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Article *in* Geochemical journal GJ · January 2000

DOI: 10.2343/geochemj.34.155

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Geochemical Journal, Vol. 34, pp. 155 to 173, 2000

Geochemistry of Lower Cretaceous sediments, Inner Zone of Southwest Japan: Constraints on provenance and tectonic environment

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(*Received October 31, 1998; Accepted November 2, 1999*)

A geochemical study was carried out on Lower Cretaceous sedimentary rocks of the Wakino Subgroup, Kenseki Formation, and Sasayama Group, distributed in the Inner Zone of Southwest Japan. The chemical characteristics of the Lower Cretaceous sediments indicate that these rocks are immature first-order sediments derived from igneous and/or meta-igneous rocks of predominantly felsic composition. The sediments from the Kenseki Formation and the Sasayama Group, however, show high Cr and Ni abundances, suggesting a significant contribution of detritus from ultramafic rocks. Weathering at the source areas was moderate.

The high Th/U ratios (mostly > 3.8), negative Eu anomalies (Eu/Eu* between 0.67 and 0.93) and Th/Sc ratios (mostly between 0.5 and 1) of the Lower Cretaceous sediments suggest their derivation dominantly from an old upper crust with minor amounts of young arc-derived detritus. The major, trace and rare earth element compositions imply that deposition took place in an active continental margin environment. The small amounts of young arc-derived material in the sediments support the inference by other workers that arc magmatism was not so prominent in Southwest Japan during the early Cretaceous.

INTRODUCTION

It is widely accepted that during the pre-Tertiary period, the Japanese Islands had been located much closer to, or in contact with, the eastern margin of the ancient Asian continent. The paleogeography and tectonic environment of the Japanese Islands during this period have been a primary focus for Japanese researchers in recent years (e.g., Okada and Sakai, 1993; Taira and Tashiro, 1987). Previous researches related to the Cretaceous paleoenvironments of the Japanese Islands are principally based on provenance (e.g., Seo *et al.*, 1992; Ishiga *et al.*, 1997), biostratigraphy (e.g., Mizutani, 1995; Matsukawa *et al.*, 1997), type of sedimentary basins (e.g., Okada and Sakai, 1993; Sakai and Okada, 1997), and tectonics (e.g., Taira *et al.*, 1983). Very few studies, however, applied geochemical techniques to the evaluation of provenance and tectonic history of sediments from the Cretaceous basins distributed in the Inner Zone of Southwest Japan (i.e., the Kanmon Group, Kenseki Formation, Sasayama Group and a part of the Tetori Group; Fig. 1). In addition, provenance and paleogeographic studies to date have largely concentrated in the Kanmon Group (e.g., Seo *et al.*, 1992; Okada and Sakai, 1993) and the Tetori Group (e.g., Matsukawa *et al.*, 1997), but, on the other hand, the Kenseki Formation and the Sasayama Group have received little attention.

The composition of detrital sediments is primarily controlled by the composition of the source rocks, and to a minor extent, by weathering and diagenetic processes. The tectonics of the source area and the depositional basin impose constraints on sediment compositions insofar as they determine the kinds of source rocks exposed and the

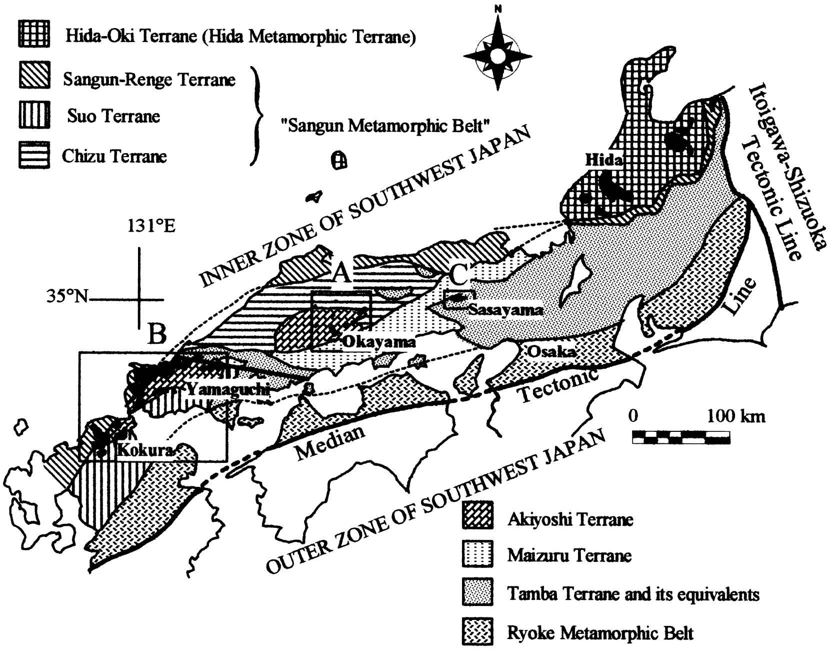

Fig. 1. Tectonic framework of the Inner Zone of Southwest Japan (after Nishimura, 1990) showing the distribution of Lower Cretaceous sedimentary rocks (solid black). A, Kenseki Formation; B, Kanmon Group; C, Sasayama Group.

rate at which the sediments are buried. One of the approaches in provenance studies is, therefore, to consider sediment compositions in the context of plate tectonic framework (e.g., Dickinson and Suczek, 1979). The reason for this approach is that the tectonic setting exerts first-order control on sediment compositions and overwhelms the effects of all other variables such as climate and depositional environment (Crook, 1974; Dickinson and Suczek, 1979; Valloni and Maynard, 1981).

In this paper, we examine the geochemical characteristics of Lower Cretaceous sediments from the Kenseki Formation, Wakino Subgroup (i.e., the lower part of the Kanmon Group) and Sasayama Group in an attempt to reveal a geochemical linkage, if any, among the sediments from these three basins; and from a geochemical perspective, we infer their provenance and the tectonic environment of the Inner Zone of Southwest Japan during the early Cretaceous.

GENERAL GEOLOGY

Southwest Japan is tectonically divided into the Inner Zone (continental side) and Outer Zone (oceanic side) by the Median Tectonic Line (Fig. 1). Cretaceous sedimentary basins are widely distributed in Southwest Japan. A striking contrast is noticed for the depositional environments between the continental side and oceanic side of Southwest Japan; i.e., non-marine sediments characterize the continental side, whereas marine sediments the oceanic side (Tanaka, 1977). For the present study, we collected sandstone and mudstone (i.e., siltstone and claystone) samples from three main Lower Cretaceous sedimentary basins of the Inner Zone; these are labeled as A (Kenseki Formation), B (Wakino Subgroup of the Kanmon Group) and C (Sasayama Group), respectively, in Fig. 1. A brief review of the geology of each of the three studied areas is presented below.

Kenseki Formation

The Lower Cretaceous Kenseki Formation is composed mostly of limestone breccia, conglomerate, sandstone, and red shale, with minor thin alternating beds of sandstone and mudstone. Conglomerate dominates in the lower part of this formation whilst mudstone dominates in the upper part. The Kenseki Formation is correlative with a part of the Wakino Subgroup, Sasayama Group, and the Kyeongsang Supergroup of southeastern Korea on the basis of non-marine fauna and nonvolcanic lithology (Matsumoto *et al*., 1982). Total thickness of the Formation is up to about 400 m. The Formation was deposited in a fluvial environment (Asiedu and Suzuki, 1995).

Basement rocks of the Kenseki Formation in the study area consist mainly of Paleozoic, nonmetamorphic sedimentary rocks and schists, with minor amounts of pre-Cretaceous igneous rocks and Triassic to Jurassic sedimentary rocks.

Sasayama Group

The Lower Cretaceous Sasayama Group is distributed in the Sasayama district, Hyogo Prefecture, and is stratigraphically divided into the Lower and Upper Formation (Kurimoto *et al*., 1992). The Lower Formation, about 1,300 m thick, is composed mainly of brownish red and green sandstone and siltstone, brown and red mudstone, and conglomerate, with rhyolitic tuff beds at the basal part (Kurimoto *et al*., 1992). The Upper Formation is about 250 m thick; it is composed of hornblende andesite lapilli tuff, tuff breccia and fine-grained tuff at the lower part, and tuffaceous sandstone, mudstone and conglomerate at the upper part. Fission-track datings indicate Tithonian and Berriasian age for the Lower Formation and Aptian to Albian age for the Upper Formation (Matsuura and Yoshikawa, 1992).

The Sasayama Group was deposited in a nonmarine environment and Sakaguchi (1959) has suggested a probable lacustrine environment. The Upper Formation can be correlated to the Upper Zone of the Wakino Subgroup of northern Kyushu on the basis of molluscan fossils (Ota, 1960). The Sasayama Group unconformably overlies the Ajima and Hikami sedimentary formations.

Wakino Subgroup

The Lower Cretaceous Wakino Subgroup forms the lower part of the Kanmon Group; the upper part is called the Shimonoseki Subgroup (Ota, 1953). The Wakino Subgroup, about 1200 m thick, is composed mainly of black shale interbedded with conglomerate, sandstone, shale and rhyolitic tuff. It is stratigraphically subdivided into three formations; i.e., Sengoku, Nyoraida, and Wakamiya, respectively, from bottom to top (Seo *et al*., 1992). The Sengoku Formation is composed of thickly bedded conglomerate, sandstone, black to purple shale, with minor intercalations of rhyolitic tuffs. The Nyoraida and Wakamiya Formation are characterized by evenly interbedded sandstone rhythmite and massive black shale with minor conglomerate. The Shimonoseki Subgroup, about 300 m thick, is largely composed of andesitic volcanic lavas and volcaniclastic sediments. Sedimentary facies analysis on the Wakino Subgroup indicates deposition in alluviofluvial, marginal lacustrine, and offshore lacustrine environments (Seo *et al*., 1994).

The basement rocks of Wakino Subgroup in northern Kyushu are crystalline schists of the Sangun Metamorphic Rocks, Carboniferous to Permian sedimentary rocks, and serpentinized peridotites. In the Yamaguchi area, the basement rocks include the Sangun crystalline schists, Carboniferous to Permian sediments, serpentinites, Triassic sediments and Jurassic to lowermost Cretaceous sediments.

PETROGRAPHY

The sandstones of the Kenseki Formation have matrix contents ranging from 15 to 30 modal %; those of the Wakino Subgroup and Sasayama Group have matrix contents ranging from 6 to 20 modal %. The matrix of the Kenseki, Wakino and Sasayama sandstones are composed mainly of clay minerals and altered, unstable grains. On the QFR plot of Okada (1971), the sandstones from the Kenseki Formation can be classified as lithic wacke (Fig. 2). Those from the Wakino Subgroup can be classified as lithic arenite/wacke whereas those from the Sasayama Group can be classified

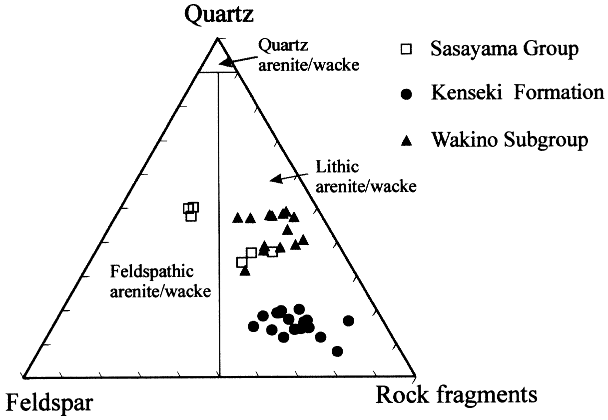

Fig. 2. Petrographic classification of the Lower Cretaceous sandstones (after Okada, 1971).

Table 1. Summary of modal analyses of the Lower Cretaceous sandstones

Constituent	Kenseki Formation		Sasayama Group		Wakino Subgroup	
	mean	s.d.	mean	s.d.	mean	s.d.
Monocrystalline quartz	6.5	1.8	21.5	9.1	22.3	3.5
Polycrystalline quartz	4.3	0.9	13.4	4.0	15.1	4.9
K-feldspar	1.4	1.1	6.9	3.3	6.6	3.1
Plagioclase feldspar	15.0	3.7	14.1	4.8	6.3	3.2
Igneous fragment	25.2	5.0	10.2	2.0	1.2	3.8
Felsic	[19.1]		[11.1]		[1.1]	
Mafic-ultramafic	[6.1]		[4.2]		[0.1]	
Metamorphic fragment	3.4	2.0	7.5	4.4	23.0	4.9
Schist	[2.1]		[5.7]		[19.6]	
Phyllite/slate	[1.3]		[1.8]		[3.4]	
Sedimentary fragment	6.2	1.6	2.6	1.7	3.7	0.8
Chlorite	9.3	3.8	3.8	5.3	0.0	0.0
Heavy minerals	3.0	1.5	3.1	1.8	0.4	0.3
Calcite	2.9	1.1	2.6	2.9	0.0	0.0
Matrix	22.8	3.7	14.4	3.8	12.4	3.7

s.d. = standard deviation.

as either feldspathic arenite/wacke or lithic arenite/wacke (Fig. 2).

The modal analysis of the sandstones from the Lower Cretaceous sediments is summarized in Table 1. The sandstones from the three basins are all generally rich in rock fragments; however, the proportions of rock types in lithic fragments differ from basin to basin (Fig. 2). Igneous lithic fragments for the Kenseki Formation and the Sasayama Group are mostly felsic volcanics (rhyolites?) with minor black colored mafic-ultramafic rocks. The metamorphic lithic fragments from the Sasayama and Wakino sandstones are composed mainly of schists. The chemistry of the chlorite from the Kenseki Formation and the Sasayama Group (MgO, 21.8–32.5 wt%; Cr ~ 2030 ppm) suggest their source from mafic-ultramafic rocks (Wrafter and Graham, 1989).

The heavy minerals in sandstones from the Sasayama Group are mostly epidote, magnetite and garnet, but also include, in decreasing order of abundance, chromian spinel, zircon, hematite,

Table 2. Unnormalized X-ray fluorescence-determined major element abundances (S, sandstone; M, mudstone)

Sample No.		SiO_2	TiO_2	Al_2O_3	FeO*	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5
Kenseki Formation (wt%)											
941006-3	S	59.62	0.64	12.13	4.94	0.10	4.41	7.47	1.75	2.92	0.09
941006-5	S	66.98	0.71	15.21	6.58	0.03	1.56	0.95	0.85	2.23	0.09
941006-8	S	55.19	0.85	12.98	7.15	0.11	3.38	8.33	1.29	1.13	0.10
941006-9	S	61.11	0.78	11.91	5.58	0.10	2.99	6.90	1.79	0.89	0.07
941006-11	S	53.30	0.94	12.40	5.50	0.13	4.27	10.65	1.81	0.79	0.11
960118-2	S	57.49	0.83	14.97	8.25	0.06	4.32	4.39	0.23	1.72	0.07
960613-2	S	56.26	1.00	11.83	6.99	0.14	7.01	6.34	2.73	0.37	0.08
960613-8	S	54.55	1.04	12.32	7.28	0.09	7.26	6.27	1.02	0.64	0.08
960613-9	S	47.89	0.81	10.34	6.35	0.11	4.85	14.68	0.31	0.90	0.09
951207-6	S	53.70	0.75	11.37	7.58	0.09	3.50	11.02	0.05	0.89	0.05
951207-8	S	54.78	0.75	13.14	6.24	0.10	3.22	9.38	0.18	1.78	0.07
951207-7B	S	62.91	0.80	13.88	4.48	0.07	2.30	5.36	1.40	1.99	0.08
951207-7	S	58.69	0.77	11.69	4.51	0.08	2.46	9.36	1.25	1.44	0.09
951207-10	S	52.04	0.69	11.69	6.32	0.14	3.64	12.14	0.58	1.32	0.06
951207-9	S	66.61	0.69	17.51	2.58	0.07	1.85	1.32	2.51	2.80	0.10
960613-1	S	57.49	0.95	13.09	9.30	0.09	5.48	3.90	2.11	1.27	0.06
Sasayama Group (wt%)											
960319-1	M	62.80	1.06	16.89	4.56	0.06	2.14	3.13	4.56	1.68	0.24
960319-s22	M	60.41	0.81	12.70	5.68	0.10	7.47	3.42	2.42	1.07	1.11
960319-1B	M	65.82	0.80	13.70	4.71	0.05	2.23	3.37	2.18	2.62	0.17
960319-2B	M	56.61	0.69	13.16	4.95	0.19	4.34	7.15	1.69	2.68	0.15
Wakino Subgroup (wt%)											
KK08B	S	77.49	0.38	9.60	4.07	0.07	1.62	1.30	2.18	1.28	0.07
KK11	S	64.44	0.53	15.08	4.41	0.10	1.75	5.26	2.94	2.43	0.15
KK02	S	61.87	0.72	14.06	4.55	0.08	2.22	5.69	2.34	2.12	0.16
KN04	S	78.91	0.39	10.02	2.69	0.05	1.23	1.31	2.83	1.17	0.07
KN07B	S	77.99	0.37	9.26	3.81	0.08	1.62	2.14	1.80	1.22	0.07
KN12	S	67.03	0.67	15.59	4.39	0.06	2.52	1.65	2.82	2.97	0.11
KN16	S	70.24	0.55	15.27	2.74	0.04	2.01	1.31	2.56	2.92	0.09
KN01	S	80.05	0.35	8.63	3.37	0.07	1.30	1.59	1.90	1.00	0.07
KK06B	M	43.32	0.39	8.34	3.40	0.08	3.46	23.27	1.05	0.96	0.13
KK06	M	55.66	0.74	15.66	6.00	0.07	3.31	5.31	1.31	3.16	0.12

**Total Fe as FeO.*

ilmenite, sphene, rutile and tourmaline. The heavy minerals of sandstones from the Kenseki Formation include, in decreasing order of abundance, epidote, opaque minerals, chlorite, chromian spinel, Ti-oxide minerals (anatase?), zircon, sphene, and garnet. The heavy mineral assemblage of the Wakino sandstones is dominated by opaque minerals (mostly ilmenite and magnetite with minor rutile and chromian spinel) and zircon. Other heavy minerals include, in order of decreasing abundance, garnet, aluminum silicate minerals (andalusite?), chromian spinel, rutile, epidote and pyroxene.

METHODS

Major element analysis by XRF

Thirty representative sandstone (all medium-grained) and mudstone (i.e., siltstone and shale) samples were analyzed for major elements by the X-ray fluorescence spectrometry (XRF). The preparation of samples is as follows: about 0.5 g of rock powder was weighed, and lithium tetraborate flux was added to give a flux to rock ratio of 10:1. The mixture was then fused in an induction furnace and the resulting melt was cooled to form a glass disk. The XRF analysis was carried out at the Kusatsu-Shirane Volcano Observatory, Tokyo Institute of Technology, with a Phillips PW1480 automated spectrometer. Calibration was performed using GSJ and USGS geochemical standards. Loss on ignition (LOI) was not determined. Table 2 shows the unnormalized chemical compositions.

Table 3. Representative ICP-determined major, trace and rare earth element abundances for the Lower Cretaceous sandstones and mudstones (i.e., siltstone and shale)

Sample No. Formation Rock type	D.L.	960319-2 Sasayama siltstone	960705-8 Sasayama shale	960705-2 Sasayama sandstone	961107-2b Kenseki siltstone	960613-7 Kenseki shale	941006-1 Kenseki sandstone	961108-4 Wakino shale	961109-4 Wakino sandstone	961109-10 Wakino siltstone
Major elements (wt%)										
SiO_2	0.01	58.36	60.35	65.88	68.95	63.79	60.95	70.29	62.23	66.81
Al_2O_3	0.01	13.10	10.66	13.45	14.46	12.21	12.35	16.02	15.06	13.59
Fe_2O_3*	0.01	7.88	5.55	4.74	5.33	6.05	8.23	3.96	5.74	4.90
MnO	0.01	0.12	0.11	0.08	0.04	0.12	0.12	0.05	0.10	0.05
MgO	0.01	6.56	6.74	1.78	1.26	3.62	6.18	0.77	3.31	1.41
CaO	0.01	3.21	4.95	3.54	1.40	4.67	3.06	0.69	6.63	2.43
Na_2O	0.01	2.18	2.02	2.19	2.70	3.23	2.84	1.05	3.98	2.76
K_2O	0.01	1.85	1.26	2.15	2.23	1.45	0.46	3.60	1.61	2.10
TiO_2	0.01	0.73	0.89	0.57	0.91	0.79	0.81	0.66	0.70	0.57
P_2O_5	0.01	0.11	0.13	0.11	0.11	0.12	0.14	0.10	0.13	0.09
LOI	0.01	6.36	7.85	5.53	2.35	4.66	4.62	2.86	1.48	4.19
Total		100.46	100.51	100.02	99.75	100.71	99.75	100.06	100.97	98.89
Trace elements (ppm)										
Sc	2.00	17	15	10	14	15	23	10	17	10
V	5.00	110	74	64	117	90	144	92	110	77
Cr	10.00	348	359	69	98	237	597	49	46	51
Co	0.50	26.0	23.0	12.0	13.0	28.0	35.0	5.0	14.0	11.0
Ni	10.00	288	226	47	64	268	395	24	17	11
Cu	10.00	12	26	20	16	26	11	<10	17	16
Rb	0.10	70	47	89	89	62	13	132	62	75
Sr	0.01	158	143	296	174	345	184	153	480	242
Y	0.10	23	33	22	22	28	18	19	16	16
Zr	0.10	166	262	177	195	212	94	181	97	149
Nb	0.50	10.0	12.0	13.0	13.0	14.0	5.9	7.7	8.6	6.8
Cs	0.10	4.1	2.7	14.0	15.0	3.1	0.8	6.5	4.5	5.0
Ba	0.10	414	232	567	411	449	136	771	503	459
Hf	0.10	4.8	7.4	5.5	6.0	6.4	2.8	5.3	2.9	4.5
Ta	0.01	0.74	0.95	1.10	1.01	1.28	0.43	0.66	0.65	0.57
Tl	0.05	1.10	0.46	0.91	0.54	0.57	0.19	1.58	1.42	1.18
Th	0.05	8.42	8.73	13.40	8.28	11.80	3.53	8.47	5.44	7.15
U	0.05	1.81	1.83	2.56	1.83	2.38	0.90	1.57	1.42	1.52
Rare earth elements (ppm)										
La	0.010	26.1	30.1	31.8	18.7	32.6	9.9	24.1	18.3	19.2
Ce	0.010	51.8	61.9	63.9	36.5	65.8	20.9	49.1	37.4	45.0
Pr	0.005	5.72	6.73	6.73	4.10	7.27	2.55	5.54	4.21	4.73
Nd	0.010	22.5	27.3	25.4	16.3	28.2	10.9	21.6	17.0	18.8
Sm	0.010	4.85	5.90	5.17	3.59	5.94	2.77	4.35	3.67	4.11
Eu	0.005	1.05	1.31	1.04	1.00	1.16	0.84	0.97	0.96	0.91
Gd	0.010	4.07	5.26	4.09	3.40	4.90	2.75	3.29	2.95	3.13
Tb	0.010	0.73	0.98	0.72	0.66	0.87	0.54	0.60	0.52	0.55
Dy	0.010	4.11	5.50	4.05	3.86	5.00	3.21	3.41	2.91	2.95
Ho	0.010	0.83	1.13	0.80	0.84	1.00	0.67	0.71	0.61	0.59
Er	0.010	2.50	3.32	2.37	2.63	3.06	2.11	2.18	1.80	1.74
Tm	0.005	0.36	0.45	0.34	0.39	0.44	0.30	0.32	0.24	0.26
Yb	0.010	2.20	2.88	2.18	2.54	2.75	1.92	2.12	1.55	1.66
Lu	0.002	0.34	0.45	0.34	0.41	0.44	0.30	0.36	0.26	0.26
Total REE		127.2	153.2	148.9	94.9	159.4	59.7	118.7	92.4	103.9

**Total Fe as Fe_2O_3; D.L. = Detection limit.*

Major and trace elements by ICP-OES and ICP-MS

Twenty representative samples, 8 from the Kenseki Formation (3 sandstones and 5 mudstones), 4 from the Sasayama Group (3 mudstones and 1 sandstone), and 8 from the Wakino Subgroup (3 sandstones and 5 mudstones) were analyzed for major and trace elements at the Activation Laboratories Ltd., Ontario, Canada. Major elements were analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES) and trace elements and rare earth elements (REE) by inductively coupled plasma mass spectrometry (ICP-MS). Loss on ignition (LOI) was determined.

Samples were prepared and analyzed in a batch system. Each batch contains a method reagent blank, certified reference material and samples with 17% replicates. Samples were mixed with a flux of lithium metaborate and lithium tetraborate and fused in an induction furnace. The molten melt was immediately poured into a solution of 5% nitric acid containing an internal standard, and mixed continuously until completely dissolved (~30 minutes). The sample solution obtained was divided into two parts. One part was run for major oxides and the trace element Sc, on a combination simultaneous/sequential Thermo Jarrell-Ash ENVIRO II ICP. Detection limits are 0.01% for all the major elements and 2 ppm for Sc. Calibration was performed using 7 prepared USGS and Canmet certified reference materials. The other part of the sample solution was spiked with internal standards to cover the entire mass range, and was further diluted and introduced into a Perkin Elmer SCIEX ELAN 6000 ICP-MS using a proprietary sample introduction method. Representative major element, trace element, and REE data are shown in Table 3.

RESULTS AND OBSERVATIONS

The overall range of compositions for the sandstones and mudstones (i.e., siltstone and shale) from each of the three sedimentary basins is relatively narrow (Tables 2 and 3). The Kenseki sandstones show low to moderate SiO_2 concentrations (55–70%; on average 62%), TiO_2 concentrations averaging *ca*. 0.9%, Fe_2O_3 (i.e., total Fe as Fe_2O_3) + MgO concentrations of around 11%, Al_2O_3/SiO_2 ratios of around 0.22, and high and erratic K_2O/Na_2O ratios. The high K_2O/Na_2O ratios may be due to generally low and erratic concentrations of Na_2O. The reason for the erratic nature of Na_2O concentration is not clear, but may probably be related to its depletion in the source area by weathering or to its mobility at the depositional site. The moderately high contents of K_2O suggest that it is incorporated mainly in felsic volcanic fragments, since potassium-rich minerals like K-feldspars, muscovite and illite are generally poor in the sandstones. The Kenseki mudstones have compositions similar to those of the Kenseki sandstones, though the former shows a lower Fe_2O_3 + MgO content (around 9%). The sandstones and mudstones of the Sasayama Group are similar in composition to those of the Kenseki Formation but with slightly lower TiO_2 contents (on average 0.8%). The Wakino sandstones appear to be more mature than the others; i.e., the Wakino sandstones are characterized by higher SiO_2 contents (on average 74%), lower Fe_2O_3 + MgO contents (on average 6%), and lower Al_2O_3/SiO_2 ratios (around 0.17). The K_2O/Na_2O ratios of the Wakino sandstones, however, are mostly < 1 (on average 0.68) and less than those of the Kenseki and Sasayama sandstones. Compared to the Wakino sandstones, the Wakino mudstones have lower SiO_2 (on average 62%), higher Fe_2O_3 + MgO contents (on average 8%) and K_2O/Na_2O ratios of mostly > 1. Trace element and REE compositions are generally similar for all the sedimentary basins with few exceptions; e.g., the Kenseki and Sasayama sediments have elevated Cs, Cr and Ni concentrations.

Chemical classification

Various authors have devised classification schemes for sandstones based on their geochemical compositions (e.g., Pettijohn *et al*., 1972; Blatt *et al*., 1980; Crook, 1974). In the (Fe_2O_3* + MgO)-Na_2O-K_2O ternary diagram of

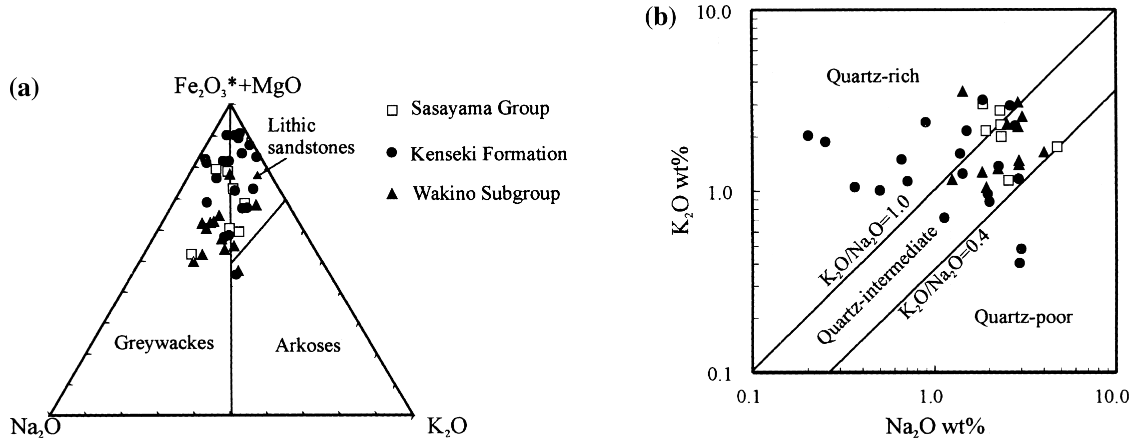

Fig. 3. Major element chemical classification of the sandstones and siltstones (diagrams from Blatt et al., 1980; Crook, 1974).

Blatt *et al.* (1980), the Wakino sandstones and siltstones plot in the greywacke field whereas the Kenseki sandstones and siltstones plot astride the greywacke and lithic sandstone fields (Fig. 3(a)). On the Na_2O-K_2O diagram, the Sasayama and Wakino data plot in the quartz-intermediate field whereas the data from the Kenseki Formation plot astride the quartz-rich and quartz-intermediate fields (Fig. 3(b)). The Kenseki sandstones, however, have lower quartz values (average of 17%) and SiO_2 contents (average of 62%) than those of typical quartz-rich sandstones (quartz values > 65%, average SiO_2 content: 89%; Crook, 1974).

Mineral controls on major and trace element distribution

The TiO_2 contents of the Wakino sediments show a strong correlation with the Al_2O_3 contents (Table 4). This correlation suggests that Ti is contained mainly in phyllosilicates (Dabard, 1990; Condie *et al.*, 1992). In contrast to the Wakino sediments, the TiO_2-Al_2O_3 correlations in the sediments from the Kenseki and Sasayama basin are poor, suggesting that TiO_2 may be concentrated in other minerals such as black mafic rock fragments and oxides (e.g., ilmenite, rutile, and sphene). There appears to be little or no positive correlation between Fe_2O_3 + MgO and Al_2O_3 contents of the Sasayama and Kenseki sediments (Table 4). Thus, this leads to the inference that both Fe and Mg contents are controlled by non-aluminous phases such as black mafic-ultramafic rock fragments and/or accessory oxide minerals. A weak to moderate correlation between Fe_2O_3 + MgO and Al_2O_3 in the Wakino sediments (Table 4) implies that both Fe and Mg are in part controlled by clay minerals and/or micas.

Clear positive correlations between K contents and the abundances of Al, Cs, Ba, total REE, Th and U in the Wakino sediments (Table 4) suggest that concentrations of these trace elements are controlled by clay minerals and mica (McLennan *et al.*, 1983). For the Kenseki and Sasayama sediments, the correlations between the K content and these trace elements are not as strong as those of the Wakino sediments, suggesting contributions from other minerals. Moderate correlation of Al_2O_3 with total REE in the Wakino sediments (Table 4) indicate that the REE are controlled in part by clay minerals and mica. Correlation of Al_2O_3 with total REE is weak in the Kenseki and Sasayama sediments (Table 4), suggesting that minerals other than clays and mica control their REE. For the Kenseki and Sasayama sediments, the correlations of Zr with total REE are moderately strong, but the Wakino sediments show weak correlations of Zr with total REE (Table 4). These facts may indicate that for the Kenseki and

Table 4. Linear correlation coefficients for selected elements of the Lower Cretaceous sediments

	Kenseki Formation	Sasayama Group	Wakino Subgroup
K_2O-Al_2O_3	0.67	0.71	0.80
K_2O-Ba	0.81	0.95	0.86
K_2O-Cs	0.49	0.82	0.76
K_2O-total REE	0.44	–0.26	0.68
K_2O-Th	0.56	0.54	0.64
K_2O-U	0.55	0.48	0.79
P_2O-Cs	–0.57	–0.38	–0.14
Th-Cs	0.19	0.83	0.11
Al_2O_3-Fe_2O_3+Mg	–0.39	–0.58	0.41
Al_2O_3-TiO_2	–0.18	0.34	0.91
Th-total REE	0.87	0.67	0.83
Al_2O_3-total REE	–0.19	0.19	0.65
Zr-total REE	0.68	0.80	0.35
La-Al_2O_3	–0.15	0.42	0.53
Yb-Zr	0.77	0.98	0.39
Yb-Al_2O_3	0.29	–0.43	0.61
La-Zr	0.68	0.58	0.29
Zr-Eu/Eu*	–0.42	0.14	0.01
TiO_2-Al_2O_3	–0.18	0.34	0.91
Fe_2O_3-Al_2O_3	–0.28	0.16	0.55
Sc-Al_2O_3	–0.31	–0.27	0.70
Ni-Al_2O_3	–0.44	0.02	–0.06
Co-Al_2O_3	–0.54	–0.19	0.30
Cr-Ni	0.94	0.97	0.77
Cr-Co	0.88	0.95	–0.17
Ni-Co	0.97	0.97	0.02

Sasayama sediments, zircon is more important than clay/mica in housing all REE whereas for the Wakino sediments clay and mica are more important than zircon in housing REE (Condie *et al.*, 1992). A moderate positive correlation of Th with total REE for the Lower Cretaceous sediments may indicate allanite-monazite control on REE (Condie *et al.*, 1992).

Large-ion-lithophile (LIL) elements (e.g., K, Rb, Cs, Sr, and U), the REE, and high field strength elements (e.g., Zr, Nb, Hf, Y) generally show consistent inter-relationships. Also, ferromagnesian trace elements (Cr, Ni, V, Co, and Sc) in the Kenseki and Sasayama sediments show strong interrelationships. These trace element relationships illustrate the chemical coherence and uniformity of the sediments from the three Lower Cretaceous basins on the whole, and those of the Kenseki and Sasayama sediments in particular. The Co and Sc in the Kenseki and Sasayama sediments show a strong positive correlation with Cr, Ni, and V but no correlation with Al_2O_3, whereas Co and Sc in the Wakino sediments show no correlation with Ni, Cr, and V but show a moderate correlation with Al_2O_3. We thus infer that for the Wakino sediments Co and Sc are chiefly controlled by chlorite, but, on the other hand, for the Sasayama and Kenseki sediments Co and Sc are concentrated in oxides and other accessory non-aluminous silicate minerals.

DISCUSSIONS

Sorting and weathering effects

The Th/U ratio in most upper crustal rocks is typically between 3.5 and 4.0 (McLennan *et al.*, 1993). In most cases, weathering and sedimentary recycling typically result in loss of U, leading to an elevation in the Th/U ratio. In sedimentary rocks, Th/U values higher than 4.0 may indicate

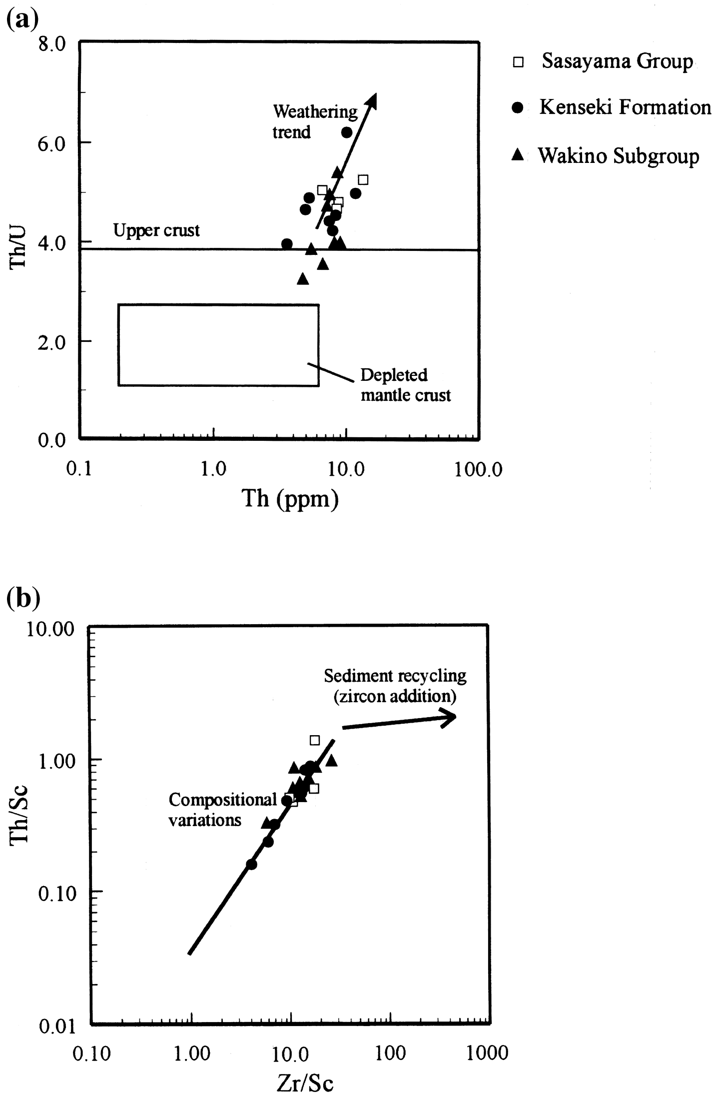

Fig. 4. Plots of (a) Th/U versus Th and (b) Th/Sc versus Zr/Sc for the Lower Cretaceous sediments (after McLennan et al., 1993).

intense weathering in source areas or sediment recycling (i.e., derivation from older sedimentary rocks). The Th/U versus Th plot for the Lower Cretaceous sediments (Fig. 4(a)) shows a typical distribution similar to the average values of fine-grained sedimentary rocks reported by Taylor and McLennan (1985) and follows the normal weathering trend (McLennan *et al.*, 1993). It follows from this result that the sources for the Lower Cretaceous sediments were recycled sediments and/or might have undergone some degree of weathering.

The sedimentary sorting and recycling can be monitored by a plot of Th/Sc against Zr/Sc (McLennan *et al.*, 1993). First-order sediments show a simple positive correlation between these ratios (i.e., Th/Sc and Zr/Sc) whereas recycled sediments show a substantial increase in Zr/Sc with far less increase in Th/Sc. On the Th/Sc versus Zr/Sc diagram, the Lower Cretaceous

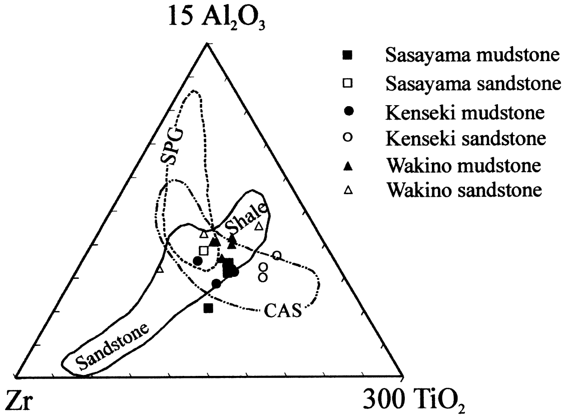

Fig. 5. Al-Ti-Zr plot for the Lower Cretaceous sediments. The solid contour refers to the observed range of compositions in clastic sediments. CAS refers to the fields of calc-alkaline suites and SPG refers to fields of strongly peraluminous granites (after Garcia et al., 1994).

sediments follow a general trend consistent with their direct derivation from igneous rocks (Fig. 4(b)). It can be, therefore, inferred from Figs. 4 (a) and (b) that the bulk of the Lower Cretaceous sediments were directly derived from igneous rocks that had undergone some degree of weathering.

The chemical index of alteration (CIA) defined as: CIA = $100 \times Al_2O_3/(Al_2O_3 + CaO + Na_2O + K_2O)$ has been established as a general guide to the degree of weathering in the provenance regions (Nesbitt and Young, 1982). High values (i.e., 76–100) indicate intensive chemical weathering in the source areas whereas low values (i.e., 50 or less) indicate unweathered source areas. The Kenseki, Sasayama, and Wakino mudstones with LOI values lower than 10% have CIA values of 57–70 (average of 65), 56–65 (average of 63), and 57–75 (average of 66), respectively. The reported CaO content includes non-silicate Ca; therefore, the CIA values reported are likely to represent minima. The CIA values suggest moderate weathering at the source areas of these sediments; however, the CIA values should be interpreted with caution as it relies heavily on the elements that are readily mobilized during transport and burial (i.e., alkali elements). The Rb/Sr ratios of sediments also monitor the degree of source-rock weathering (McLennan *et al.*, 1993). The Kenseki, Sasayama, and Wakino sediments have average Rb/Sr ratios of 0.40, 0.37 and 0.34, respectively, and these values are close to that of the average upper continental crust (0.32) but significantly lower than the average post-Archean Australian shale (0.80; McLennan *et al.*, 1983). This suggests that the degree of source area weathering was most probably moderate rather than intense.

The Al-Ti-Zr ternary diagram monitors the effects of sorting processes (Garcia *et al.*, 1994). On this diagram, mature sediments consisting of both sandstones and shales show a wide range of TiO_2/Zr variations whereas immature sediments of sandstones and shales show a more limited range of TiO_2/Zr variations. On the Al-Ti-Zr diagram, the Lower Cretaceous sediments are confined in the center with a limited range of TiO_2/Zr variations, suggesting poor sorting and rapid deposition of the sediments (Fig. 5).

Source rock compositions

The Lower Cretaceous sediments have high K_2O and Rb concentrations and a uniform K/Rb ratio of 260 that lies close to a typical differentiated magmatic suite or “main trend” with a ratio of 230 (Fig. 6(a); Shaw, 1968). This feature emphasizes the chemically coherent nature of the sediments and derivation mainly from acidic to intermediate rocks. The K-Rb coherence, however, could also result from their redistribution by hydrothermal fluids during regional metamorphism and metasomatism (Floyd and Leveridge, 1987). The Kenseki Formation and the Sasayama Group are unmetamorphosed but the Wakino Subgroup is weakly metamorphosed. Nevertheless, the elemental redistribution is unlikely in the light of element ratios that are unaffected by element mobility (Floyd and Leveridge, 1987).

A plot of La/Th against Hf (Fig. 6(b)) provides a useful tool for bulk rock discrimination between different arc compositions and sources. Felsic composition-dominated arcs have low and uniform La/Th ratios (less than 5) and Hf contents of about

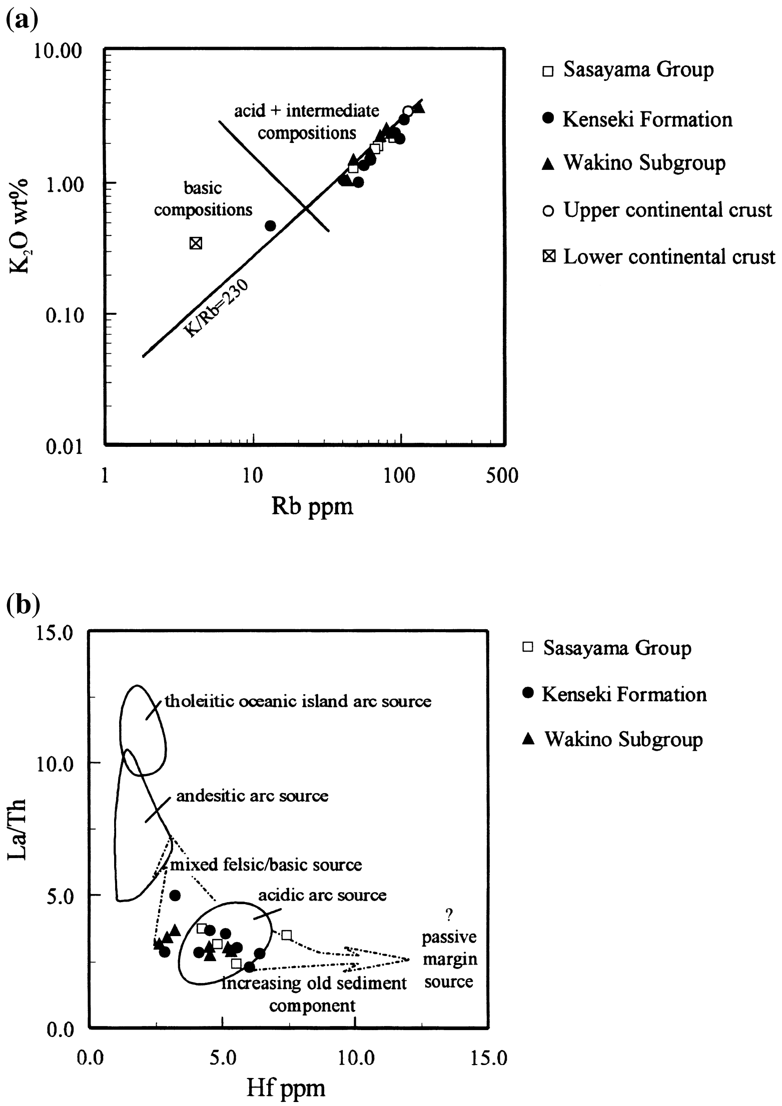

Fig. 6. (a) Distribution of K and Rb in the Lower Cretaceous sediments relative to a K/Rb ratio of 230 (=main trend of Shaw, 1968). Average Upper and Lower Continental Crust from Taylor and McLennan (1985). (b) Plot of La/Th versus Hf for the Lower Cretaceous sediments (compositional fields are after Floyd and Leveridge, 1987).

3–7 ppm. With the progressive unroofing of the arc and/or incorporation of sedimentary basement rocks, the Hf content increases via the release of zircon (Floyd and Leveridge, 1987). The compositions of the Lower Cretaceous sediments suggest derivation mainly from felsic igneous rocks with minor mafic input (Fig. 6(b)).

The chondrite-normalized REE distribution patterns are about the same for the three studied areas and are similar to that of the average Post-Archean Australian Shale (PAAS; Taylor and McLennan, 1985). The predominantly felsic composition of the Lower Cretaceous sediments is supported by the REE plots which show enriched light REE, negative Eu anomaly and flat or uniform heavy REE (Fig. 7).

Very high levels of Cr and Ni have been used by various authors (e.g., Hiscott, 1984; Wrafter

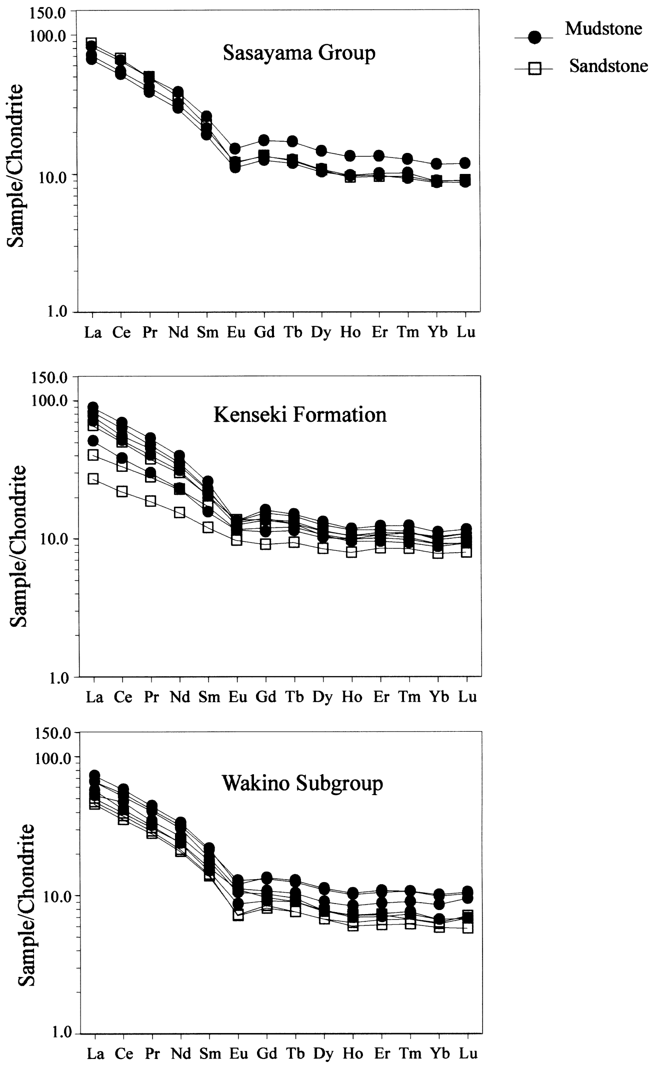

Fig. 7. Chondrite normalized REE diagrams for the Lower Cretaceous sediments (normalizing factors from Taylor and McLennan, 1985).

and Graham, 1989) to infer an ultramafic provenance for sediments. Graver *et al*. (1994) have suggested that (1) elevated concentrations of Cr and Ni, (2) a high correlation coefficient between Cr and Ni and corresponding high correlation of both elements with Co, and (3) a Cr/Ni ratio of about 1.2 to 1.5 can be used to infer an ultramafic source, and therefore presumably ophiolites, in sediments. Higher Cr/Ni ratios probably indicate derivation of these elements from mafic volcanic rocks (Garver and Scott, 1995). The Sasayama and Kenseki sediments have high Cr (69–359 and 67–597 ppm, respectively) and Ni (47–288 and 25–395 ppm, respectively) concentrations, Cr/Ni ratio of 1.42 and 1.46, respectively, and a high correlation coefficient between Cr and Ni and corresponding high correlation of both elements with Co (Table 4). These relationships indicate a significant contribution of detritus from ophiolitic rocks. The Wakino sediments have low levels of Cr (45–83 ppm) and Ni (11–45 ppm), Cr/Ni ratio of 2.66, a medium, positive correlation coefficient

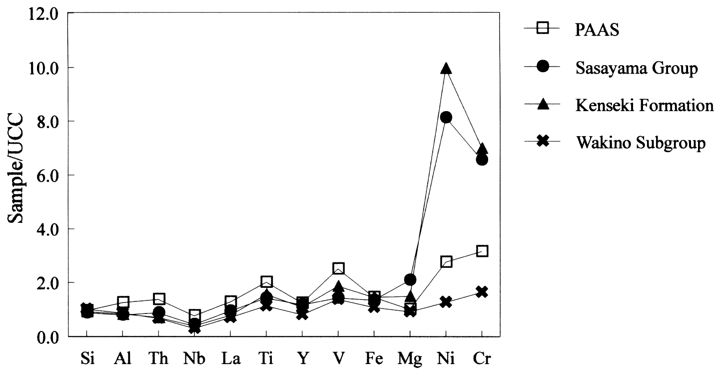

Fig. 8. Multi-element diagram of the average Lower Cretaceous sediments normalized to average Upper Continental Crust (UCC). The elements are arranged in such a way that those elements that are derived mainly from felsic source rocks are plotted on the left-hand side and those from mafic and ultramafic source rocks are plotted on the right-hand side. The average post-Archean Australian Shales (PAAS) and UCC data are from Taylor and McLennan (1985).

Table 5. Geochemical characteristics of sediment derived from different provenance types (after McLennan et al., 1993; Girty et al., 1996)

Provenance type	Eu/Eu*	Th/Sc	Th/U	Others
Old Upper Continental Crust	0.6–1.1	~1.0	>3.8	Evolved major element compositions (e.g., high Si/Al, CIA); high LILE abundances; uniform compositions
Recycled Sedimentary Rocks	0.6–1.1	≥1.0	>3.8	Evidence of heavy mineral concentration from trace elements (e.g., Zr, Hf for zircon)
Young Differentiated Arc	0.5–0.9	~1.0 to <0.01	<3.0	Evolved major element compositions (e.g., high Si/Al, CIA); high LILE abundances; variable compositions
Young Undifferentiated Arc	~1.0	~1.0 to <0.01	<3.0	Unevolved major element compositions (e.g., low Si/Al, CIA); low LILE abundances; variable compositions

of Cr-Ni, and no correlation among Cr, Ni and Co. This may suggest either a minor amount of ultramafic input into the depositional system or else that trace elements could have traveled into the depositional basin as adsorbed ions on clays (McCann, 1991). Vanadium concentrations (48–110 ppm) are relatively higher than the levels commonly recorded in sediments (about 20 ppm) and given that V is concentrated in mafic rocks, they suggest some mafic input into the depositional system (McCann, 1991).

Provenance

Figure 8 shows a multi-element diagram of the Lower Cretaceous sediments normalized to the average upper continental crust UCC (Taylor and McLennan, 1985). The figure shows that, with exception of the high Cr and Ni values for the Kenseki and Sasayama samples, the Lower Cretaceous sediments have compositions similar to those of the average UCC and PAAS. This feature indicates that the sediments were derived mainly from the upper continental crust. As discussed earlier, the high Cr and Ni values for the Kenseki and Sasayama sediments reflect a significant contribution from ultramafic rocks.

McLennan *et al.* (1993) have defined five distinct provenance components on the basis of geochemical compositions. These components include the following: Old Upper Continental Crust (OUC), Recycled Sedimentary Rocks (RSR), Young Undifferentiated Arc (YUA), Young Differentiated Arc (YDA) and Exotic components. The Old Upper Continental Crust components

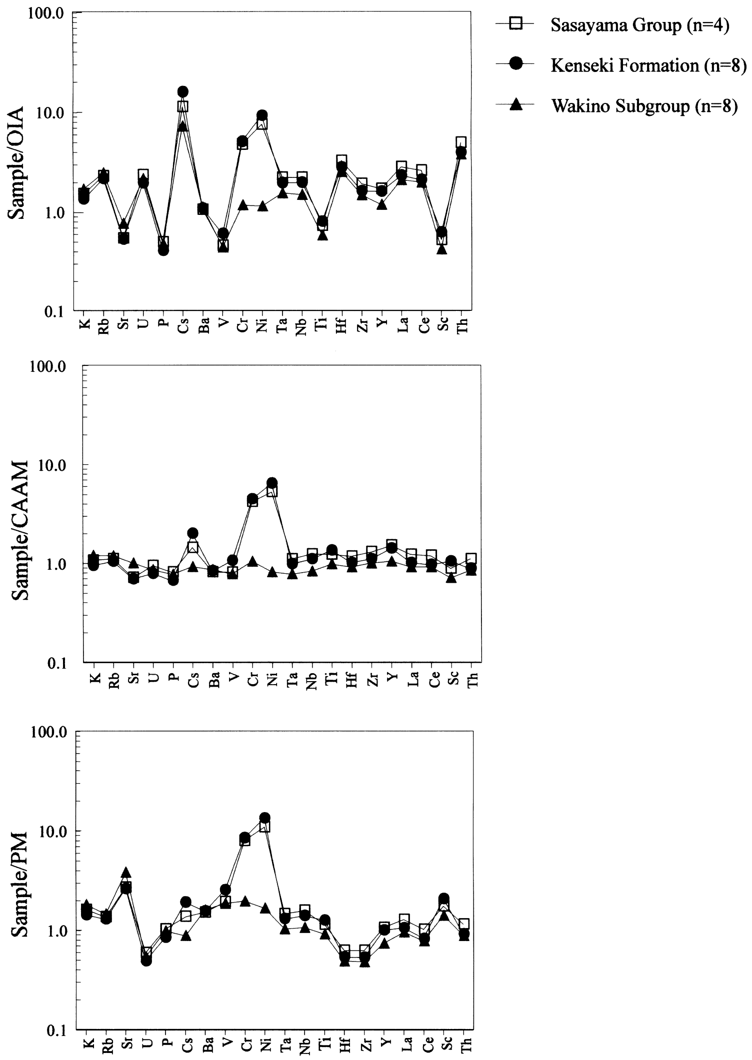

Fig. 9. Multi-element patterns for the Lower Cretaceous sediments. Data are normalized to the passive margin (PM), continental arc + active margin (CAAM), and oceanic island arc (OIA). Standards from Floyd et al. (1991).

constitute the old, well-differentiated upper continental crust that is characterized by a substantial Eu anomaly. The Recycled Sedimentary Rocks component includes recycled sedimentary and metasedimentary rocks. The Young Undifferentiated Arc component represents the young (dominantly mantle derived) igneous arc material (volcanic or plutonic) that has not undergone significant intracrustal differentiation (i.e., it has not undergone plagioclase fractionation and therefore show no Eu anomalies). The Young Differentiated Arc provenance component constitutes the young (mantle derived) volcanic or plutonic igneous rocks from island and continental arcs that have undergone significant intracrustal differentiation. The general geochemical characteristics of sediments derived from these four provenance types are summarized in Table 5.

The chemical characteristics of the Lower Cretaceous sediments, i.e., fairly uniform compositions, high Th/U ratios (>3.0; Fig. 4), negative Eu anomalies (Eu/Eu* 0.67–0.93; Fig. 7) and Th/Sc

Table 6. Rare earth element calculated elemental ratios and anomalies

		La_N/Yb_N	La_N/Sm_N	Eu/Eu*	Ce/Ce*
Wakino Subgroup					
WYL-2	S	7.98	2.08	0.89	1.00
WYM-3	M	7.82	2.18	0.78	1.11
WYS-1	M	7.68	3.49	0.78	1.00
WYS-4	M	6.49	3.08	0.77	1.04
WKL-1	S	7.78	1.89	0.67	0.99
WKL-2	S	7.40	1.96	0.67	1.01
WKS-3	M	7.37	3.33	0.70	1.02
WKS-4	M	8.51	3.80	0.74	0.98
Average		7.63	2.73	0.75	1.02
Kenseki Formation					
KNS-4	M	7.05	3.47	0.75	0.96
KNS-5	M	8.01	3.45	0.66	1.00
KHS-2	M	7.85	3.41	0.71	0.95
KHM-1	M	9.49	3.42	0.73	1.00
KHIROM-3	M	4.98	2.12	0.87	0.98
KNL-6	S	3.48	1.12	0.93	0.97
KNL-7	S	4.41	1.67	0.81	1.00
KNL-8	S	7.21	2.73	0.81	1.00
Average		6.56	2.68	0.78	0.98
Sasayama Group					
SSS-3	M	7.06	3.21	0.72	1.02
SSM-1	M	7.67	2.76	0.72	1.02
SSM-2	M	8.02	2.96	0.72	0.99
SSL-4	S	9.86	3.61	0.69	1.02
Average		8.15	3.13	0.71	1.01
PAAS		9.15	4.33	0.65	1.02
UCC		9.21	3.40	0.65	1.03

Ratios determined from chondrite-normalized values using equations in McLennan (1989). PAAS (post-Archean shales) and UCC (upper continental crust) data from McLennan (1989). M, mudrocks; S, sandstones.

ratios (mostly less than 1.0; Fig. 4), favor the OUC provenance for the Lower Cretaceous sediments. The SiO_2/Al_2O_3, Th/Sc (mostly < 1) and La/Sc (<4.0) ratios are, however, slightly lower than typical OUC, and these ratios may suggest a minor contribution of young arc-derived material.

Tectonic environment

Major elements and immobile trace elements are useful for tectonic setting determination. Useful major element discriminating plots include a SiO_2 versus K_2O/Na_2O diagram (Roser and Korsch, 1986) and, useful trace element plots include La-Sc-Th and La-Th-Zr triangular diagrams (Bhatia and Crook, 1986). Although these discrimination diagrams have been utilized with some success, Floyd *et al*. (1991) have indicated that the diagrams that rely on only few diagnostic elemental ratios may be adversely affected by sorting, heavy mineral contents and proportions of mafic input in such a way that sedimentary series spread across a number of geologically unrelated tectonic fields. They, therefore, proposed the use of the full range of elemental compositions for tectonic setting discrimination. The advantage here is that such a compositional plot will show the effect of variable mafic and heavy mineral inputs within the sedimentary suite and in addition provide a pointer to the tectonic environment.

Figure 9 shows the averages of the data from each sedimentary basin normalized to the average composition of greywackes from different tectonic environments (after Floyd *et al*., 1991). Apart from elevated Cs, Cr, and Ni abundances for the Kenseki and Sasayama sediments, the patterns are closest to those of continental arc/active margin tectonic environment (Fig. 9). The elevated Cs abundances reflect their derivation from felsic igneous rocks. The high abundances of Cr and Ni in the Sasayama and Kenseki sediments indicate a significant addition of ultramafic component.

A number of studies have used the REE distribution in sediments to determine or infer their plate tectonic settings (e.g., Bhatia and Crook, 1986; McLennan *et al*., 1990; McLennan and Taylor, 1991; Girty and Barber, 1993). These studies show that sediments deposited in the continental margin are characterized by LREE enrichment (indicated by high La/Sm) and high total rare earth elements (∑REE), but, on the other hand, those from young, undifferentiated oceanic arcs have lower La/Sm, lower ∑REE, and lack an Eu anomaly. In general, therefore, the REE patterns of sediments deposited in continental margins can be differentiated from those derived from undifferentiated oceanic arcs. Continental margins can be classified into passive and active types. Passive margin provenance is characterized by REE patterns being uniform and similar to PAAS (McLennan, 1989; Bhatia, 1985). Sediments deposited at active continental margins generally show a REE pattern intermediate between a “typical andesite pattern” and PAAS or in some cases indistinguishable from PAAS itself (McLennan, 1989). Thus, most active continental margin sediments display intermediate REE abundances, variable LREE enrichments and variable negative Eu-anomalies, with Eu/Eu* in the range of 0.6–1.0 (McLennan, 1989). Compared to the average PAAS, the Lower Cretaceous sediments have relatively lower LREE (i.e., La_N/Sm_N), lower La_N/Yb_N ratios and higher Eu/Eu* ratios (Table 6), suggesting deposition in an active continental margin rather than passive margin tectonic environment (McLennan, 1989).

SYNTHESIS AND CONCLUSIONS

The major, trace, and REE data presented in the previous sections suggest an active continental margin environment for the Lower Cretaceous sediments. Active margin sediments typically have the Young Undifferentiated Arc (YUA) and/or Young Differentiated Arc (YDA) provenance, whereas passive margin sediments typically have the Old Upper Continental Crust (OUC) provenance (McLennan *et al*., 1990, 1993). Contrary to this general tendency, however, the chemical compositions of the Lower Cretaceous sediments suggest their derivation dominantly from the Old Upper Continental Crust with minor amounts of young arc-derived detritus. These observations thus place constraints on the nature of plate boundary in Southwest Japan during the early Cretaceous. If the nature of plate boundary was the normal subduction type, it will be expected that the magmatic arc developed at the plate margin would supply the bulk of the detritus in the adjacent depositional basin, and hence that the derived sediments would contain a substantial amount of young magmatic arc component (i.e., YUA and/or YDA). Apparently, this does not apply to the present case since YUA and/or YDA components contribute only in minor amounts to the Lower Cretaceous sediments. Okada and Sakai (1993) have indicated that the Kanmon basin began to form at the time of oblique subduction of the Izanagi plate beneath the eastern margin of the Asian continent. If this is the case, then a minor contribution of YUA and/or YDA components to the Lower Cretaceous sediments with the dominantly OUC provenance suggests that the oblique subduction of the Izanagi plate during the early Cretaceous was not so pronounced. This interpretation is in agreement with Takahashi (1983) and Sakai and Okada (1997), who have suggested that the nature of plate boundary during the early Cretaceous was oblique-slip type and that plate subduction and arc magmatism occurred in Southwest Japan during the Late Cretaceous. It is, however, also possible that the plate margin was the normal subduction type, but the depositional basins were located far from the plate margin and were in close proximity to the ancient Asian continent. Such a depositional basin (e.g., the Sea of Japan) may be dominated by the OUC component (McLennan *et al*., 1990).

Acknowledgments—This work was undertaken by D.K.A. as a part of his Ph.D. Project financed by a graduate fellowship of the Japanese Ministry of Education, Science, Culture and Sports (Monbusho). We thank M. Usui, T. Saito and N. Ando for their technical assistance. We also thank K. Yamamoto, K. Sugitani and H. Shimizu for thoughtful and constructive reviews that helped improve the original version of the manuscript.

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