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Chemical Variation of Paleozoic-Cenozoic Sandstone and Shale across the Western Shikoku District, Southwest Japan

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Abstract: Sandstones (17 samples), shales (26 samples) and metamorphic rocks (3 samples) of late Paleozoic to Tertiary age collected across the western Shikoku district were analyzed for major elements and CO_2, C, S, Li, Zn, Cu, Pb, Th, U, Sn and As by conventional wet methods and atomic absorption spectrometry.

Sedimentary rocks of the Izumi Group (late Late Cretaceous) are generally rich in K_2O reflecting their main origin of felsic igneous rocks. Rocks of the Chichibu belt (late Paleozoic-Jurassic) are most sodic indicating fragments of spilitic rocks involved in the clastics. Those of the Lower Shimanto Group (Cretaceous) are somewhat rich in femic components, but the Upper Shimanto Group (lower Tertiary) is extremely high in SiO_2 and low in Al_2O_3, reflecting high maturity of the clastics which may have been derived mainly from sedimentary and metamorphic terranes.

In the Izumi belt where the sedimentation mechanism has been well established, femic components decrease but SiO_2/Al_2O_3 ratio increases stratigraphically upward. The observation indicates that the upper rocks are more matured than the lower ones, because these components constitute unstable minerals of mafic silicates and feldspars during weathering and sedimentation. The same relationship is seen on the Chichibu-Sambosan pair, and Lower and Upper Shimanto Groups pair of the Chichibu and Shimanto terranes, respectively; thus indicating source of the clastics existed to the north of each terrane. The southernmost belt of the Upper Shimanto Group with high FeO/Fe_2O_3 ratio and C content on the shales is in accord with the distribution of the most reduced, S-type ilmenite-series granitoids of Miocene age in the Shimanto terrane.

Introduction

The western Shikoku district is underlain by upper Paleozoic to Paleogene metamorphic and sedimentary rocks accumulated in the Chichibu and Shimanto geosynclines. These rocks are distributed in an east-northeast direction parallel to lineaments of Southwest Japan Arc, a major one of which is shown as the Median Tectonic Line. Most of the rocks occur to the south of the Median Tectonic Line; thus geotectonically belonging to the Outer Zone of Southwest Japan.

Several unilateral variations have been recognized across the Outer Zone. Age of the geosynclinal sediments becomes younger oceanward from the Median Tectonic Line. Sandstone of the Shimanto geosynclinal sediments called the Shimanto Supergroup, which occupies a great part of the Outer Zone, contains an increasing amount of quartz toward the same direction (TERAOKA, 1977, 1979). Miocene igneous rocks intruding the metamorphic and sedimentary rocks reveal

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an S-type ilmenite-series characteristic in the south but an I-type ilmenite-series one in the north (NAKADA and TAKAHASHI, 1979; TAKAHASHI *et al.*, 1980; MURATA, M., 1982).

Recently much attention has been focussed on the Shimanto Supergroup as to the origin of the clastic materials. They could have been derived from presently seen land area to the north (TERAOKA, 1979), but partly from a missing continental mass to the south (HARADA *et al.*, 1970). The clastics are interpreted as an accretional prism of trench and deep ocean-floor sediments (KANMERA, 1976; TAIRA *et al.*, 1980).

Miocene igneous rocks occur mostly as granitoids in the Shimanto terrane and partly in the Sambagawa and other terranes from Shikoku to Kyushu (Fig. 1). All but one (Ashizuri) granitic plutons belong to ilmenite-series which have been reduced by sedimentary carbon from the wall rocks or their basement (ISHIHARA, 1977).

In spite of these arguments developed in recent years, petrochemical data of the sediments are scarse. In this paper, we describe chemical data of the sedimentary rocks of the western Shikoku district, and discuss their variation in time and space, and their bearing on petrogenesis of the Miocene granitic activity.

Geology and analyzed samples

The studied transect covers, from north to south, the Ryoke, Sambagawa, Chichibu and Shimanto terranes (Fig. 1). In the Ryoke terrane, the Cretaceous Ryoke granitic and metamorphic rocks are unconformably overlain by the Izumi Group of late Late Cretaceous, mostly Campanian age, which is a flysh-type sequence composed of alternating sandstone and shale with basal conglomerate and thin layers of felsic tuff at some horizons (HARADA, 1965; NISHIMURA, 1984).

Bounded by the Median Tectonic Line,

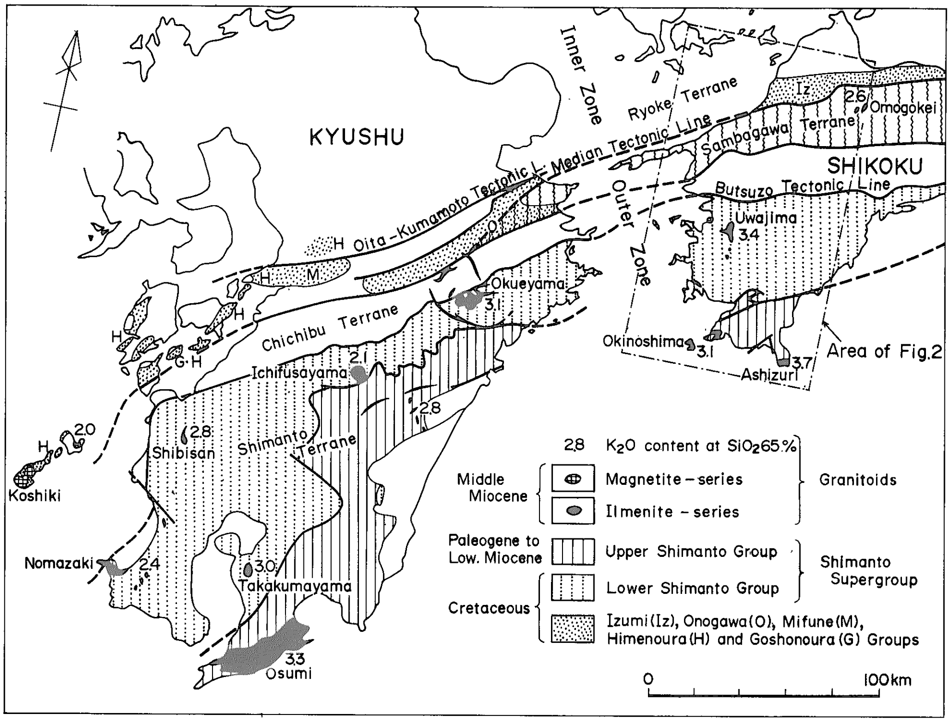

Fig. 1 Sedimentary and tectonic units and locality of Miocene granitoids in the Outer Zone of Southwest Japan. K_{65} is taken from NAKADA and TAKAHASHI (1979), except Koshiki Island which was calculated from ISHIHARA *et al.* (1984).

high P/low T-type metamorphic rocks of the Sambagawa terrane occur to the south of the Izumi Group. The metamorphic rocks are originated dominantly in mafic volcanic rocks but partly sandstone and shale of probable late Paleozoic to middle Mesozoic age. SUYARI *et al.* (1980) found Late Triassic conodonts in lenticular bodies of calcareous schist at north of Yawatahama. Mixed samples of representative rock types at three localities were chosen for the chemical analyses. The Mikabu green rocks occurring to the south of the Sambagawa metamorphic rocks are characterized by mafic submarine effusive rocks (SUZUKI, 1972).

The Sambagawa metamorphic rocks change southward into non- or weakly metamorphosed rocks in the Chichibu and Sambosan belts of the Chichibu terrane. They consist of sandstone, shale, chert, limestone and mafic volcanic rocks, and range in age from late Paleozoic to Jurassic (KASHIMA, 1969, 1983; MURATA, A., 1982; NAKATANI, 1983; TERAOKA *et al.*, 1985). Chert is occasionally associated with bedded manganese deposits. Samples for this study were collected from probable Jurassic strata of clastic facies, mainly along the Uchiko-Nomura route (Fig. 2).

The southern half of the western Shikoku district is a terrane of Shimanto Supergroup, which is separated from the Chichibu terrane by the Butsuzo Tectonic Line, a large low-angle thrust fault dipping north. The Shimanto Supergroup is a thick geosynclinal sequence composed of sandstone and shale alternating in various manners with minor amounts of conglomerate, limestone, chert and submarine volcanic rocks, and is intensely folded and faulted. It comprises the Lower Shimanto Group (Cretaceous) and the Upper Shimanto Group (lower Tertiary).

The Shimanto terrane of this transect is divided into five belts as illustrated in Figure 2. Judging from recent studies (e.g., TANAKA, 1980; TAIRA *et al.*, 1980; TERAOKA *et al.*, 1985), the age of strata in the respective belts is considered as follows: North Uwa belt, late Neocomian to Santonian; South Uwa belt, Coniacian to Campanian; Nakasuji belt, Coniacian to Maastrichtian and Eocene to early Oligocene; Hiromi belt, Eocene; and Shimizu belt, Eocene to early Oligocene.

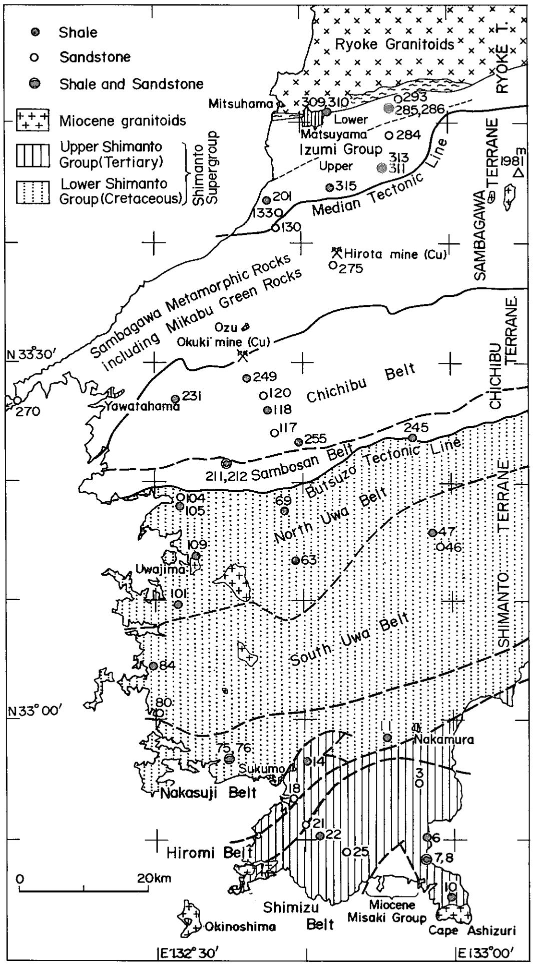

Fig. 2 Geotectonic division and locality of the analyzed samples in the western Shikoku district. Two copper mines are of the Besshi-type stratabound deposits.

Samples for chemical analyses were collected from the most common rock types at given outcrops to obtain a regional average composition of each geotectonic terrane. The sample localities are illustrated in Figure 2. A fist size of the samples, except for the Sambagawa samples which were chosen from about 30 cm wide zone across the schistosity, was sliced normal to the bedding plane by diamond cutter, then crushed and powdered for the analyses. About sandstone, fine- to mediumgrained ones were selected, although coarse-grained sandstone is not rare in some parts of the studied area. Shale is composed of siltstone and claystone which are sometimes alternated finely. No red shale which could have been pelagic sediments of deep ocean (SAITO and TANAKA, 1983) were selected, because they are very minor in the area. Analytical methods are conventional wet methods for major elements, atomic absorption spectrometry for Li, Cu, Pb and Zn (TERASHIMA, 1971), As (TERASHIMA, 1976), and Sn (TERASHIMA, 1975).

Microscopically, sandstones of Cretaceous to early Tertiary age contain clayey matrix more than 15 volume percent in most cases, and vary in modal composition stratigraphically and regionally (TERAOKA, 1977, 1979). The Cretaceous sandstones tend to become poorer in feldspars and richer in volcanic fragments stratigraphically upward. The Tertiary ones are characterized by the predominance of quartz.

Table 1 Chemical compositions of sandstones from

Terrane	Ryoke Terrane				
Belt	Izumi Group, Lower		Izumi Group, Upper		
Sample No.	70S–293	70S–286	70S–284	70S–313	70S–133
SiO_2(%)	74.06	74.38	72.36	73.89	76.34
TiO_2	0.32	0.20	0.23	0.25	0.35
Al_2O_3	13.77	12.73	11.02	12.31	11.92
Fe_2O_3	0.76	0.52	0.28	1.00	0.92
FeO	1.40	1.26	0.97	0.90	0.75
MnO	0.02	0.02	0.05	0.04	0.02
MgO	0.48	0.54	0.28	0.33	0.43
CaO	0.36	1.43	4.30	1.51	1.04
Na_2O	3.13	2.93	2.67	1.95	3.03
K_2O	3.36	2.83	2.26	2.86	1.94
P_2O_5	0.04	0.04	0.04	0.05	0.05
$H_2O(+)$	1.67	1.69	1.54	2.23	2.17
$H_2O(-)$	0.32	0.34	0.32	0.46	0.42
CO_2	0.01	0.65	3.23	1.73	0.33
C	0.15	0.11	0.10	0.21	0.11
S	<0.01	<0.01	0.06	0.09	0.02
Total	99.84	99.73	99.71	99.81	99.84
Li(ppm)	44	43	44	57	57
Zn	35	35	36	42	185
Cu	5	6	6	6	8
Pb	14	16	13	13	11
Th	7	8	9	9	8
U	2.1	2.1	2.1	1.7	1.7
Sn	1.5	1.8	1.6	1.4	1.7
As	4.2	3.7	4.3	7.5	6.0

For Tables 1–4, SiO_2 through S were analyzed by Tokyo Analytical Lab for Coal & Minerals; Li through As, by S. TERASHIMA; Th and U from ISHIHARA *et al.* (1981)

Chemical compositions

Analytical results and sample description are given in Tables 1–4. The sample localities are shown in Figure 2 and also listed in the appendix.

Sandstone

Sandstones of the Izumi Group are lithic ones rich in felsic volcanic fragments. Other sand grains are quartz and feldspars with minor amounts of granitic, sedimentary and metamorphic rocks. Sandstone petrography and paleocurrent analysis indicate that the sediments of the group were supplied from a source area in the Inner Zone of Southwest Japan where felsic volcanic and granitic rocks were widely exposed (HARADA, 1965; TERAOKA, 1977; NISHIMURA, 1984).

Analytical data of sandstones reveal a contrasting compositional difference between the lower part of the group (Lower Subgroup of

the Ryoke, Sambagawa and Chichibu terranes.

Sambagawa Terrane			Chichibu Terrane		
			Chichibu Belt		Sambosan Belt
70S–130	70S–275	70S–270	70S–120	70S–117	70S–212
54.23	63.70	69.93	71.83	73.99	74.99
0.81	0.61	0.41	0.40	0.34	0.34
14.01	16.15	14.92	14.08	12.84	12.64
1.89	1.16	0.80	0.72	0.40	0.36
3.51	3.66	2.60	1.87	2.08	1.76
0.09	0.13	0.06	0.06	0.04	0.04
3.20	2.14	1.32	1.01	0.92	0.52
10.85	0.99	1.00	1.23	0.95	0.70
2.67	2.58	2.62	4.33	5.36	3.04
1.45	3.26	2.60	1.27	0.58	2.33
0.19	0.11	0.10	0.08	0.10	0.05
1.76	2.66	2.00	2.17	1.82	1.40
0.20	0.24	0.26	0.24	0.04	0.12
4.69	1.18	0.37	0.19	0.28	1.32
0.14	0.69	0.42	0.13	0.09	0.15
0.02	0.47	0.24	0.27	<0.01	0.13
99.71	99.73	99.85	99.88	99.83	99.89
17	33	37	21	30	19
70	80	63	42	42	43
34	38	19	7	7	4
13	19	16	10	11	14
3	9	8	6	6	10
0.6	2.1	2.1	1.5	1.3	2.6
0.9	1.9	2.3	0.5	1.0	1.8
6.0	4.0	8.5	4.5	5.0	4.5

The Sambagawa samples: 70S–130, Epidote green schist more than black schist; 70S–275, Quartz schist more than black schist; 70S–270, Quartz schist much more than black schist.

Table 2 Chemical compositions of sandstones from the Shimanto terrane.

Group	Lower Shimanto Group					Upper Shimanto Group			
Belt	North Uwa Belt	South Uwa Belt		Nakasuji Belt		Hiromi Belt	Shimizu Belt		
Sample No.	70S–104	70S–80	70S–46	70S–75	70S–18	70S–21	70S–3	70S–25	70S–8
SiO_2(%)	65.96	69.47	69.78	68.89	76.96	67.07	75.76	77.93	79.85
TiO_2	0.42	0.43	0.50	0.33	0.43	0.43	0.43	0.44	0.24
Al_2O_3	15.23	14.70	14.44	12.68	10.69	8.86	11.20	9.61	8.80
Fe_2O_3	1.68	0.20	0.76	0.16	0.72	0.16	0.12	0.36	0.48
FeO	2.01	3.05	2.26	2.30	1.94	1.87	2.77	2.12	1.44
MnO	0.05	0.04	0.06	0.28	0.03	0.21	0.02	0.03	0.04
MgO	1.88	1.18	1.09	1.01	0.88	0.75	1.06	1.03	0.54
CaO	3.05	2.30	1.65	3.77	0.87	7.96	1.15	1.12	1.93
Na_2O	3.57	3.98	4.38	2.84	3.00	1.94	2.75	2.39	3.13
K_2O	2.02	2.41	2.23	2.24	1.83	1.84	1.14	1.66	0.58
P_2O_5	0.11	0.09	0.08	0.06	0.06	0.09	0.09	0.08	0.05
$H_2O(+)$	2.75	1.49	1.81	2.04	1.50	1.97	1.89	1.53	1.36
$H_2O(-)$	0.62	0.02	0.16	0.02	0.12	0.16	0.08	0.18	0.08
CO_2	0.43	0.22	0.64	2.85	0.57	6.29	1.23	1.17	1.29
C	0.08	0.05	0.04	0.12	0.09	0.06	0.04	0.06	0.05
S	<0.01	0.30	0.05	0.22	0.25	0.01	0.01	0.02	0.03
Total	99.86	99.93	99.79	99.81	99.94	99.67	99.74	99.73	99.89
Li(ppm)	23	37	26	28	29	20	36	20	32
Zn	51	77	45	43	36	30	50	38	24
Cu	13	8	7	9	4	6	9	5	8
Pb	14	19	14	15	11	13	13	12	10
Th	4	6	6	8	7	7	6	7	5
U	1.1	1.6	1.4	1.8	1.3	1.4	1.7	1.4	1.2
Sn	1.2	0.8	0.9	1.6	1.0	0.7	0.7	0.7	0.5
As	3.5	1.6	5.0	3.0	6.1	5.2	2.7	5.7	3.3

Table 3 Chemical compositions of shales from the Ryoke and Chichibu terranes

Terrane	Ryoke Terrane						Chichibu Terrane					
Group	Izumi Group, Lower			Izumi Group, Upper			Chichibu Belt				Sambosan Belt	
Sample No.	70S–285	70S–309	70S–310	70S–311	70S–315	70S–201	70S–255	70S–118	70S–231	70S–249	70S–211	70S–245
SiO_2 (%)	61.99	61.75	65.77	65.72	68.03	70.38	61.45	62.96	65.53	66.61	63.01	66.81
TiO_2	0.53	0.44	0.51	0.53	0.46	0.33	0.77	0.70	0.65	0.67	0.61	0.59
Al_2O_3	17.86	17.82	14.96	19.34	16.65	15.30	17.24	17.15	16.59	15.85	17.83	15.86
Fe_2O_3	2.04	1.60	1.52	0.64	1.04	1.60	0.80	2.49	0.80	2.51	2.08	1.72
FeO	1.87	2.73	2.52	0.86	0.57	0.72	4.93	3.03	4.00	2.05	2.66	2.19
MnO	0.03	0.06	0.06	0.01	0.01	0.01	0.08	0.06	0.11	0.05	0.06	0.09
MgO	1.43	1.58	1.39	0.99	0.73	0.78	2.61	2.20	2.00	1.61	1.59	1.72
CaO	0.70	1.60	2.13	0.11	0.42	0.39	1.57	0.98	0.90	0.50	1.26	1.51
Na_2O	1.52	1.58	2.13	1.20	1.77	1.09	2.42	3.35	2.15	2.30	2.10	2.05
K_2O	4.29	3.52	3.01	4.36	3.74	3.38	2.65	2.05	3.02	3.06	3.70	2.93
P_2O_5	0.06	0.07	0.10	0.06	0.05	0.06	0.19	0.19	0.11	0.13	0.11	0.13
H_2O (+)	4.77	4.30	2.96	3.94	4.01	3.94	3.78	3.67	3.01	3.38	3.68	3.32
H_2O (−)	1.32	1.00	0.64	1.02	1.22	0.98	0.28	0.48	0.14	0.44	0.76	0.87
CO_2	0.03	0.47	0.75	0.03	0.04	0.02	0.39	0.06	0.47	0.01	0.02	<0.01
C	1.12	0.63	0.68	1.21	1.21	0.93	0.73	0.48	0.41	0.57	0.34	0.15
S	0.02	0.68	0.78	0.03	0.04	0.03	0.17	0.09	0.13	0.04	0.03	0.01
Total	99.94	99.82	99.91	100.05	99.99	99.94	99.99	99.94	100.02	99.78	99.84	99.96
Li (ppm)	40	65	60	34	39	32	28	41	50	35	40	28
Zn	87	89	66	52	75	37	93	107	84	77	78	73
Cu	31	19	18	16	40	21	40	23	31	23	22	16
Pb	26	25	21	16	32	14	14	23	20	46	22	34
Th	14	13	11	16	13	13	7	11	13	12	18	11
U	3.4	3.9	2.9	4.0	3.6	3.3	2.1	2.8	2.4	2.8	4.1	2.5
Sn	3.6	3.6	2.8	4.4	3.7	3.2	1.9	3.0	2.0	2.7	4.3	2.1
As	15.5	19.5	34.0	16.0	9.5	12.0	11.0	13.0	8.0	12.0	5.5	1.2

70S–285, claystone, Formation B; 70S–309, sandy siltstone, Formation A; 70S–310, sandy siltstone, Formation A; 70S–311, claystone, Formation D of Harada (1965); 70S–315, silty claystone; 70S–201, silty claystone; 70S–255, phyllitic sandy siltstone, Nomura Formation; 70S–118, siltstone of Oyabu Formation; 70S–231, phyllitic claystone more than siltstone, Futaiwa Formation; 70S–249, silty claystone of Nomura Formation; 70S–211, siltstone of Itagatani Formation of Kashima (1969); 70S–245, siltstone of Itagatani equivalent formation.

Table 4 Chemical compositions of

Belt	North Uwa Belt					South Uwa Belt	
Sample No.	70S–101	70S–105	70S–63	70S–69	70S–109	70S–47	70S–84
SiO_2(%)	58.39	60.36	61.40	66.38	66.68	61.36	64.75
TiO_2	0.65	0.67	0.55	0.61	0.54	0.64	0.53
Al_2O_3	19.69	17.82	17.99	14.32	15.32	19.23	15.94
Fe_2O_3	1.52	3.89	1.12	1.44	2.15	1.76	1.56
FeO	3.07	1.80	4.72	2.79	1.94	1.80	2.83
MnO	0.05	0.08	0.20	0.03	0.04	0.02	0.04
MgO	2.16	2.38	2.19	1.67	1.40	1.40	1.43
CaO	1.15	0.31	0.34	2.13	1.46	0.31	1.18
Na_2O	2.04	1.61	1.66	2.70	1.62	1.71	1.98
K_2O	4.56	3.05	3.22	2.10	2.72	4.02	3.10
P_2O_5	0.11	0.10	0.07	0.08	0.08	0.08	0.07
$H_2O(+)$	4.26	5.89	4.60	2.96	3.25	4.94	3.09
$H_2O(-)$	0.96	1.04	0.66	0.72	0.68	1.24	0.52
CO_2	0.07	0.06	0.53	1.00	1.22	0.03	0.79
C	0.72	0.87	0.70	0.55	0.55	1.29	1.36
S	0.64	0.04	0.14	0.51	0.30	0.06	0.81
Total	100.04	99.97	100.09	99.99	99.95	99.89	99.98
Li(ppm)	48	42	61	42	31	21	65
Zn	95	111	100	75	80	66	75
Cu	35	50	66	15	15	40	27
Pb	26	20	16	12	12	14	23
Th	12	11	11	8	8	14	10
U	3.3	1.8	1.6	1.5	1.7	2.6	2.2
Sn	2.1	1.8	2.4	1.4	1.3	3.0	2.3
As	23.0	10.0	14.0	4.0	7.5	16.5	15.5

70S–101, claystone much more than siltstone; 70S–105, claystone; 70S–63, claystone; 70S–69, sandy siltstone; 70S–109, sandy siltstone; 70S–47, claystone; 70S–84, siltstone,

HARADA, 1965) and upper part (Middle and Upper Subgroups). Sandstones of the upper part, as compared with those of the lower part, are depleted in those components that constitute feldspars and ferromagnesian silicates, such as Al_2O_3, Na_2O, K_2O, Pb, FeO and MgO, while the SiO_2/Al_2O_3 ratio as well as CO_2 content increase upward (Fig. 3, Tables 1, 2). Thus, the chemical data suggest that unstable minerals of feldspars and mafic silicates were deposited preferentially in the early stage of the sedimentation, while stable quartz was accumulated in the later stage. The chemical difference can best be explained by difference in maturity of the sandstones; those of the upper part appear to be more matured. Sulfur content is high in the upper part.

On sandstones of the Chichibu belt of the studied area, KASHIMA (1969) concluded that the clastics were chiefly derived from volcanic rocks and subordinately from sedimentary, granitic and metamorphic rocks. For the shales from the Shimanto terrane.

Nakasuji Belt			Shimizu Belt			
70S–76	70S–11	70S–14	70S–10	70S–22	70S–7	70S–6
66.13	71.17	65.50	63.47	63.91	65.69	68.09
0.57	0.41	0.60	0.79	0.74	0.52	0.50
16.21	13.83	16.59	18.07	17.01	16.07	14.79
0.68	0.72	0.48	1.28	0.72	1.24	1.56
4.12	2.37	4.29	4.30	5.09	3.83	3.15
0.10	0.02	0.04	0.06	0.08	0.03	0.03
2.01	1.17	1.96	2.23	2.22	1.82	1.70
0.50	0.73	0.73	0.22	0.34	0.45	0.73
2.22	2.38	2.16	1.21	1.69	1.88	2.31
2.82	2.61	2.35	2.94	2.81	2.81	2.33
0.10	0.07	0.09	0.12	0.09	0.10	0.08
2.73	2.18	3.08	3.92	3.56	3.12	2.35
0.48	0.96	0.92	0.76	0.66	0.54	1.04
0.12	0.25	0.40	0.03	0.13	0.17	0.45
0.79	0.71	0.50	0.37	0.63	0.96	0.42
0.32	0.36	0.32	0.05	0.29	0 75	0.56
99.90	99.94	100.01	99.82	99.97	99.98	100.09
59	47	65	64	60	58	57
98	72	118	105	93	87	80
57	18	35	18	49	33	18
28	11	14	23	18	8	14
12	10	10	14	12	11	10
2.2	2.0	2.2	2.5	1.8	2.0	2.2
2.3	1.7	2.0	3.1	2.2	2.1	2.1
18.0	5.5	15.5	9.5	10.5	8.0	7.0

70S–76, silty claystone; 70S–11, sandy siltstone (poorly sorted); 70S–14, sandy siltstone (poorly sorted); 70S–10, Siltstone; 70S–22, silty claystone; 70S–7, sandy claystone with siltstone fragments; 70S–6, sandy siltstone

Uwagawa Formation in which two analyses are given in this study, he gave the following percentages on rock fragments of sandstones: 91% basalt and andesite, 3% sedimentary rocks, 1% metamorphic rocks and 1% granitic rocks.

Chemical composition of sandstones of the Uwagawa Formation is characterized by a high content of components that form sodic plagioclase and ferromagnesian minerals. As compared with sandstones of the Izumi Group, they are depleted in K_2O, Li, Pb, Th and U (ISHIHARA *et al.*, 1981), which are concentrated generally in ilmenite-series granitic or felsic igneous rocks. Thus these sandstones must have had sodic and mafic igneous rocks in their source areas. Sandstones of the Itagatani Formation (KASHIMA, 1969), which is distributed in the Sambosan belt, are more siliceous and felsic in composition than those of the Uwagawa Formation.

Sandstones of the Shimanto Supergroup

Table 5 Average compositions of sandstones of the Shimanto Supergroup (data from TERAOKA, 1977, 1979).

Age and Delt	Quartz	K-feldspar	Plagioclase	Rock fragments
Paleogene				
Shimizu Belt (n=42)	42.6%	6.5%	24.5%	26.4%
Nakasuji Belt (n=21)	32.8	12.7	21.2	33.3
Cretaceous				
South Uwa and Nakasuji Belts (n=88)	23.7	8.2	27.6	40.5
North Uwa Belt (n=171)	29.7	10.2	33.2	26.9

Number of analyses in parethesis.

have various ratios in their modal composition with respect to age and belt. The average compositions are recalculated from TERAOKA (1977, 1979) and are listed in Table 5. It is clear from the statistics that rock fragments of the Cretaceous sandstones increase stratigraphically upward and regionally southward in compensating decreasing of feldspars and quartz. The rock fragments are mostly volcanic rocks of felsic to intermediate composition, and basalt is very rare. Paleogene sandstones of the Shimizu belt are characterized by a high content of quartz and a low content of feldspars in particular.

Chemical compositions of the sandstones are averaged in each studied belt and selected components are shown in Figure 3. An interesting tendency is observed in the respective terranes of the Ryoke, Chichibu and Shimanto that the femic total (TiO_2+Fe_2O_3+FeO+MnO+MgO) decreases from north to south or stratigraphically upward, and SiO_2/Al_2O_3 ratio increases toward the same direction. These parameters are known to reveal maturity of the sandstones in the Izumi Group. It follows that sandstones of the Sambosan belt in the Chichibu terrane and the Upper Shimanto Group in the Shimanto terrane are more matured than those of the Chichibu and Lower Shimanto Groups, respectively, and indicate that these clastics are probably derived from north, as pointed out by TERAOKA (1979) in the case of the Shimanto Supergroup.

Content of CO_2 is generally higher in the upper unit in each terrane. Sulfur is relatively abundant in the Nakasuji belt.

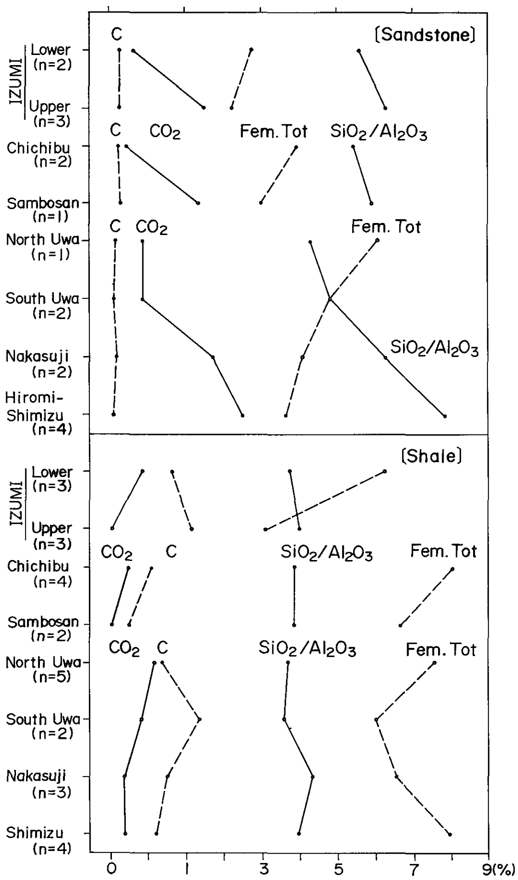

Fig. 3 Time-space variation of selected chemical components, as shown by average values of individual sedimentary units. Fem. Tot. implies sum of TiO_2, Fe_2O_3, FeO, MnO and MgO.

Average compositions of representative rocks in each belt are listed in Table 5, together with a Japanese average of geosynclinal sandstones by KATADA and ONO (1978). The average of the Izumi Group is higher in K_2O and SiO_2 and lower in femic components and CaO than those of the Japanese average, indicating felstic igneous rocks involved in the source area for the sandstones. The average of the Chichibu belt is most characterized by high Na_2O/K_2O ratio. Thus spilitic rocks must have been abundant in volcanic rocks of the source area. The average of the Lower Shimanto Group is lowest in SiO_2 and highest in femic components among the studied belts. Rock fragments are known to be mostly felsic to intermediate volcanic rocks (TERAOKA, 1979), so that basaltic interminglings must be present in the matrix. The average of the

Table 6 Average compositions of sandstones from western Shikoku district.

	1	2	3	4	5
SiO_2(%)	74.21	72.91	68.53	77.85	72.27
TiO_2	0.27	0.37	0.42	0.37	0.39
Al_2O_3	12.35	13.46	14.26	9.87	11.91
Fe_2O_3	0.70	0.56	0.70	0.32	0.71
FeO	1.06	1.98	2.41	2.11	2.14
MnO	0.03	0.05	0.11	0.03	0.06
MgO	0.41	0.97	1.29	0.88	1.18
CaO	1.73	1.09	2.69	1.40	2.43
Na_2O	2.74	4.85	3.69	2.76	3.22
K_2O	2.65	0.93	2.23	1.13	2.04
P_2O_5	0.04	0.09	0.09	0.07	0.08
$H_2O(+)$	1.86	2.00	2.02	1.59	1.72
$H_2O(-)$	0.37	0.14	0.21	0.11	0.23
CO_2	1.19	0.24	1.03	1.23	2.31
C	0.14	0.11	0.07	0.05	0.11
S	0.03	0.14	0.14	0.02	n.g.
Total	99.78	99.89	99.89	99.79	100.80
Li(ppm)	49	26	29	29	19*1
Zn	67	42	54	37	45*1
Cu	6	7	9	7	7*1
Pb	13	11	16	12	13*1
Th	8	6	6	6	n.g.
U	1.9	1.4	1.5	1.4	n.g.
Sn	1.6	0.8	1.1	0.6	n.g.
As	5.1	4.8	3.3	3.9	n.g.

(1) Izumi Group, average of 70S–293, 286, 284, 313 and 133.
(2) Chichibu belt, average of 70S–120 and 117.
(3) Lower Shimanto Group, average of 70S–104, 80, 46 and 75.
(4) Upper Shimanto Group, average of 70S–3, 25 and 8.
(5) Average of Japanese geosynclinal sandstones by KATADA and ONO (1978). Li, Zn, Cu and Pb are averages of Northern Kitakami and Iwaizumi belts (TERASHIMA and ISHIHARA, 1974). n.g., not given.

Table 7 Average compositions of shales from western Shikoku district.

	1	2	3	4	5	6
SiO_2	65.61	64.14	62.64	65.85	65.29	64.78
TiO_2	0.47	0.70	0.60	0.54	0.64	0.65
Al_2O_3	16.99	16.71	17.03	16.30	16.49	16.48
Fe_2O_3	1.47	1.65	2.02	1.18	1.20	1.63
FeO	1.55	3.50	2.86	2.78	4.09	3.65
MnO	0.03	0.08	0.08	0.05	0.05	0.07
MgO	1.15	2.11	1.96	1.50	1.99	2.01
CaO	0.89	0.99	1.08	0.68	0.44	0.78
Na_2O	1.55	2.56	1.93	2.07	1.77	2.30
K_2O	3.72	2.69	3.13	3.14	2.72	3.40
P_2O_5	0.07	0.16	0.09	0.08	0.10	0.14
$H_2O(+)$	3.97	3.46	4.19	3.24	3.24	2.77
$H_2O(-)$	1.03	0.34	0.81	0.80	0.75	0.51
CO_2	0.22	0.23	0.58	0.30	0.20	0.09
C	0.96	0.55	0.68	1.04	0.60	0.80
S	0.26	0.11	0.33	0.39	0.41	n.g.
Total	99.94	99.98	100.01	99.94	99.97	100.06
Li(ppm)	45	39	45	48	60	41
Zn	68	90	92	74	91	75
Cu	24	29	36	36	30	23
Pb	22	26	17	19	16	18
Th	13	11	10	12	12	n.g.
U	3.0	2.5	2.0	2.3	2.1	2.7
Sn	3.6	2.4	1.8	2.3	2.4	n.g.
As	17.8	11.0	11.7	13.9	8.8	n.g.

(1) Izumi Group, average of 70S–285, 309, 310, 311, 315 and 201.
(2) Chichibu belt, average of 70S–255, 118, 231 and 249.
(3) North Uwa belt, Lower Shimanto Group, average of 70S–101, 105, 63, 69 and 109.
(4) South Uwa and Nakasuji belts, Lower Shimanto Group, average of 70S–47, 84, 76 and 11.
(5) Shimizu belt, Paleogene Shimanto Supergroup, average of 70S–6, 7, 10 and 22.
(6) Weighted average of Japanese geosynclinal mudstone by KATADA and ONO (1978). Li, Zn, Cu and Pb are averages of northern Kitakami and Iwaizumi belts (n=16) taken from TERASHIMA and ISHIHARA (1974). U is average of various upper Paleozoic to middle Mesozoic formations (n=57) taken from ISHIHARA *et al.* (1969). n.g., not given.

Upper Shimanto Group has unique composition of high SiO_2 and low Al_2O_3, which may have been resulted from polycyclic sedimentation background of the sandstones derived from sedimentary and metamorphic terranes or longer distance of the transportation.

Shale

Chemical variation of shales in the Izumi Group is similar to that of sandstones, except for K_2O (Fig. 3), i.e., ferromagnesian components decrease but SiO_2/Al_2O_3 ratio increase in the upper part. However, no such relationship is observed among the Oyabu, Futaiwa, Uwagawa and Nomura Formations (KASHIMA, 1969) of the Chichibu belt, in which one of representative samples for each formation is analyzed (Table 3). Shales of the Sambosan belt are less mafic than those of the Chichibu belt.

Shales of the Shimanto Supergroup have rather homogeneous compositions. However, the southward increasing of SiO_2/Al_2O_3 ratio, which is very distinct in sandstones, is not evident on shales (Fig. 3). Ferromagnesian components are relatively high throughout the units.

Among other elements, manganese is very low in the upper part of the Izumi Group. As compared with geosynclinal sediments of other belts, the Izumi Group is unique in the sense that the shales contain abundant carbon and have high C/S ratio (Fig. 4). This group contains coaly seams in places, but a rapid sedimentation may have inhibited to fix seawater sulfate sulfur into the shales. Geosynclinal shales of the Shimanto Supergroup have a positive correlation between C and S (Fig. 4). However, shales of the Chichibu and Sambosan belts are low in sulfur contents and have high C/S ratio, which is somewhat similar to the tendency seen in the Izumi Group.

In Table 7, averages of the studied shales are given for the respective belts and compared with a Japanese weighted average for shales. The average of the Chichibu belt is most similar to the Japanese average, except for K_2O and CO_2 contents. The other belts give slightly different compositions from the

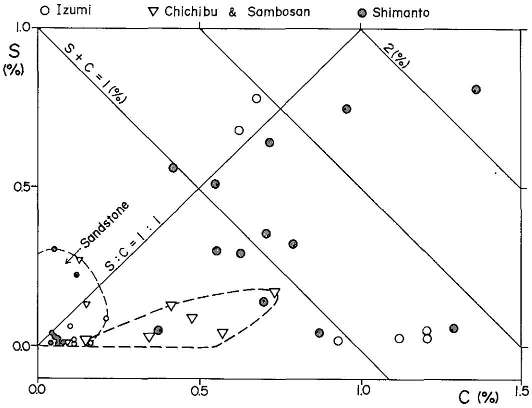

Fig. 4 Carbon-sulfur plot of sedimentary rocks.

Japanese average. Characteristic feature observed in sandstones of the respective belts are also seen slightly on the averaged shales.

Bearing of the chemistry on the genesis of Miocene granitoids

The studied sedimentary and metamorphic rocks have been intruded by Miocene plutons of various size (up to 400 km^2 in exposed area, see Fig. 1). They are generally high-level ones of ilmenite-series granitoids associated with cassiterite, chalcopyrite, arsenopyrite and pyrrhotite mineralizations (ISHIHARA, 1977, 1978; TERASHIMA and ISHIHARA, 1976), except for the Ashizuri alkaline complex which contains magnetite and has very low initial strontium ratio (0.7035, SHIBATA and ISHIHARA, 1979). Composite samples of sandstone and shale of the Shimanto Supergroup from the North and South Uwa belts give the initial ratio of 0.7090 and isochron age of 81.0 Ma. Thus the complex appears to be an independent, hot-spot type intrusion.

The ilmenite-series granitoids can be divided into I-type in the north and S-type in the south of the Outer Zone, based on absence and presence respectively of Al-silicates (NAKADA and TAKAHASHI, 1979). There are two possibilities about the origin of the two types of granitoids. One is magmatism of originally andesitic activity from the upper mantle mixed in different ratio with the continent-born magmas (TAKAHASHI, 1980;

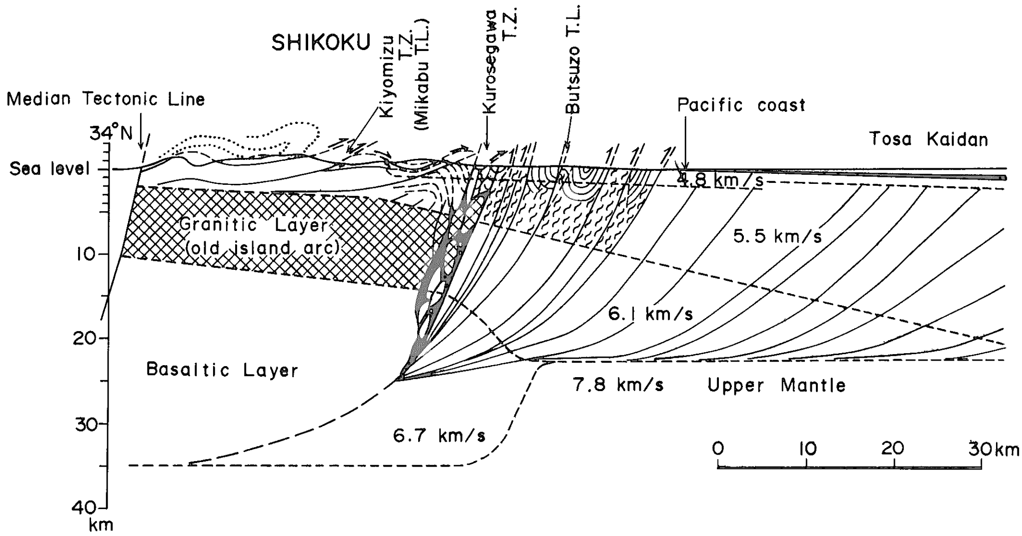

Fig. 5 Schematic crustal section of the Outer Zone of Southwest Japan across the central Shikoku. After HADA *et al.* (1982).

NAKADA, 1983), whereas the other possibility is I-type and S-type originated respectively in igneous and sedimentary source rocks of the continental crust (MURATA, 1984).

Seismic profile across the central Shikoku district indicates that so-called basaltic layer by P-wave velocity of 6.7 km/s disappears at the northern edge of the Shimanto terrane to the south (Fig. 5). Granitic layer of 6.1 km/s overlies above it in varying thickness. Focal distribution of shallow earthquake within the granitic layer and upper continental crust abruptly changes underneath the Butsuzo tectonic line or Kurosegawa tectonic zone, and structure of sedimentary and metamorphic terranes is steeply dipping north at south of these tectonic lines, while gently undulated to the north of the tectonic lines (Fig. 5). HADA *et al.* (1982), who emphasize significant bearing of the Kurosegawa tectonic zone in the tectonic development of Southwest Japan, therefore, consider that the continental crust materials are completely different on both sides of the Kurosegawa tectonic zone, i.e., the granitic layer to the north is older island arc basement but that to the south is highly metamporphosed part of the Shimanto Supergroup. Their model implies that Miocene granitoids of the Shimanto terrane should reflect directly composition of the sedimentary rocks.

Among major components, K_2O content of sandstone decreases steadly southward, as mentioned previously (ISHIHARA *et al.*, 1981). The content of shale is highest in the South Uwa belt (Fig. 6). All together, southward increasing of K_2O content is not clearly observed. Regional variation of major chemistry of the Miocene granitoids is reexamined recently by SATO and ISHIHARA (1983), and is found that an excess alumina expresses better I and S-type distinction, although it varies among individual plutons. Ratio of sandstone and shale in the Shimanto Supergroup is roughly unity, coeval submarine basalts occur only locally. Combined average of sandstone and shale gives the following normative corundum:

North Uwa belt: 6.5 wt.%
Shimizu belt: 7.4 wt.%
(Chichibu belt: 6.0 wt.% or less because of abundant basalts)
(Sambagawa terrane: 6.0 wt.%)

Effect of wall rock assimilation can be best shown by minor elements whose abundance difference between the wall rocks and magmas is large, an example of which is best shown on bulk S content and $\delta^{34}S$ value of the Miocene granitoids (ISHIHARA *et al.*, 1983). Table 8

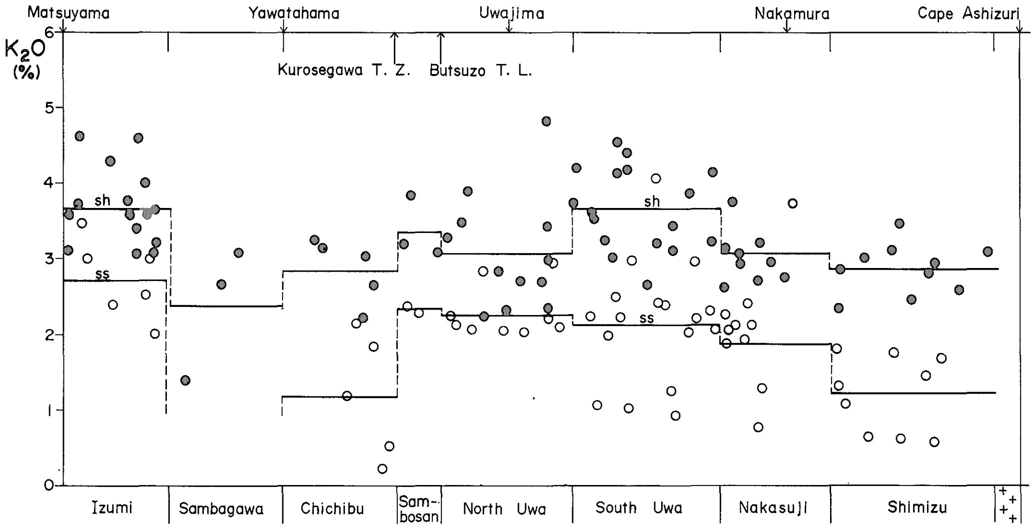

Fig. 6 Lateral variation of K_2O contents as plotted geographically against N20°W direction. Open circle, sandstone; Solid circle, shale and slate. Lateral bar indicates average content in each belt.

Table 8 Selected components of average shales from five sedimentary and metamorphic belts.

Terrane and Belt	n	FeO/Fe_2O_3	C(%)	S(%)	Cu(ppm)	As(ppm)
Sambagawa	3	2.6	0.42	0.24	30.3	6.2
Chichibu-Sambosan	6	1.8	0.45	0.08	25.8	8.5
North-South Uwa	7	1.4	0.86	0.36	35.4	12.9
Nakasuji	3	5.7	0.67	0.33	36.7	13.0
Shimizu	4	3.4	0.60	0.41	30.0	8.8

gives several other components which are critical to the genesis of the granitic magmatism and related mineralization.

Reducing agent of bulk FeO/Fe_2O_3 ratio and carbon content are higher in the southern, Nakasuji-Shimizu belt. Sulfur and arsenic which are rich in the Miocene granitoids are preponderant in rocks of the Shimanto terrane, as compared with those in the other terranes. There is, thus, consistency on the chemical characteristics of the Miocene granitoids and intruded sedimentary rocks. However, degree of melting and mode of emplacement may give crucial control on the chemical compositions of each pluton. Further, detailed studies are needed in the individual pluton areas.

Conclusions

Late Paleozoic to Tertiary sedimentary rocks of the western Shikoku district have unique chemical characteristics depending upon the geotectonic units, and maturity of the clastics appears to be high toward south in each terrane. The northernmost, Izumi Group is most potassic indicating that the clastics were derived from north of felsic igneous and metamorphic terrane. The Chichibu rocks are, on the other hand, sodic implying mafic and sodic igneous source rocks; similar rocks are distributed in just north of the Chichibu terrane. In the Shimanto terrane, the northern Lower Shimanto Group has an intermediate character between the two terranes mentioned above, with somewhat higher femic components, but the southernmost, Upper Shimanto Group is characterized by high silica and low alumina contents. Clastics of the Lower Shimanto Group is considered to have derived from igneous, metamorphic and sedimentary terranes to the north, including especially felsic-intermediate igneous rocks of the Inner Zone. Extremely high maturity of the Upper Shimanto Group may largely be due to its source materials of polycyclic sedimentary and metamorphic rocks which were derived from north.

Chemical similarity between the sedimentary rocks and intruding Miocene granitoids is not consistently seen on the analyzed components, but high reducing capacity as shown by high FeO/Fe_2O_3 ratio and C content, is observed on shales of the southern part of the Shimanto terrane where the most reduced, S-type ilmenite-series granitoids are distributed. Carbon, sulfur and other mobile components contained in the granitoids that have high concentration gradient between the magma and wall rocks may have been effected, at least, by the chemistry of sedimentary rocks in the Shimanto terrane. However, consistently reduced character of the Miocene granitoids even on those intruded into the Sambagawa metamorphic terrane (e.g., Omogokei, Fig. 1) indicates another essential reasoning for the reduction of the Miocene granitoids.

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付録　分析試料の採取地点

試料番号		試料採取地点
	和泉層群	
70S–133	(砂岩)	愛媛県伊予市大平四ッ松　南方直 700m
70S–201	(頁岩)	愛媛県伊予市下三秋　北東端
70S–284	(砂岩)	愛媛県温泉郡重信町大畑　南方直 500m．原田 (1965) の D 累層 (下部)
70S–285	(頁岩)	愛媛県温泉郡重信町岡　北方直 500m．原田 (1965) の B 累層
70S–286	(砂岩)	愛媛県温泉郡重信町岡　北方直 500m．原田 (1965) の B 累層
70S–293	(砂岩)	愛媛県温泉郡重信町神子野　北方直 900m．原田 (1965) の B 累層最上部
70S–309	(頁岩)	愛媛県松山市道後　伊佐爾波神社裏
70S–310	(頁岩)	愛媛県松山市道後　伊佐爾波神社裏
70S–311	(頁岩)	愛媛県温泉郡重信町横根—八幡間．原田 (1965) の D 累層 (中・上部)
70S–313	(砂岩)	愛媛県温泉郡重信町横根—八幡間．原田 (1965) の D 累層 (中・上部)
70S–315	(頁岩)	愛媛県伊予郡砥部町千足　東方直 500m．国道33号線旧道
	三波川変成帯	
70S–130	(片岩)	愛媛県伊予郡中山町犬寄　犬寄峠北方直 500m．国道56号線旧道．双海町境
70S–270	(片岩)	愛媛県西宇和郡三崎町名取　国道197号線沿いの崖．釜木への町道分岐点北方直 500m
70S–275	(片岩)	愛媛県伊予郡広田村猿谷子猿　旧日本鉱業広田鉱山ズリ
	秩父—三宝山帯	
70S–117	(砂岩)	愛媛県東宇和郡野村町栗の木　背間行．1.5m の砂岩層．全体として粘板岩≫砂岩 宇和川累層
70S–118	(頁岩)	愛媛県喜多郡肱川町硯　北端．鹿野川湖畔．互層．砂岩はわずか．KASHIMA (1969) の小藪累層
70S–120	(砂岩)	愛媛県喜多郡肱川町鹿野川　部落西端．橋の東側．KASHIMA (1969) の宇和川累層
70S–211	(頁岩)	愛媛県東宇和郡野村町出合．鹿島 (1967) の未詳古生層 (三宝山)
70S–212	(砂岩)	愛媛県東宇和郡野村町出合．鹿島 (1967) の未詳古生層 (三宝山)
70S–231	(頁岩)	愛媛県大洲市　鳥坂峠近く．鹿島 (1967) の久米古生層双岩相
70S–245	(頁岩)	高知県高岡郡梼原(ゆずはら)村梼原　北端 (三宝山)
70S–249	(頁岩)	愛媛県大洲市成能(なるの)　鳥首．KASHIMA (1969) の宇和川累層
70S–255	(頁岩)	愛媛県東宇和郡城川町辰の口
	四万十帯	
70S– 3	(砂岩)	高知県土佐清水市市野瀬　北東方．伊豆田峠西方 500m の大カーブ
70S– 6	(頁岩)	高知県土佐清水市久百々(くもも)　南端．N70°E–80°S のスレート劈開面
70S– 7	(頁岩)	高知県土佐清水市大岐　南端
70S– 8	(砂岩)	高知県土佐清水市大岐　南端．レンズ状～塊状で少量
70S– 10	(頁岩)	高知県土佐清水市　足摺半島中央部の新観光道路．451m 最高点の北西直頁 500m
70S– 11	(頁岩)	高知県中村市楠島．郵便局の対面
70S– 14	(頁岩)	高知県宿毛市昭和．バラス用砕石場
70S– 18	(砂岩)	高知県宿毛市小筑紫町伊与野．"石塔石"石切場
70S– 21	(砂岩)	高知県宿毛市石原　東端．砂岩のみ
70S– 22	(頁岩)	高知県宿毛市船ノ川　東端．泥岩≫砂岩
70S– 25	(砂岩)	高知県土佐清水市宗呂内　小場島．砂岩のみ
70S– 46	(砂岩)	高知県幡多郡大正町木屋が内　西方約 1 km
70S– 47	(頁岩)	高知県幡多郡大正町下道．小学校対岸．泥岩≫砂岩
70S– 63	(頁岩)	愛媛県北宇和郡松野町．予土線吉野駅東方．トンネル上
70S– 69	(頁岩)	愛媛県北宇和郡広見町広見　中組の対岸

試料番号		試料採取地点
70S- 75	(砂岩)	愛媛県南宇和郡一本松町満倉．中川トンネルの西方 500m の大屈曲部．泥岩＞砂岩の互層
70S- 76	(頁岩)	愛媛県南宇和郡一本松町満倉．中川トンネルの西方 500m の大屈曲部．泥岩＞砂岩の互層
70S- 80	(砂岩)	愛媛県南宇和郡内海村柏　脇田の南約 1 km．砂岩のみ．ホルンフェルス．磁硫鉄鉱―斧石プールあり
70S- 84	(頁岩)	愛媛県北宇和郡津島町　嵐坂トンネル西側入口
70S-101	(頁岩)	愛媛県宇和島市祝森　柿の木南西方約 1 km．"mud ball"
70S-104	(砂岩)	愛媛県北宇和郡吉田町．法花津坂最初の大カーブ．66m 水準点の北 400m
70S-105	(頁岩)	愛媛県北宇和郡吉田町引地．泥岩のみ
70S-109	(頁岩)	愛媛県宇和島市．市内北端．北宇和島駅南西方の路傍

四国西部の南北断面における古生代—新生代砂岩・頁岩の化学成分変化

石原舜三・寺岡易司・寺島　滋・坂巻幸雄

要　旨

四国西部を横断する地域から得られた砂岩(17個)，頁岩(26個)，変成岩(3個)の試料について，主成分のほか，CO_2, C, S, Li, Zn, Cu, Pb, Sn, As 等を分析し，南北変化，時代的変化を検討し，地殻上部物質との反応が指摘されている西南日本外帯花崗岩類との関連性を追求した．堆積岩類にみられる主成分の特徴は，砂岩に最もよくあらわれる．和泉層群は全般的に K_2O に富み，その後背地に珪長質火成岩類が多かったことが予想される．秩父帯では砂岩が Na_2O に富む特徴を示し，その供給源にスピライト等が多かったことを暗示する．上部四万十層群は特徴的に SiO_2 に富み，Al_2O_3 に乏しく，これは砂岩の熟成度が高いためと考えられ，その後背地は主に堆積岩(と変成岩)であったと推定される．

熟成度を示すパラメータとして，苦鉄質成分量 ($TiO_2+Fe_2O_3+FeO+MnO+MgO$) と SiO_2/Al_2O_3 比を用いると，和泉層群では下部層(北部)から上部層(南部)へ，前者の減少と後者の増加，すなわち熟成度の上昇が認められる．同様の傾向は秩父—三宝山帯，四万十帯の宇和—清水帯でもみられ，いずれも南側の時代的に若い地層で熟成度が高い．この事実はこれら砕屑物が一般に北方から供給されたことを示しているものと考えられる．

砂岩・頁岩比を 1 と仮定して地帯別の変化を見ると，K_2O は，Sタイプ-チタン鉄鉱系花崗岩類が広く分布する四万十帯南部へ増加する傾向を示さない．しかし，過剰 Al_2O_3 は南部の清水帯で最も多い．酸化—還元条件を最も規制すると思われる頁岩中の Fe_2O_3/FeO 比は，清水帯・中筋帯など南部で大きく，C含有量は四万十帯全体で高い．S, Cu, As なども四万十帯に多い．これらは，花崗岩類にみられる広域的化学成分変化と巨視的には一致する．

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