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NORM activity concentration in sediment cores from the Peninsular Malaysia East Coast Exclusive Economic Zone

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Abstract Study for distribution of Naturally Occurring Radioactive Materials (NORM) i.e. ^{226}Ra, ^{228}Ra and ^{40}K in the east coast of Peninsular Malaysia Exclusive Economic Zone (EEZ) was carried out as part of the national marine environment project. Sixteen marine sediment cores from selected locations within the EEZ were collected for determination of NORM activity concentrations using high-purity germanium (HPGe) gamma spectrometer. From the measurement, the activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K is ranged from 16 ± 4 Bq/kg to 46 ± 6 Bq/kg (total mean 30), 28 ± 7 Bq/kg to 87 ± 11 Bq/kg (total mean 56) and 171 ± 33 Bq/kg to 690 ± 89 Bq/kg (total mean 420), dry wt., respectively. The activity concentrations of radionuclides in most of the core were quite uniform suggesting that there were thorough vertical mixed of sediment throughout the core. The results obtained were also in good agreement with those previous reported from other countries in the region and therefore can be used to enhance present radioactivity database. The calculated external hazard values were ranged from 0.25 to 0.51 with the mean of 0.38 (less than unity) showed little risk of external hazard to the workers handling the sediments and it was likely low level of the mainland natural gamma-radiation in the east coast of Peninsular Malaysia.

Keywords NORM · Exclusive Economic Zone · External hazards value

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Introduction

Radioactivity monitoring in marine environment normally based on analyses of specific nuclides such as NORM in seawater, suspended particulate matters and sediments [1]. Two main sources of marine radioactivities are coming from the weathering and mineral recycling of the terrestrial rocks.

Thorium-232 (^{232}Th), uranium-235 (^{235}U) and uranium-238 (^{238}U) are three major radionuclides precursor in the decay series, which produces other radionuclides such as radium, radon, actinium, protactinium, lead and polonium. These progenies first appear in the lithosphere level may contaminate ground, water, air and the biological organisms. Most of the radioactivity deposited on the surface soil is washed and drained through rivers transport and finally ended in the estuary and entering the marine environment and also through several pathways such as weathering, erosion, fallout, rainwater and human activities [2–6].

Natural existing primordial radionuclides such as uranium-238 ($T_{1/2} = 4.47 \times 10^9$ years) series, thorium-232 ($T_{1/2} = 1.41 \times 10^{10}$ years) series and potassium-40 ($T_{1/2} = 1.277 \times 10^9$ years) are present everywhere in the earth's crust far before mankind and make the biggest contribution to the total background dose [1]. Most of these radionuclides are long half-lives which will remain in the environment even after millions of years. Some of these radionuclides and their progenies are emitting gamma rays, which become one of the major sources for external exposure. Among those, radium-226 (^{226}Ra, uranium series progeny), radium-228 (^{228}Ra, thorium series progeny) and potassium-40 (^{40}K) are at most concern due to their high solubility and mobility.

Measurement of these three radionuclides can be carried out easily by using a high resolution HPGe detector gamma

(γ)-spectrometry that can determine a number of γ-ray emitting radionuclides simultaneously without requiring tedious and complicated chemical separation procedures. In most marine studies, HPGe γ-spectrometry has always be the first choice for the measurement of γ-ray emitting radionuclides [7–9]. Moreover, the activity concentration of the progenies from the natural decay series in the marine environment may not necessary be in equilibrium with its parents [10].

Normally, in coastal areas and on the continental shelf which are not very far from the land-based pollutants, is reasonable to assume that the sediments have a range of concentration of natural radionuclides similar to that of terrestrial rocks, and the radioactive equilibrium is maintained.

The knowledge of the activity concentrations and distributions of these radionuclides are of interest since it provides useful information in the monitoring of environmental contamination of natural radioactivity. The distribution of activity concentrations for ^{226}Ra, ^{228}Ra and ^{40}K on terrestrial especially in the areas of Peninsular Malaysia had been widely reported elsewhere [11–15]. In fact, several studies on NORM and their progenies had been also reported on the Malaysian marine ecosystems [16–20] but did not cover the whole Exclusive Economic Zone (EEZ).

EEZ in the east coast of Peninsular Malaysia is located on the 1°14.04′ to 7°48.92′ N latitude, and 102°5.03′ to 105°48.77′ E longitude. It has a length of approximately 1,150 km and a maximum width of 417 km, with an area of around 130,000 km^2. The zone is relatively shallow, with an average depth of 60–70 m deep. Furthermore, the seawater in the respective area is generally well mixed throughout the water column while the prevailing surface currents are closely associated with the monsoon seasons [21].

Experimental

Sampling, preparation and measurement

The east coast of Peninsular Malaysia EEZ was the designated area for this study. Samples were collected during sampling expedition on board the K.L. PAUS (owned by the Malaysian Fisheries Institute) from June 11th to 30th, 2008. The locations chosen were extended from previous study (28,052 km^2) [11], where the sampling locations for this study were covered an area of 140,972 km^2 [22]. In these area, a total of thirty locations had been identified to be a sampling point as can be seen in Fig. 1 and; location and sampling dates are given in Table 1.

The samplings were carried out systematically according to a grid within Malaysian EEZ which covered shallow coastal, near-shore and off shore zones of the east coast of Peninsular Malaysia. Physical and chemical parameters such as salinity, temperature and dissolved oxygen (DO) of the water column were also measured as supporting parameters (Table 1).

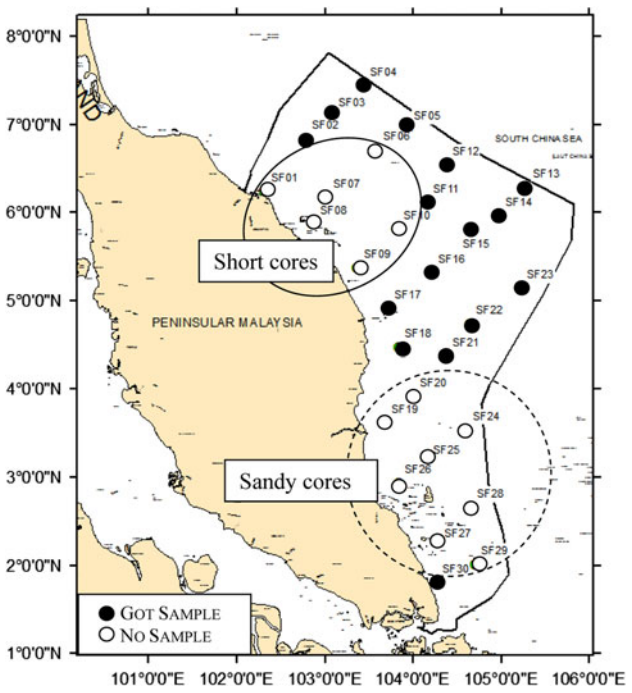


Fig. 1 Sampling locations in east coast of Peninsular Malaysia EEZ

The sediment cores were collected using a 12 cm diameter multicorer device and sliced into 2 cm sections at site and kept in a pre-weighted HDPE container. During the sampling expedition, some of the cores showed thorough vertical mixing of fine and sandy mud. However, some cores are too short (<20 cm) and too sandy as illustrated in Fig. 1. Therefore, only sixteen sediment cores with enough length and low sand content are being reported in this paper (marked • close circle in Fig. 1).

In the laboratory, all sediment samples were dried at 60 °C for minimum 72 h until reached a constant weight, then ground to pass through a 200-mesh sieve. Samples were then transferred into 200 ml marinelli beaker, sealed with thick PVC tape to inhibit radon from escaping. All samples were stored for a period in excess of 30 days [1, 23] to establish secular equilibrium between ^{226}Ra and ^{228}Ra and their respective radioactive progeny prior to gamma counting.

Then, all samples were counted for 86,400 s using high-purity germanium (HPGe) spectrometer and corrected to the date of sampling. Count times were long enough to ensure a 2σ counting error of less than 10%. The ^{226}Ra and ^{228}Ra were measured through the photopeaks of their progenies; mean activity of ^{214}Pb (295.22 keV and

Table 1 Surrounding physical parameters and coordinates of sampling locations

Station	Date	Latitude	Longitude	Water depth (m)	Surface seawater temperature (°C)	Distance from shore (nautical miles)
SF01	18.06.08	06°13.99′ N	102°19.00′ E	13.0	30.87	2.7
SF02	17.06.08	06°50.04′ N	102°47.04′ E	46.5	30.51	50
SF03	17.06.08	07°05.03′ N	103°04.99′ E	50.0	30.32	73
SF04	17.06.08	07°25.98′ N	103°26.01′ E	61.0	30.03	100
SF05	16.06.08	06°56.09′ N	103°56.04′ E	52.0	30.01	108
SF06	16.06.08	06°42.14′ N	103°35.17′ E	52.0	30.13	80
SF07	16.06.08	06°10.00′ N	103°01.00′ E	45.0	30.25	40
SF08	18.06.08	05°52.10′ N	102°51.92′ E	34.0	30.41	15
SF09	20.06.08	05°22.06′ N	102°21.97′ E	47.0	28.98	14
SF10	14.06.08	05°48.20′ N	103°48.98′ E	55.0	30.27	48
SF11	14.06.08	06°06.16′ N	104°09.11′ E	72.0	30.04	75
SF12	14.06.08	06°32.01′ N	104°22.11′ E	59.0	29.58	101
SF13	13.06.08	06°16.98′ N	105°16.99′ E	55.0	29.87	139
SF14	13.06.08	05°57.15′ N	104°58.13′ E	56.0	29.63	115
SF15	12.06.08	05°29.08′ N	104°29.02′ E	60.7	29.62	80
SF16	12.06.08	05°18.50′ N	104°12.60′ E	60.0	29.65	56
SF17	20.06.08	04°54.12′ N	103°42.98′ E	54.0	29.93	17
SF18	11.06.08	04°28.14′ N	103°49.98′ E	40.0	29.79	20
SF19	22.06.08	03°37.07′ N	103°41.08′ E	23.0	29.50	15
SF20	22.06.08	03°55.10′ N	104°00.05′ E	50.0	29.74	40
SF21	23.06.08	04°22.16′ N	104°22.07′ E	65.0	29.63	52
SF22	23.06.08	04°44.19′ N	104°38.44′ E	66.0	29.63	70
SF23	12.06.08	05°08.10′ N	105°12.90′ E	67.2	29.89	109
SF24	23.06.08	03°32.08′ N	104°36.00′ E	62.0	29.75	70
SF25	24.06.08	03°09.14′ N	104°09.04′ E	41.0	29.03	42
SF26	26.06.08	02°56.13′ N	103°49.97′ E	20.0	29.44	24
SF27	24.06.08	02°16.94′ N	104°16.97′ E	30.0	29.21	19.5
SF28	24.06.08	02°39.18′ N	104°38.91′ E	58.0	29.30	47
SF29	25.06.08	02°00.55′ N	104°41.97′ E	46.0	29.73	35
SF30	25.06.08	01°48.04′ N	104°15.03′ E	14.0	29.47	4.5

351.93 keV) and ^{214}Bi (609.31 keV) for ^{226}Ra [24, 25] and ^{228}Ac (911.20 keV) for ^{228}Ra [26, 27]. Meanwhile, ^{40}K content was measured directly via its 1460 keV energy peak [1, 24, 27].

Counting system

The high-purity germanium (HPGe) detector was characterized to provide 25% 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 source (comprising 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. This source was purchased from Isotope Products Laboratories, USA (source no. 1290-84). Reference material (IAEA, Soil-6) in the same counting geometry (hence efficiency), was used to check the energy and the efficiency calibration for the system. The performance of this instrument is monitored regularly to ensure it is fit for the purpose [28]. The radioactivity concentrations in the environmental samples were calculated using equation as reported in previous paper [1, 29].The minimum detectable activity (MDA) for both ^{226}Ra and ^{228}Ra was quantified at 2 Bq/kg per dry weight (dry wt.), while ^{40}K was quantified at 5 Bq/kg after considering the size and counting time of the sample.

Results and discussion

Surrounding physical conditions at sampling locations were summarized in Table 1. The activity concentration

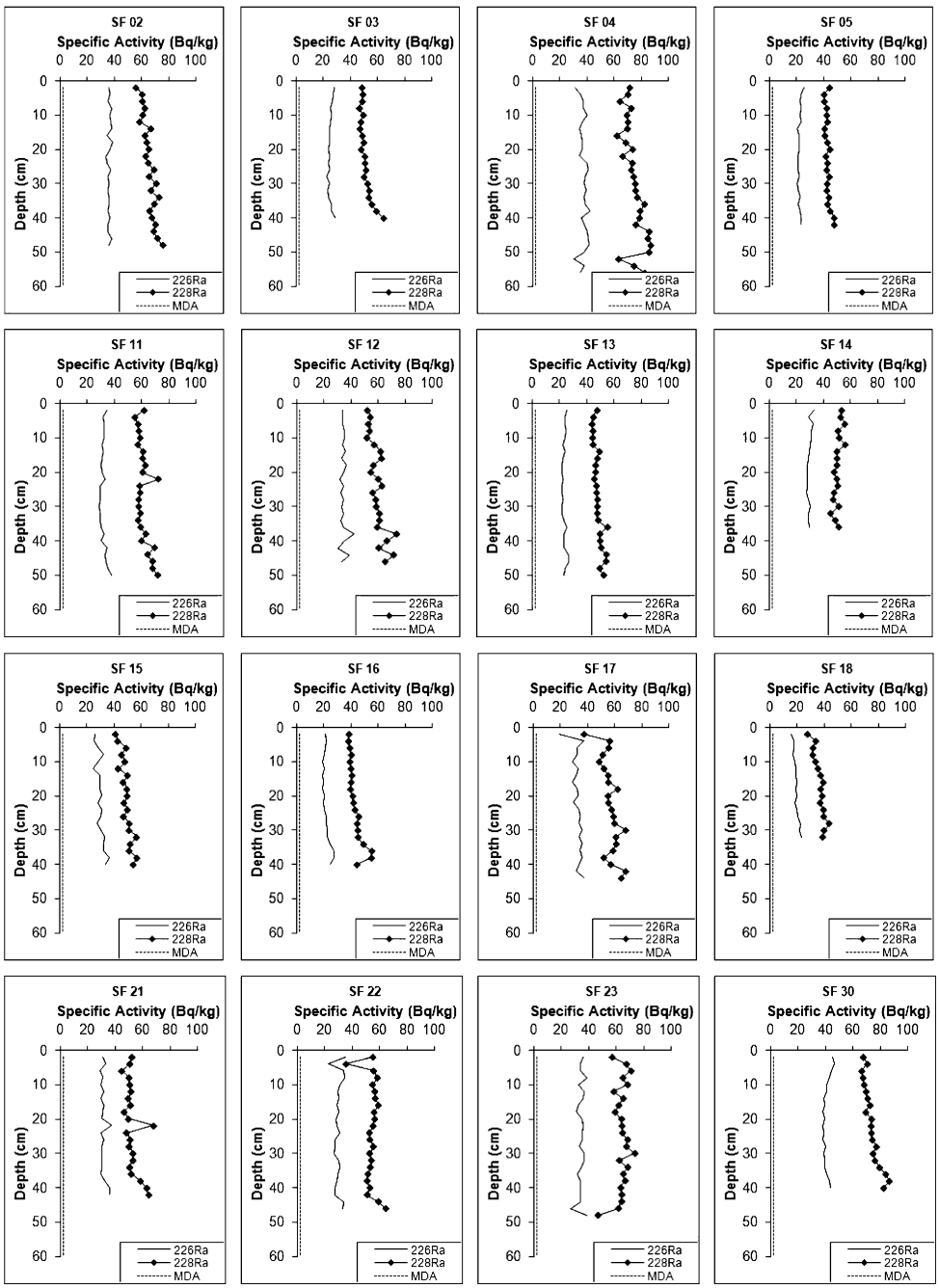


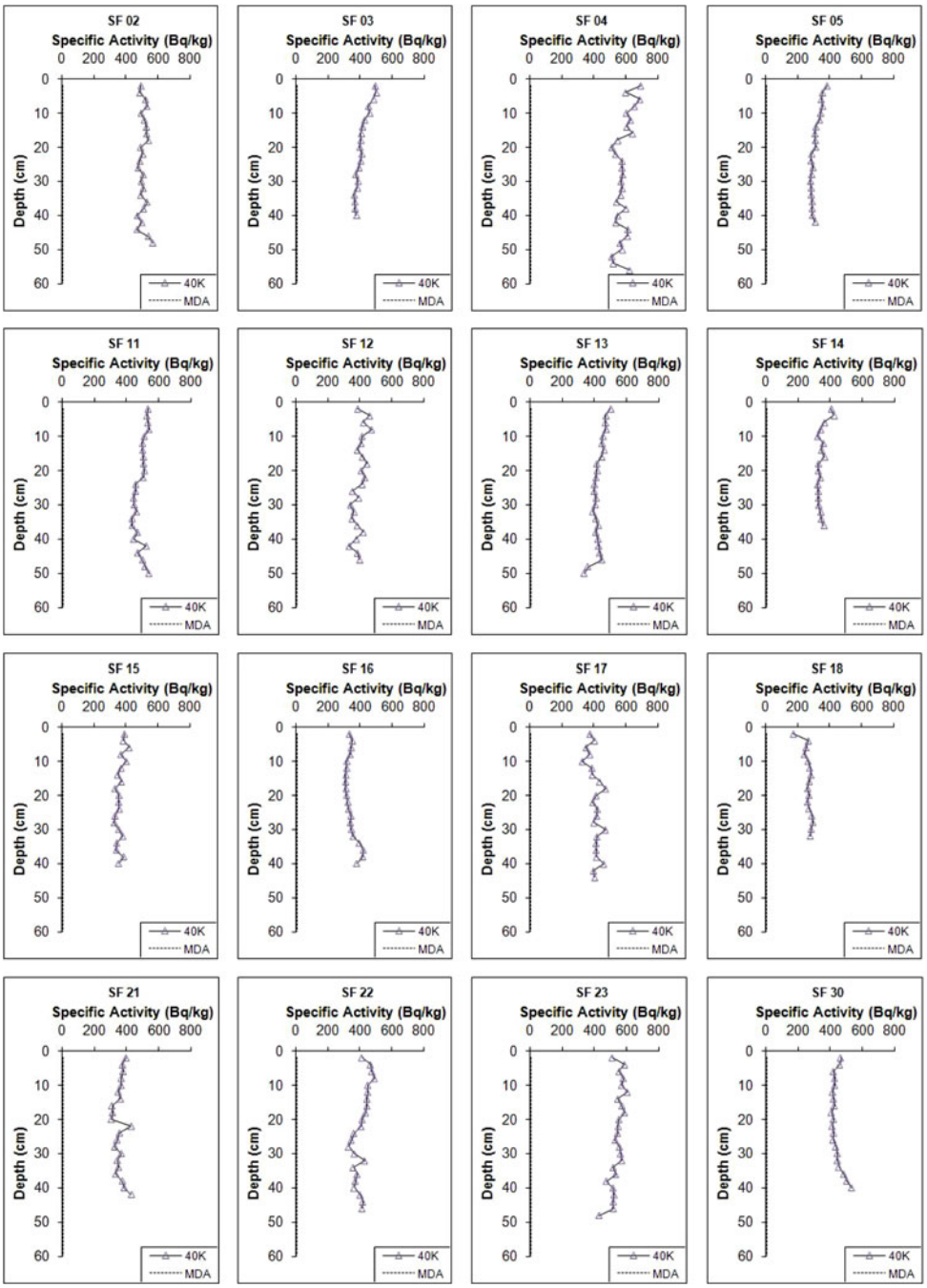
Fig. 2 Vertical profile of ^{226}Ra, ^{228}Ra and ^{40}K activity concentration

levels of ^{226}Ra, ^{228}Ra and ^{40}K in the marine sediment core in this study are shown in Figs. 2, 3 and summarized in Table 2. All the values of samples are found to be greater than the minimum detectable activity (MDA). It is evident that the activity concentrations of naturally occurring radionuclides vary significantly from place to place indicating their presence in the marine environment depends on their physical, chemical and geochemical properties and the pertinent environment in the biological process [30].

Generally, the activity concentration of ^{226}Ra is ranged between 16 and 46 Bq/kg with a mean value of 30 ± 6 Bq/kg; the activity of ^{228}Ra varies from 28 to 87 Bq/kg with a mean value of 56 ± 11 Bq/kg; and that of ^{40}K from 171 to 690 Bq/kg with a mean of 420 ± 90 Bq/kg. The large area of sampling probably could be explained this observation. The activity concentrations of radionuclides in most of the cores were quite uniform suggesting that there were thorough vertical mixing of sediment throughout the cores. However, the activity concentrations in some cores appear to be higher compared to other stations.

Higher activities of ^{226}Ra were found in stations SF02, SF04, SF11, SF12, SF17, SF21, SF22, SF23 and SF30, with

Fig. 2 continued



their average activity recorded were 36.22 Bq/kg, 37.00 Bq/kg, 31.33 Bq/kg, 34.12 Bq/kg, 31.66 Bq/kg, 31.06 Bq/kg, 30.07 Bq/kg, 34.67 Bq/kg and 40.55 Bq/kg, respectively. While station SF02, SF04, SF11, SF12, SF17, SF22, SF23 and SF30 were also recorded higher activity for ^{228}Ra with their average activity of 63.51 Bq/kg, 70.44 Bq/kg, 59.81 Bq/kg, 57.05 Bq/kg, 55.53 Bq/kg, 54.14 Bq/kg, 64.97 Bq/kg and 71.15 Bq/kg, respectively. Meanwhile, higher activity concentrations of ^{40}K were recorded at station SF02, SF03, SF04, SF11, SF13 and SF23 in respectively with 505.28 Bq/kg,

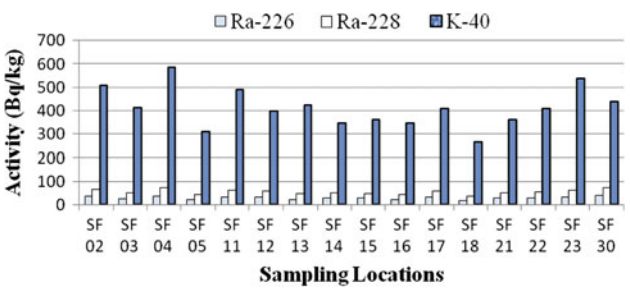


Fig. 3 ^{226}Ra, ^{228}Ra and ^{40}K mean activity concentration obtained in this study

Table 2 Specific activity and external hazard index in the east coast Peninsular Malaysia EEZ sediment cores

Station	Specific activity (Bq/kg, dry wt.)						$^{228}Ra/^{226}Ra$ ratio[a]	H_{ex}[a]
	^{226}Ra		^{228}Ra		^{40}K			
	Range	Mean	Range	Mean	Range	Mean		
SF 02	34–39	36 ± 1	56–76	66 ± 5	468–567	506 ± 25	1.83	0.46
SF 03	22–29	25 ± 2	46–64	51 ± 4	360–499	411 ± 43	2.04	0.35
SF 04	30–43	38 ± 3	62–87	75 ± 7	510–690	584 ± 47	1.97	0.51
SF 05	20–25	22 ± 1	40–48	43 ± 2	278–381	310 ± 30	1.95	0.29
SF 11	29–38	32 ± 2	55–72	61 ± 5	431–537	488 ± 36	1.91	0.42
SF 12	31–42	34 ± 2	52–74	60 ± 6	331–472	397 ± 37	1.76	0.41
SF 13	22–27	23 ± 1	44–55	48 ± 3	335–498	423 ± 36	2.09	0.34
SF 14	27–33	30 ± 2	45–56	51 ± 3	320–422	346 ± 28	1.70	0.35
SF 15	24–37	30 ± 3	41–57	49 ± 4	324–416	360 ± 25	1.63	0.35
SF 16	19–27	22 ± 2	38–55	43 ± 5	309–420	344 ± 34	1.95	0.30
SF 17	19–37	33 ± 4	38–68	57 ± 7	325–473	406 ± 35	1.73	0.39
SF 18	16–23	19 ± 2	28–43	36 ± 4	171–294	264 ± 28	1.89	0.25
SF 21	29–38	31 ± 2	45–68	53 ± 6	305–427	358 ± 33	1.71	0.36
SF 22	23–36	30 ± 3	36–64	54 ± 5	327–489	408 ± 44	1.80	0.38
SF 23	27–39	34 ± 2	47–74	64 ± 5	424–601	537 ± 39	1.88	0.45
SF 30	38–46	41 ± 3	66–86	74 ± 6	404–529	438 ± 33	1.80	0.49
Average	16–46	30 ± 6	28–87	56 ± 11	171–690	420 ± 90	1.87	0.38

[a] $^{228}Ra/^{226}Ra$ ratio and H_{ex} value was calculated using the mean value of each sediment core

424.43 Bq/kg, 598.13 Bq/kg, 495.83 Bq/kg, 436.19 Bq/kg and 556.05 Bq/kg.

High activity of 86.49 ± 12.98 Bq/kg for ^{228}Ra and 46.48 ± 6.41 Bq/kg for ^{226}Ra was measured at station SF30 which is located at shallower water of 14 m and near to mainland (4.5 nautical miles). Therefore, this was probably due to this station received more input of sediments and particulate matters from terrestrial which are contained high activity concentration of radium [31]. Weathering process, shoreline erosion and rapidly development process along the shoreline make possible for radium leached out from all the processes occurred. On the other hand, due to shoreline erosion of the granite rocks and monazites; and fertilizer application in agricultural areas of drainage basin [32] would also encourage the increase of radium isotope concentration at station SF30.

Higher activity concentration of radionuclides in some locations may be attributed to geological areas consisting of granites and gneisses which contain higher concentration of uranium and thorium series as well as potassium. This assumption was supported in previous report that higher activity concentration of uranium, thorium and potassium in soil at Johore state were found in the area, which are covered by granites and metamorphic rocks [33].

The phenomena of changing in radionuclides distribution believed to be caused by several factors such as chemical and physical properties, precipitation, movement and transportation of the radionuclide. This statement was also agreed that the re-suspension, transportation and re-sedimentation of sediment when moving from higher ground or during flooding might be caused the inconsistency of the radium distribution [34]. On the other hand, the distributions of radium were also affected by two other main processes that the sedimentation of the suspended solid and re-suspension of the bottom sea sediments, in which both processes were, depends on the conditions such as current flow and choppiness of the river or sea [35].

The comparison of ^{226}Ra, ^{228}Ra and ^{40}K activity concentration with other reported values in other areas are summarized in Table 3. The activity concentration of ^{226}Ra and ^{228}Ra recorded in this study was comparable with the value reported from the nearby regions. However, the values were higher when compared to other areas of the world such as in Red sea [30], Sudan harbour [36] and Algerian coast [37] but ^{226}Ra and ^{228}Ra was much lower than those observed in such area [38, 39]. Meanwhile, the activity concentration of ^{40}K was lower compared to those reported in Vietnam [40] and Fujian [8] but much higher that reported in Margarita Island [41]. However, the level of ^{226}Ra, ^{228}Ra and ^{40}K activity concentrations in generally found were quite closed and comparable to the activity concentration in most environments of the world. Therefore, these values can be used as a reference for Malaysian marine environment.

Table 3 Comparison of activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K in marine sediment

Region	Activity concentration (Bq/kg)			Reference
	^{226}Ra	^{228}Ra	^{40}K	
EEZ Peninsular Malaysia	16–46 (30)	28–87 (56)	171–690 (420)	Present work
Coastal of Sarawak	23–41 (30)	27–45 (39)	142–680 (462)	[17]
Coastal of Sabah	16–30 (23)	23–45 (35)	402–842 (577)	[17]
South China Sea (Sarawak)	11–36 (22)	21–65 (39)	149–517 (309)	[17]
South China & Sulu Sea (Sabah)	9–31 (14)	10–48 (21)	140–580 (269)	[17]
East coast Peninsular Malaysia	46.23–121.49	–	–	[18]
Vietnam	27.88–54.80	30.86–59.61	477.4–1254	[40]
East Coast Peninsular Malaysia	19.84–73.57	35.16–92.95	–	[16]
West Coast Peninsular Malaysia	14.25–38.90	16.91–97.18	–	[16]
Vietnam	24.6–39.7	–	–	[43]
Xinghua Bay mouth, Fujian, China	22.4–27.9	54.4–71.9	684–795	[8]
North east coast of Tamilnadu, India	< 5.5–30.42 (8.39)	< 5.5–218.64 (24.52)	212–423 (275)	[39]
Gulf of Mexico	11.8–97.3 (28.9)	7.8–64.8 (33.3)	–	[42]
Red Sea	2.4–59.9 (11.6)	0.22–19.29 (6.02)	23.7–515.0 (158.4)	[30]
Port Sudan Harbour	5.00–13.90 (11.05)	4.47–12.99 (10.35)	95–434 (311)	[36]
Sawakin harbour	6.41–26.68 (12.61)	2.83–9.99 (6.18)	59–341 (192)	[36]
Algerian Coast	17–26	18–32	311–690	[37]
Margarita Island	–	–	12.2–211.7	[41]
Krka River estuary	45–662	–	–	[38]

Note: Values given in the parenthesis are the mean/median values

The computed statistical analyses of Pearson correlation for the mean values of the activity concentration of naturally occurring radionuclides are plotted in Fig. 4. The results found that there has a strongly significant correlated between ^{226}Ra and ^{228}Ra with the Pearson coefficient, $r = 0.95$ at 90% confident level. Meanwhile, ^{226}Ra and ^{228}Ra were also strongly significant correlated ($p < 0.010$) with ^{40}K with $r = 0.71$ and 0.86, respectively. According to previous reporter [44], those relationships revealed that both radium isotopes and ^{40}K were originated from the same source of environment and had same chemical properties as well as their behaviors.

^{238}U and ^{232}Th commonly occur together in nature. This frequently leads to a relatively constant ^{238}U/^{232}Th or ^{228}Ra/^{226}Ra ratio in many natural systems. This ratio may vary from the original value if the sample was subjected to physical or chemical interaction that may affect one series more or less than the other one. The mean value of calculated ^{228}Ra/^{226}Ra ratio was 1.83 (1.42–2.33) (Table 2). This indicated that the activity concentration of ^{228}Ra is approximately doubled the value of ^{226}Ra. Smaller ratio range suggesting interaction on these two series with the surrounding were very similar.

Normally, nearby terrestrial mainland was the main contributor of NORM to the marine sediment. Therefore, calculating the external hazard index (H_{ex}) will enable to have a rough idea to evaluate the radiological hazard of natural gamma-radiation on mainland as well as the hazard to the workers whose deal and surrounded by the sediments

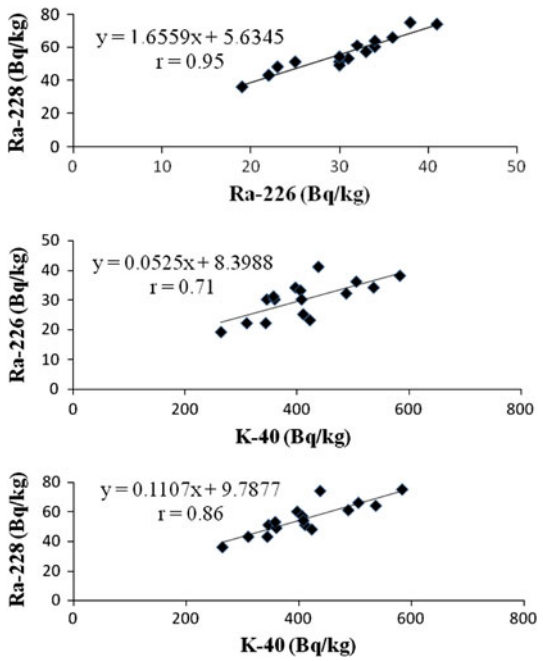


Fig. 4 Correlation between ^{226}Ra, ^{228}Ra and ^{40}K (mean value activity concentration) in marine sediment core

daily. The calculation of H_{ex} was performed using equation as reported by such reporter [1, 45]. The calculated external hazards values are found to be between values 0.25–0.51 with the mean of 0.38 and this is less than unity (Table 2). This indicated that there was only little risk of external hazard and may not harm the workers who surrounded and deal with the sediments. Also, there is high possibility that the mainland natural gamma-radiation on east coast Peninsular Malaysia was also low. The result found that H_{ex} value from this study was higher compared to the value reported in the East Malaysia marine sediment of 0.17–0.33 [17]. This is reasonable and align with the background radiation on terrestrial in East Malaysia (Sabah and Sarawak) which was lower compared to the Peninsular Malaysia mainland [17].

Conclusion

NORM activity concentrations in sixteen marine sediment cores in the east coast Peninsular Malaysia were measured. Generally, the activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K were ranged between 16–46 Bq/kg (mean 30 ± 6 Bq/kg), 28–87 Bq/kg (mean 56 ± 11 Bq/kg) and 171–690 Bq/kg (mean 420 ± 90 Bq/kg), respectively. The activity concentrations of radionuclides in most of the core were quite uniform suggesting that there were thorough vertical mixing of sediment throughout the core. Data reported in this paper was in good agreement with those reported by other countries in the region as well as the previous study in East Malaysia and therefore can be used to enhance the present database. The calculated external hazard values were between 0.25 and 0.51 with the mean of 0.38 (less than unity) showing little risk of external hazard to the workers handling the sediments and it is likely that the mainland natural gamma-radiation in the East Coast of Peninsular Malaysia was also low as it was the main contributor for NORM in that area.

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