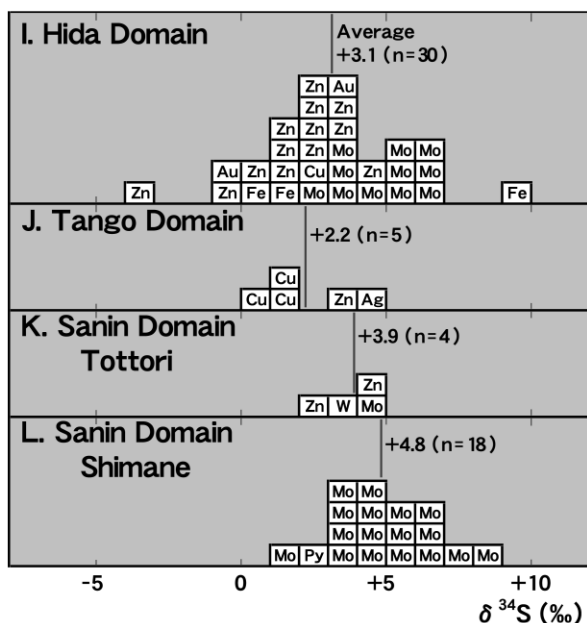


Fig. 6 Histograms of  $\delta^{34}\text{S}$  ‰ values of ore deposits in the magnetite-series granitic terrane. The abbreviations the same as of Fig. 5. Py, pyrite. Average values of each domain are indicated by bar.

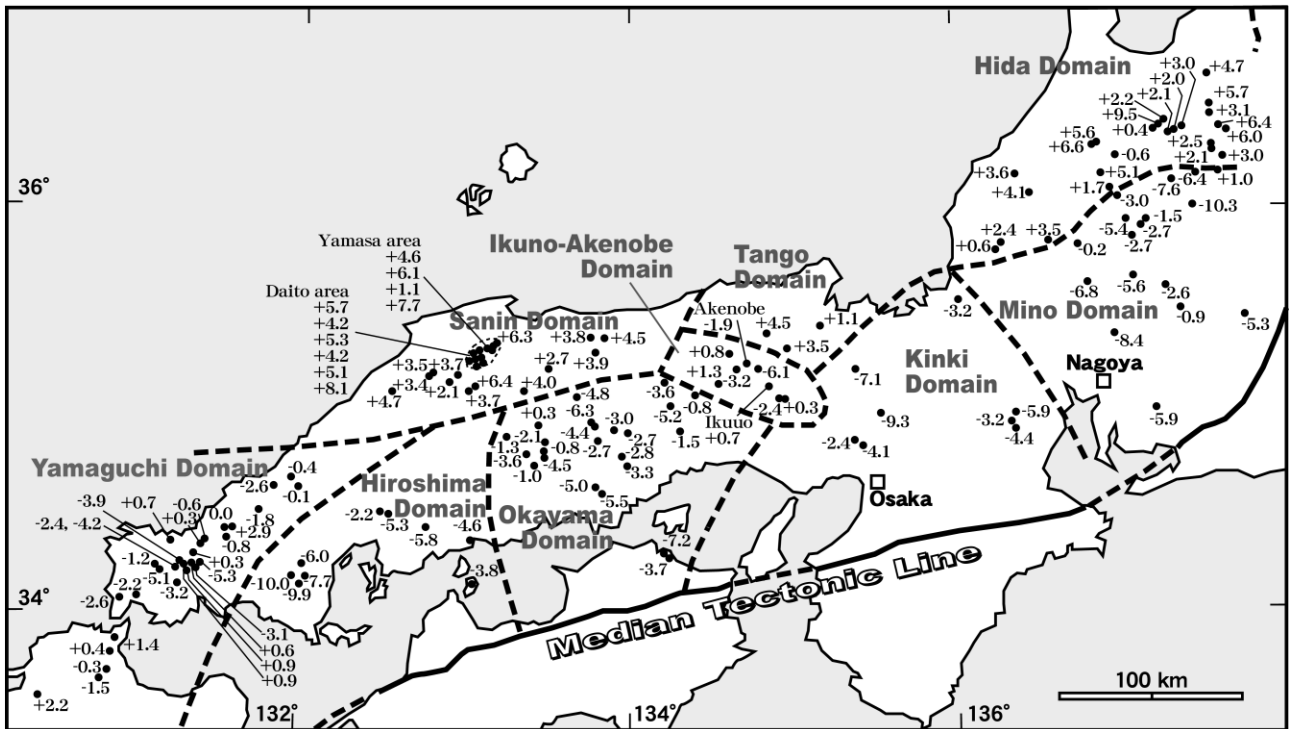


Fig. 7 Regional distribution of  $\delta^{34}\text{S}$  ‰ values of ore deposits and proposed domains in the Inner Zone of Southwest Japan.

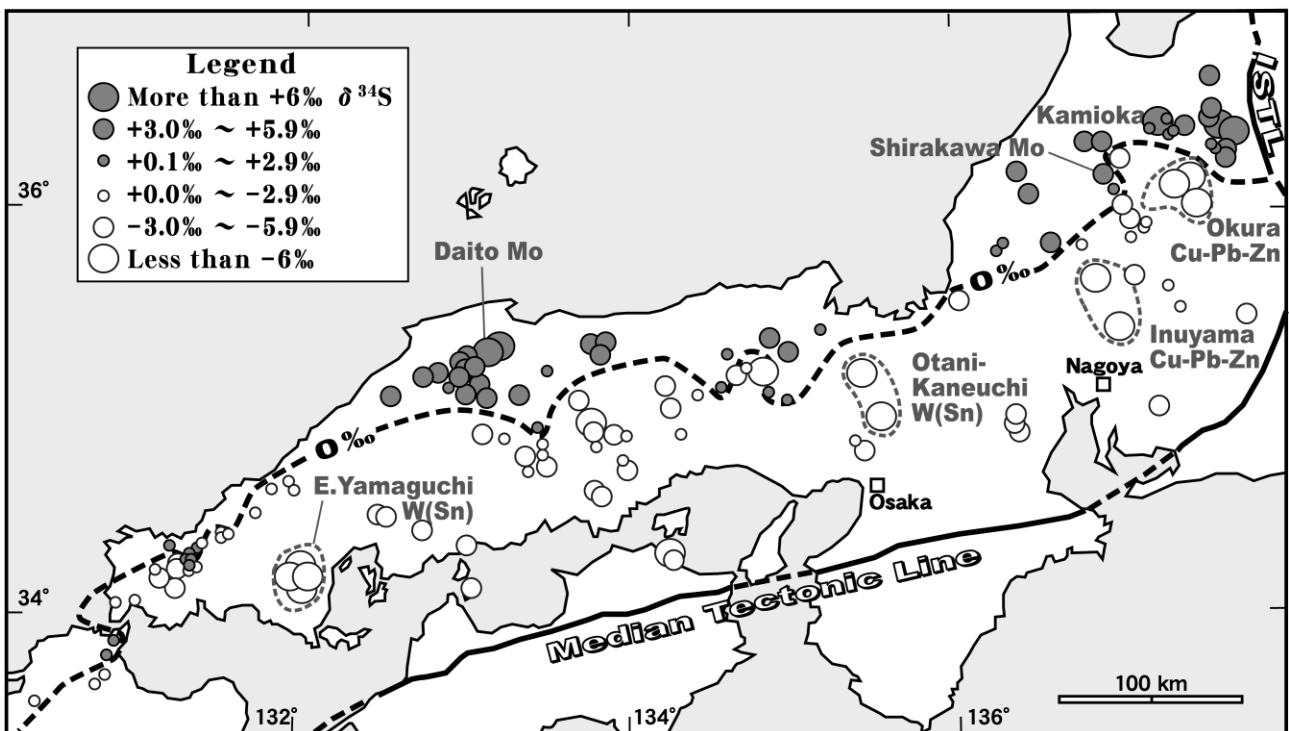


Fig. 8 Regional variation of  $\delta^{34}\text{S}$  ‰ values of ore deposits of the Inner Zone of Southwest Japan. The magnetite-series/ilmenite-series granitoids boundary corresponds roughly to the ore  $\delta^{34}\text{S}$  value of 0 ‰ line.

deposits so far mentioned, subvolcanic Sn-W-bearing base metal deposits occurring at the northern margin of the Sanyo-Naegi belt in Hyogo Prefecture have higher  $\delta^{34}\text{S}$  values around 0 permil. This is a unique feature of the Ikuno-Akenobe domain.

The Ikuno veins (average +0.7 ‰) are hosted in late Mesozoic volcanic rocks composed of mostly felsic but partly basaltic compositions. On the other hand, the Akenobe veins are hosted in sedimentary and some volcanic rocks of the late Paleozoic Maizuru Group. The veins are composed of distinct two stages of the early Cu-Zn vein and the later Cu-Sn vein. They have negative average  $\delta^{34}\text{S}$  values of -3.3 and -1.6 permil, respectively. Matsuhisa *et al.* (1980) found  $^{18}\text{O}$  depleted oxygen isotopic ratios on the vein quartz of both the early Cu-Zn veins and the later Cu-Sn veins, and suggested that 80-100 percent of the ore fluids originated in meteoric water. It follows that some part of the ore sulfur at Akenobe could have been extracted from the sedimentary wall rocks and mixed with magmatic sulfur during the hydrothermal circulation.

## 7. Concluding Remarks

$\delta^{34}\text{S}$  values of ore sulfur from the late Cretaceous-Paleogene granitic terranes in Southwest Japan show a similar asymmetric pattern to that of the late Cenozoic mineralizations (Ishihara *et al.*, 2000b). Before the late Cenozoic mineralizations, Cretaceous-Paleogene fore-arc sediments were accreted into the Shimanto Supergroup of Southwest Japan. When the accretion was terminated and the tectonic setting was converted to extensional, the Miocene granitic magmatism triggered by basaltic injection (Ishihara and Matsuhisa, 1999) occurred in the whole Outer Zone of Southwest Japan. Hence, the Outer Zone granitoids and related ore sulfur were strongly influenced by biogenic sulfur in the accretionary sediments. (Ishihara *et al.*, 2000b).

Similarly, negative  $\delta^{34}\text{S}$  values of the ore sulfur from the Ryoke and Sanyo-Naegi Belts are considered originating from the negative  $\delta^{34}\text{S}$  values of the related ilmenite-series granitoids, and in turn of the crustal components of possibly Jurassic accretionary complex of the Mino Terrane (Ishihara and Matsuhisa, 2002). Magnetite-series ore sulfur is generally positive in  $\delta^{34}\text{S}$  values reflecting their source granitoids. The magnetite-series granitoids occur generally in none-accretionary terranes, which contain little or no biogenic sulfur. The  $^{34}\text{S}$  enrichment may be accomplished by addition of seawater sulfate through subduction processes (Sasaki and Ishihara, 1979).

The paired belts of  $\delta^{34}\text{S}$  values are obvious in the late Cretaceous-Paleogene metallogenic terrane, as well as in the late Cenozoic terrane. This asymmetry pattern may be called the Japanese type, which is char-

acteristic of an island arc with accretionary wedges and varying tectonic settings. This is so contrasting when compared with stable continental country such as the Korean Peninsula where no accretion and generally positive  $\delta^{34}\text{S}$  values are observed (Ishihara *et al.*, 2000a).

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## 硫黄同位体比の対配列: 西南日本内帯の白亜紀後期—古第三紀鉱床

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### 要 旨

西南日本内帯の白亜紀後期—古第三紀の鉱床と岩石産の硫化物を領家帯5個, 山陽—苗木帯18個, 羽越—関東帯15個, 山陰—白川帯47個について  $\delta^{34}\text{S}$  値を測定した. 既発表資料と合わせて鉱床別平均値を, チタン鉄鉱系花崗岩地帯で109個, 磁鉄鉱系花崗岩地帯で56個求めた.  $\delta^{34}\text{S}$  値は鉱床のタイプや鉱種よりも先ず花崗岩系列により変化し, チタン鉄鉱系地帯では0パーミルよりも軽く, 磁鉄鉱系地帯では0パーミルよりも重い. この対配列は日本列島において特徴的なもので, “日本型”と呼ばれたが, その成因は前弧の圧縮場において形成された付加体と背弧の張力的な構造場においてマグマ活動が生じた結果として説明される. 各系列では地域別に  $\delta^{34}\text{S}$  値と鉱種に変化が見られることがあり, これはその地域の成因的背景を暗示するものとして, ドメインと呼ばれた. チタン鉄鉱系地帯では著しく  $\delta^{34}\text{S}$  値が低い目玉が現れ, いずれも美濃—丹波帯で代表される付加体で認められる. 付加体は深所まで達するために花崗岩マグマの発生に関与し, 泥質岩からの還元性硫黄がマグマを経由して鉱床に反映したものと考えられる. 京都の大谷—鐘打と山口県東部の目玉では鉱種がタングステンである共通性があり, Sと共にWも堆積岩に由来する可能性がある. 生野—明延多金属型鉱床は+1パーミル前後の値を持つ珪長質マグマから形成されたが, 明延では母岩からの硫黄の供給の可能性も考えられる.