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POTENTIAL RADIOLOGICAL SIGNIFICANCE OF NORM IN MARINE SEDIMENTS FROM THE EAST COAST OF PENINSULAR MALAYSIA

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ABSTRACT

The NORM radioactivities of ²³⁸U, ²³²Th and ⁴⁰K in the marine surface sediments collected from 20 sampling points in the east coast of Peninsular Malaysia using a Ponar grab sampler and measured using HPGe gamma-ray spectrometer was studied. The objective of this work is to evaluate the radiological health hazard effect of natural radioactivity associated with sediments at study area. Generally, the mean radium equivalent activity concentration index and other radiological hazard parameter were found to be lower than their maximum permissible limits. The conclusion is made that the sediments in the region confirmed were not posed radiological risks to the public and; sediments are suitable and safe to use in any activities. Furthermore, the results of this study could serve as an important radiometric baseline data upon which future epidemiological studies and environmental monitoring initiatives could be based.

Keywords: Dose, marine sediment, Peninsular Malaysia, radiological hazard effect, radionuclides

INTRODUCTION

The radioactivity level of the natural radionuclides such as ²³⁸U, ²³²Th and ⁴⁰K are termed as background radiation, which will depend on the amount of the radioactive materials in the environment. The background radiation can be high if the environment is polluted either from man-made or natural activities (Chandrasekaran et al., 2014). These radionuclides are widely distributed in the environment as a result of their natural occurrence in the earth's environment or the atmosphere (Botwe et al., 2017). These radionuclides exist with the high radiation levels are due to the presence of large quantities in various geological formations like soils, rocks, plants, sand, sediment, water and air (Singh et al., 2007; SureshGandhi et al., 2014). Although, these radionuclides and their decay products present at trace levels in all ground formations (Tzortzis et al., 2004), however, they constitute an important source of ionising radiation exposure to human and non-human populations (Kam and Bozkurt, 2007; UNSCEAR, 2000).

Since natural radioactivity of these radionuclides has 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 ²³⁸U, ²³²Th and their progenies and ⁴⁰K present in the sediment, which in turns depends upon the geology of the regions (Malik, 2014; Quindos et al., 1994; Radhakrishna et al., 1993).

The ⁴⁰K activity concentration on average by taking into account the value of the halflives of ²³⁸U (uranium-238; $t_{1/2} = 4.5 \times 10^9$ years), ²³²Th (thorium-232; $t_{1/2} = 1.4 \times 10^{10}$ years), and ⁴⁰K (K-40; $t_{1/2} = 1.3 \times 10^9$ years) is about one order of magnitude higher than that of ²³⁸U and ²³²Th (Kovler et al., 2017). Due to such characteristic, thus monitoring the releases main sources of gamma radiation are important to protect the humans (Chandrasekaran et al., 2014) because these radionuclides may cause harmful biological effects such as DNA damage and cancer (Chandrasekaran et al., 2014; Little, 2003; Ravanat et al., 2014; Schmid and Schrader, 2007).

In the recent years, studies on the high background radiation areas in the world have been of prime importance for risk estimation due to long term low-level whole body exposures to the public. Furthermore, 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). Thus, the potential radiological health hazard effects of ²³⁸U, ²³²Th and ⁴⁰K associated with surface sediments collected at near shore on the east coast of Peninsular Malaysia were studied. 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 aim of the study is 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 in 4.439 - 6.089 °N and longitude of 102.542 - 103.610 °E and all points are situated about 9 - 20 km from mainland and water depth ranged from 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 natural 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 and preparation

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, the samples were dried at 60° C in an electric

oven until a constant weight for 2-5 days. All the dried samples were ground into powder form to homogeneity and then, the samples were kept in an airtight container. 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 ²³⁸U and ²³²Th and their respective radioactive progenies prior to gamma counting.

Sample counting

All samples were counted for 54000 seconds using High-Purity Germanium (HPGe) gammaray 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%. The activity concentration of ⁴⁰K was calculated through its single photopeak at gamma energy line of 1460.7 keV (IAEA, 1989a). Meanwhile, under the assumption that secular equilibrium was reached between ²³⁸U and ²³²Th 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) ²²⁶Ra (186.21 and 241.98 keV), ²¹⁴Pb (295.21, 351.92 keV) and ²¹⁴Bi (609.31 keV) for uranium-238 (IAEA, 1989b).
- (ii) 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).

Location	Station ID	Latitude, ⁰N	Longitude, °E	Sampling Date	Water Depth (m)
Melawi	ML01	6.086	102.574	23 Sept. 2017	22.1
Tok Bali	TB 01	5.947	102.542	23 March 2018	15.5
	TB03	5.968	102.594	04 May 2018	23.4
Pulau	PP02	5.993	102.754	04 May 2018	35.4
Perhentian	PP04	5.944	102.631	04 May 2018	25.7
Kuala	KT01	5.558	103.148	04 July 2018	30.7
Terengganu	KT02	5.541	103.220	04 July 2018	42.9
	KT03	5.476	103.298	04 July 2018	46.6
	KT04	5.403	103.340	04 July 2018	44.4
Marang	MG01	5.293	103.378	05 July 2018	42.1
	MG02	5.224	103.408	05 July 2018	44.7
	MG03	5.143	103.445	05 July 2018	41.8
	MG04	5.068	103.480	05 July 2018	41.4
Redang -	RB01	5.690	103.110	09 Aug. 2018	42.1
Bidong	RB03	5.747	102.941	09 Aug. 2018	34.8
Dungun	DG02	4.906	103.554	03 Oct. 2018	44.2
	DG03	4.796	103.610	03 Oct. 2018	40.5
Paka - Kerteh	PK01	4.611	103.626	04 Oct. 2018	39.6
	PK02	4.525	103.655	04 Oct. 2018	39.0
	PK03	4.439	103.632	04 Oct. 2018	37.8

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



Figure 1: Map shows the points for sediment sampling

The HPGe detector was characterized to provide 25% relative efficiency and 1.8 keV at FWHM for the 1332 keV gamma ray line of ⁶⁰Co. It was calibrated using a customized gamma multinuclides standard solution which comprise of ²¹⁰Pb, ²⁴¹Am, ¹⁰⁹Cd, ⁵⁷Co, ^{123m} Te, ⁵¹Cr, ¹¹³Sn, ⁸⁵Sr, ¹³⁷Cs, ⁸⁸Y and ⁶⁰Co in the same geometry with the samples. The 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 of 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 ²³⁸U, ²³²Th ²²⁶Ra and ⁴⁰Kwere quantified at 1.0, 1.0 and 5.0 Bq/kg dry weight (dw.), respectively.

RESULTS AND DISCUSSION

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 ²³⁸U (or²²⁶Ra), ²³²Th and ⁴⁰K (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 these radionuclides in sediment 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 of all materials (Berekta and Matthew, 1985; Sivakumar et al., 2014). It was calculated according to equation below:

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

Where, A_U , A_{Th} and A_K are the activity concentrations (Bq/kg) for ²³⁸U, ²³²Th and ⁴⁰K, respectively. It has been assumed here that 370 Bq/kg of ²³⁸U or ²⁵⁹ Bq/kg of ²³²Th or 4810 Bq/kg of ⁴⁰K produces the same radiation dose rate.

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. (Table 2). 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 was 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 in any activities 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 238 U (or 226 Ra), 232 Th and 40 K in the sediment (Erenturk et al., 2014). The absorbed dose rates due to radiation of the uniform distribution of these 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):

$$\mathbf{D}_{adr} \left(nGy/h \right) = 0.462 \mathbf{A}_{U} + 0.604 \mathbf{A}_{Th} + 0.042 \mathbf{A}_{K}$$
(2)

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

The absorbed dose rate (D_{adr}) values ranged from 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 dose in the studied locations. The results for the relationship of the radionuclides contributed to the dose in the sediments are shown in Table 3. The positive correlation was observed between ²³⁸U and ²³²Th 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 ²³²Th as a key natural radionuclide played an important role to contribute the emission of radiation in all the studied locations compared to ²³⁸U.

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):

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

The calculated D_{aedr} values were ranged from 0.01 - 0.08 mSv/y with a mean value of 0.05 mSv/y (Table 2), 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 varies 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 ²³²Th. This finding is strictly supported by Pearson's correlation coefficient as summarized in Table 3. Furthermore, the finding also indicated that relatively higher concentrations of ²³²Th (mean value: 37.90 Bq/kg dw.) exist than ²³⁸U (mean value: 18.20 Bq/kg dw.) in the sediment. This is could be due to the presence of the input detrital mineral particles associated with sediments at studied locations which have a relatively high concentration of ²³²Th.

iii) External hazard index

In order to assess the health effects of the radioactivity of the earth's surface materials containing 238 U, 232 Th and 40 K to the people at surrounding marine environment, a single quantity termed as an external hazard index (H_{ex}) should be quantified (SureshGandhi 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:

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

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

Location S	Station	Activity Concentration (Bq/kg dw.)			Radium Equivalent	Absorbed Dose	Annual Effective Dese	External
	ш –	²³⁸ U	²³² Th	$^{40}\mathrm{K}$	Concentration Index, Ra _{eq} (Bq/kg dw.)	(nGy/h)	Rate, D _{aedr} (mSv/y)	Index, H _{ex}
Melawi	ML01	31.68 ± 3.57	52.59 ± 6.94	162.85 ± 11.17	118.28	53.24	0.07	0.32
Tok Bali	TB 01	32.58 ± 3.70	63.29 ± 8.24	218.25 ± 14.52	138.36	62.45	0.08	0.38
	TB03	21.19 ± 2.98	28.71 ± 3.77	99.18 ± 6.85	69.19	31.30	0.04	0.19
Pulau Perhentian	PP02	18.60 ± 3.75	38.12 ± 5.03	272.08 ± 18.49	92.16	43.05	0.05	0.25
	PP04	32.50 ± 2.66	33.93 ± 4.48	110.59 ± 7.67	88.76	40.15	0.05	0.24
Kuala	KT01	24.32 ± 3.24	31.28 ± 4.31	326.76 ± 6.51	91.92	43.85	0.05	0.25
Terengganu	KT02	17.73 ± 0.48	37.26 ± 4.92	321.97 ± 6.57	93.55	44.22	0.05	0.26
	KT03	16.30 ± 0.52	33.38 ± 4.42	324.53 ± 6.23	86.75	41.32	0.05	0.24
	KT04	9.76 ± 2.05	18.51 ± 2.44	255.30 ± 5.11	54.10	26.41	0.03	0.15
Marang	MG01	3.81 ± 0.29	12.51 ± 1.65	21.10 ± 1.72	23.18	10.20	0.01	0.06
	MG02	16.10 ± 1.19	42.84 ± 5.65	323.15 ± 8.41	99.98	46.89	0.06	0.28
	MG03	11.94 ± 1.39	32.31 ± 4.62	265.00 ± 20.91	76.69	36.16	0.04	0.21
	MG04	12.90 ± 1.72	18.80 ± 2.80	91.52 ± 7.34	46.19	21.16	0.03	0.13
Redang-Bidong	RB01	18.59 ± 3.90	35.25 ± 4.58	425.11 ± 8.33	97.00	46.92	0.06	0.27
	RB03	16.83 ± 3.72	31.91 ± 4.15	322.24 ± 6.86	72.00	34.57	0.04	0.20
Dungun	DG02	14.28 ± 0.39	41.45 ± 5.74	228.22 ± 38.70	89.53	41.22	0.05	0.25
	DG03	10.19 ± 1.31	26.31 ± 3.47	194.32 ± 9.85	61.42	28.76	0.04	0.17
Paka-Kerteh	PK01	4.88 ± 0.36	27.98 ± 3.69	166.67 ± 7.01	56.56	26.15	0.03	0.16
	PK02	22.37 ± 2.87	33.96 ± 4.48	105.59 ± 6.32	78.32	35.28	0.04	0.21
	PK03	15.62 ± 0.45	26.97 ± 3.94	246.67 ± 4.98	71.45	33.87	0.04	0.20

Table 2: Radiological hazard parameters of natural radionuclides in surface sediment

The range calculated external hazard values 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 is 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 Pearson's correlation coefficient, r = 0.8285 (Figure 2) 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.

In the other word, the good strong 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, the 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 a., 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 the harmful effects of ionizing radiation from the natural radionuclides in sediment.

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

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

Cluster analysis of dendrogram

Cluster analysis is a method to classify similar observations into a number of clusters based on the observed value of several variables for each individual or average. Each individual within a cluster is same but dissimilar from each other (Sinharay, 2010). Thus, type of hierarchical cluster is a statistical technique for grouping same variable within cluster based on dissimilarities or distance between variable (Yim and Ramdeen, 2015). It can be generated via dendrogram to display the linkage within variable at increasing dissimilarity.

In this study, the averages for 7 parameters i.e. ²³⁸U, ²³²Th, ⁴⁰K, Ra_{eq}, D_{adr}, D_{aedr} and H_{ex}were classify into three major clusters. Cluster I consisted of ²³⁸U, ²³²Th, D_{adr}, D_{aedr} and H_{ex}; the cluster II set for ²³⁸U and Ra_{eq} and cluster III only for single member of ⁴⁰K. The most significant way to show dissimilarities each average of parameter are by compute square Euclidean distance and the derived dendrogram is shown in Figure 4. This showed the parameter of cluster I was closely distance of result, followed by cluster II. Meanwhile, ⁴⁰K was identified in cluster III which is far distance result from other parameter. Interpretation from this cluster analysis can be made that D_{adr}, D_{aedr} and H_{ex} are the main contributions hazard effects to the human beings are due to ²³⁸U and ²³²Th present in the study area, while Ra_{ex} due to clearly high concentration of ²³⁸U. Generally, ⁴⁰K found to be higher at study area, rather it was not significant in contributing radiation dose to study area. Therefore, in this current study found that ²³⁸U is a key radionuclide has a possibility contributed the radiation dose hazard effect to the study area.



Figure 4: Dendrogram shows the clustering of radionuclides, Ra_{eq} and radiological hazard index

CONCLUSION

The evaluation of radium equivalent activity (R_{aeq}) , absorbed dose rate (D_{adr}) , annual effective dose rate (D_{aedr}) and external hazard index (H_{ex}) were lower than their maximum

permissible limits. This indicates that the potential dose rates to human from the surface sediment radioactivity levels of ²³⁸U, ²³²Th and ⁴⁰K in the east coast of Peninsular Malaysia have strictly been not present significant risks to human health. Absolutely, it can be confirmed that the surrounding people, fisherman, divers or who-else presence in this area were not posed radiological risks owing to harmful effects of ionizing radiation from the natural radionuclides in sediment. Furthermore, these sediments are suitable and safe to use for any activity purposes.

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