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RESEARCH ARTICLE

Geochemical constraints on the depositional environment of Upper Devonian radiolarian cherts from Loei, north-eastern Thailand

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Abstract Late Devonian radiolarian chert sequences in the Indochina block of north-eastern Thailand are exposed in a narrow belt located to the east of Loei province. The analyzed radiolarian cherts were collected from Chiang Klom, Sumnoi and Pha Samyod localities. They are characterized by high silica content (>90 wt.%), high aluminum content and low iron content ($Al_2O_3/(Al_2O_3+Fe_2O_3)$: geomeans 0.91, 0.88 and 0.92). The $Al/(Al+Fe+Mn)$ values are high in both cherts and interbedded shales (geomean 0.89). High $Si/(Si+Al+Fe)$ ratios are observed in cherts (geomean 0.97) and slightly low in shales (geomean 0.69), whereas Fe_2O_3/TiO_2 values are low (geomean 5.91). For rare earth element (REE) analysis, the cherts exhibit low La abundances (geomean 4.31, 3.59 and 4.22), slightly negative Ce anomalies (Ce/Ce*: geomean 0.81, 0.76 and 0.93), intermediate ratios of North American Shale Composite (NASC) normalized La_n/Ce_n (geomean 1.33, 1.37 and 1.12) and intermediate La_n/Yb_n values (0.79, 0.94 and 1.22). In the interbedded shales, REE characteristics are more or less equal to the cherts. The results indicate that these late Devonian radiolarian cherts were deposited in a continental margin environment. High $Si/(Si+Al+Fe)$ values indicate a biogenic origin of the cherts, however, additional silica content in the cherts could be the result of diagenetic alteration. Intermediate positive Eu anomalies (Eu/Eu*: geomean 1.32, 1.25 and 1.44) are interpreted as the result of detrital feldspar contribution corresponding to the distinctive low content of Fe. Geological evidences from the field support volcanic activities during the Late Devonian–Early Carboniferous. Weathered materials and fragments of basalts and andesites would possibly be supplied to the basin during chert sedimentation. These geochemical constraints indicate that, in western portion of Indochina, deep marine basin was closed before Late Devonian which was followed by the incursion of Early Carboniferous orogeny.

Keywords Devonian, geochemistry, rare earth element (REE), radiolarian chert, depositional environment

1 Introduction

Analysis of rare earth element (REE) signatures in radiolarian cherts and shales has been proved to be a useful tool for determining the depositional environment of marine deposits (e.g., Jones and Murchey, 1986; Murray et al., 1990; Murray, 1994; Owen et al., 1999). It has also been reported that REE concentration in shale partings exhibited a similar pattern to in cherts (Murray et al., 1991, 1992). REE analysis of chert is widely used because of their immobility during diagenesis in comparison to most of major elements (Murray, 1994; Chen et al., 2006). However, some major elements such as Al, Fe and Ti are relatively unaffected by the diagenetic fractionation process. These major elements have been used together with REEs in order to elucidate the origin of cherts (e.g., Murray, 1994; Kunimaru et al., 1998; Halamić et al., 2001; Kato et al., 2002; Zhu et al., 2006; Thassanapak et al., 2011). Differences in REE compositions in cherts and shales are generally related to continental and spreading ridge influences, so the proximity of the sequences to these environments can be deduced (Murray, 1994; Owen et al., 1999). The main source of REEs is from land. When the

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rocks were weathered, REEs were transported to the sea by rivers. In river and shallow marine environments, light rare earth elements (LREE) and heavy rare earth elements (HREE) are exhibit relatively equally, without apparent fractionation. However, anomalies of some REEs can be observed in sediments deposited in a relatively deeper environment, some distant from the continent. The major elements, including Al, Ti and K are derived mainly from land. Fe is more related to hydrothermal activity, mainly from spreading ridges (Murray, 1994).

In Thailand, for the last several decades there have been some previous studies on radiolarian cherts for biostratigraphic correlation, palaeogeographic and tectonic interpretations. The former aspect has concerned the use of radiolarians as a tool for temporal investigation of sequences. However, most of the studies relevant to the latter aspects largely misinterpreted radiolarian cherts as the evidence of oceanic setting especially for the Triassic sequences. This interpretation is controversial in relation to the spatial distribution of the main Palaeotethys in Thailand. Recently, Thassanapak et al. (2011) reports on geochemistry of Triassic radiolarian cherts from the north of Thailand suggested that these radiolarian chert sequences were deposited on a continental margin environment. This first report on geochemistry of cherts in Thailand indicates that the main Palaeotethys should be located further to the west of Nan–Uttaradit Suture. Beside the Triassic cherts, there are Devonian, Carboniferous and Permain radiolarian cherts which have been reported in northern, western, southern and north-eastern Thailand (e.g., Caridroit, 1993; Sashida and Igo, 1999; Sashida et al., 2000; Sashida and Salyapongse, 2002; Feng et al., 2002, 2004, 2005, Thassanapak et al., 2007, 2009).

The study area is located in north-eastern province of Loei. It belongs to western margin of Indochina continental block/terrane. This terrane is bounded to the west and the north by Shan–Thai along Nan–Uttaradit suture and South China along Song Ma suture, respectively (Bunopas, 1992; Metcalfe, 2002, 2011). Indochina as well as South China, North China and Tarim were separated from Gondwana during Early Devonian. As a consequence of this separation, Palaeotethys basin was created during Middle Devonian. The existence of this basin was evidenced by geological data such as the occurrence of deep marine radiolarian cherts (Metcalfe, 1997, 2011). In northern extension of Indochina, the basin was closed along Song Ma suture by amalgamation of Indochina and South China during Carboniferous (Metcalfe, 2002). This process was followed by Middle/Upper Carboniferous and Permian subsidence which indicated by thick siliciclastic sequences overlain by thick carbonate sequences in North Vietnam (Mouret, 1994). However, tectonic evolution along this suture zone was complex and controversial. It was reinterpreted that this oceanic basin was consumed by the south–directed subduction of South China underneath Indochina along Dian–Qion suture during Late Permian–Early Triassic (Cai and Zhang, 2009).

In the western margin of Indochina, the existence of oceanic basement was supported by the occurrence of Late Devonian–Early Carboniferous MOR–basalts (MORBs) and volcanic island–arc lavas (Panjasawatwong et al., 2006). These volcanic rocks could be a younger seafloor spreading evolved after termination of the older basin by suturing process. This process was interpreted from geochemistry of rhyolites (Intasopa and Dunn, 1994; Chonglakmani and Helmcke, 2001). Angular unconformity between the Upper Carboniferous–Permian and the lower strata suggested that the younger seafloor was closed during Late Devonian–Early Carboniferous (Helmcke, 1994; Chonglakmani and Helmcke, 2001). This scenario is compatible with Early Carboniferous orogeny which was possibly followed by Late Carboniferous rifting (Kozar et al., 1992; Mouret, 1994). This concept is in agreement with the occurrence of thick Upper Carboniferous–Permian strata in Loei. These strata were much less deformed in comparison to the underlying sequences (Chonglakmani and Helmcke, 2001). However, the other concept suggested that orogenic event in this region took place before Late Silurian and resumed during Middle Devonian with its climax in Late Carboniferous before the inception of magmatic activity during Late Permian–Triassic (Chairangsee et al., 1990). To test these concepts, geochemical constraints on depositional environment of Upper Devonian cherts were carried out. In Loei area, Upper Devonian radiolarian cherts were first reported in the vicinity of Pak Chom (Sashida et al., 1993). It was previously assumed that these Devonian cherts were hydrothermal in origin according to their closed occurrence with basalts (Chairangsee et al., 1990). However, geochemical composition of these cherts has never been revealed and this assumption has never been tested. The origin and depositional environment of chert sequences as well as other geological disciplines could provide more understanding on tectonic evolution of this area. Our study is aimed at clarifying the origin and the depositional environment of these cherts using rare earth element together with major element geochemistry.

2 Geological setting

Thailand consists of two main continental blocks/terranes (Fig. 1), Indochina and Shan–Thai which are located, respectively, to the east and the west of the Nan–Uttaradit suture (Bunopas, 1981). This suture was previously interpreted as the remnant of the main Palaeotethys (e.g., Bunopas, 1992; Metcalfe, 1997; Charusiri et al., 1997). However, this suture was possibly closed during the Middle–Late Permian by amalgamation of Indochina and Shan–Thai as a consequence of the Late Variscan orogeny

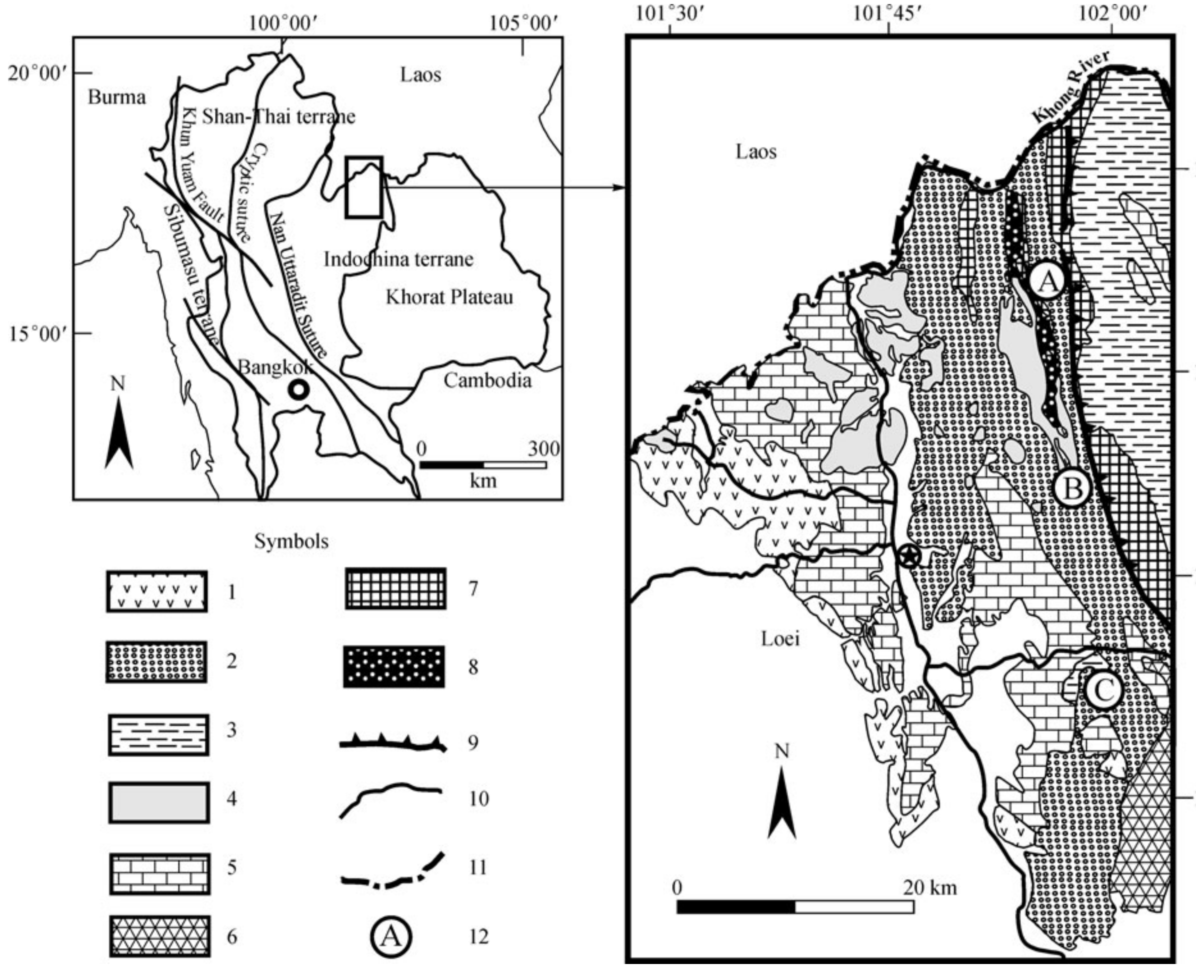

Fig. 1 Simplified geological map of Loei showing distribution of sedimentary sequences and volcanic rocks. The study sections are located to the east of Loei. Legend: 1–Permo–Triassic volcanic rocks including rhyolite, andesite, tuff, agglomerate; 2–Carboniferous rocks including conglomerate, sandstone, shale, slate, chert and limestone; 3–Permo–Trassic rocks including sandstone, argillaceous limestone, rhyolitic tuff, shale, limestone, and chert; 4–Permo–Trassic granite; 5–Permian rocks including limestone, shale, sandstone; 6–Triassic rocks including sandstone, siltstone, mudstone; 7–Devonian rocks including chert, shale and tuff; 8–Devono–Carboniferous volcanic rocks including basalt, andesite, tuff; 9–trust fault; 10–road, 11–country boundary; 12– the locations of study section including A = Chiang Klom, B = Sumnoi, C = Pha Samyod

(Helmcke and Kraikhong, 1982; Helmcke and Lindenberg 1983; Chonglakmani, 1999). Similarity of marine faunal assemblages together with geological evidence between these terranes supports their closed relationship of Permo – Carboniferous palaeogeography (Fontaine et al., 1993; Chonglakmani, 2002). Then the so-called “the main Palaeotethys” should be located to the west of Shan–Thai (Helmcke, 1985; Ueno, 1999; Chonglakmani, 1999, 2002). The amalgamation of Sibumasu (Cimmerian continent) with Shan–Thai was taken place during Late Triassic (Ferrari et al., 2008; Metcalfe, 2011). Sibumasu terrane was referred as the eastern portion of the Cimmerian continent in this region (Sengör, 1979; Metcalfe, 1997). This terrane shows distinctive Lower Permian glaciogene diamictite underlying Middle Permian platform carbonates in western and peninsular Thailand and western Malaysia. Fossil assemblages observed in the upper sequence are unique and difference to those reported from Shan–Thai and Indochina. These evidences reveal their different climatic and biogeographic proximities (Wang et al., 2001; Ueno, 2003).

In the Indochina terrane, the Silurian metamorphic rocks known as Na Mo Formation in the Loei area are the lowermost strata exposed in Thailand. This formation is unconformably overlain by marine Devonian sequences known as Pak Chom Formation (Chairangsee et al., 1990). The Pak Chom Formation is overlain by the Late Devonian–Carboniferous Nong Dok Bua Formation and Wang Sa Phung Formation which are in– turn overlain by Lower and Middle Permian sequences (Charoenprawat et al., 1976; Chonglakmani et al., 1979). In Pak Chom area, the Upper Devonian and Lower Carboniferous rocks can be subdivided into three main categories including oceanic basin cherts, island–arc basalts and turbidites (Feng et al., 2009). The cherts are tuffaceous (silicified tuff; in Panjasawatwong et al., 2006) and closely related to tuffaceaous shales (turbidites). By the occurrence of radiolarians including *Palaeoscenidium* sp. and *Helenifore* sp, the sequence was determined to be of late Devonian age (Sashida et al., 1993). According to regional geologic investigation, seismic survey and drilling exploration, these marine Palaeozoic sequences are unconformably overlain by non–marine Mesozoic strata. These strata belong to the Khorat Group which was mainly exposed in the Khorat Plateau (Sattayalak et al., 1989; Mouret, 1994; Meesook et al., 2002).

The volcanic rocks in Loei can be separated into three sub-belts; eastern rhyolites, western andesites and central basalts/hyaloclastites/braccias (Panjasawatwong et al., 2006). The eastern and central rocks are of late Devonian to early Carboniferous age determined by the whole rock K–Ar and Rb–Sr techniques (Intasopa and Dunn, 1994). The western andesites are Permo–Triassic in age (Bunopas, 1981). These Permo–Triassic rocks were also reported from the Petchabun area south of Loei, which dated by the Ar^{40}/Ar^{39} method (Intasopa, 1993). By geochemical analysis, the Central Loei volcanic rocks can be subdivided into three groups including Group A (transition tholeiitic basalt), Group B (tholeiitic micrograbbro) and Group C (calc–alkalic basalt/andesite). These rocks were interpreted as MOR–basalts (MORBs) and volcanic island–arc lavas with dike rocks, possibly by Group B, cut across the basin basement (Panjasawatwong et al., 2006).

3 Study sections and sample collection

Three chert sections were selected mainly for REEs geochemical discrimination. These investigated sections were composed of Chiang Klom, Sumnoi and Pha Samyod located to the north, central and south of the study area, respectively. The study area consists mainly of Devonian and Carboniferous rocks. The Devonian rocks include slatety shales, cherts, tuffs, limestones, conglomerates, rhyolites, basalts and andesites. The Carboniferous strata consist mainly of shales, sandstones and limestones (Charoenprawat et al., 1976).

3.1 Chiang Klom section (CK)

The chert sequence is exposed to the east of route no.2108 in the area south of Ban Klang. On topographic map sheet 5344I (Ban Na Kho) at scale 1∶50000, it lies at latitude 17° 50′ 30″ N and longitude 101° 56′ 20″ E. The section is characterized by chert sequences overlain by shale and limestone strata. Cherts are dark gray with an average bed thickness of 5 cm. The whole thickness of the chert sequences is difficult to estimate due to strongly folded character, but the thickness of the study section was about 10 m. Five samples of cherts were collected for geochemistry analysis and age determination. Preliminary study of the radiolarians from these cherts showed that they consist of *Astroentactinia multispinosa* (Won 1983), *Stigmosphaerostylus variospina* (Won 1983), *Trilonche davidi* (Hinde 1899), *Trilonche echinata* (Hinde 1899), *Trilonche elegans* Hinde 1899, *Trilonche guangxiensis* (Li and Wang 1991), *Trilonche hindea* (Hinde 1899), *Trilonche minax* (Hinde 1899), *Trilonche vetusta* Hinde 1899, *Bisyllentactinia arrhinia* (Foreman 1963) and *Palaeoscenidium cladophorum* Deflandre 1953, referred to late Devonian (Thassanapak et al., in preparation). The shales were brown with approximately 5–10 cm of bed thickness, brachiopods and trilobites were common while coral fragments and bryozoans were occasionally observed. Turbidite sequences, indication of slope deposits, were commonly observed in these shale strata. Thin–bedded limestones are uncomformably overlain by shales. These limestones are in-turn overlain by thick-bedded limestones in which corals and stromatoporoids are prolific. The Devonian corals including *Heliolites* sp., *Phillipsastrea* sp. were found associated with stromatoporoids and these built-up were interpreted as patchy reef environment (Fontaine, 1990).

3.2 Sumnoi section (SN)

The study section was found to the west of the route No. 4023, located between Ban Wang Yen and Ban Huai Toei. On topographic map sheet 5344II (Ban Sup) at scale 1∶50000, this section is located at latitude 17° 35′ 55″ N and longitude 101° 57′ 45″ E. The section consists of bedded cherts, dark gray in color, with a bed thickness of 5 cm. The chert beds are steeply inclined, nearly in vertical orientation. The study section is approximately 10 m thick. On a geological map, scale 1:250000, this area is covered by Devonian and Carboniferous rocks (Charoenprawat et al., 1976). Five samples of cherts were collected from this section for radiolarian and geochemistry analysis. Preliminary result on radiolarians suggests late Devonian by the occurrence of *Trilonche guangxiensis* (Li and Wang), *Archocyrtium* sp., *Stigmosphaerostylus* sp., *Trilonche vetusta*, *Trilonche* sp., and *Polyentactinia* sp.

3.3 Pha Samyod section (PSY)

The chert section is exposed by road-cut on top of a small hill located to the west of route No. 4033. It lies at latitude 17°23′ 45″ N and longitude 102° 02′ 20″ E on topographic map sheet 5443III (Amphoe Pha Khao) of scale 1:50000. The cherts are intensively folded with approximately 15 m of thickness. At the foot of the hill in this section, there were loose blocks of fossiliferous limestones with characteristic Devonian faunas as found in Chiang Klom section. Preliminary data on radiolarians from cherts includes *Trilonche* sp., *Stigmosphaerostylus variospina* (Won 1983), *Stigmosphaerostylus* sp., and *Protoholoeciscus* sp. suggesting late Devonian. However, on geological map scale 1:250000, the area was mapped as Carboniferous (Charoenprawat et al., 1976). Cherts in this section are dark gray and found with interbedded green shales. Five samples of cherts together with five samples of shales are collected for geochemical analysis.

4 Materials and methods

Collected samples of radiolarian cherts and shales were crushed into small fragments and then selected by handpicking under a stereomicroscope to avoid contamination of Fe and Mn coatings and calcite veins. Selected fragments were pulverized in a vibratory cup mill. Rock powder was passed through No. 200 standard sieve for geochemical analysis.

For geochemical method, X–ray fluorescence (XRF) technique was used to analyze major element concentrations. Fused–glass bead were selected for sample preparation. This process was conducted by PANalytical model “Axios advanced” instrument at the Department of Mineral Resources, Bangkok, Thailand.

The content of REEs was determined by inductively coupled plasma–mass spectrometry (ICP–MS) (Agilent 7500a) at State Key Laboratory, China University of Geosciences (Wuhan). Range of detection limit is 0.003 to 0.1 μg/g for heavier mass elements and 0.01 to 1 μg/g for lighter elements. Analytical precision is generally better than 5% for REEs. REE abundances were normalized to North American Shale Composite (NASC) with normalized values as proposed by Gromet et al. (1984). Cerium anomalies (Ce/Ce*) were calculated from $Ce/Ce^* = Ce_n/(La_n \times Pr_n)^{1/2}$ and europium anomalies (Eu/Eu*) were deduced from $Eu/Eu^* = Eu_n/(Sm_n \times Gd_n)^{1/2}$ (Taylor and McLennan 1985). NASC–normalized La_n/Yb_n ratio was used to represent HREE relative to LREE (Chen et al., 2006). Depositional environment of cherts discriminate from La_n/Ce_n vs. $Al_2O_3/(Al_2O_3 + Fe_2O_3)$ ratio (Murray, 1994).

5 Results

5.1 Major elements

The analyzed radiolarian cherts reported from all localities show high silica contents (Table 1) range from 93.37 to 94.62 wt.%, 93.67 to 95.80 wt.% and 92.18 to 94.60 wt.% from Chiang Klom, Sumnoi and Pha Samyod, respectively. By contrast, interbedded green shales from Pha Samyod locality exhibit relatively low SiO_2 content (51.27–68.60 wt. %). Most of them are lower than that of NASC (64.80 wt.%) as proposed by Gromet et al., 1984.

The aluminum abundances in cherts from all localities are high, ranged from 2.74 to 3.48 wt.%, 2.17 to 3.17 wt.% and 2.53 to 3.92 wt.% on Chiang Klom, Sumnoi and Pha Samyod, respectively. The aluminum contents are more pronounced and much higher in shales than those found in cherts (17.53–24.96 wt.%).

The titanium concentrations in cherts are very low and more or less equal in all localities, they are recorded as 0.05 to 0.07 wt.% and 0.04 to 0.09 wt.% on Chiang Klom and Pha Samyod, respectively. They are recorded 0.04 wt% in each analyzed samples from Sumnoi locality. The shale samples show higher and more variable TiO_2 contents than those of the cherts (0.33–1.33 wt.%).

The potassium contents in cherts are relatively equal in all localities. These are reported from 0.58 to 0.75 wt.%, 0.47 to 0.55 wt.% and 0.57 to 0.82 wt.% on Chiang Klom, Sumnoi and Pha Samyod, respectively. The potassium contents were significantly higher in shales and ranged from 3.56 to 7.98 wt.%.

Total iron contents ($Fe_2O_3 + FeO$) in cherts from all localities were low. They range from 0.23 to 0.47 wt.%, 0.19 to 0.63 wt.% and 0.19 to 0.55 wt.% on Chiang Klom, Sumnoi and Pha Samyod, respectively. The concentrations are higher in shales (1.36–5.42 wt.%).

The phosphorus contents in all localities are low in both cherts and shales. They are reported from 0.02 to 0.12 wt.% on cherts from Chiang Klom, while cherts from Sumnoi and Pha Samyod are of 0.02 to 0.54 wt.% and 0.01 to 0.16 wt.%, respectively. While shales from Pha Samyod exhibit from 0.01 to 0.26 wt.%.

Most of the manganese concentrations as well as some of the magnesium and sodium concentrations in cherts from these localities are very low. They are lower than 0.01 wt.% for manganese and less than 0.10 wt.% for magnesium and sodium (Table 1).

The $Al_2O_3/(Al_2O_3 + Fe_2O_3)$ values are high and more or less equal in both cherts and shales from all localities. These values are of 0.87 to 0.93, 0.81 to 0.94 and 0.88 to 0.93 as registered on cherts from Chiang Klom, Sumnoi and Pha Samyod, respectively. They are of 0.81 to 0.93 as reported in shales. The $Si/(Si + Al + Fe)$ ratios from cherts from all samples are high and range from 0.95 to 0.98.

5.2 Rare earth elements

REE concentrations in chert samples are very low in comparison to NASC values and mostly lower than 1 mg/kg (Table 2). While the interbedded shale samples show relatively high values compared with the cherts. Total REE concentrations (ΣREE) of these shales are high and range from 41.11 to 339.04 mg/kg. By contrast, ΣREE values in cherts from all localties are low and range from 12.81 to 50.08 mg/kg.

The concentrations of La and Ce in all samples are relatively high in comparison to the other REEs. Cerium anomalies (Ce/Ce*) in all samples are intermediate to high

Table 1 Major element concentrations as weight percent (wt.%) and major element ratios(Fe_2O_3 = Fe_2O_3 + FeO; NA = not available)

Locality/ sample No.	Rock type	Color	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	Al/(Al + Fe)	Si/(Si + Al + Fe)	Fe/Ti
Chiang Klom															
CK1	chert	dark gray	94.00	0.06	3.02	0.47	0.00	0.12	0.09	0.17	0.61	0.02	0.87	0.96	7.83
CK3	chert	dark gray	93.37	0.07	3.48	0.37	0.00	0.16	0.10	0.21	0.75	0.03	0.90	0.96	5.29
CK4	chert	dark gray	94.43	0.05	2.81	0.34	0.00	0.09	0.09	0.20	0.60	0.12	0.89	0.97	6.80
CK6	chert	dark gray	94.62	0.05	2.74	0.23	0.00	0.10	0.10	0.17	0.58	0.06	0.92	0.97	4.60
CK8	chert	dark gray	94.21	0.06	3.15	0.25	0.00	NA	0.11	0.22	0.68	0.05	0.93	0.97	4.17
Sumnoi															
SN4	chert	dark gray	95.80	0.04	2.17	0.19	0.00	NA	0.10	0.15	0.47	0.00	0.92	0.98	4.75
SN5	chert	dark gray	95.63	0.04	3.17	0.20	0.00	NA	0.09	0.15	0.48	0.08	0.94	0.97	5.00
SN6	chert	dark gray	94.86	0.04	2.54	0.45	0.00	NA	0.10	0.16	0.52	0.32	0.85	0.97	11.25
SN7	chert	dark gray	94.68	0.04	2.30	0.47	0.00	NA	0.19	0.15	0.48	0.54	0.83	0.97	11.75
SN8	chert	dark gray	93.67	0.04	2.77	0.63	0.00	NA	0.16	0.19	0.55	0.02	0.81	0.96	15.75
Pha Samyod															
PSY3	chert	dark gray	94.60	0.04	2.53	0.30	0.02	NA	0.21	0.24	0.57	0.00	0.89	0.97	7.50
PSY4	chert	dark gray	93.88	0.06	3.02	0.37	0.01	0.13	0.14	0.45	0.64	0.01	0.89	0.97	6.17
PSY-7A	chert	dark gray	94.39	0.05	2.71	0.19	0.01	0.11	0.20	0.21	0.70	0.02	0.93	0.97	3.80
PSY-7B	chert	dark gray	93.52	0.06	3.36	0.25	0.00	0.16	0.15	0.21	0.73	0.16	0.93	0.96	4.17
PSY10	chert	dark gray	92.18	0.09	3.92	0.55	0.00	0.20	0.12	0.19	0.82	0.09	0.88	0.95	6.11
PSY11	shale	green	52.72	0.90	21.68	4.72	0.05	2.83	0.13	NA	7.98	0.12	0.82	0.67	5.24
PSY13	shale	green	56.45	0.33	22.31	2.56	0.02	2.20	0.05	NA	7.75	0.01	0.90	0.69	7.76
PSY14	shale	green	68.60	0.45	17.53	1.36	0.01	1.17	0.03	NA	3.56	0.01	0.93	0.78	3.02
PSY101	shale	green	51.27	1.33	22.71	5.42	0.05	2.54	0.07	0.13	7.35	0.26	0.81	0.65	4.08
PSY121	shale	green	53.12	0.54	24.96	2.41	0.02	2.68	0.11	NA	7.62	0.00	0.91	0.66	4.46

Table 2 Rare earth element concentrations in mg/kg (NASC = North American Shale Composite; Geomean = geometric mean; Stdev = standard deviation; Ce/Ce* = $Ce_n/(La_n \times Pr_n)^{1/2}$; Eu/Eu* = $Eu_n/(Sm_n \times Gd_n)^{1/2}$)

	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	ΣREE	Ce/Ce*	Eu/Eu*	La_m/Yb_n	La_n/Ce_n
NASC	**32**	**73**	**7.9**	**33**	**5.7**	**1.24**	**5.2**	**0.85**	**5.2**	**1.04**	**3.4**	**0.5**	**3.1**	**0.48**	**172.61**				
CK1	2.82	4.79	0.55	2.00	0.38	0.12	38.00	0.064	0.45	0.11	0.46	0.081	0.55	0.10	12.86	0.83	1.41	0.50	1.34
CK3	3.90	6.84	0.78	2.85	0.53	0.18	0.51	0.092	0.72	0.17	0.61	0.098	0.68	0.12	18.09	0.85	1.53	0.55	1.30
CK4	5.08	9.00	1.24	4.56	0.90	0.21	0.67	0.10	0.59	0.12	0.44	0.066	0.50	0.09	23.56	0.78	1.22	0.98	1.29
CK6	6.88	10.9	1.34	4.71	0.89	0.23	0.77	0.11	0.68	0.16	0.50	0.072	0.50	0.08	27.82	0.78	1.21	1.33	1.44
CK8	3.88	6.92	0.86	3.13	0.67	0.17	0.52	0.088	0.54	0.12	0.41	0.071	0.44	0.07	17.91	0.82	1.27	0.85	1.28
Geomean	**4.31**	**7.41**	**0.91**	**3.29**	**0.64**	**0.18**	**0.55**	**0.09**	**0.59**	**0.14**	**0.48**	**0.08**	**0.53**	**0.09**	**19.38**	**0.81**	**1.32**	**0.79**	**1.33**
Stdv	**1.54**	**2.34**	**0.33**	**1.14**	**0.22**	**0.04**	**0.15**	**0.02**	**0.11**	**0.03**	**0.08**	**0.01**	**0.09**	**0.02**	**5.76**	**0.03**	**0.14**	**0.34**	**0.06**
SN4	3.04	5.37	0.70	2.56	5.00	0.12	0.44	0.075	0.46	0.095	0.31	0.051	0.35	0.05	14.15	0.80	1.16	0.84	1.29
SN5	3.25	5.75	0.75	2.71	0.47	0.14	0.46	0.077	0.45	0.11	0.33	0.054	0.34	0.05	14.95	0.80	1.35	0.94	1.29
SN6	2.99	5.51	0.78	3.04	0.70	0.23	0.67	0.10	0.59	0.12	0.32	0.051	0.31	0.05	15.44	0.79	1.50	0.94	1.24
SN7	3.76	5.45	0.82	3.30	0.73	0.20	0.68	0.11	0.67	0.14	0.42	0.057	0.46	0.07	16.87	0.68	1.26	0.79	1.57
SN8	5.35	8.14	1.14	4.27	0.87	0.20	0.84	0.12	0.80	0.15	0.43	0.067	0.41	0.07	22.86	0.72	1.04	1.26	1.50
Geomean	**3.59**	**5.96**	**0.82**	**3.12**	**0.64**	**0.18**	**0.60**	**0.10**	**0.58**	**0.12**	**0.36**	**0.06**	**0.37**	**0.06**	**16.60**	**0.76**	**1.25**	**0.94**	**1.37**
Stdv	**0.98**	**1.18**	**0.17**	**0.68**	**0.16**	**0.05**	**0.17**	**0.02**	**0.15**	**0.02**	**0.06**	**0.01**	**0.06**	**0.01**	**3.50**	**0.06**	**0.18**	**0.18**	**0.15**
PSY3	2.77	5.19	0.58	2.19	0.41	0.13	0.39	0.063	0.41	0.078	0.25	0.037	0.24	0.05	12.81	0.89	1.38	1.10	1.22
PSY4	2.83	5.30	0.61	2.34	0.44	0.15	0.41	0.063	0.45	0.087	0.27	0.039	0.30	0.04	13.34	0.88	1.59	0.92	1.22
PSY-7A	4.39	9.35	1.03	3.90	0.70	0.21	0.55	0.088	0.51	0.11	0.32	0.049	0.33	0.05	21.61	0.96	1.45	1.28	1.07
PSY-7B	4.29	8.99	0.97	3.66	0.68	0.24	0.60	0.086	0.51	0.10	0.31	0.046	0.31	0.05	20.83	0.96	1.62	1.34	1.09
PSY10	9.07	20.4	2.37	10.0	2.46	0.61	2.07	0.28	1.33	0.22	0.56	0.078	0.55	0.09	50.08	0.96	1.18	1.59	1.02
Geomean	**4.22**	**8.60**	**0.97**	**3.74**	**0.74**	**0.23**	**0.65**	**0.10**	**0.58**	**0.11**	**0.33**	**0.05**	**0.33**	**0.06**	**20.75**	**0.93**	**1.44**	**1.22**	**1.12**
Stdv	**2.58**	**6.20**	**0.73**	**3.23**	**0.86**	**0.20**	**0.71**	**0.09**	**0.39**	**0.06**	**0.13**	**0.02**	**0.12**	**0.02**	**15.29**	**0.04**	**0.18**	**0.25**	**0.09**
PSY11	65.1	133	16.7	68.7	14.5	3.70	12.7	1.78	9.85	1.86	4.89	0.68	4.49	0.66	339.04	0.88	1.02	1.40	1.11
PSY13	36.8	75.6	8.47	32.2	5.99	1.50	5.83	1.03	7.17	1.51	4.45	0.69	1.72	0.75	186.62	0.93	0.97	0.75	1.11
PSY14	11.5	24.9	2.93	11.3	2.01	0.46	1.94	0.29	1.72	0.35	0.92	0.13	0.90	0.14	59.46	0.93	0.97	1.24	1.06
PSY101	12.3	27.7	3.27	14.3	3.44	0.83	3.11	0.40	1.81	0.32	0.72	0.10	0.62	0.10	69.08	0.95	1.00	1.93	1.02
PSY121	7.79	17.2	1.80	7.44	1.50	0.46	1.57	0.23	1.31	0.26	0.70	0.11	0.67	0.11	41.11	1.00	0.93	1.13	1.03
Geomean	**19.26**	**41.25**	**4.76**	**19.26**	**3.90**	**0.99**	**3.71**	**0.54**	**3.11**	**0.60**	**1.59**	**0.23**	**1.51**	**0.24**	**101.33**	**0.94**	**0.98**	**1.23**	**1.07**
Stdv	**24.33**	**49.14**	**6.20**	**25.28**	**5.32**	**1.36**	**4.59**	**0.66**	**3.90**	**0.77**	**2.14**	**0.31**	**2.13**	**0.32**	**125.58**	**0.04**	**0.03**	**0.43**	**0.04**

negative with geomeans of 0.81, 0.76 and 0.93 in cherts from Chiang Klom, Sumnoi and Pha Samyod, respectively and 0.94 in shale from Pha Samyod. Europium anomalies (Eu/Eu*) in all chert samples show positive values (>1), while highly negative values are observed in the shale samples (geomean 0.98).

NASC-normalized values of $\mathrm{La_n/Ce_n}$ from all samples are closed to 1 (1.03–1.57). The relative ratios of LREE and HREE which are represented by NASC-normalized values of $\mathrm{La_n/Yb_n}$ show slightly high and close to 1 in most of the samples. These ratios consist of 0.79, 0.94 and 1.22 registered from cherts in Chiang Klom, Sumnoi and Pha Samyod, respectively. The ratio is 1.23 in shales from Pha Samyod.

The relationship between LREE and HREE can also be determined from the diagram of NASC-normalized relative REE abundances (Fig. 2). The result shows that REE distribution patterns from all sections are basically similar. They display a slightly flat pattern showing comparable amounts of LREE in relation to HREE. The Ce anomalies are slightly low in samples from Sumnoi and Chiang Klang, but no significant amount has found in Pha Samyod samples. Apparently positive Eu anomalies are observed particularly in the cherts from all localities. The most convex REE pattern shows in chert samples from Pha Samyod whereas the pattern from shales are less convex.

6 Discussion

6.1 Depositional environment and origin of cherts

Rare earth geochemistry of cherts has been widely applied for depositional environment and paleogeographic discriminations in particular areas or regions. REEs are less affected by post-burial diagenesis, whereas some of the major and trace elements appear to be modified (Murray et al., 1991). Chronological determination of the sequences by radiolarians was undertaken in conjunction with this geochemical analysis in order to determine their temporal aspect. The depositional environment of cherts and shale can be interpreted using Ce anomalies in which extremely low anomalies were observed in the spreading ridge (Ce/Ce* ~ 0.29), not as low from the oceanic basin (Ce/Ce* ~ 0.55) and highly negative to lowly positive values from continental margins (Ce/Ce* ~ 0.90 to 1.30) (Murray et al., 1990; Murray, 1994). In our study, the Devonian cherts and interbedded shales are closely related to the continental margin regime (Ce/Ce* geomean 0.81, 0.76, 0.93 and 0.94).

Removal of $\mathrm{Ce^{4+}}$ through a water column induces negative Ce anomalies in deep seawater, while river water, the main source of REEs, shows no apparent fractionation of REEs and mostly displays positive Ce anomalies. Thus, there are distinctive differences among $\mathrm{La_n/Ce_n}$ values in cherts from different environmental settings. The continental margin environment shows no apparent fractionation among them ($\mathrm{La_n/Ce_n} \sim 1$). By contrast, sites closing to the spreading ridge have a high ratio ($\mathrm{La_n/Ce_n} \sim 3.5$) due to strong Ce depletion, whereas intermediate values are observed in the ocean basin (Murray et al., 1990; 1991). In our study, these ratios are closed to 1 which represented a continental margin depositional environment.

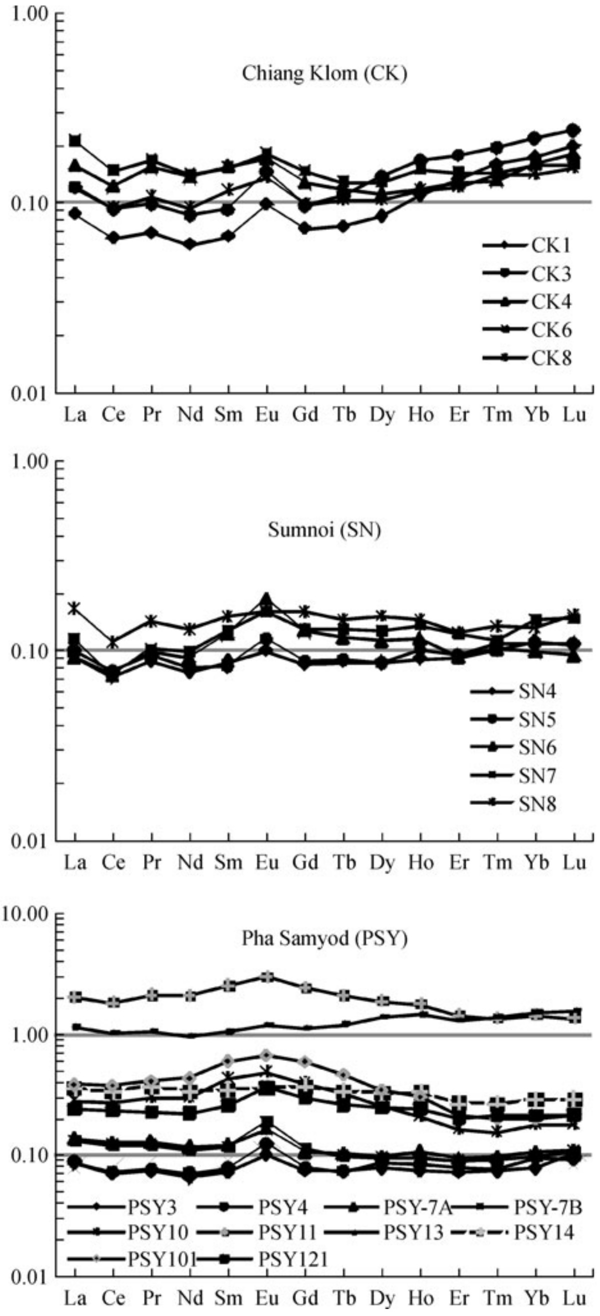

Fig. 2 NASC-normalized REE distribution diagrams of bedded chert and interbedded shale samples

As mentioned above, there is no apparent fractionation between LREE and HREE in rivers and continental margin regimes. Relative abundance between LREE and HREE can be obtained from the shale-normalized La_n/Yb_n ratio. It was reported that if significant amounts of terigenous input is reached, the ratio range from 1.0 to 1.3 (Sholkovitz, 1990; Condie, 1991). In our study, most of samples from Pha Samyod show significant signatures of terrigenous sediments relative to the water masses (La_n/Yb_n; geomeans 1.22 and 1.23 in cherts and shales, respectively). However, slight removal of LREE relative to HREE are detected in our samples from Chiang Klang and Samnoi (geomeans 0.79 and 0.94, respectively), these negative values indicates relative distant from terrigenous sources.

Radiolarian cherts deposited in continental margins show profound flat patterns in NASC-normalized REE distribution diagrams without significance negative Ce anomalies (Armstrong et al., 1999). These patterns are observed in both cherts and shale partings of our samples. In the Chiang Klang and Sumnoi diagrams, Ce anomalies are slightly negative which indicate spatial reduction of terrigenous inputs, but not as lower as that observed in deep marine deposits (Murray et al., 1990; 1991; Murray, 1994).

The convex pattern of the NASC-normalized REE distribution diagram by positive europium anomalies is normally related to hydrothermal influence (e.g., Chen et al., 2006). However, positive Eu/Eu* in cherts can also be the result of detrital feldspar contribution (Owen et al., 1999). Upper Devonian/Lower Carboniferous volcanic rocks in Loei including tuff, tuffaceous shale/mudstone, basalts and andesites from the contemporaneous volcanic activities suggests the source of Eu content in the cherts. These basalts and andesites are dark greenish gray and green in colors and consisted of high amounts of plagioclase matrix (Panjasawatwong et al., 2006). The weathering materials and fragments of these rocks were possibly supplied to the depositional basin during accumulation of cherts.

As mentioned above, Eu enrichments generally indicate mid-oceanic ridge proximity, but it is not the case since most of the REE data from our analyzed cherts reveal a continental margin environment. Moreover, the ratio of aluminum and iron from our study is very high and closely related to the field of continental margin as proposed by Murray, (1994) ($Al_2O_3/(Al_2O_3 + Fe_2O_3)$; geomeans 0.91, 0.88, 0.92 and 0.91). Previous work has been reported that well-known cherts depositions which are closely related to the spreading centers characterized by the enrichment of total Fe_2O_3 and this ratio would be decreased with increasing distant away from the hydrothermal source (Adachi et al., 1986; Murray, 1994). Al_2O_3 and TiO_2 are related to detrital components which show increasing content with decreasing distant to the terrigeneous sources. The ratios of these elements from cherts show positive correlations to each other (Fig. 3). Among the major elements, only the contents of Fe, Al and Ti are hardly affected by diagenetic processes (Yamamoto et al., 1997;

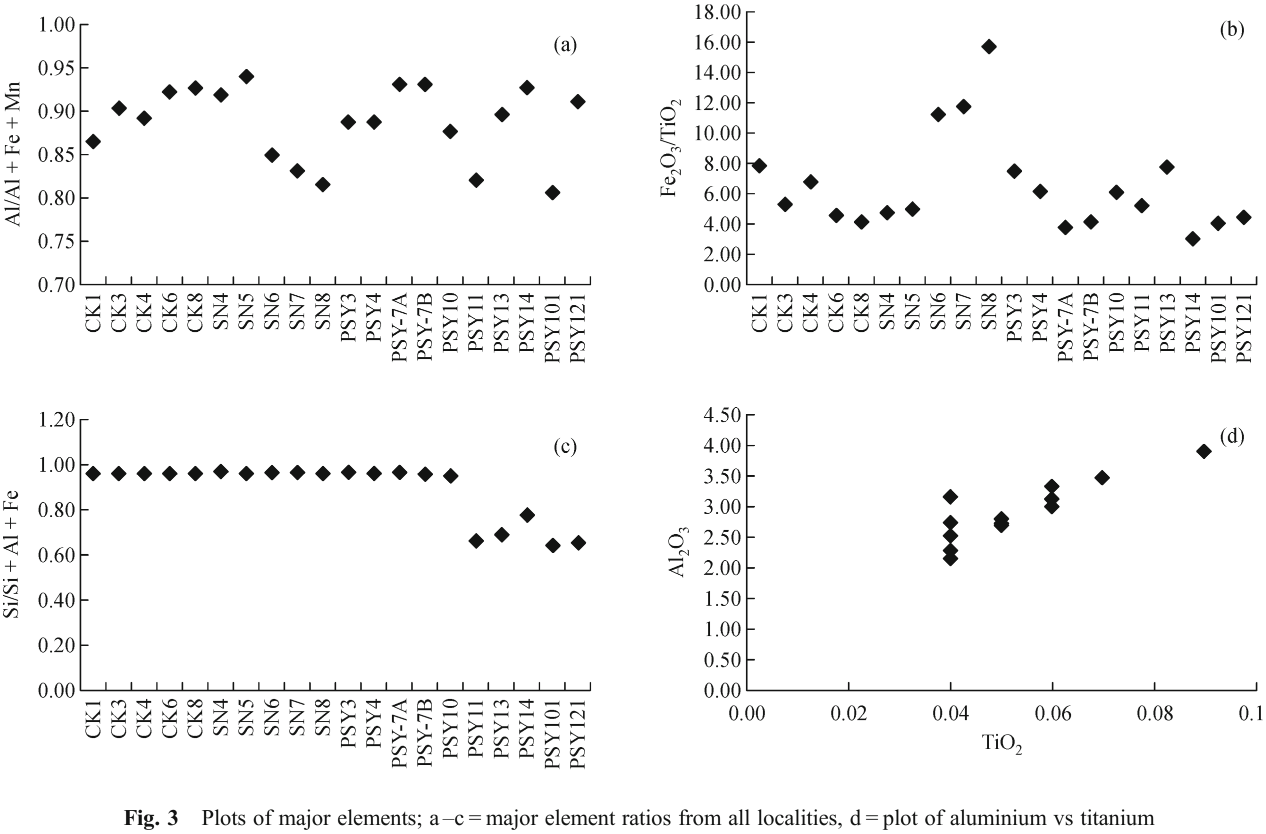

Fig. 3 Plots of major elements; a–c = major element ratios from all localities, d = plot of aluminium vs titanium

Murray, 1994). The Al/(Al + Fe + Mn) ratio can be used for determining relative distant from hydrothermal source as it shows decreasing values with increasing hydrothermal input (Boström and Peterson, 1969; Yamamoto, 1987; Aitchison and Flood, 1990). The ratios from our cherts and shale samples are relatively high, indicating a relative long distant from the hydrothermal source. This interpretation is in agreement with low Fe_2O_3/TiO_2 ratios from most of the samples. These data are also compatible with the biogenic origin of cherts which is determined from Si/(Si + Al + Fe) ratio (Rangin et al., 1981). From these analysis, our chert samples display high values and indicate biogenic origin, but not mainly from a hydrothermal origin as previously assumed (e.g., Chairangsee et al., 1990). However, diagenetic processes can cause some additional silica content in cherts.

6.2 Tectonic evolution of western Indochina

Indochina was separated from Gondwana during Early Devonian and Palaeotethys basin was created subsequent to this separation (Metcalfe, 1997, 2011). In western part of Indochina, tectonic evolution was complex and it would be possibly more than single suturing process (Chonglakmani and Helmcke, 2001). There were two different magmatic phases in this area which evidenced by Loei rhyolites (374±33 Ma) and Loei oceanic floor tholeiites (361± 11 Ma). The former was interpreted as the older suturing process and the latter was an indication of a younger seafloor spreading (Intasopa and Dunn, 1994). However, these ages should be used more carefully since they were derived from K–Ar and Sr–Nd dating techniques which could provide older ages than modern technique (Ferrari et al., 2008). By our result on geochemical constraints, it indicates that Upper Devonian chert sequences from eastern part of Loei were accumulated in continental margin environment. It can be suggested that if the above mentioned age constraints are correct, the second phase of sea–floor spreading which took place after Middle Devonian was closed during Late Devonian. This scenario is in agreement with Hercynian tectonic phase as indicated by compressional deformation during Early Carboniferous in this area (e.g., Mouret, 1994; Chonglakmani and Helmcke, 2001; Ferrari et al., 2008). By contrast, if climax compressional deformation occurred in Late Carboniferous as proposed by Chairangsee et al., (1990), geochemical analysis of Upper Devonian cherts should reveal pelagic depositional environment. Subsequent to compressional deformation event, there was rifting event as indicated by the occurrence of Lower Carboniferous basalts in southern Yunnan (Chonglakmani and Helmcke, 2001). This interpretation was supported by thick sequences of siliciclstics overlain by thick limestone sequences of Middle Carboniferous–Permian. These sequences were observed in western and eastern portion of Indochina which indicate rifting event or subsidence phase commencement after Early Carboniferrous orogeny (Mouret, 1994).

7 Conclusions

Upper Devonian radiolarian cherts from the east of Loei exhibit high silica content with more than 90% in all analyzed samples. They show high values of the Si/(Si + Al + Fe) ratios indicating biogenic origin, however, diagenetic processes could enhance the original content of silica. Moderate positive Eu anomalies caused convex patterns in NASC–normalized REE distribution diagrams possibly reflect detrital feldspar contribution. These minerals were assumed to be the weathered products of basalts/andesites from the Central Loei volcanic sub-belt. Low content of Fe in all chert samples in conjunction with REE discrimination supports this scenario. Depositional environment discrimination by major elements and REE analysis indicates that these cherts and shales were deposited in a continental margin regime. This interpretation is supported by high ratios of $Al_2O_3/(Al_2O_3 + Fe_2O_3)$, high negative to low positive Ce anormalies, more or less equal amounts between NASC-normalized La and Ce ($La_n/Ce_n \sim 1$), no apparent fractionation between LREE and HREE (La_n/Yb_n) and being without pronounced negative Ce anomalies in NASC-normalized REE distribution diagrams. These geochemical analyses indicated that, in western portion of Indochina, deep marine basin was closed before Late Devonian which was followed by Early Carboniferous orogeny.

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