# A possible mechanism of rhythmic alternation of bedded cherts revealed by their chemical composition

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#### **ABSTRACT**

Detailed analyses for major, minor, and rare earth elements in bedded cherts from the Shimanto Terrane, Southwest Japan, show that bedded cherts just above a basalt are dominated by hydrothermal (Fe + Mn) component. The mixing ratio of clastic materials and hydrothermal emanations in the cherts and shale partings suggests that the sedimentation rate of clastics controlled the formation of rhythmic alternation of the bedded cherts.

#### INTRODUCTION

Many geologists have emphasized that the depositional environment of bedded cherts in orogenic belts, especially those associated with greenstones, is deep pelagic and that the cherts are accreted to island arc regions (e.g. Taira et al., 1988). Geochemical data inconsistent with this view, however, have accumulated: Sugisaki et al. (1982) and Yamamoto (1983) proposed that the TiO<sub>2</sub>-normalized values of hydrogeneous elements, such as Mn, Ni and Co, are a useful tool to identify the depositional environment of siliceous sedimentary rocks. Using the TiO<sub>3</sub>-normalized values as a criterion, they concluded that the Triassic bedded cherts in the Mino Terrane, central Japan, are not pelagic. Similar conclusion was also given by the rare earth element (REE) study of Shimizu and Masuda (1977); they showed that Ce anomaly in REE pattern of cherts is an environmental indication of their formation and regarded the Triassic bedded cherts in the Mino Terrane as being of hemipelagic origin. Moreover, Shimizu et al. (1991) reported that La-Ce and Sm-Nd systematics are useful for the study of sedimentary environments and the Mino bedded cherts were formed in a hemipelagic region.

The formation mechanism of rhythmic alternation of chert beds and thin shale partings has not satisfactorily been explained. Since the main components of bedded chert are siliceous organisms and clastic materials (Sugisaki et al., 1982), the rhythmicity of bedded cherts could have been due to rhythmic changes of either the sedimentation rate of siliceous organisms or clastic materials, or to diagenetic segregation (e.g. McBride and Folk, 1979). Chemical data of bedded cherts show mixing ratios of siliceous organisms and clastic materials but cannot be critically used to determine formative mechanism. In contrast, data from

bedded cherts that rest on basalt and consist of hydrothermal, biogenic, and clastic components may indicate their formative mechanism.

In this study, I have analyzed major and minor elements, including rare earths, in cherts and shale partings from the Shimanto Terrane in the Southwest Japan and examined the process of rhythmic alternation of bedded cherts.

## SAMPLE AND ANALYTICAL METHOD

The samples were collected from a bedded chert outcrop in Cretaceous Shimanto Terrane, western part of Kii Peninsula, Japan (Fig. 1). Three pairs of a chert and

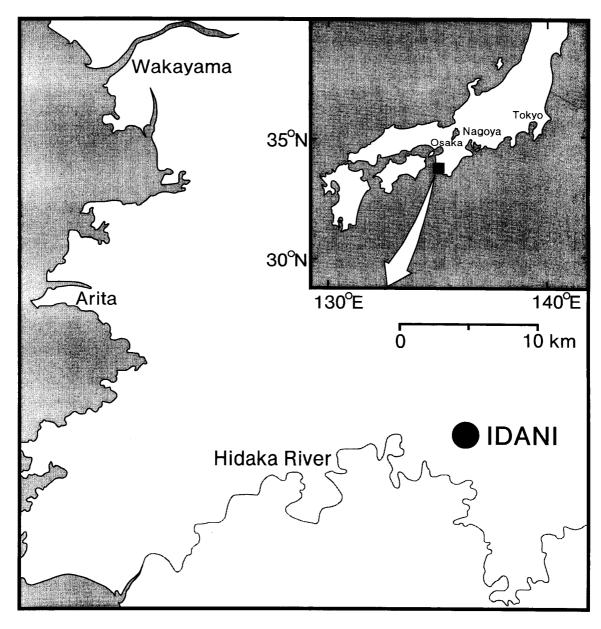


Fig. 1. Sampling location of chert-shale pairs from the Shimanto terrane, Southwest Japan.

an adjacent shale parting were collected from different horizons above basal basalts. The basal basalt just below the bedded cherts was also sampled. REE of a recent radiolarian sample and a hydrothermal chert were also analyzed for reference. The radiolarians were separated from radiolarian ooze collected from the central Pacific Ocean during the Cruise of Hakurei, Geological Survey of Japan. The hydrothermal chert is an akashiro silica-stone just above pillow basalt in the Franciscan terrane (sample No.10 in Yamamoto, 1987).

Analytical procedure for major and minor elements is the same as that described by Yamamoto (1987). Rare earth elements (REE) were analyzed by the isotope dilution mass spectrometry (IDMS) using JEOL JMS 05RB according to the method of Nakamura (1974).

The analytical data (Table 1) were recalculated on water-, carbonate-, and "others"-free basis. Their averages are listed in Table 2 with those of bedded cherts from the Shimanto and the Mino Terranes.

Table 1. Analytical results for bedded cherts and basal basalt from the Shimanto Terrane. Major elements are in %, and minor and rare earth elements in ppm. Residual materials are calculated by subtracting  $\rm H_2O$  from ignition loss and may contain sulfur, organic materials and others. Major and minor element compositions of recent radiolaria and hydrothermal chert (\*) are quoted from Yamamoto (1983) and Yamamoto (1987), respectively.

Sample No.	1	2	3	4	5	6	7	8		
Lithology	Chert	Shale	Chert	Shale	Chert	Shale	Basalt	Basalt	Recent* Radiolaria	Hydrothermal* Chert
SiO,	91.83	66.27	94.85	79.56	92.90	80.77	50.08	50.64	95.50	88.70
$ ext{TiO}_2^2 \\  ext{Al}_2  ext{O}_3 \\  ext{FeO}  ext{}$	0.090	0.63	0.044	0.26	0.046	0.31	2.18	2.29	tr.	0.02
$Al_{3}O_{3}$	1.57	11.92	0.53	3.67	0.88	4.98	11.97	12.63	1.18	0.18
FeO	0.76	0.84	0.16	0.99	0.41	0.10	7.88	6.68	tr.	0.16
Fe,O,	1.60	6.50	2.01	6.11	1.90	2.40	4.49	6.13	0.002	9.22
MnO"	0.37	0.40	0.28	0.55	0.72	0.78	0.26	1.03	0.007	0.07
MgO	1.10	2.80	0.78	1.90	1.30	2.21	6.23	6.39	0.11	tr.
CaO	0.39	1.58	0.13	0.74	0.19	0.31	8.04	5.90	0.42	0.09
Na <sub>a</sub> O	0.55	1.65	0.38	0.98	0.60	0.66	3.51	2.89	0.59	tr.
K,Ő	0.20	1.58	0.09	0.36	0.05	0.48	0.08	0.36	0.26	0.06
$P_2^2O_5$	0.083	0.22	0.032	0.23	0.040	0.067	0.18	0.21	0.030	0.01
H <sub>0</sub> O(~)	0.72	1.53	0.49	1.01	0.86	1.87	0.71	1.00		0.23
H <sub>2</sub> O(+)	0.72	1.76	0.67	1.59	1.03	1.40	1.94	2.03		0.14
Residuals	0.53	1.14	0.48	0.44	0.14	0.26	0.83	0.25		1.19
Cr	1	9	tr.	4	7	13	68	46	1	
Co	10	7	11	16	10	15	29	28	5	5
Ni	20	21	18	58	24	31	48	51	24	7
Cu	91	51	3	47	31	55	68	79	tr.	
Zn	80	140	70	140	82	99	130	160	31	770
Rb	10	52	6	15	7	20	3	14	4	tr.
Sr	39	230	48	61	41	53	230	140	31	770
Y	10	19	9	31	8	14	52	55	tr.	1
Zr	23	76	9	45	6	63	120	140	4	7
Nb	tr.	1	6	5	tr.	tr.	5	9	1	2
Mo	8	3	5	3	3	6	9	tr.	8	4
Pb	tr.	4	2	14	tr.	9	6	1	3	53
Ba	86	510	39	120	690	250	86	501		250
Ga	4	15	4	7	2	6	16	15		tr.
La	11.14	15.13	7.813	30.13			4.363	6.446	1.809	2.050
Ce	13.67	26.00	6.096	24.47			13.77	16.71	0.447	1.557
Nd	13.08	20.93	9.234	32.46			13.47	14.79	2.255	2.496
Sm	2.787	4.792	2.036	6.948			4.751	4.989	0.537	0.607
Eu	0.666	1.337	0.489	1.655			1.611	1.721	0.138	0.480
Gd	2.647	4.721	2.075	6.987			6.676	7.050	0.601	0.740
Dy	2.313	4.455	1.762	6.110			8.139	8.598	0.595	0.862
Er	1.261	2.236	0.948	3.285			5.148	5.501	0.355	0.547
Yb	1.054	1.988	0.753	2.699			4.871	5.217	0.346	0.611
Lu	0.149	0.295	0.106	0.381			0.731	0.772	0.0681	0.0982
	0.130	0.200	0.100	0.001			0.701	0.112	0.0001	0.0002

Table 2. Averaged chemical composition of cherts and shales from the Shimanto, Franciscan (Yamamoto, 1987), and Mino Terranes (Yamamoto, 1983) on the water and others free basis. Elements are in ppm and oxides in %.

	Shin	nanto	Franc	csican	Mino		
	$\overline{ ext{Chert}}$	Shale	$\overline{ ext{Chert}}$	Shale	Chert	Shale	
$SiO_2$	94.12	79.58	92.63	56.95	95.19	63.96	
$TiO_2$	0.06	0.42	0.09	1.21	0.10	0.75	
$\text{Al}_{2}\hat{\text{O}}_{3}$	1.00	7.23	1.41	12.15	1.97	15.97	
$Al_2O_3$ $Fe_2O_3^*$	2.36	6.60	2.97	15.82	1.19	6.21	
MnO	0.46	0.61	0.80	2.27	0.03	0.26	
MgO	1.07	2.43	0.33	3.03	0.53	2.47	
CaO	0.24	0.93	0.11	1.59	0.38	0.26	
Na <sub>2</sub> O	0.51	1.16	0.16	0.59	0.09	0.31	
$\mathbf{K}_{2}\mathbf{\mathring{O}}$	0.11	0.85	0.42	3.87	0.51	4.68	
$P_{0}^{2}O_{5}$	0.05	0.18	0.03	0.14	0.06	0.27	
$P_2^{^{\prime}}O_5$ FeO	0.35	0.68	0.26	1.26	0.50	2.35	
$\mathrm{Fe_2O_3}$	1.86	5.85	2.67	14.69	0.63	3.62	
Cr	4	8	10	90	40	68	
Co	11	13	4	18	3	17	
Ni	21	39	16	70	14	66	
Zn	78	130	43	160	30	160	
Rb	8	31	9	110	29	210	
$\operatorname{Sr}$	43	120	25	41	39	53	
Y	9	22	9	63	7	38	
$\mathbf{Zr}$	13	65	27	180	23	160	
Nb	2	2	3	13	3	14	
Mo	7	3	2	4	9	9	
Ga	3	10	2	19	3	25	

<sup>\*</sup> Total iron as  $Fe_2O_3$ 

## RESULTS AND DISCUSSION

## Major and minor element features

Bedded cherts in the Shimanto and the Franciscan Terranes are accompanied by basalt whereas those in the Mino terrane are accompanied by clastic rocks. Chemical compositions of shales are important for estimating lithogenic portions of the cherts (Sugisaki et al., 1982; Yamamoto, 1983). Shale partings from the Shimanto terrane contain more SiO<sub>2</sub>, and less TiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, than do those from other terranes (Table 2); this may be due to a higher proportion of radiolarian silica relative to clastics. Shimanto shales have higher Na<sub>2</sub>O/K<sub>2</sub>O and lower Cr contents than Franciscan and Mino shales. These differences may be attributed to the compositional difference of the clastic materials in them.

In spite of the dilution by siliceous organisms, Shimanto cherts and shale partings are more enriched in total iron as  $Fe_2O_3$  (hereafter  $Fe_2O_3^*$ ) and MnO than are Mino shale partings. This feature is also observed in Franciscan shale partings and is regarded as a general character of shales resting on basalt (Yamamoto, 1987).

## Rare earth element features

Cherts and shale partings from the Shimanto Terrane, hydrothermal chert in the Franciscan terrane (Yamamoto, 1987), and recent radiolarians (Yamamoto, 1983) were examined on the basis of their REE (Fig. 2).

Radiolarians, a component of chert, have light REE (LREE)-enriched pattern and show a large negative Ce anomaly of 0.11. The Ce anomaly is defined as the ratio of observed abundance to that which would fall on the La-Nd join in the REE pattern. This REE pattern of a Recent radiolarians is similar to those reported by Shimizu and Masuda (1977), although the absolute REE abundances are variable.

The hydrothermal chert from the Franciscan terrane also shows LREE-en-

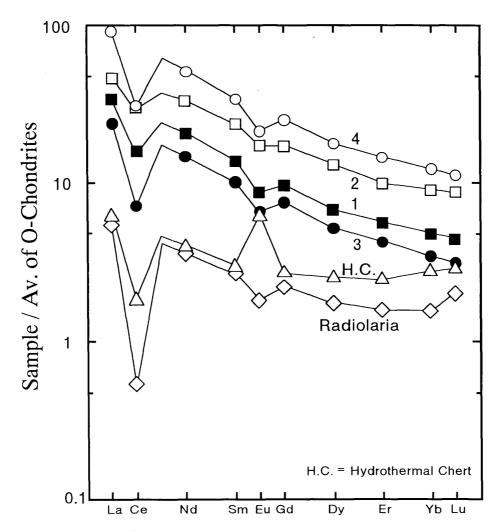


Fig. 2. Abundance patterns for Shimanto bedded cherts, recent radiolarian, and hydrothermal chert normalized to the average of ordinary chondrites (Nakamura, 1974). Sample symbols: pair of open and closed symbols represents paring shale and chert from the Shimanto Terrane, respectively; open triangle = hydrothermal chert from the Franciscan Terrane; open diamond = recent radiolarian from the central Pacific Ocean. Numerics show the sample number.

riched pattern with a large negative Ce anomaly (0.34). Whereas hydrothermal water generally does not show a Ce anomaly (Michard et al., 1983), the Ce anomaly observed in the hydrothermal chert is attributed to scavenging REE from sea water. The scavenging process is due to absorption on Mn and Fe hydroxide derived from hydrothermal emanation (e.g. Ruhlin and Owen, 1986). On the other hand, this sample has a positive Eu anomaly; this may have been derived from hydrothermal water that normally shows positive Eu anomaly of about 10 (e.g. Michard and Albarede, 1986). Thus the presence of both anomalies shows that the hydrothermal chert has acquired its REE composition not only from sea water but also from hydrothermal water, although the magnitude of these contributions cannot be estimated because the REE abundances of these components fluctuate widely.

Two sets of REE patterns of the Shimanto shales are similar to those of the associated cherts. Shimanto cherts and shales show LREE-enriched patterns with large negative Ce anomalies (0.35–0.74). Shale partings, composed mainly of clastic materials, have about 8–15 times higher REE abundances than the radiolarian and the hydrothermal components. As mentioned above, shale partings contain radiolarian tests and hydrothermal components with low REE abundances, which diluted the clastics in respect of REE composition. Therefore, clastic components originally must have higher REE abundances than those observed in the present shale partings. Sugisaki et al. (1982) pointed out that the lithogenic portion of chert and associated shale are consanguineous. These facts suggest that clastic materials play an important role in the REE abundances of cherts.

Shimizu and Masuda (1977) pointed out that the degree of Ce anomaly can be an indication of depositional environment of rocks. According to them, sedimentary rocks of pelagic origin have large negative Ce anomalies whereas those of hemipelagic do not. Cherts and shales from the Shimanto terrane showing large Ce negative anomalies might therefore be regarded as pelagic.

Hydrothermal sediments around active ridges, however, also have large negative Ce anomalies in their REE patterns (e.g. Barrett and Jarvis, 1988). This suggests that hydrothermal components can scavenge seawater REE with a Ce anomaly, perhaps even in hemipelagic sediments. Therefore the Ce anomaly in the REE pattern is not always useful for the deduction of depositional environment.

## Hydrothermal contributions

Enriched Fe and Mn contents are indicative of hydrothermal contribution, which is examined by Al-Fe-Mn diagram (Boström & Peterson, 1969). Cherts, shale partings, and basalts are plotted on the Al-Fe-Mn diagram as Fig. 3. Also plotted on this figure are marine sediments from various environments such as nearshore, marginal sea, and mid-ocean, and metalliferous sediments around active mid-ocean ridges.

The Shimanto cherts and shale partings under consideration plot in an Alpoor and Fe- and Mn-rich field, which is far from that of pelagic sediments that

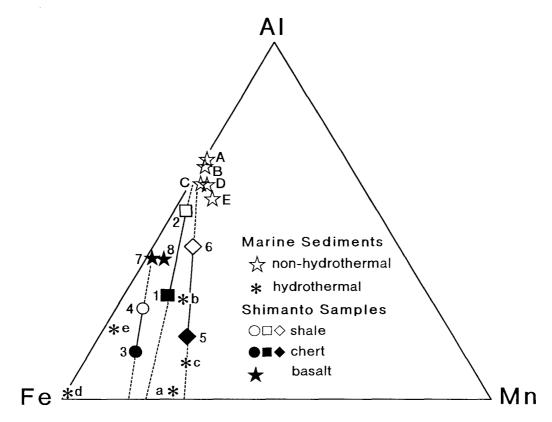


Fig. 3. Al-Fe-Mn diagram showing the effect of clastics and hydrothermal emanations. Sample symbols: Pair of open and closed symbols represents pairing shale and chert from the Shimanto Terrane, respectively; closed stars = basal basalts from the Shimanto Terrane; open stars = averaged values of non-hydrothermal modern marine sediments - A = the Nishitsugaru basin in the Japan Sea, 81 samples (Sugisaki, in press); B = the Yamato Bank in the Japan Sea, 53 samples (Sugisaki, 1979); C = the Japan Trench, DSDP Legs 56 and 57, 53 samples (Sugisaki, 1980); D = around the Izu-Ogasawara Trench, 21 samples (Sugisaki and Kinoshita, 1981); E = the central Pacific transect, Wake to Tahiti, 65 samples (Sugisaki and Kinoshita, 1982); asterisks = averaged values of hydrothermal deposits around active ocean ridge - a = East Pacific Rise deposit, group b, 9 samples (Bostrom and Peterson, 1969); b = East Pacific Rise deposit, group a, 5 samples (Bostrom and Peterson, 1969); c = East Pacific Rise deposit, 18 samples (Piper, 1973); d = nontronite in Galapagos Mounds sediment, DSDP Leg 70, 88 samples (Moorby and Cronan, 1979); e = sediment from the TAG field, Mid-Atlantic Ridge, 24 sample (Shearme et al., 1983).

are enriched in hydrogeneous materials represented by micro-manganese nodules (Ohashi, 1985; Sugisaki et al., 1987). Therefore the Fe and Mn enrichment in the Shimanto cherts and shale partings may not be due to the hydrogeneous contribution. The field of metalliferous deposits around active ocean ridge such as East Pacific Rise overlaps with that of some Shimanto cherts and shale partings enriched in Fe and Mn. This suggests that cherts and shale partings from the Shimanto terrane have been subjected to hydrothermal activities and hence contain hydrothermal components to some extent.

Cherts and shale partings from the Shimanto terrane contain radiolarian tests and clastic materials as well as hydrothermal components. As Yamamoto (1983) mentioned, the  $Al_2O_3$  content of radiolarian tests (1.18%; Table 1) were

attributed mostly to the contamination of clastic materials and radiolarians comprise little Al, Mn, and Fe. Thus the relative abundances of Al, Mn, and Fe in the cherts and shale partings are determined by the proportions of hydrothermal and clastic components. Moreover, clastic components are derived from not only terrigenous materials but also basaltic fragments, because the bedded cherts rest on the basal basalt.

The proportions of the terrigenous and the basaltic clastics can be estimated by the  ${\rm Al_2O_3/TiO_2}$  ratio of a sample (e.g. Yamamoto, 1987); marine sediments containing "usual" terrigenous clastics show a uniform  ${\rm Al_2O_3/TiO_2}$  ratio of about 22–24 (Sugisaki et al., 1982), whereas the Shimanto basalts show the ratio of about 5.5. The ratio, therefore, tends to be smaller in samples of less terrigenous contribution and more basaltic debris. The  ${\rm Al_2O_3/TiO_2}$  ratio of the Shimanto cherts and shale parting ranges from 12.0 to 19.3, and the mixing ratio of terrigenous and basaltic clastics fluctuates from sample to sample.

On the Al-Fe-Mn diagram (Fig. 3), chert and associated shale parting are connected with each other by a line; if we extend this line in both directions as shown by dotted lines, two lines (Nos. 1 and 2; Nos. 5 and 6) converge on the marine sediment field, and one (Nos. 3 and 4) reaches the basal basalt field. Both No.3 chert and No.4 shale parting show low  $Al_2O_3/TiO_2$  ratios around 13, and hence their clastic component is inferred to be largely basaltic fragments. In contrast, clastics in other pairs with higher  $Al_2O_3/TiO_2$  ratios of about 18 may be derived from mainly usual terrigenous materials. A hydrothermal end member may be plotted on the Fe-Mn axis, because of its negligible amounts of Al (e.g. Boström and Peterson, 1969; Adachi et al., 1986). The three extended lines on the diagram, thus, can be regarded as mixing lines for clastic and hydrothermal components.

## Possible Mechanism for the Formation of Bedded Cherts

A possible mechanism for the rhythmic alternation of cherts and shales in a bedded chert sequence emerges from the Al-Fe-Mn relation described above.

The rhythmic alternation of bedded cherts may be interpreted differently (e.g. McBride and Folk, 1979): (1) episodes of rapid and slow production of radiolarians in surface waters with a constant rate of clastic deposition; (2) episodes of current deposition of clastics during constant production of radiolarians; (3) diagenetic segregation of silica from initially sub-homogeneous silica mud. I prefer the second process as described below.

The position of chert and shale partings on the mixing line in Fig. 3 indicates the mixing ratio of the hydrothermal and clastic components. Three cherts (Nos. 1, 3, and 5) are close to hydrothermal end member compared with shale partings (Nos. 2, 4, and 6). This suggests that the cherts with abundant radiolarian tests contain more hydrothermal component than do the shale partings. This feature can be explained by the following processes: When the sedimentation rate of clastic materials is higher, shales with higher Al/(Al+Fe+Mn) ratio and with lower SiO<sub>2</sub> content are formed. At a lower rate of clastic sedimentation, radiolarian tests and hydrothermal emanations become predominant and consequently

chert is formed. Thus alternation of bedded cherts can be considered to be controlled by the sedimentation rate of clastics. This process is identical to that of (2) by McBride and Folk (1979).

On the other hand, the explanation (1) describes that the rhythmic alternation is controlled by production rate of radiolarians. If so, cherts and shale partings should be plotted at the same point on the Al-Fe-Mn diagram, because radiolarians hardly contain these three elements. Nevertheless, cherts are plotted in a field close to Fe-Mn axis, representing hydrothermal components. This evidence excludes the process (1).

If third explanation is correct, chert and shale are not plotted in different points on the Al-Fe-Mn diagram (Fig. 3) unless segregation of clastic component from hydrothermal one occurred. Little evidence for such a segregation, however, is available.

The bedded cherts from the Franciscan terrane were also examined on an Al-Fe-Mn diagram (Fig. 4). Each pair of chert and shale parting is connected with a line. The extended lines for some pairs (Nos. 1, 3, 4, 5, 6, and 8) approach the field of basal basalt; others (Nos. 2, 7, and 9) extend to a field between the basal

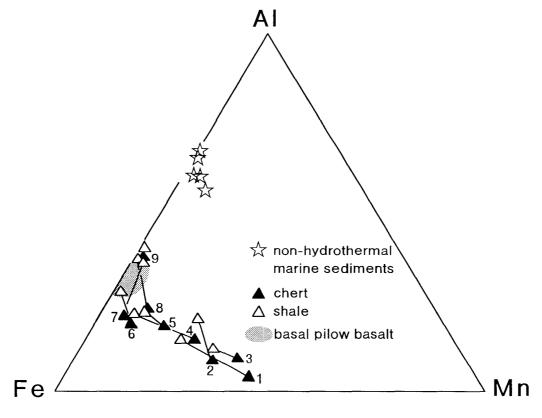


Fig. 4. Al-Fe-Mn diagram for the Franciscan bedded cherts. Sample symbols: closed triangle = chert; open triangle = shale; star = averaged values of non-hydrothermal modern marine sediment (data sources are the same as Fig. 3); dotted area is the field of basal basalt. Numerics beside the closed triangle show the sedimentary horizon from basal basalt in the order from lower to upper. The averaged composition of cherts just above and below of a shale parting was used for comparison with that of the shale parting.

basalt and marine sediment. As to Franciscan bedded cherts, Yamamoto (1987) concluded that the hydrothermal activity was intense and the contribution of basaltic clastics was large. Thus, most mixing lines for Franciscan sample pairs approximately converge on basal basalt field, in contrast with only one of three for Shimanto. Franciscan cherts fall mostly closer to the hydrothermal Fe-Mn axis and are more enriched in hydrothermal components than shales in the same manner as Shimanto sample pairs. This result is not inconsistent with the formation mechanisms of the Shimanto bedded cherts.

#### CONCLUSION

In this study, I suggested that the rhythmic alternation of chert beds and thin shale partings in a bedded chert sequence is controlled mainly by sedimentation rate of clastic materials. The geochemical data of bedded cherts up to date are not sufficient to confirm this model. As clarified by the present study, geochemical analyses of a bedded chert sequence on basal basalt are crucial to the formation mechanisms of the sequence. These types of the sequence may not be unusual in orogenic belts. They should be applied to the mechanical problems of these rhythmic sediments.

#### **ACKNOWLEDGEMENT**

I thank Drs. R. Sugisaki and M. Adachi, Nagoya University, for their continuous encouragement and valuable suggestions. I am indebted to Drs. N. Nakamura and J. Matsuda for their valuable comments. This work was supported by the Grant-in-Aid for Encouragement of Young Scientists from The Ministry of Education, Science and Culture, Japan (No. 63740440).

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