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RADIATION HAZARD FROM NATURAL RADIOACTIVITY IN THE SEDIMENT OF THE EAST COAST PENINSULAR MALAYSIA EXCLUSIVE ECONOMIC ZONE (EEZ)

(Hazard Sinaran Dari Radionuklid Tabii Dalam Sedimen Di Zon Ekslusif Ekonomi (ZEE) Perairan Pantai Timur Semenanjung Malaysia)

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Abstract

Sixteen marine sediment cores from selected locations within the EEZ were collected for determination of NORM concentrations. The samples were dried, finely ground, sealed in a container and stored for more than 30 days to establish secular equilibrium between ^{226}Ra and ^{228}Ra and their respective radioactive progenies. They were counted and quantified using high-purity germanium (HPGe) detector coupled to spectrometer at respective progeny energy peak. Three calculated parameters from NORM concentrations, i.e. the Radium equivalent (Ra_{eq}), Representative level index ($I_{\gamma r}$), External hazard index (H_{ex}) are ranged between 68.6 – 210.5 Bq/kg (mean 143.1 ± 27.7 Bq/kg), 0.50 – 1.54 (mean 1.04 ± 0.20) and 0.19 – 0.57 (mean 0.39 ± 0.07 Bq/kg), respectively. This is well below the recommended limit value of 370 Bq/kg (for Ra_{eq}) and unity (for H_{ex}). It is also slightly less than the background level radiation from soil in Peninsular Malaysia, $I_{\gamma r}$ ~ 1.5. Therefore, the additional radiation exposure to peoples handling the samples is small, when compared to the background radiation received by them. The data is discussed and compared with those given in the literature.

Keywords: EEZ, sediment, radium equivalent activity, representative level index, external hazard index.

Abstrak

Enam belas turus sedimen dari lokasi terpilih dalam ZEE telah diambil untuk pengukuran kepekatan NORM. Sampel tersebut dikeringkan, dikisar halus, dilakri dalam bekas dan disimpan untuk tempoh melebihi 30 hari bagi mencapai keseimbangan sekular antara ^{226}Ra dan ^{228}Ra bersama progeni radioaktif mereka. Mereka diukur dan dikuantitikan pada puncak tenaga progeni masing-masing dengan menggunakan pengesan germanium lampau tulin yang disambung kepada spektrometer. Tiga parameter kiraan dari kepekatan NORM iaitu, Radium setaraan (Ra_{eq}), Indeks tahap perwakilan ($I_{\gamma r}$), Indeks hazard luaran (H_{ex}) adalah masing-masing dalam julat 68.6 – 210.5 Bq/kg (purata 143.1 ± 27.7 Bq/kg), 0.50 – 1.54 (purata 1.04 ± 0.20) dan 0.19 – 0.57 (purata 0.39 ± 0.07 Bq/kg). Nilai tersebut adalah di bawah had yang dicadangkan iaitu 370 Bq/kg (untuk Ra_{eq}) dan uniti (bagi H_{ex}). Ia juga kurang berbanding tahap sinaran latar belakang dari tanah di Semenanjung Malaysia, $I_{\gamma r}$ ~ 1.5. Oleh itu, pendedahan sinaran tambahan kepada orang yang mengendalikan sampel adalah kecil jika dibandingkan dengan sinaran latar belakang yang diterima oleh mereka. Data keputusan akan dibincang dan dibanding dengan nilai dari rujukan.

Kata kunci: ZEE, sedimen, radium setaraan, indeks tahap perwakilan, indeks hazard luaran.

Introduction

Two prominent sources of external radiation are cosmic rays and terrestrial gamma rays. Cosmic rays are mostly been shielded off by the ozone and contribute less radiation hazard to human beings. Meanwhile, the primary source of terrestrial radiation received by humans is from the store of natural radioactivity in earth crust, which derives essentially from ^{40}K and the progenies of ^{238}U and ^{232}Th decay series such as radium, radon, actinium, protactinium, lead and polonium. These progenies first appear in the lithosphere level, deposited on the soil surface, then washed and drained through rivers transport and finally end up in the estuary and entering the marine environment. It also has gone through several pathways such as weathering, erosion, fallout, rainwater and human activities [1, 2, 3, 4].

When the marine sediments were excavated for study, the researchers whose working environment was surrounded by these sediment samples daily are exposed to additional radiation from these natural radioactivities. Therefore, the radiation hazards to these people are of interests in order to ensure that no additional doses are imposed on to them. 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 theirs high solubility and mobility as well as emitting gamma energies. In most literatures, radiation hazards were calculated from the specific activity of these three radionuclides [5, 6, 7, 8].

Koide *et al.* (1973) [9] specified that the radioactivity of the progenies from the natural decay series in the marine environment may not necessary be in equilibrium with it parents. Two main sources of marine radioactivities are coming from the weathering and mineral recycling of the terrestrial rocks. In other word, sediment in the coastal is mainly contributed from materials on terrestrial. Baseline information on the spatial variation of radionuclide concentration in the environment is necessary to evaluate any change induced by humans in the future [10]. In addition, these data are required to trace the movement of radionuclides from original source into environmental and biological systems [11].

In coastal areas and on the continental shelf, it 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 concentrations of ^{226}Ra, ^{228}Ra and ^{40}K on terrestrial especially in the areas of Peninsular Malaysia had been widely reported [12, 13, 14, 15, 16]. Several studies on NORM and their progenies had also been reported on the Malaysian marine ecosystems [17, 18] but none specify on the radiation hazard.

The main scope of this work is to determine the additional radiation hazards received by the researchers while carry out they daily routine. This enable a dosimetric evaluation for hazard to these people were estimated. Additionally, these data will indirectly give a brief idea of the radiation levels in the surrounding area on terrestrial at East Coast Peninsular Malaysia.

Experimental

Sampling, preparation and measurement

The east coast of Peninsular Malaysia Exclusive Economic Zone, 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. A total of thirty locations had been identified to be sampling site. Location and sampling dates are as given in Table 1 while Fig. 1 marks the location of each sampling points. The samplings were done systematically according to a grid within Malaysian EEZ which covers 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).

The core sediments were collected using a 12 cm diameter multicorer device and sliced into 2 cm sections at site and sealed in a pre-weighted HDPE container. During sampling expedition, some of the cores showed thorough vertical mixing of fine and sandy mud, however, some cores are too short (< 20 cm) and while others are too sandy (as illustrated in Fig. 1) and all these cores are not processed for analysis. Only good sediment cores (sixteen in total) with enough length and low sand content (marked • close circle as in Fig.1) were processed and analyzed.

Inside the laboratory, all these selected core samples were dried at 60^{o}C for minimum 72 hours until they reached a constant mass, then ground to pass through a 200 mesh sieve for radiochemical analysis. Samples were transferred into 200 mL marinelli beaker, sealed with thick PVC tape to inhibit radon from escaping. All samples were stored for a period in excessive of 30 days [5, 19] to establish secular equilibrium between ^{226}Ra and ^{228}Ra and their respective radioactive progeny prior to gamma counting.

Table 1: Surrounding physical conditions and coordinates of sampling locations

Station	Date	Latitude	Longitude	Water depth (m)	Surface Seawater temp.(^oC)	Distance from shore (nautical miles)
SF01	18.06.08	06^o 13.99' N	102^o 19.00' E	13.0	30.87	2.7
SF02	17.06.08	06^o 50.04' N	102^o 47.04' E	46.5	30.51	50
SF03	17.06.08	07^o 05.03' N	103^o 04.99' E	50.0	30.32	73
SF04	17.06.08	07^o 25.98' N	103^o 26.01' E	61.0	30.03	100
SF05	16.06.08	06^o 56.09' N	103^o 56.04' E	52.0	30.01	108
SF06	16.06.08	06^o 42.14' N	103^o 35.17' E	52.0	30.13	80
SF07	16.06.08	06^o 10.00' N	103^o 01.00' E	45.0	30.25	40
SF08	18.06.08	05^o 52.10' N	102^o 51.92' E	34.0	30.41	15
SF09	20.06.08	05^o 22.06' N	102^o 21.97' E	47.0	28.98	14
SF10	14.06.08	05^o 48.20' N	103^o 48.98' E	55.0	30.27	48
SF11	14.06.08	06^o 06.16' N	104^o 09.11' E	72.0	30.04	75
SF12	14.06.08	06^o 32.01' N	104^o 22.11' E	59.0	29.58	101
SF13	13.06.08	06^o 16.98' N	105^o 16.99' E	55.0	29.87	139
SF14	13.06.08	05^o 57.15' N	104^o 58.13' E	56.0	29.63	115
SF15	12.06.08	05^o 29.08' N	104^o 29.02' E	60.7	29.62	80
SF16	12.06.08	05^o 18.50' N	104^o 12.60' E	60.0	29.65	56
SF17	20.06.08	04^o 54.12' N	103^o 42.98' E	54.0	29.93	17
SF18	11.06.08	04^o 28.14' N	103^o 49.98' E	40.0	29.79	20
SF19	22.06.08	03^o 37.07' N	103^o 41.08' E	23.0	29.50	15
SF20	22.06.08	03^o 55.10' N	104^o 00.05' E	50.0	29.74	40
SF21	23.06.08	04^o 22.16' N	104^o 22.07' E	65.0	29.63	52
SF22	23.06.08	04^o 44.19' N	104^o 38.44' E	66.0	29.63	70
SF23	12.06.08	05^o 08.10' N	105^o 12.90' E	67.2	29.89	109
SF24	23.06.08	03^o 32.08' N	104^o 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

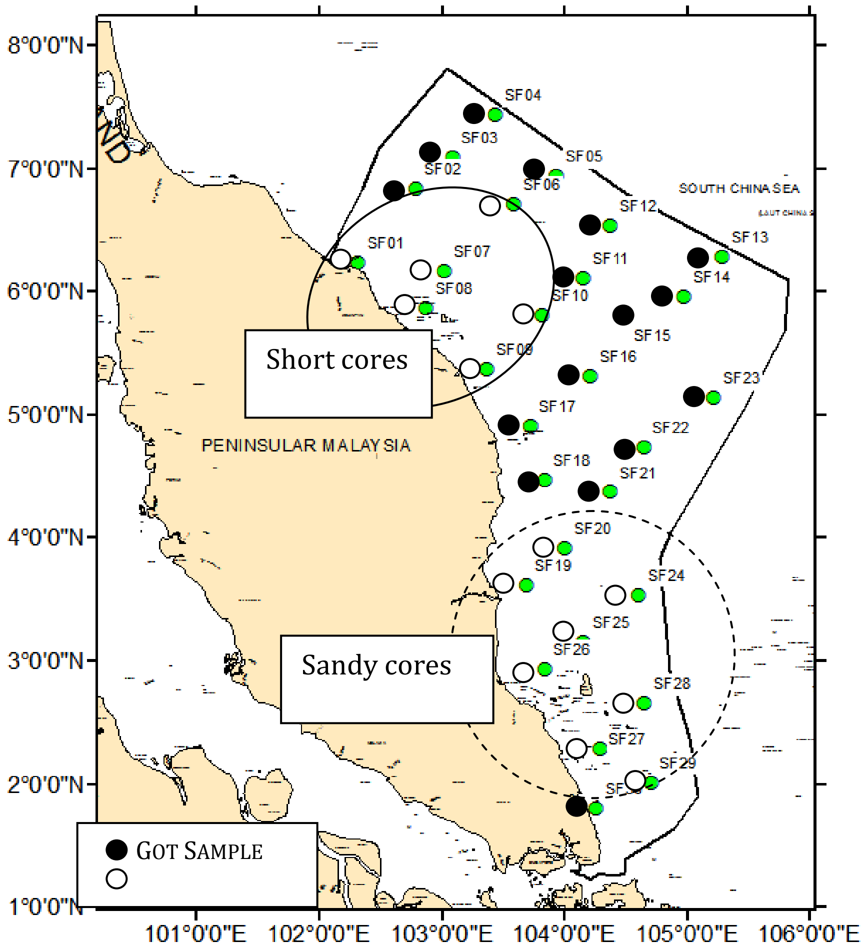

Fig. 1: Sampling locations in east coast of Peninsular Malaysia EEZ

All samples were counted for 86,400 seconds using HPGe spectrometer and corrected to the date of sampling. Counting times were long enough to ensure a 2σ counting error of less than 10%. The energy peaks used are the same as reported by Yii *et al.* 2009 [20].

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 procedures as reported earlier by Yii *et al.* 2009 [20].

The radioactivity concentrations in the environmental samples were calculated using equation as reported by Yang *et al.* (2005) [5] and Chen *et al.* (2005) [21]. 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 the counting time of the sample.

Results and Discussion

Surrounding physical conditions at sampling locations were summarized in Table 1. All the values of samples are found to be greater than the minimum detectable activity (MDA). From the measurement, it found that the concentrations of naturally occurring radionuclides vary significantly from place to place as their presence in the marine environment depends on their physical, chemical and geochemical properties and the pertinent environment in the biological process [22]. In total, from all sediment cores, the concentration of ^{226}Ra is ranged between 16 – 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 activity concentrations of radionuclides in most cores are quite uniform suggesting that there is thorough vertical mixing of sediment throughout the core. However, the activity concentrations in some cores appear to be higher when compared to other stations. The large area of sampling, i.e. EEZ could be the reason why the concentrations are not uniformly distributed.

Calculation of radiological effects

The most widely used radiation hazard index is called the radium equivalent activity, Ra_{eq}. The radium equivalent activity is a weighted sum of activities of the ^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) and ^{40}K radionuclides based on the assumption that ^{226}Ra, 259 Bq/kg of ^{228}Ra and 4810 Bq/kg of ^{40}K produce the same gamma ray dose rate [5, 8, 23]. Radium equivalent activity can be calculated from the following relation:

$$Ra_{eq} = A_{Ra} + 1.43\, A_{Th} + 0.077 A_K \qquad (1)\ [24]$$

where A_{Ra}, A_{Th}, A_K are the activity concentration of ^{226}Ra, $^{232}Th(^{228}Ra)$ and ^{40}K, respectively. To be non-hazardous, the Ra_{eq} should not exceed a maximum of 370 Bq/kg [25].

Another radiation hazard index called the representative level index, $I_{\gamma r}$, is defined from the following formula [26].

$$I_{\gamma r} = \frac{1}{150\, Bq/kg} A_{Ra} + \frac{1}{100\, Bq/kg} A_{Th} + \frac{1}{1500\, Bq/kg} A_K \qquad (2)$$

where A_{Ra}, A_{Th}, A_K having the same meaning as in Eq. (1).

External hazard index (H_{ex}) is another assumption that enables to evaluate the additional radiological hazard of natural gamma-radiation to the workers whose deal and surrounded by the sediments daily. The calculation was performed using equation as reported by Yang *et al*. (2005) [5] and Nabil *et al*. (2010) [6].

$$H_{ex} = \frac{Q_U}{370} + \frac{Q_{Th}}{259} + \frac{Q_K}{4810} \le 1 \qquad (3)$$

where Q_U, Q_{Th}, and Q_K are the activity concentrations of $^{238}U(^{226}Ra)$, $^{232}Th(^{228}Ra)$ and ^{40}K, respectively.

The Radium equivalent activity (Ra_{eq}), Representative level index ($I_{\gamma r}$), External hazard index (H_{ex}) for each individual 2 cm layer section sediment of the core had been calculated from the NORM activity and shown as in Figs. 2 – 4 and summarized in Table 2.

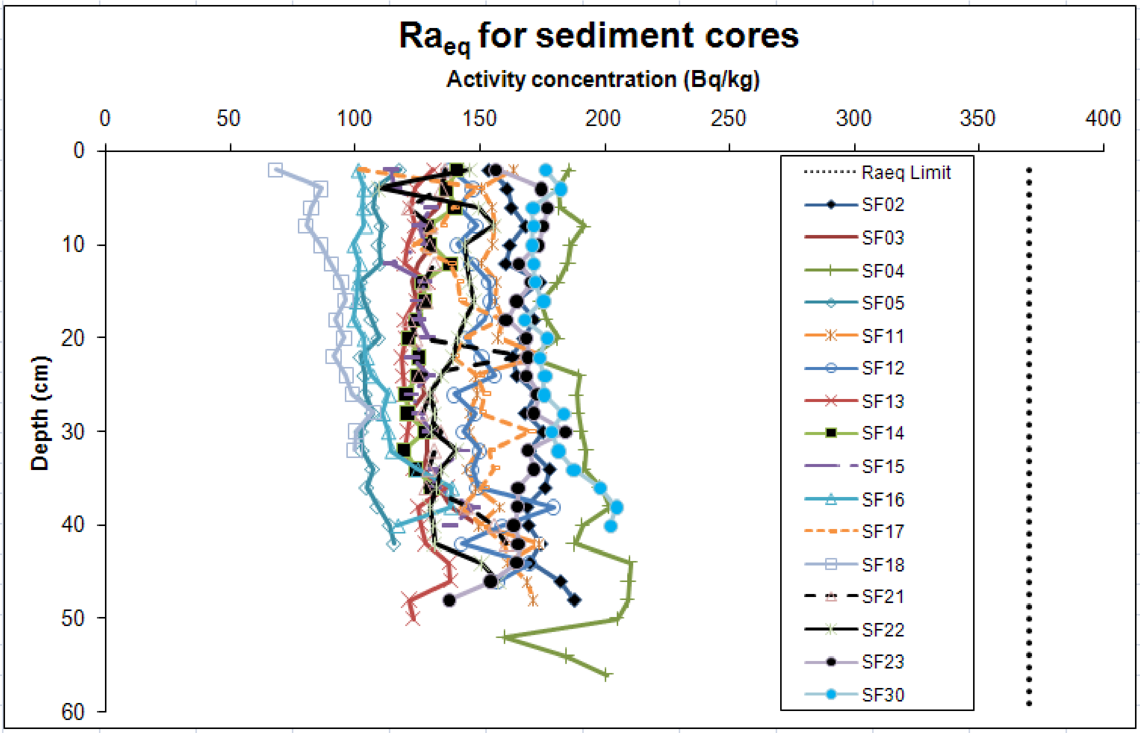

Fig. 2**:** Radium Equivalent in core sediment from east coast of Peninsular Malaysia EEZ

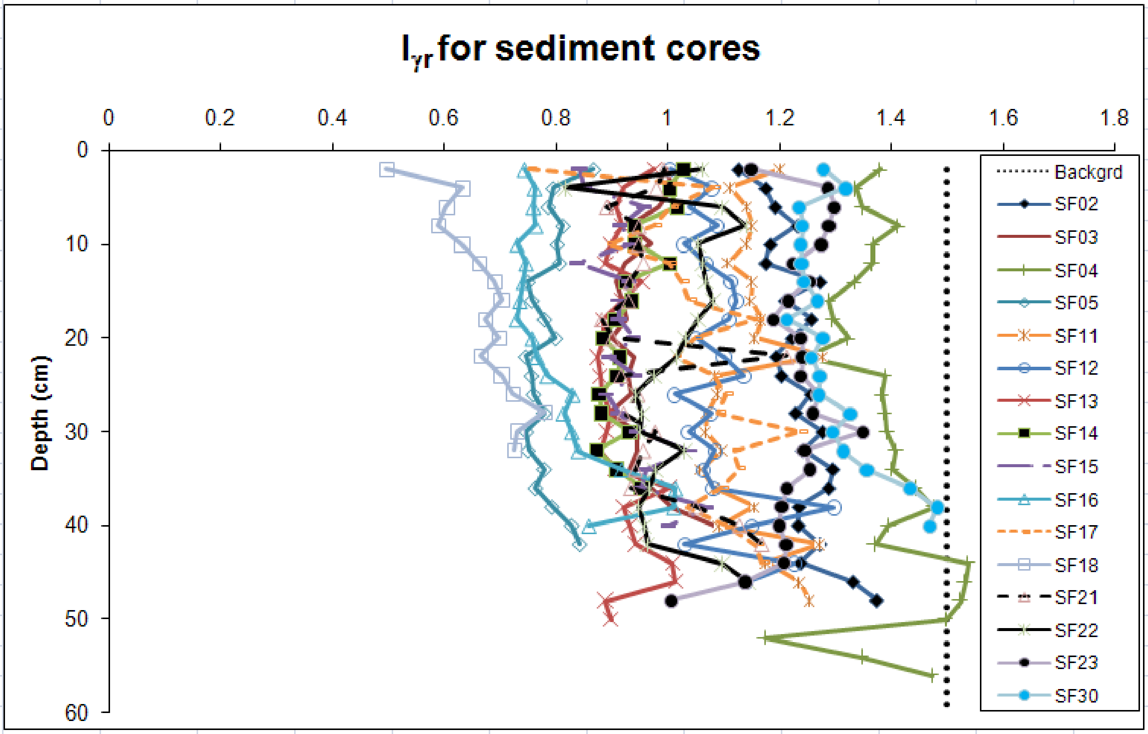

Fig. 3: Representative Level Index in core sediment from east coast of Peninsular Malaysia EEZ

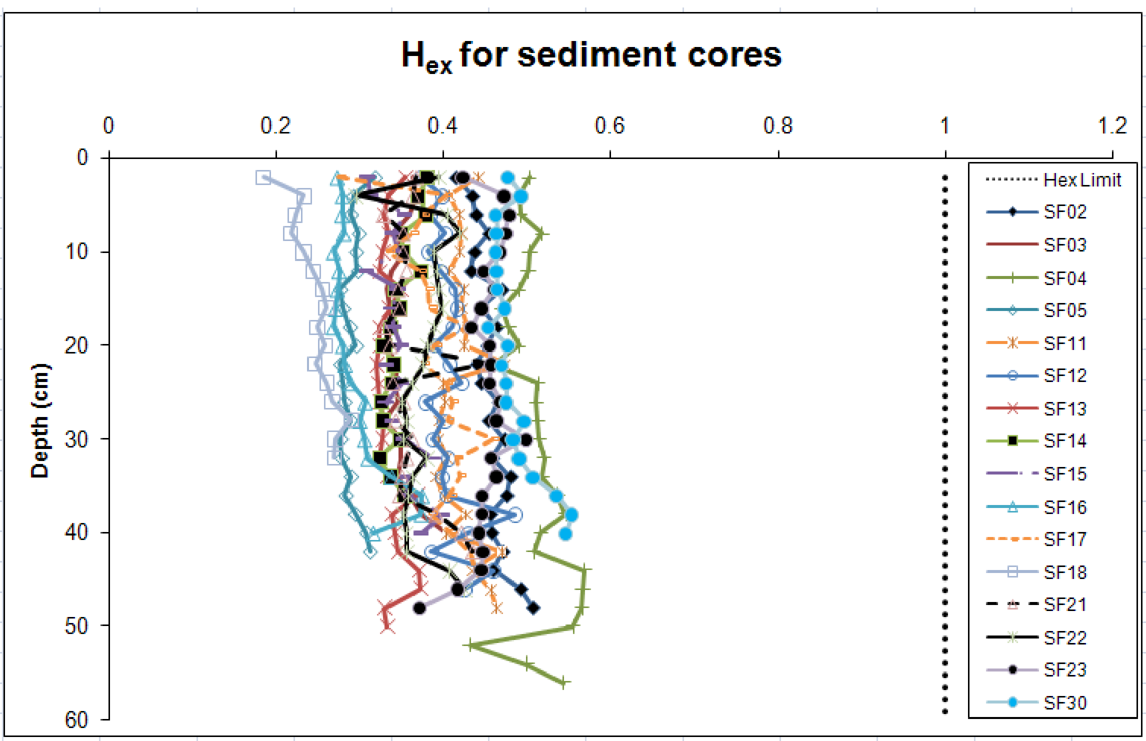

Fig. 4: External Hazard Index in core sediment from east coast of Peninsular Malaysia EEZ

Table 2: Ra_{eq}, $I_{\gamma r}$, H_{ex} in the east coast Peninsular Malaysia EEZ sediment cores

Station	Ra_{eq}, (Bq/kg)		$I_{\gamma r}$		H_{ex}	
	Range	Mean	Range	Mean	Range	Mean
SF 02	153.6 – 187.8	169.4 ± 7.6	1.13 – 1.37	1.24 ± 0.05	0.41 – 0.51	0.46 ± 0.02
SF 03	121.9 – 149.4	129.7 ± 6.6	0.89 – 1.08	0.95 ± 0.05	0.33 – 0.40	0.35 ± 0.02
SF 04	160.0 – 210.5	189.4 ± 11.9	1.17 – 1.54	1.39 ± 0.08	0.43 – 0.57	0.51 ± 0.03
SF 05	102.5 – 118.0	107.7 ± 4.4	0.75 – 0.87	0.79 ± 0.03	0.28 – 0.32	0.29 ± 0.01
SF 11	144.9 – 175.0	156.3 ± 8.9	1.06 – 1.28	1.15 ± 0.06	0.39 – 0.47	0.42 ± 0.02
SF 12	138.0 – 179.7	150.5 ± 9.6	1.00 – 1.30	1.09 ± 0.07	0.37 – 0.49	0.41 ± 0.03
SF 13	118.6 – 138.1	124.8 ± 5.8	0.87 – 1.01	0.92 ± 0.04	0.32 – 0.37	0.34 ± 0.02
SF 14	120.0 – 140.9	128.7 ± 6.6	0.87 – 1.03	0.93 ± 0.05	0.32 – 0.38	0.35 ± 0.02
SF 15	114.3 – 147.7	128.1 ± 8.5	0.84 – 1.07	0.93 ± 0.06	0.31 – 0.40	0.35 ± 0.02
SF 16	99.6 – 138.6	110.1 ± 11.8	0.73 – 1.01	0.81 ± 0.09	0.27 – 0.37	0.30 ± 0.03
SF 17	101.5 – 169.8	145.6 ± 14.3	0.75 – 1.24	1.06 ± 0.10	0.27 – 0.46	0.39 ± 0.04
SF 18	68.6 – 107.2	91.8 ± 9.3	0.50 – 0.78	0.67 ± 0.07	0.19 – 0.29	0.25 ± 0.03
SF 21	121.9 – 167.4	134.6 ± 12.4	0.88 – 1.21	0.98 ± 0.09	0.33 – 0.45	0.36 ± 0.03
SF 22	109.4 – 157.6	139.3 ± 10.6	0.82 – 1.14	1.02 ± 0.08	0.30 – 0.43	0.38 ± 0.03
SF 23	137.9 – 184.4	167.2 ± 9.0	1.01 – 1.35	1.23 ± 0.07	0.37 – 0.50	0.45 ± 0.02
SF 30	167.9 – 204.8	180.0 ± 10.6	1.21 – 1.48	1.30 ± 0.08	0.45 – 0.55	0.49 ± 0.03
*Overall	68.6 – 210.5	143.1 ± 27.7	0.50 – 1.54	1.04 ± 0.20	0.19 – 0.57	0.39 ± 0.07

*Overall value given was the range and average mean for all reported sediment cores

From the data, it is obvious that for each sediment section of the cores, the calculated value for that of radium equivalent activity and external hazard index are well below the recommended value, showing that the sediment samples does not necessary adding extra radiation hazards to the workers even though they are surrounded by these samples daily. Meanwhile, calculated representative level index ($I_{\gamma r}$ ~ 1.0) was also less than the background level ($I_{\gamma r}$ ~ 1.5) in Peninsular Malaysia's soil. In all cores, only layer 42 – 44 cm of the SF 04 core sediment having values slightly more than that of the background level.

From the pictorials in Figs. 2 – 4, we can conclude that the hazard index is low especially when dealing with sediments from stations SF 05, SF 16 and SF 18. Whilst, stations SF 04 and SF 30 are contributing more radiation hazards compared to the sediments from other locations.

The results for the radium equivalent activity, representative level index, and external hazard index of the present work and other studies are compared in Table 3. It is found that , in general, the value of Ra_{eq}, $I_{\gamma r}$ and H_{ex} calculated for the sediments in this study is comparable to those reported elsewhere and less than recommended figures, indicating that the hazard of radiation exposure for the workers whose handle the samples are less when compared to the Peninsular Malaysia's background radiation received by them.

Table 3**:** Comparison of Ra_{eq}, $I_{\gamma r}$, H_{ex} of the present work and other studies.

Country	Sample	Ra_{eq} (Bq/kg)	$I_{\gamma r}$	H_{ex}	References
Egypt	Beach sand	182.0	1.3[a]	0.5[a]	Ref. [27]
Egypt	Nile island's soil	152.9	1.3	0.4[a]	Ref. [8]
Brazil	Soil	147.8[a]	1.1[a]	0.4[a]	Ref. [28]
India	Soil	86.7[a]	0.6[a]	0.2[a]	Ref. [7]
Thailand	Soil	204.6[a]	1.4[a]	0.6[a]	Ref. [12]
Malaysia (Peninsula)	Soil	208.1[a]	1.5[a]	0.6[a]	Ref. [12]
World average	Soil	118.5[a]	0.9[a]	0.3[a]	Ref. [29]
World average	Soil	129.7[a]	1.0[a]	0.4[a]	Ref. [12]
Red Sea	Sediment	32.4[a]	0.2[a]	0.1[a]	Ref. [22]
South China Sea (Sarawak)	Sediment	101.6[a]	0.7[a]	0.3	Ref. [20]
South China & Sulu Sea (Sabah)	Sediment	64.7[a]	0.5[a]	0.2	Ref. [20]
Peninsular EEZ	Sediment	143.1	1.0	0.4	Present work

[a]Calculated by the author using data given in the reference.

Conclusion

Core sediments from stations SF 04 and SF 30 are contributing more radiation hazards whiles core sediments from stations SF 05, SF 16 and SF 18 are contributing less radiation hazards than most of the average cores. Overall, the Radium equivalent activity (Ra_{eq}), Representative level index ($I_{\gamma r}$), External hazard index (H_{ex}) is ranged between 68.6 – 210.5 Bq/kg (mean 143.1 ± 27.7 Bq/kg), 0.50 – 1.54 (mean 1.04 ± 0.20) and 0.19 – 0.57 (mean 0.39 ± 0.07 Bq/kg), respectively. This value is less than the recommended limit value of 370 Bq/kg (for Ra_{eq}) and unity (for H_{ex}). It is also slightly less than the background level radiation from soil in Peninsular Malaysia, $I_{\gamma r}$ ~ 1.5. Therefore, the additional radiation exposure to the workers while handling the samples is small, as compared to the background radiation received by them.

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