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Enrichment of natural radium isotopes in the southern South China Sea surface sediments

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Abstract—Surface sediment samples were collected at 31 stations in the east and west coasts of Peninsular Malaysia on August 2003 and February 2004, respectively for determine the level of natural radium isotopes. The concentration levels of ^{228}Ra and ^{226}Ra varied at east coast of Peninsular Malaysia with ranging from 64 to 145 Bqkg^{-1} dry and 22 to 124 Bqkg^{-1} dry, respectively. Meanwhile, in the west coast stations were ranging from 35 to 65 Bqkg^{-1} dry for ^{228}Ra and 22 to 36 Bqkg^{-1} dry for ^{226}Ra. The statistical analysis with an ANOVA at 95% confidence level was also proved that the concentration level of radium at sampling stations was different between east and west coast regions. These probably related to the sources of radium which is discharge from the neighboring countries and western Pacific region during monsoon seasons. The distribution ratios of ^{228}Ra/^{226}Ra were slightly higher than the natural value and fluctuated in the east and west coasts of Peninsular Malaysia with an average about 1.8.

Key words: ^{228}Ra; ^{226}Ra; Activity ratio; Distribution; Surface marine sediment

Introduction

Radium isotopes such as ^{228}Ra ($t_{1/2}$=5.75 years) and ^{226}Ra ($t_{1/2}$=1602 years) are radioactive members of the ^{232}Th and ^{238}U decay series, respectively (Cochran, 1979). Both are important tracers in oceanographic issues on time-scales from months to years. Additionally, ^{226}Ra with a deep-sea source has been suggested as a tracer for ocean mixing processes. Many scientists found nature radioisotope such as ^{228}Ra and ^{226}Ra in marine environment are not strongly particle reactive and not scavenged from the water column into particles, these indicating their supply in surface sediment come from bottom sediments, was not relate on the thickness of water column (e.g., Schmidt et al., 1998). However, both isotopes are released into the water column from the sediment through thorium isotope decay from different parents and half life, ^{226}Ra is liberated rather from deep-sea sediments while ^{228}Ra accumulates to higher activities in shallow water regions. The ^{228}Ra/^{226}Ra ratio may vary greatly because a large difference in their half-lives and was due to high regeneration of ^{228}Ra by thorium at the bottom sediments (Moore, 1997).

High activity concentrations of radium have been found at the water-sediment interface, where the porewater plays as a media to transfer radium into the sediment (Cochran, 1979) and same character shows by sediment located in coastal region (Key et al., 1985). The distribution of radium isotopes in marine sediment environment are mostly related to their physical, chemical and geochemical properties (Khatir et al., 1998).

The study on natural radium such as ^{228}Ra and ^{226}Ra in Malaysia coastal waters is not well documented but the paper in generally related to distribution of naturally occurring radionuclides was published by Yii et al. (2008). However, some papers related to distribution of radium in petroleum industry waste and its terrestrial radioactivity in Malaysia was published by Malaysian researchers (e.g., Abu et al., 2000; Omar et al., 2004; Muhamad-Samudi et al., 2006). Then the main objective of this report is to describe the potential of southern South China Sea area as an enrichment region for natural radionuclide such as radium.

Materials and Methods

Analytical procedures

About 31 stations of surface marine sediments were collected at the east and west coasts of Peninsular Malaysia on August 2003 and February 2004 using gravity corer (Table 1, Fig. 1). Sediment samples were kept into the sample container and brought to the laboratory for further analyses. Briefly, all the sediment samples were dried in an oven at 60°C until a constant weight and ground properly to homogeneity.

Then about 300–350 g of homogenous dried sediments were transferred into the 350 ml polyethylene containers, sealed and kept for four weeks to reach secular equilibrium between radium and their progenies. The specific activities of ^{228}Ra and ^{226}Ra were measured through their gamma emitting daughters using high-purity vertical germanium detectors (HpGe) with model E&G ORTEC for 15 hours. The HpGe detector energy and efficiency were calibrated using a several source of nuclide (e.g., ^{60}Co, ^{137}Cs) and mix standard of gamma radionuclide. The quality control was confirmed using the International Atomic Energy Agency (IAEA) standard reference material (SRM Soil 6) at same geometry with measured samples. A relative efficiency about 25% and ranged of energy from 1.95 keV to 1332 keV was use to estimate the activity of radium (Dukat and Kuehl, 1995; Brunskill et al., 2004; El Memoney and Khater, 2004). The activities of ^{226}Ra were calculated from the measurement of granddaughters photopeak of ^{214}Pb at energy of 295.2 and 351.9 keV; and photopeak of ^{214}Bi at energy of 609 keV (Brunskill et al., 2004), meanwhile, ^{228}Ra activities were determined from their daughter ^{228}Ac at energy of 911 keV (Dukat and Kuehl, 1995).

Table 1. Sampling stations obtained during this cruises.

Region	Location	Station	Latitude, °N	Longitude, °E	Water depth (m)	Distance from land (miles nautical)
East coast of Peninsular Malaysia	Kota Bharu	EC 01	06°38.67′	103° 36.18′	45.8	32.5
	Kuala Terengganu	EC 02	05°36.14′	103° 24.25′	51.6	14.5
	Kuantan	EC 03	03°58.30′	104° 07.05′	50.1	34
	Pulau Tioman	EC 04	02°52.82′	104°14.05′	44.1	–
	Desaru	EC 05	02°08.44′	104°30.44′	42.0	–
	Muara Sungai Kelantan	EC 06	06°16.58′	102°08.71′	9.2	2.7
	Muara Sungai Besut	EC 07	05°50.61′	102°36.40′	8.3	2.3
	Kuala Terengganu	EC 08	05°21.65′	103°09.30′	8.7	1.6
	Muara Sungai Dungun	EC 09	04°47.23′	103°26.74′	17.3	1.4
	Muara Sungai Kemaman	EC 10	04°13.45′	103°27.60′	9.5	1.3
	Muara Sungai Kuantan	EC 11	03°47.63′	103°24.13′	11.9	2.3
	Muara Sungai Pahang	EC 12	05°28.95′	103°30.39′	11.4	1.6
	Muara Sungai Rompin	EC 13	02°48.55′	103°32.25′	8.2	1.7
	Muara Sungai Sedili Besar	EC 14	01°52.24′	104°13.07′	15.1	2.9
	Tanjung Datok	EC 15	01°23.87′	104°18.36′	13.5	–
West coast of Peninsular Malaysia	Kuala Kedah	WC 01	06°06.60′	099°58.30′	83.0	48.0
	Pulau Pinang	WC 02	05°56.70′	099°29.80′	50.0	51.0
	Kuala Terung	WC 03	05°28.35′	099°19.00′	77.5	50.0
	Sabak Bernam	WC 04	03°21.45′	100°22.94′	69.9	48.4
	Pulau Langkawi	WC 07	06°09.90′	099°51.40′	9.8	2.3
	Kuala Kedah	WC 08	06°01.79′	100°11.52′	7.4	4.5
	Sungai Merbok	WC 09	05°46.88′	100°10.57′	19.0	7.9
	Kuala Terung	WC 10	04°40.52′	100°22.78′	12.9	12.3
	Kuala Perak	WC 11	03°55.57′	100°39.03′	15.2	4.4
	Sungai Bernam	WC 12	03°42.38′	100°47.59′	20.8	4.9
	Kuala Selangor	WC 13	03°19.43′	101°08.97′	13.2	4.7
	Tanjung Ru	WC 14	02°40.81′	101°26.87′	34.2	1.6
	Sungai Linggi	WC 15	02°18.10′	102°03.00′	7.9	3.2
	Sungai Muar	WC 16	01°58.54′	102°30.54′	6.4	4.9
	Sungai Batu Pahat	WC 17	01°48.84′	102°44.85′	14.6	3.2
	Sungai Benut	WC 18	01°36.41′	103°08.40′	5.2	2.3

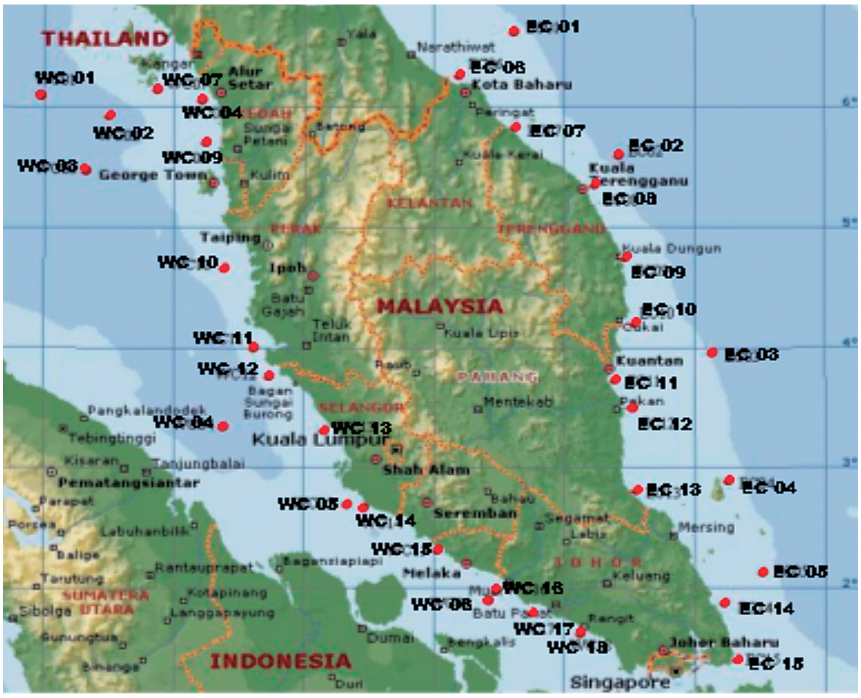

Fig. 1. Sediment sampling conducting during this study.

Result and Discussion

Distribution of ^{228}Ra and ^{226}Ra in surface marine sediment

The ranged concentration of ^{228}Ra and ^{226}Ra (Bqkg^{-1} dry) in surface marine sediments collected at 31 stations of the east and west coasts of Peninsular Malaysia were from 35 to 145 Bqkg^{-1} dry and 22 to 124 Bqkg^{-1} dry, respectively (Table 2). It was found that the activities concentration of those radionuclide in surface marine sediment are varied with sampling stations and it was proved by an ANOVA analysis that have significant difference at 95% confidence level with $p<0.050$. The activities of both radionuclide at the east coast were higher than west coast of Peninsular Malaysia because the east coast will classify as semi-enclosed system receive large input of natural radioisotope sources from the neighboring countries and western Pacific waters as external sources. In this case physical process such as remobilization, re-suspension and re-deposition due to heavy rain, strong wave, turbulent and current during monsoon season play important role to transport sediments and particles which contained radium and other natural radioisotopes. Internal sources from the biological remobilization as the alternative way to bring radium at the study area.

Additionally, the sampling stations at the east coast region was enclosed mostly by the landmasses receive large input of soil which are known as sources of radium and others natural radioisotopes came from soil erosion especially during monsoon season. High concentration activity of radium at the east coast region probably also related to their parents (^{232}Th and ^{238}U) from the semi-enclosed marginal sea of South China Sea (SCS), bottom sediment and *in situ* decay (Table 2). Meanwhile, activities of ^{228}Ra and ^{226}Ra obtained in the west coast of Peninsular Malaysia is slightly low because Malacca Straits is an enclosed system and received less input from the original sources or land (Nozaki and Yamamoto, 2001; Mohamed et al. 2006). Furthermore, the sampling stations at the west coast region located far from the continent of Peninsular Malaysia, Sumatra Island and other islands (Table 1), thus, received a small input of sediment which contained less the concentration of ^{228}Ra and ^{226}Ra from the land.

Generally, activities concentration of ^{228}Ra were high compared to ^{226}Ra at all sampling stations of both regions (Fig. 2-a), was expected relate to the high detrital source from the coastal area (Schmidt et al., 1998). It was supported by Krest et al. (1999) that regeneration of ^{228}Ra by its parent, ^{232}Th was 280 times higher compared to ^{226}Ra. In Table 3 shows a positive Pearson correlation between ^{228}Ra with ^{232}Th ($r=0.810$; $p<0.050$) and between ^{228}Ra with ^{232}Th/^{238}U (0.790; $p<0.050$) as seen especially at stations EC 06, EC 12 and EC 13 (Table 2).

Moreover, a positive statistical correlation between ^{228}Ra and water depth at east coast ($R=0.886$; Fig. 3-a) will reflects to high regeneration of ^{228}Ra from its parent (^{232}Th) in water column towards to enrich of ^{228}Ra on the water-sediment interface. Meanwhile, a negative correlation between ^{226}Ra with water depth ($R=0.837$) in the east coast region, indicating that ^{226}Ra was regenerated by ^{238}U in water column. In the case of low activity is probably due to the diffu-

Table 2. Activity concentration of ^{228}Ra and ^{226}Ra in the surface marine sediment at the east and west coast of Peninsular Malaysia.

Region	Station	Activity concentration (Bq/kg dry wt)		^{232}Th/^{238}U activity ratio	Activity concentration (Bq/kg dry wt)		^{228}Ra/^{226}Ra activity ratio
		^{228}Ra	^{226}Ra		^{232}Th	^{238}U	
East coast of Peninsular Malaysia	EC 01	33±2	21±2	1.6±0.2	66.0±14.5	22.7±1.6	2.9±2.0
	EC 02	12±1	8±1	1.4±0.2	98.8±21.7	41.8±2.9	2.4±2.0
	EC 03	77±5	24±2	3.2±0.3	132.1±29.1	62.2±4.2	2.1±1.8
	EC 04	23±2	17±3	1.4±0.2	76.8±16.9	30.3±2.1	2.5±1.9
	EC 05	54±4	22±2	2.5±0.3	130.9±28.8	98.0±6.9	1.3±1.2
	EC 06	130±10	44±3	3.0±0.3	131.2±28.8	57.1±9.2	2.3±1.8
	EC 07	82±6	35±3	2.3±0.2	65.4±14.4	50.0±8.1	1.3±1.1
	EC 08	62±4	28±2	2.2±0.2	108.8±23.9	81.5±7.7	1.3±1.1
	EC 09	76±6	31±2	2.5±0.3	98.9±15.9	82.7±7.9	1.2±1.0
	EC 10	71±5	30±2	2.4±0.2	100.0±16.1	65.5±6.2	1.5±1.2
	EC 11	61±4	22±2	2.8±0.3	144.6±31.8	123.7±11.8	1.2±1.0
	EC 12	110±8	48±4	2.3±0.2	98.4±15.8	69.2±6.6	1.7±1.3
	EC 13	113±8	48±4	2.4±0.2	110.2±24.2	57.3±5.3	1.9±1.6
	EC 14	61±4	26±2	2.4±0.2	99.5±16.0	79.4±7.6	1.2±1.0
	EC 15	78±6	33±2	2.4±0.2	86.7±19.1	64.2±6.1	1.4±1.1
West coast of Peninsular Malaysia	WC 01	69±5	28±2	2.5±0.3	65.0±14.3	26.4±1.4	2.5±2.1
	WC 02	75±6	24±2	3.1±0.3	52.2±11.5	24.7±1.8	2.1±1.7
	WC 03	54±4	21±2	2.6±0.3	52.8±11.6	28.0±2.6	1.9±1.5
	WC 04	36±3	13±1	2.7±0.3	41.2±4.9	24.8±1.7	1.7±1.3
	WC 07	87±6	33±2	2.6±0.3	43.0±9.1	28.6±2.1	1.5±1.2
	WC 08	89±5	64±5	1.4±0.1	44.5±9.8	29.0±1.1	1.5±1.2
	WC 09	44±3	41±3	1.1±0.1	35.1±5.6	23.0±1.7	1.5±1.2
	WC 10	67±5	50±4	1.3±0.1	47.7±7.7	24.8±2.7	1.9±1.6
	WC 11	81±6	28±2	2.9±0.3	50.0±11.0	29.0±3.2	1.7±1.4
	WC 12	76±6	24±2	3.2±0.3	52.8±11.6	35.6±2.2	1.5±1.2
	WC13	71±5	17±1	4.2±0.4	59.5±13.1	27.2±3.0	2.2±1.8
	WC 14	40±3	19±1	2.1±0.2	44.0±9.7	27.1±2.3	1.6±1.3
	WC 15	52±4	16±1	3.4±0.4	35.7±7.9	23.0±2.5	1.6±1.3
	WC 16	61±4	19±1	3.2±0.3	47.0±10.3	28.0±2.1	1.7±1.4
	WC 17	62±4	19±1	3.2±0.3	58.2±12.8	26.7±2.9	2.2±1.8
	WC 18	48±2	15±1	3.2±0.3	41.7±9.2	30.4±2.1	1.4±1.1

sion processes (Hancock and Murray, 1996). Then no correlation occurs between ^{226}Ra, ^{238}U and ^{232}Th/^{238}U (Table 3) will revealed that ^{226}Ra was not regenerated from *in situ* decay of ^{238}U in surface sediment but might be regenerated from ^{238}U at bottom sediment, then transferred through porewater to surface sediment.

However, the west coast of Peninsular Malaysia, displayed strongly positive correlation between ^{228}Ra and water depth (R=0.715; Fig. 3-b) probably due to actively and effectively regenerated of ^{228}Ra by its parent (^{232}Th) in water column. Then correlation between ^{228}Ra, ^{232}Th and ^{232}Th/^{238}U (Table 3) was not obtained at the west coast region, indicating ^{228}Ra was not regenerated from *in situ* decay of ^{232}Th in surface sediment but might be supplied from water column and bottom sediment. Meanwhile, ^{226}Ra displayed strongly negative correlation with water depth (R=0.849) where most of ^{226}Ra in marine environment are not strongly particle reactive and not scavenge from the water column into particles, these indicating their supply in the surface sediment come from bottom sediments.

A statistical analysis has proved that strong positive correlated between ^{228}Ra and ^{226}Ra at the east (R=0.710) and west (R=0.747) coasts region (Fig. 4 a-b). A strong positive correlation at both region revealed that both radium isotopes was supplied from the same environmental origin as well described by Moore (1997). Meaning their parents (^{232}Th and ^{230}Th) was associated with particle in water column before deposited on to sediment, thus the particle also consist radium isotopes (^{228}Ra and ^{226}Ra) which generated from their parents decay.

^{228}Ra/^{226}Ra ratio in surface sediment

The activity ratios of ^{228}Ra and ^{226}Ra were uniformed in surface sediments of the east and west coasts ranged from 1.17 to 2.87 with an average of 1.8 (Table 2-b). Moreover, it was supported by ANOVA analysis found that there was no significant different at 95% confidence level (p=0.885) at all sampling stations.

Most of the sampling stations found slightly high activity ratio of ^{228}Ra/^{226}Ra will indicate the enrichment of ^{228}Ra on the sediment was occurred (Fig. 2-b, Table 2). The statistical analysis also proved has a strong positive correlation between ^{228}Ra/^{226}Ra and water depth (Fig. 3-c) with R=0.885 for east coast and 0.937 for west coast. The strong correlation suggesting ^{228}Ra at both regions was totally supplied by the water column. It also supporting that ^{228}Ra at both regions has come from the coastal, and bottom sediments,

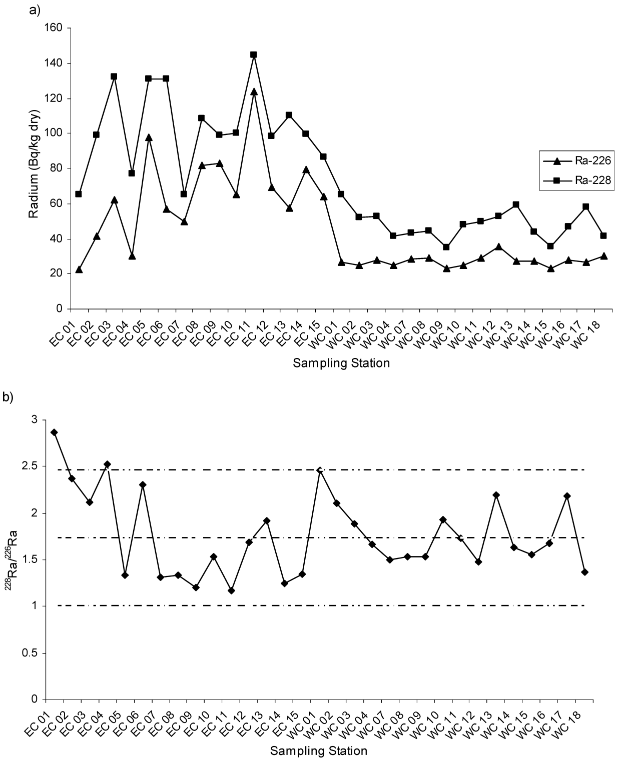

Fig. 2. Distribution of ^{228}Ra, ^{226}Ra and ^{228}Ra/^{226}Ra in surface marine sediments of Peninsular Malaysia.

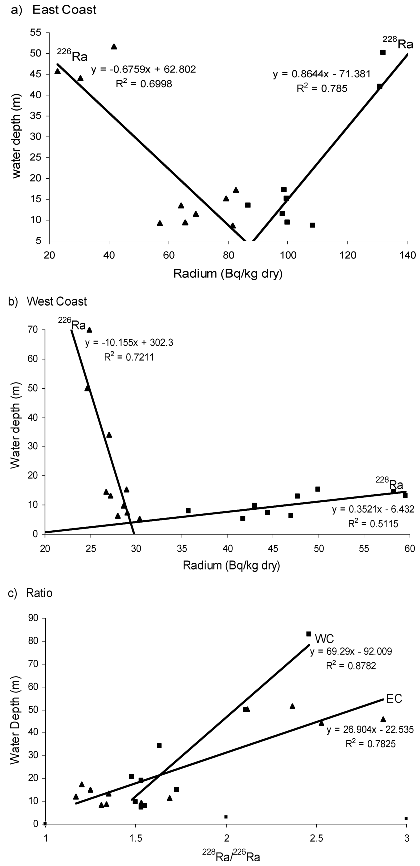

Fig. 3. Relationship between water depth and radium activity and activity ratio in surface marine sediments of Peninsular Malaysia.

Table 3. Correlation between ^{228}Ra, ^{226}Ra, ^{228}Ra/^{226}Ra and their parents in surface marine sediments of Peninsular Malaysia.

Region	Activity concentration (Bq/kg dry)	Activity concentration (Bq/kg dry) ratio		^{232}Th/^{238}U activity ratio	^{228}Ra/^{226}Ra activity ratio
		^{232}Th	^{238}U		
East coast of Peninsular Malaysia	^{228}Ra	r=0.810** p=0.004		r=0.790** p=0.007	r=−0.733** p=0.016
	^{226}Ra		r=0.235 p=0.419	r=0.563 p=0.071	r=−0.531 p=0.093
	^{228}Ra/^{226}Ra			r=−0.947** p=0.000	
West coast of Peninsulara Malaysia	^{228}Ra	r=0.465 p=0.094		r=0.557 p=0.152	r=−0.016 p=0.969
	^{226}Ra		r=0.154 p=0.599	r=0.075 p=0.861	r=0.704 p=0.051
	^{228}Ra/^{226}Ra			r=0.470 p=0.240	

** Correlation is significant at the 0.01 level (2-tailed).

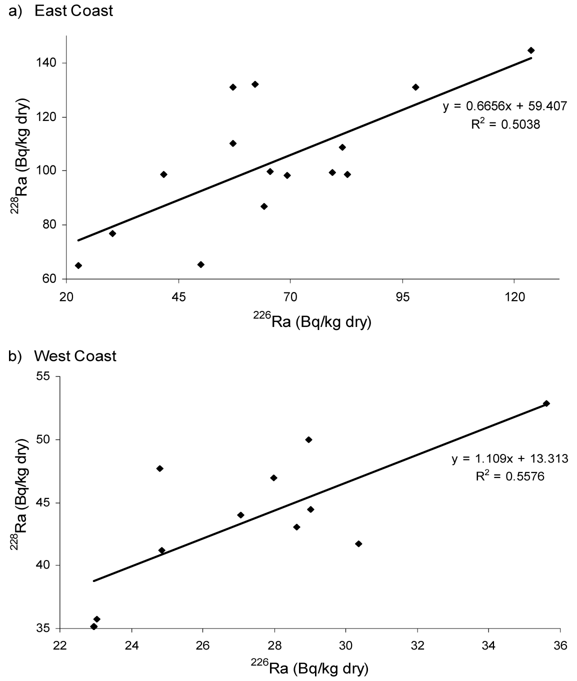

Fig. 4. Correlation between ^{228}Ra and ^{226}Ra in surface marine sediment at east (a) and west coast (b) of Peninsular Malaysia.

thereafter increases the activity ratio of water-sediment interface (Schmidt et al., 1998). This finding also was supported by the published report (Hancock and Murray, 1996), where the enrichment of ^{228}Ra at near-shore environments is often much greater than the long-lived ^{226}Ra ($t_{1/2}=1602$ years).

High activity ratio which is more than 1.0, indicating that ^{228}Ra ($t_{1/2}=5.75$ years) was actively and rapidly regenerated by their parent (^{232}Th) compared to the ^{226}Ra from ^{230}Th (Moore 1997) as well found in this study (Table 2). This idea was strictly support by statistical analysis with strong negative correlation between ^{228}Ra/^{226}Ra and ^{232}Th/^{238}U ($R=-0.947$; $p<0.050$) at the east coast region which reflects to the enrichment of ^{238}U in the water-sediment interface and sediment layer (IAEA, 1990; El Memoney and Khater, 2004).

Conclusions

The concentration levels of ^{228}Ra and ^{226}Ra in surface marine sediment at the east and west coasts of Peninsular Malaysia varied with sampling sites. The difference of activity concentration of radium between east and west coast was related to the sources during monsoon seasons. The statistical analysis with an ANOVA at 95% confidence level also proved that the activity concentration of radium was different at both regions. The distribution of ^{228}Ra/^{226}Ra ratios was slightly high but uniformed in the east and west coast of Peninsular Malaysia with an average of 1.8.

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