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Level, trends and effects of natural radionuclides in sediment from the east coast of Peninsular Malaysia

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Abstract

Naturally occurring radionuclides ^{238}U, ^{232}Th, ^{226}Ra and ^{210}Po present in the marine surface sediments collected in the east coast of Peninsular were measured using HPGe gamma-ray spectrometer and alpha spectrometer. The objectives of this work are to quantify the level of those radionuclides, to study their trend and distribution in studied area and to evaluate the radiological health hazard effect due to natural radioactivity associated with sediments at studied area. The results found the level of ^{238}U, ^{232}Th, ^{226}Ra and ^{210}Po in surface sediment were ranged of 3.81 ± 0.29 – 32.58 ± 7.63 Bq/kg dw., 12.51 ± 1.65 – 63.29 ± 8.24 Bq/kg dw., 7.21 ± 0.59 – 64.93 ± 7.63 Bq/kg dw. and 22.28 ± 0.99 – 69.14 ± 3.09 Bq/kg dw., respectively. Meanwhile, their radioactivity trends and distribution observed to be higher toward the coastal area. This result due to terrestrial play an important role as the main sources for supplying these radionuclides to studied area. The mean radium equivalent activity concentration index and other radiological hazard parameter were less than their maximum permissible limits and the world averages. Therefore, the occurrence of those radionuclides in surface sediment were not posed radiological health hazard to anyone presence in the surrounding area, rather these sediments are suitable and safe to use as a road or building construction materials.

Keywords: Natural radionuclides, sediment, level, trends and distribution, radiological effect

Introduction

The uranium and thorium decay series have numerous radionuclides with half-lives ranging from fractions of a second to a few years. The radionuclides in both decay series with respectively half-lives that makes them useful in tracing a variety of oceanic process. The ^{238}U (uranium-238; $t_{1/2}$: 4.468 x10^9 years) and ^{226}Ra (radium-226; $t_{1/2}$: 1620 years) are alpha radioactive emitter and; more mobile and soluble as a result tend to remain in solution. Meanwhile ^{232}Th (thorium-232; $t_{1/2}$: 1.4 x 10^{10} years) and ^{210}Po (polonium-210; $t_{1/2}$: 138.4 days) are particle-reactive which are scavenged in water column before deposited in the seabed. Uranium-238 behaves as a nearly conservative element in oxygenated seawater, but it is precipitated under chemically reducing conditions that occur in sediments underlying low-oxygen bottom water or in sediments receiving high fluxes of particulate organic carbon (Zheng et al. 2002a). In other word, the primary source of ^{238}U accumulated in sediments is diffusion across the sediment-water interface into reducing sediments (Barnes and Cochran, 1990). Other source of ^{238}U to the sediment is particulate non-lithogenic uranium formed in

surface ocean waters in conjunction with marine particulate matter and provides a small and variable flux of ^{238}U to sediments (Zheng et al., 2002b).

Meanwhile, ^{232}Th is a primer radionuclide of thorium decay series. It is transported from catchment areas and entered into the marine environment through the form of weathered detrital minerals (Goldberg and Koide, 1962) and associated with particles and removed from seawater by adsorption, coprecipitation or biological processes (Sam, 2003). Because of ^{232}Th has a unique behaviour of strong particle reactive, extremely non-conservative and very insoluble in seawater, thus it is rapidly scavenged onto settling particles and removed from the water column, accumulated and buried onto sediment (Bacon and Anderson, 1982) resulted to contribute of higher activity concentration in coastal sediment.

Naturally occurring ^{210}Po and its grandparent ^{226}Ra are members of the ^{238}U decay series. Radium-226 in marine environment are not particle reactive and not scavenged from the water column into particles, these indicating its supply in surface sediment come from bottom sediment. Thus, its distribution in sediment is not related to water depth or the thickness of water column (Schmidt et al., 1998). Radium-226 is relatively mobile radionuclide that is produced from above mentioned highly insoluble parents of ^{238}U (Rutgers Van Der Loeff et al., 2004). High activity concentrations of ^{226}Ra have been found at the water-sediment interface, where the pore-water play as a media to transfer ^{226}Ra into the sediment (Cochran, 1980) and same character shows by sediment located in coastal region (Key et al., 1985). Key et al. (1985) also reported that the estuarine bottom sediments is significant supply radium includes ^{226}Ra to the coastal and continental shelf sediments. Generally, the distributions of ^{226}Ra in marine sediment are mostly related to its physical, chemical and geochemical properties (Khatir et al., 1998a).

A small amount of ^{210}Po in the marine environment i.e. seawater originates from the atmospheric deposition of polonium itself (Cochran, 1992; Nozaki, 1991), but most ^{210}Po is largely produced by the decay, *in situ* of ^{210}Pb ($t_{1/2}$: 22.3 years) deposited from the atmosphere (Meli et al., 2013). Polonium-210 can be transported from the water to the sediment phases by physical (e.g. sedimentation), chemical (e.g. ion-exchange, polymerisation, colloid aggregation) and biological (e.g. detris) processes. Physical remobilization of contaminants from sediments to water can occur due to natural or anthropogenic re-suspension of the sediments, e.g. during flooding, erosion or estuary dredging; chemical mobilisation includes ion-exchange, leaching, and dissolution (Rudjord et al., 2019). Po-210 is a strong particle reactive which is scavenged and removed from water column to the sediment as a last compartment for the primary repository of its deposition. Furthermore, Bacon et al. (1976) also documented that ^{210}Po in the oceans was more rapidly removed from surface waters to sediment. Other than that, the activity concentration of ^{210}Po can be locally enhanced by the impact of industrial emissions, in particular the discharges from factories that produce phosphoric acid used in the manufacturing of fertiliser and that generate phosphatic gypsum wastes (Stepnowski and Skwarzec, 2000; Al-Masri et al., 2002; Jia et al., 2003), oil and gas exploring and processing, etc.

Natural radioactivities of ^{238}U, ^{232}Th, ^{226}Ra and ^{210}Po have great contributions in ionizing radiations to the world population due to their presence in surrounding such as marine environment compartment includes sediment. Therefore, these radionuclides can be of great concern from the standpoint of radiation protection because of their radio-toxicity. Natural radioactivity in sediment is mainly due to those radionuclides which cause external and internal radiological hazards due to emission of gamma rays and inhalation of radon and

its daughters (UNSCEAR, 1988). Measurement of external radiation dose from marine sources is also necessary not only due to its contributions to the collective dose but also due to variations of the individual dose related to the pathway. These doses strongly depend on the concentrations of ^{238}U, ^{232}Th and their progenies, present in sediment, which in turns depends upon the geology of the regions (Malik, 2014; Quindos et al., 1994; Radhakrishna et al. 1993). Since sediment plays a predominant role in aquatic radioecology and plays a role in accumulating and transporting contaminants within the geographic area, thus it is the basic indicator of radiological contamination in the environment (Suresh et al., 2011).

In this study, ^{238}U, ^{232}Th, ^{226}Ra and ^{210}Po activity concentrations in surface sediment are determined at nearshore in the east coast of Peninsular Malaysia. This study was conducted as a part of the national project with the title of "Level, trends and effects of natural and anthropogenic radionuclides in the Malaysian marine environment" under IAEA CRP K41017 project: Behaviour and effects of natural and anthropogenic radionuclides in the marine environment and their use as tracers for oceanography studies. The aims of the study are (i) to quantify the level of those radionuclides, ii) to study their trend and distribution in study area, and iii) to evaluate the radiological health hazard effect due to natural radioactivity associated with sediments at study area.

Materials and Methods

Study area

Our study areas are located along near shore in the east coast of Peninsular Malaysia within latitude of 4.439 – 6.089 ^{o}N and longitude of 102.542 – 103.610 ^{o}E and all points are situated about 9 – 20 km from mainland and water depth ranged of 15.5 – 46.6 m (Table 1). Sediment samples were collected at different 20 points in 2017 – 2018. These sampling points were selected by presuming the mainly sources of naturally radionuclides from most of the mainland of the neighbouring countries which are able to accumulate or transfer into Malaysian waters in particular in the east coast of Peninsular Malaysia. The map of the areas for sample collection is shown in Figure 1.

Sample collection, preparation and digestion

About one (1) kg of surface sediment samples were collected using a Ponar grab sampler. All the samples were transferred into a zipped plastic bag, stored in cold storage boxes and transported to the laboratory. In the laboratory, about 2 g of sediment samples were taken for sediment particle size analysis and the rest of the samples were dried at 60^{o}C in an electric oven until a constant weight for 2 - 5 days. All the dried samples were ground to powder form to homogeneity and then, samples were kept into air tight container.

For gamma measurement, the ground samples were transferred into 300 mL counting container, sealed and stored for a period in excess of 30 days to establish secular equilibrium between ^{238}U, ^{232}Th, ^{226}Ra and their respective radioactive progenies prior to gamma counting. Meanwhile for alpha of ^{210}Po: 0.5 g of sediment sample in Teflon beaker was spiked with 0.5 mL of a known activity concentration of ^{209}Po tracer and was totally digested by adding 10 mL of concentrated HF and HNO_3 and was evaporated on a hot plate to dryness. This procedure was repeated if the samples were not totally digested. Then, 10 mL of concentrated HNO_3 and 1 mL of H_2O_2 were added and evaporate to almost dryness.

Finally, 10 mL of concentrated HCl was added into the samples, then evaporated until almost dryness and let it for a while to cool.

Table 1: Metadata of sampling collection localities, time and water depth

Location	Station ID	Latitude, ^{o}N	Longitude, ^{o}E	Sampling Date	Water Depth (m)	Sediment Grain Size (%)		
						Sand	Silt	Clay
						> 63 µm	4 - 63 µm	< 4 µm
Melawi	ML01	6.086	102.574	23 Sept. 2017	22.1	28.4	66.0	5.7
Tok Bali	TB 01	5.947	102.542	23 March 2018	15.5	17.1	82.3	0.7
	TB03	5.968	102.594	04 May 2018	23.4	86.4	13.2	0.4
Pulau Perhentian	PP02	5.993	102.754	04 May 2018	35.4	64.1	31.4	4.5
	PP04	5.944	102.631	04 May 2018	25.7	57.7	40.2	2.1
Kuala Terengganu	KT01	5.558	103.148	04 July 2018	30.7	69.2	28.1	2.8
	KT02	5.541	103.220	04 July 2018	42.9	55.1	36.6	8.3
	KT03	5.476	103.298	04 July 2018	46.6	60.6	32.5	28.0
	KT04	5.403	103.340	04 July 2018	44.4	26.0	52.8	21.3
Marang	MG01	5.293	103.378	05 July 2018	42.1	5.5	63.7	30.8
	MG02	5.224	103.408	05 July 2018	44.7	1.6	65.6	32.8
	MG03	5.143	103.445	05 July 2018	41.8	2.5	67.3	30.2
	MG04	5.068	103.480	05 July 2018	41.4	20.7	57.6	21.8
Redang - Bidong	RB01	5.690	103.110	09 Aug. 2018	42.1	65.5	31.9	2.7
	RB03	5.747	102.941	09 Aug. 2018	34.8	69.9	28.4	1.7
Dungun	DG02	4.906	103.554	03 Oct. 2018	44.2	39.3	45.5	15.2
	DG03	4.796	103.610	03 Oct. 2018	40.5	62.2	27.6	10.3
Paka - Kerteh	PK01	4.611	103.626	04 Oct. 2018	39.6	15.4	53.6	31.0
	PK02	4.525	103.655	04 Oct. 2018	39.0	5.6	70.1	24.3
	PK03	4.439	103.632	04 Oct. 2018	37.8	4.1	68.7	27.2

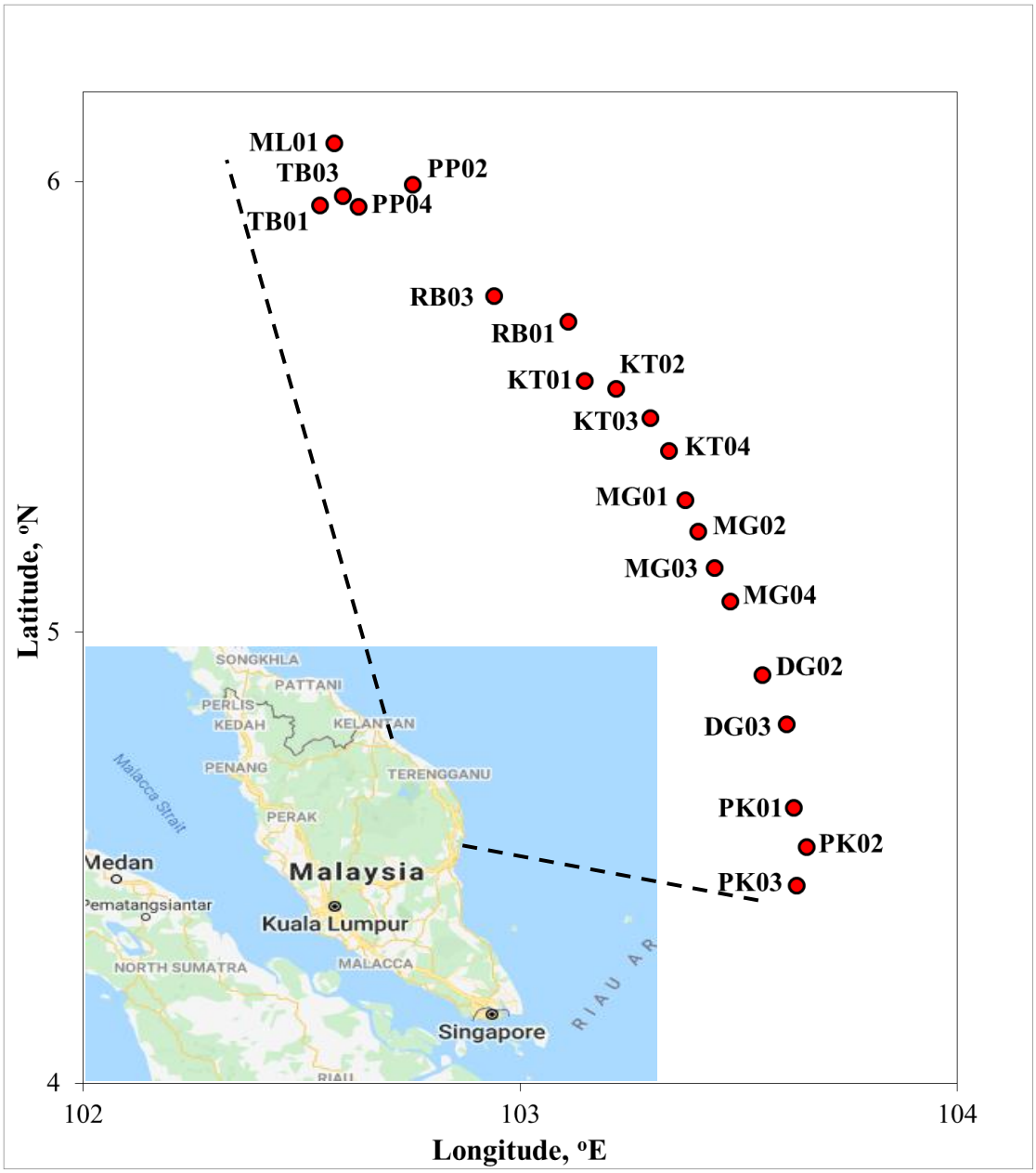

Figure 1: Map shows the points for sediment sampling

Spontaneous auto-deposition

3.3 mL of concentrated HCl was added to the dried residue and let it for a while until totally dissolved and dissolution. Then, the solution was transferred into the plating jar and added with distilled water until mark up of 80 mL. One (1) mL of stable Bi carrier (10 mg/g) solution and 1 g of hydroxylammoniumchloride were added into the solution and was heated gently until all precipitate dissolve. Polished silver disks were mounted in the plating holder and were put slowly and carefully into plating jar to avoid the solutions are splashed out. The plating jars were placed on a magnetic stirrer hot plate, stirred and heated at a temperature of 85°C for four hours. After the plating process is completed, the plating holders were taken out, washed with distilled water and rinsed with ethanol. Lastly, the discs were air dried (Zal U'yun et al., 2019).

Sample Counting

All samples were counted for 54000 seconds using High-Purity Germanium (HPGe) gamma-ray spectrometer. Their activity concentrations were corrected to the date of sampling (Noureddine and Baggoura, 1997). Counting times were long enough to ensure a 2σ counting error of less than 10%. Under the assumption that secular equilibrium was reached between ^{238}U, ^{232}Th, ^{226}Ra with their progenies, thus their activity concentrations were calculated through the photopeaks of their progenies. The γ-ray transitions to measure those concentrations of the assigned nuclides in the series are as follows:

(i) ^{226}Ra (186.21 and 241.98 keV), ^{214}Pb (295.21, 351.92 keV) and ^{214}Bi (609.31 keV) for uranium-238 (IAEA, 1989).

(ii) ^{214}Pb (295.21 and 351.92 keV) and ^{214}Bi (609.31 keV) for ^{226}Ra (Harb et al., 2008; IAEA, 1989).

(iii) ^{208}Tl (583.19 keV), ^{212}Pb (238.63 and 300.09 keV) and ^{212}Bi (727.3 keV) for ^{232}Th (Harb et al., 2008).

The HPGe detector was characterized to provide 25% relative efficiency and 1.8 keV at FWHM for the 1332 keV gamma ray line of ^{60}Co. It was calibrated using a customized gamma multinuclides standard solution which comprise of ^{210}Pb, ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m} Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co- in the same geometry with the samples. Source used was purchased from Isotope Products Laboratories, USA (source no 1290-84). IAEA Soil-6 reference material in the same counting geometry was used to check energy and efficiency calibration for the system. The performance of this instrument is monitored regularly to ensure it is fit for purpose (Yii et al., 2009). After considering the volume and the counting time of the sample, the minimum detectable activities (MDA) for thrice radionuclides of ^{238}U, ^{232}Th and ^{226}Ra were quantified at 1.0, 1.0 and 0.5 Bq/kg dry wt. (dw.), respectively.

Meanwhile, the measurement of ^{210}Po particles was carried out by using alpha spectrometry system (Ortec, Ortate Plus) for 24 hours. Polonium-210 activity concentrations were corrected to the time of sample collection. The alpha spectrometry counting system was equipped with 450 mm2 active area with alpha Passivated Implanted Planar Silicon (PIPS) detectors. The relative efficiency of each detector is ~ 25% for a detector-to-source spacing of less than 10 mm. The background count for each spectroscopy channel was less than one count per hour for energies above 3 MeV. The system was calibrated for energy and efficiency using multinuclide calibration standards comprising of ^{234}U, ^{238}U, ^{239}Pu and ^{241}Am supplied by Analytics, USA (SRS 67943-121) (Yii and Zal U'yun, 2011). The MDA for ^{210}Po was quantified at 0.2 Bq/kg dw. in sediment.

Results and Discussion

Level, trends and distribution of ^{238}U, ^{232}Th, ^{226}Ra and ^{210}Po in surface sediment

The activity concentrations range and mean values (in brackets) of ^{238}U, ^{232}Th, ^{226}Ra and ^{210}Po (Bq/kg dw.) in marine surface sediment collected in the east coast of Peninsular Malaysia were 3.81 ± 0.29 – 32.58 ± 7.63 Bq/kg dw. (18.20 Bq/kg dw.) , 12.51 ± 1.65 – 63.29 ± 8.24 Bq/kg dw. (37.90 Bq/kg dw.), 7.21 ± 0.59 – 64.93 ± 7.63 Bq/kg dw. (36.07 Bq/kg dw.) and 22.28 ± 0.99 – 69.14 ± 3.09 Bq/kg dw. (45.71 Bq/kg dw.), respectively. In general, their trends and distribution can be illustrated as plotted in Figure 2. The highest level of all radionuclides observed at TB01 due to this point situated at coastal area around 9

km from mainland and relatively shallow water of 15.5 m. This showed more input of natural radionuclides supplied from terrestrial area to this point. Furthermore, sampling at this point was conducted after heavy rain and there has rough sea with strong wave and fast bottom stream as resulted actively resuspension and mixing of sediment. Therefore, these factors most probably affected the levels, trends and distribution of these radionuclides at this sampling point.

Generally, the mean levels of ^{238}U and ^{232}Th in sediments found in this study were relatively lower than the mean value of the ^{238}U and ^{232}Th reported by Nguyen and Nguyen (2003) in Vietnam marine environment that ranged at 27.88 – 54.80 Bq/kg dw. and 30.86 – 59.61 Bq/kg dw., respectively. Other than that, the results showed that the sediment levels relatively lower for ^{238}U and relatively higher for ^{232}Th compared to the worldwide average value of 35 Bq/kg and 30 Bq/kg for ^{238}U and ^{232}Th, respectively (UNSCEAR, 2000). Meanwhile, levels of ^{226}Ra and ^{210}Po were found to be slightly higher compared to the value of 32.5 Bq/kg (^{226}Ra) and 41.6 ± 12.5 Bq/kg dw. (^{210}Po) reported by Yu et al. (1994) and Meli et al. (2013), respectively. In relation to the wide variations and high of their levels are probably due to more input were supplied in the marine environment and other factors such as geographic locations, physicochemical, chemical and geo-chemical properties (El Mamoney and Khater, 2004; Khatir et al., 1998b) were affected the levels of those radionuclides. The difference between the high activity concentrations of radionuclides in surface sediment in present work and other studies in the literature might be due to the fact that samplings were realized near the shore (Masqué et al., 2002). Furthermore, since sea is a very dynamic media, thus there are several factors that continuously change their concentrations, such as biogeochemical and biologic cycling pathways in the marine environment, scavenging, feeding of biological, re-suspension of the sediment, chemical mobilisation includes ion-exchange, leaching, and dissolution and others (Rudjord et al., 2019; Ugur and Yener, 2009).

In this study ^{210}Po found to be higher compared to other radionuclides due to ^{210}Po as one of strong particle reactive radionuclides, thus it is more rapid processes of scavenging and removal from water column to inorganic and organic particles, than deposited onto sediment. Furthermore, the facts of the change of chemical form of radionuclides due to enter in the marine environment and their different biological self-live are probably affected the level of activity concentrations (Meli et al., 2013). Moreover, other factor is mainly due to ^{210}Po half-life is much shorter compared to that of its grandparents of ^{226}Ra and ^{238}U; and ^{232}Th which are actively and fast regenerated by its grandparents in sediment (Masqué et al., 2002). Meanwhile, the level of ^{226}Ra is found to be higher than ^{232}Th and ^{238}U concentration in the sediment due to the high geochemical mobility and solubility nature of ^{226}Ra in water.

Generally, the trends and distribution of all radionuclides (Figure 2) found to be higher towards the coastal area. This finding strictly supported by Meli et al. (2013) that reported ^{210}Po concentration does not decline with increasing distance from the coast. This is as a resulted of terrestrial play an important role as a main source for supplying these radionuclides to marine environment. The similarity trends can be seen in Figure 2 whereby it can be proven that there have a significant positive correlation between radionuclides except ^{210}Po with Pearson correlation coefficient, r = 0.8285, 0.6859 and 0.5772 for ^{238}U-^{232}Th, ^{238}U-^{226}Ra and ^{232}Th-^{226}Ra, respectively (Figure 3). These good relationships reflected that ^{238}U, ^{232}Th and ^{226}Ra were supplied from the same origin source of terrestrial to study area and they were also same environmental geochemical origin. It was also reflected to high input of ^{232}Th from land towards to enrich of ^{232}Th in the sediment. Likewise, this finding strictly supported

that ^{232}Th is said to be an element which is mostly associated with terrestrial materials Other suggesting, the study areas received more input detrital river and near-shore sediment particles (Sakaguchi et al., 2004) with high level of ^{238}U, ^{232}Th and ^{226}Ra.

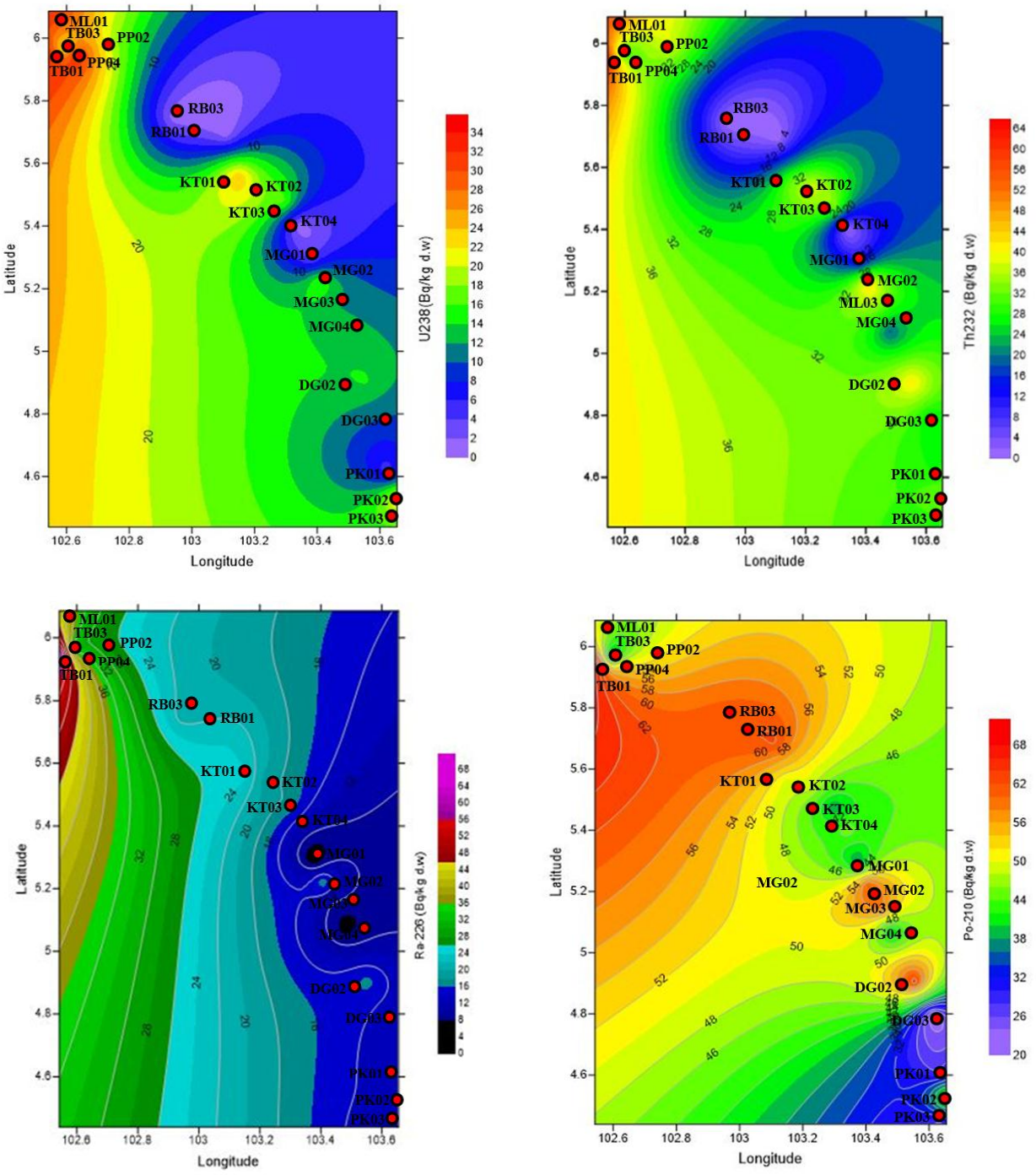

Figure 2: Trends and distribution of ^{238}U, ^{232}Th, ^{226}Ra and ^{210}Po in marine surface sediment at studied area

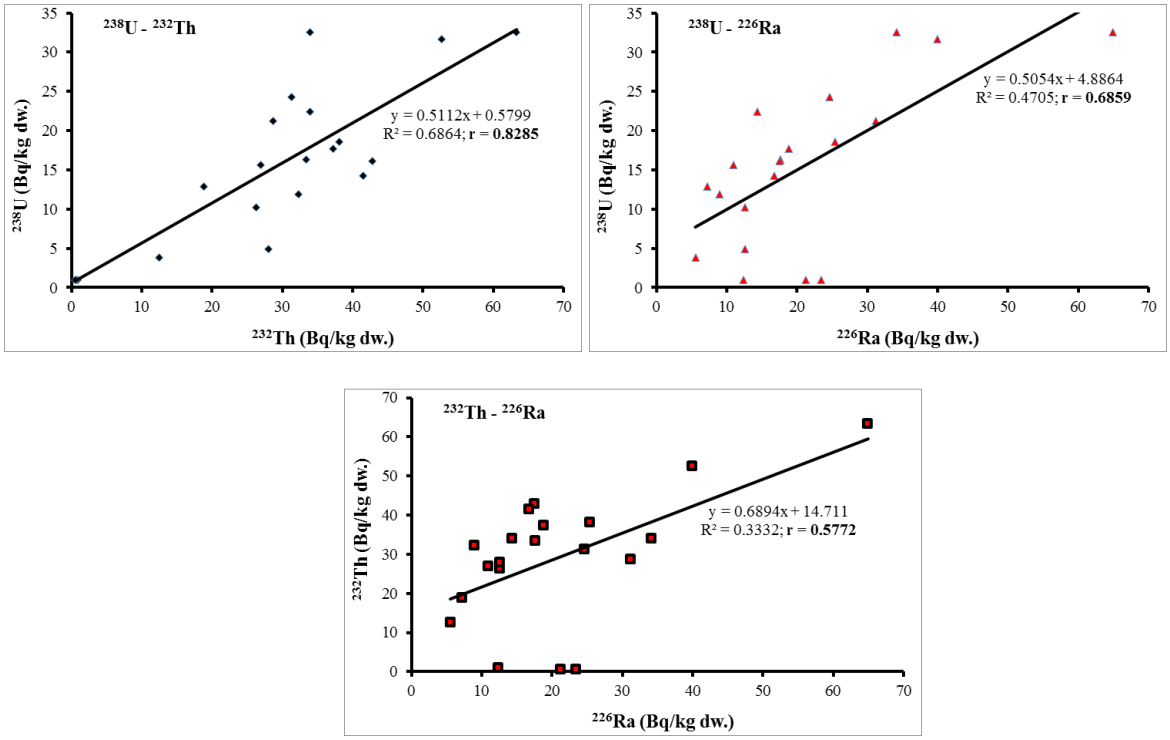

Figure 3: Significant correlation between radionuclides in sediment

Relationship curves show that strong positive correlation existed between the two radionuclides of ^{238}U and ^{226}Ra indicating they have similar behavior and characteristics in the marine environment. Furthermore, they were confirmed in the same radioactive decay series among natural radionuclides of ^{238}U and ^{226}Ra due to strong correlation that existed between them, while the behavior of ^{232}Th was identified differ due to moderate correlation with the ^{226}Ra. Ideally, a positive correlation obtained may be attributed to the retaining the sediment of these radionuclides under different seawater situations and different potential process occurred in water column before deposited onto sediment.

Statistical correlation analyses were carried out in order to clarify the relationship among the sediment variables, in particular the influence of sediment parameters on the distribution of natural radionuclides (Sivakumar et al., 2014). Normally, the relationships between the activity concentrations of the radionuclides and the clay content indicate that radionuclide activities tend to increase with increasing clay contents and adsorb to clay surfaces or become fixed within the lattice structure (Vanden Bygaart and Protz, 1995). Apparently, natural radionuclides were most abundant in the fine sediment fraction because they associated with clay. However, in this study is only distribution of ^{226}Ra which has a moderate negatively correlated (r = 0.6835) with clay content due to most of the sediment samples consist less clay with the ranged of 0.4 – 32.8% (mean: 16.6%) (Figure 4). Therefore, the range of values was not sufficiently wide to produce strong correlations with the radionuclides. Such some clay have accumulated in the deepest layers and ^{226}Ra from bottom sediment which associated with clay is most probably transported to surface layer during re-suspension of sediment as a resulted high level of this radionuclides in the surface layer. In the case of radionuclides were not correlated with fine sediment content i.e. silt and clay, it

can be explained that this feature of the radionuclides demonstrates the complexity of minerals in sediment samples (Suresh et al., 2014).

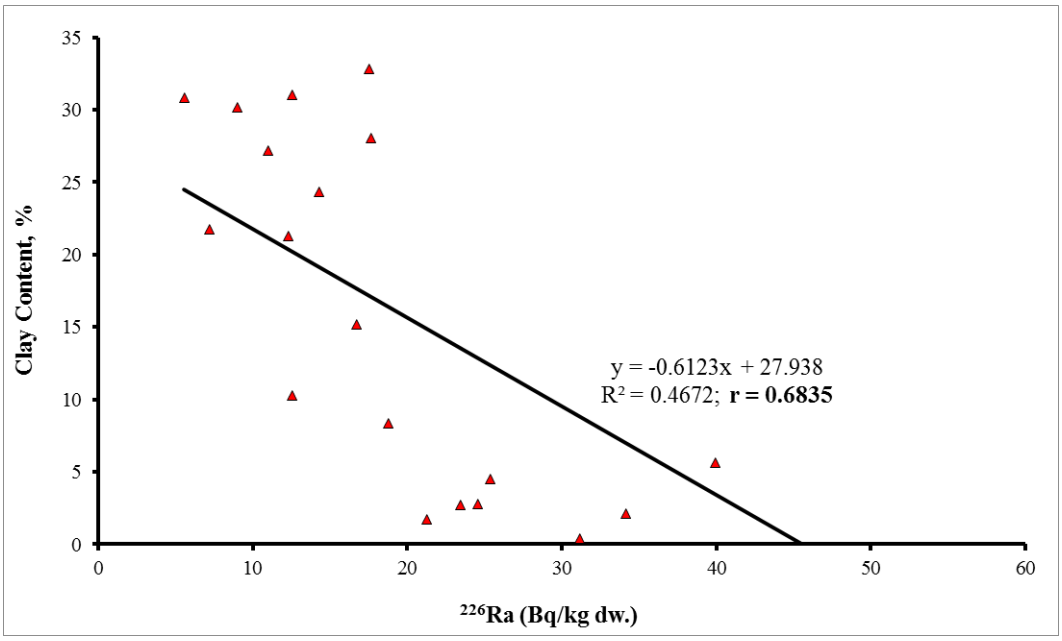

Figure 4: Relationship between ^{226}Ra (Bq/kg dw.) and clay content (%) in surface sediment

Various physico-chemical processes influenced the trends and distribution the radionuclides at the marine environment. The ^{238}U and ^{232}Th exhibited very similar spatial trends (Figure 2), which inferred a same origin source and similar responses to the sediment and marine environmental processes that affected their trends and distribution. In addition, ^{238}U was the highest level activity concentration on the coastal area and the distribution trend was quite similar to that of ^{232}Th. Thus, the similarity of the spatial trends might also explain the distributions of those radionuclides were controlled by physical processes such as sediment redistribution (deposition) and processes of sorption in the sediment. It is also can be related to the erosion of soil which are contained these radionuclides in the terrestrial area and transported by river stream and finally entered into the marine environment.

Radium equivalent activity concentration index

Due to the non-uniform distributions of radionuclides in sediment, radium equivalent activity concentration index, Ra_{eq} has been defined as a single radiological parameter that compares the specific activity of materials containing varying concentrations of ^{226}Ra (or ^{238}U), ^{232}Th and ^{40}K (Berekta and Matthew, 1985). In other word, the Ra_{eq} (Mahur et al., 2008) is a single index or number to describe the radiation output from different mixtures of those radionuclides in sediments samples from different locations and it is related to the external and internal dose due to radon and its daughters. In terms of the radiological health safety assessment, the maximum permissible limit of $Ra_{eq} \leq 370$ Bq/kg had been set for all materials (Berekta and Matthew, 1985; Sivakumar et al., 2014). It was calculated according to equation below:

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.07A_K \quad (1)$$

Where A_U, A_{Th} and A_K are the activity concentrations for ^{238}U, ^{232}Th and ^{40}K (Bq/kg), respectively. It has been assumed here that 370 Bq/kg of ^{238}U or 259 Bq/kg of ^{232}Th or 4810 Bq/kg of ^{40}K produces the same radiation dose rate. As can be seen in Table 2, the Ra_{eq} values for the sediment samples varied from 23.18 Bq/kg dw. to 138.36 Bq/kg dw.with the mean value of 80.77 Bq/kg dw. However, the calculated values of Ra_{eq} were below the suggested maximal admissible value of 370 Bq/kg at all sampling locations (Beretka and Matthew, 1985). From the radiological protection point of view, the sediments from the east coast of Peninsular Malaysia may not harm to the surrounding people, fisherman, divers or who-else presence in this area and these further confirm that the sediments are suitable to be used as building or road materials without any restrictions.

Evaluation of radiological hazard effect

i) Absorbed dose rate

The contribution of natural radionuclides to the absorbed dose rates depends on the concentrations of various radionuclides such as ^{226}Ra (^{238}U), ^{232}Th and ^{40}K in the sediment (Erenturk et al., 2014). The absorbed dose rates due to radiation of the uniform distribution of those radionuclides from the surface sediment were calculated based on the approach applied from the guidelines provided (UNSCEAR, 2000). The conversion factors used to compute absorbed dose rate (D_{adr}) in sediment per unit activity concentration in Bq/kg dw. corresponds to 0.462 nGy/h for ^{238}U, 0.604 nGy/h for ^{232}Th and 0.042 nGy/h for ^{40}K. Therefore, D_{adr} can be calculated as follows (UNSCEAR, 2000):

$$D_{adr\,(}nGy/h) = 0.462A_U + 0.604A_{Th} + 0.042A_K \quad (2)$$

Where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq/kg dw., respectively.

The absorbed dose rate (D_{adr}) values ranged of 10.20 – 62.45 nGy/h (mean: 36.33 nGy/h) (Table 2). The estimated mean value of D_{adr} contributed from the sediment collected at studied area is lower than the world average absorbed dose rate of 84 nGy/h. This might be due to there were no sources of monazite or minerals which are high content of U and Th deposited as well as no extra source of radiation from man activities that contributed a significant absorbed doses in the studied locations. The results for relationship of the radionuclides contributed to the dose in the sediments are shown in Table 3. The positive correlation was observed between ^{238}U and ^{232}Th with absorbed dose rate indicated that both radionuclides were mainly contributed to the absorbed dose in the sediment at all studied locations. Hence, this good strong relationship with r = 0.9469 showed that ^{232}Th as a key natural radionuclide played important role to contribute the emission of radiation in all the studied locations compared to ^{238}U.

Table 2: Activity concentration and radiological hazard parameters of natural radionuclides in surface sediment

Location	Station ID	Activity Concentration (Bq/kg dw.)					Radium Equivalent Activity Concentration Index, Ra_{eq} (Bq/kg dw.)	Absorbed Dose Rate, D_{adr} (nGy/h)	Annual Effective Dose Rate, D_{aedr} (mSv/y)	External Hazard Index, H_{ex}
		^{238}U	^{232}Th	^{226}Ra	^{210}Po	^{40}K				
Melawi	ML01	31.68 ± 3.57	52.59 ± 6.94	39.91 ± 5.21	48.21 ± 2.15	162.85 ± 11.17	118.28	53.24	0.07	0.32
Tok Bali	TB 01	32.58 ± 3.70	63.29 ± 8.24	64.93 ± 7.63	69.14 ± 3.09	218.25 ± 14.52	138.36	62.45	0.08	0.38
	TB03	21.19 ± 2.98	28.71 ± 3.77	31.14 ± 3.75	35.86 ± 1.60	99.18 ± 6.85	69.19	31.30	0.04	0.19
Pulau Perhentian	PP02	18.60 ± 3.75	38.12 ± 5.03	25.39 ± 3.12	51.15 ± 2.29	272.08 ± 18.49	92.16	43.05	0.05	0.25
	PP04	32.50 ± 2.66	33.93 ± 4.48	34.11 ± 4.13	51.58 ± 2.31	110.59 ± 7.67	88.76	40.15	0.05	0.24
Kuala Terengganu	KT01	24.32 ± 3.24	31.28 ± 4.31	24.58 ± 0.54	49.21 ± 2.20	326.76 ± 6.51	91.92	43.85	0.05	0.25
	KT02	17.73 ± 0.48	37.26 ± 4.92	18.80 ± 0.61	49.13 ± 2.20	321.97 ± 6.57	93.55	44.22	0.05	0.26
	KT03	16.30 ± 0.52	33.38 ± 4.42	17.63 ± 0.52	38.17 ± 1.71	324.53 ± 6.23	86.75	41.32	0.05	0.24
	KT04	9.76 ± 2.05	18.51 ± 2.44	12.32 ± 0.30	45.42 ± 2.03	255.30 ± 5.11	54.10	26.41	0.03	0.15
Marang	MG01	3.81 ± 0.29	12.51 ± 1.65	5.58 ± 0.51	37.52 ± 1.68	21.10 ± 1.72	23.18	10.20	0.01	0.06
	MG02	16.10 ± 1.19	42.84 ± 5.65	17.53 ± 0.69	60.42 ± 2.70	323.15 ± 8.41	99.98	46.89	0.06	0.28
	MG03	11.94 ± 1.39	32.31 ± 4.62	9.01 ± 0.83	58.79 ± 2.63	265.00 ± 20.91	76.69	36.16	0.04	0.21
	MG04	12.90 ± 1.72	18.80 ± 2.80	7.21 ± 0.59	38.68 ± 1.73	91.52 ± 7.34	46.19	21.16	0.03	0.13
Redang-Bidong	RB01	18.59 ± 3.90	32.25 ± 4.58	23.46 ± 0.61	62.47 ± 2.79	425.11 ± 8.33	97.00	46.92	0.06	0.27
	RB03	16.83 ± 3.72	31.91 ± 4.15	21.24 ± 0.56	61.69 ± 2.76	322.24 ± 6.86	72.00	34.57	0.04	0.20
Dungun	DG02	14.28 ± 0.39	41.45 ± 5.74	16.71 ± 2.77	63.82 ± 2.85	228.22 ± 38.70	89.53	41.22	0.05	0.25
	DG03	10.19 ± 1.31	26.31 ± 3.47	12.54 ± 0.31	22.28 ± 0.99	194.32 ± 9.85	61.42	28.76	0.04	0.17
Paka-Kerteh	PK01	4.88 ± 0.36	27.98 ± 3.69	12.58 ± 0.34	27.67 ± 1.23	166.67 ± 7.01	56.56	26.15	0.03	0.16
	PK02	22.37 ± 2.87	33.96 ± 4.48	14.31 ± 0.41	44.73 ± 2.00	105.59 ± 6.32	78.32	35.28	0.04	0.21
	PK03	15.62 ± 0.45	26.97 ± 3.94	10.98 ± 0.89	26.83 ± 1.20	246.67 ± 4.98	71.45	33.87	0.04	0.20

Table 3: Pearson's correlation coefficient (r) for relationship of radiological parameter and radionuclides

	Activity Concentration (Bq/kg dw.)		
	^{238}U	**^{232}Th**	**^{40}K**
Radium equivalent activity concentration index, Ra_{eq} (Bq/kg dw.)	r = 0.8712 (+ correlation)	r = 0.9678 (+ correlation)	r = 0.0671 (+ correlation)
Absorbed dose rate, D_{adr} (nGy/h)	r = 0.8484 (+ correlation)	r = 0.9469 (+ correlation)	r = 0.1503 (+ correlation)
Annual effective dose rate, D_{aedr} (mSv/y)	r = 0.8484 (+ correlation)	r = 0.9469 (+ correlation)	r = 0.1503 (+ correlation)
External hazard index, H_{ex}	r = 0.8645 (+ correlation)	r = 0.9633 (+ correlation)	r = 0.0894 (+ correlation)

ii) Annual effective dose rate

The annual effective dose rate, D_{aedr} (mSv/y) resulting from the absorbed dose values (D_{adr}) was simplified and calculated based on the following equation given by Ravisankar et al. (2012) and UNSCEAR (2000):

$$D_{aedr}\ (mSv/y) = D_{adr}\ nGy/h \times 8760\ h \times 0.7\ Sv/Gy \times 10^{-6} \quad (3)$$

The calculated D_{aedr} values are ranged from 0.01 – 0.08 mSv/y with a mean value of 0.05 mSv/y, which is slightly lower than the world average value of 0.07 mSv/y. The differences arising from these may be due to the influence of the geo-chemical properties of radionuclides and geological settings of the area, this vary from one place to another and from one locality to another even within the same region (Usikalu et al., 2014). As absorbed dose (D_{adr}), definitely annual effective dose rate by the human beings also due to high concentration of ^{232}Th. This finding is strictly supported by Pearson's correlation coefficient as summarized in Table 3. Furthermore, the finding also indicated that relatively higher level of ^{232}Th (mean value: 31.90 Bq/kg dw.) exist than ^{238}U (mean value: 16.79 Bq/kg dw.) in sediment. This is could be due to the presence of the input detrital mineral particles associated with sediments at studied locations which have relatively high level of ^{232}Th.

ii) External hazard index

In order to assess the health effects from the radioactivity of the earth's surface materials containing ^{238}U, ^{232}Th and ^{40}K to the people at surrounding marine environmental, a single quantity termed as external hazard index (H_{ex}) should be quantified (Suresh et al., 2014). For the radiation hazard to be negligible and for the purpose of radiological safety precautions, the H_{ex} should be less than unity (1). Radiation hazard index deals with the assessment of excess radiation originating from the sediments at the studied locations were defined as:

$$H_{ex} = (A_U/370\ Bq/kg) + (A_{Th}/259\ Bq/kg) + (A_K/4810\ Bq/kg) \quad (4)$$

Where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively. This index value must be less than unity in order to keep the radiation hazard to be insignificant.

The calculated external hazard values are ranged between 0.06 and 0.38 with a mean value of 0.22 (Table 2). The mean value of the external hazard index of 0.22 at all sampling locations is less than the recommended limit of unity (1). This deterioration values in these locations are due to the lower activity concentration of ^{238}U, ^{232}Th and ^{40}K. This relatively lower contribution to the hazard index due to the ^{238}U is lower followed by the contributions of ^{232}Th and ^{40}K. Furthermore, the less value of H_{ex} due to ^{40}K was not from the same origin source as ^{238}U and ^{232}Th. It has been proven that there is no significant and negative correlation between ^{40}K-^{238}U ($r = 0.2298$) and ^{40}K-^{232}Th ($r = 0.1192$). In estimating of H_{ex} found that ^{238}U and ^{232}Th are contributed to the significant value as a result there has a strong positive correlation with $r = 0.8285$ (Figure 3) between both radionuclides in the sediments. This can be implied that very strong relationship shows both ^{238}U and ^{232}Th contributed the emission of significant radiation to all locations.

On the other word, the high good positive correlation was observed between ^{238}U and ^{232}Th because uranium and thorium come from decay series and occur together in nature (Irena et al., 2012). While, very weak negative correlation was observed between ^{40}K-^{238}U and ^{40}K-^{232}Th due to ^{40}K origin is a primordial radionuclide and totally was not come from different decay series, which does not undergo any radioactive decay process (Chandrasekaran et al., 2014). This radionuclide was also in accordance with the results (Chen et al., 2001; Elejalde et al., 1996). Therefore, pre conclusion can be made based on our finding that these study areas confirmed not pose radiological risks to the surrounding people, fisherman, divers or who-else presence in this area owing to harmful effects of ionizing radiation from the natural radionuclides in sediment.

Conclusion

The radioactivity concentrations level of ^{238}U, ^{232}Th, ^{226}Ra and ^{210}Po in surface sediment collected in the east coast of Peninsular Malaysia were ranged of $3.81 \pm 0.29 – 32.58 \pm 7.63$ Bq/kg dw., $12.51 \pm 1.65 – 63.29 \pm 8.24$ Bq/kg dw., $7.21 \pm 0.59 – 64.93 \pm 7.63$ Bq/kg dw. and $22.28 \pm 0.99 – 69.14 \pm 3.09$ Bq/kg dw., respectively. Meanwhile, their radioactivity trends and distribution found to be higher toward the coastal area. This result due to terrestrial play an important role as a main source for supplying these radionuclides to studied area. The mean radium equivalent activity concentration index and other radiological hazard parameter were lower than their maximum permissible limits. Therefore, the sediments in the east coast of Peninsular Malaysia confirmed were not posed radiological risks to the surrounding people, fisherman, divers or who-else presence in this area owing to harmful effects of ionizing radiation from the natural radionuclides in sediment. Furthermore, these sediments are suitable and safe to use as road or building construction materials.

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